

CNWRA *A center of excellence in earth sciences and engineering*

A Division of Southwest Research Institute
6220 Culebra Road • San Antonio, Texas, U.S.A. 78228-5166
(210) 522-5160 • Fax (210) 522-5155

May 30, 2000
Contract No. NRC-02-97-009
Account No. 20-1402-561

U.S. Nuclear Regulatory Commission
ATTN: Mrs. Deborah A. DeMarco
Two White Flint North
11545 Rockville Pike
Mail Stop T8A23
Washington, DC 20555

Subject: Programmatic Review of Abstract

Dear Mrs. DeMarco:

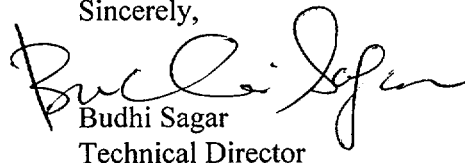
The enclosed abstract is being submitted for programmatic review. This abstract will be submitted for presentation at the Goldschmidt 2000—An International Conference for Geochemistry, to be held in September 3–8, 2000. The title of this abstract is:

“Estimating Radionuclide Release from a Uranium Deposit Through Uranium-Series Systematic in Carbonates and Opal” by David A. Pickett, Bret W. Leslie, William M. Murphy, and Melissa Nugent.

This abstract is a product of the CNWRA and does not necessarily reflect the view(s) or regulatory position of the NRC.

Please advise me of the results of your programmatic review. Your cooperation in this matter is appreciated.

Sincerely,


Budhi Sagar
Technical Director

BS: ar
Enclosure

cc: B. Meehan	S. Wastler
J. Linehan	M. Federline
J. Greeves	J. Bradbury
W. Reamer	B. Leslie
D. Brooks	N. Coleman

W. Patrick
CNWRA Directors
CNWRA Element Mgrs
T. Nagy (SwRI Contracts)



Washington Office • Twinbrook Metro Plaza #210
12300 Twinbrook Parkway • Rockville, Maryland 20852-1606

Estimating radionuclide release from a uranium deposit through uranium-series systematics in carbonates and opal

David A. Pickett, Bret W. Leslie, William M. Murphy, and Melissa Nugent

The Nopal I uranium deposit at Peña Blanca, Chihuahua, Mexico, has been studied as a natural analog to the proposed geologic repository for high-level nuclear waste at Yucca Mountain, Nevada, USA. The deposit has strong geologic, hydrologic, and geochemical analogies to Yucca Mountain. Uraninite, a spent nuclear fuel analog, was deposited at around 8 Ma (Percy et al., 1994) and has been almost completely oxidized to a series of uranyl oxide hydrates and silicates. Some late-stage uranyl minerals formed at 3 Ma (Pickett and Murphy, 1999a). The oxidized uranium has been partially redistributed by aqueous transport around the deposit into fractures filled with iron oxyhydroxide, carbonate, and silica phases (Prikryl et al., 1997; Pickett and Murphy, 1999a). Thus, Nopal I may provide insights on radionuclide release and migration at the proposed Yucca Mountain site.

Caliche and opal in and around the deposit record mobilization and deposition of uranium and may provide information on rates and episodicity of release from the deposit. Visually pure opals covering uranium mineralization have uranium as high as 6900 ppm. Fracture-filling, carbonate-rich caliches contain up to 320 ppm uranium, demonstrating near-surface mobilization. Caliche uranium contents are highest down-slope from the deposit, but reach over 100 ppm in other directions. Crystalline calcite occurs in several forms around the deposit, representing various generations. Calcite ages are uncertain relative to uranium mineralization, and field relationships suggest some may predate the deposit. Nevertheless, their uranium contents are relatively high for calcite, ranging up to 34 ppm. Partitioning of uranium into calcite can be described by a D value--the ratio of U/Ca in calcite to that in water. Using a typical Nopal I crystalline calcite uranium concentration of 10 ppm and an approximate Nopal I perched water uranium concentration of $10E-08$ molar (Pickett and Murphy, 1999b) yields a D of 0.4, consistent with experimental partitioning data (Nugent and Reeder, in preparation). Higher uranium concentrations in caliches--which clearly postdate the deposit--may be attributable to higher water concentrations and/or a fraction of uranium not in calcite. Regardless of mechanisms, uranium-rich caliches reflect mobilization from the deposit under low-temperature oxidizing conditions likely to occur over long time periods at Yucca Mountain.

Uranium-thorium ages of U-rich caliche and opal testify that release has been recent at 54 ka, and suggest episodic release. Uranium concentrations in the caliche and opal, combined with isotopic age data and estimates of material mass, can be used to calculate rates of redeposition of mobilized uranium. These estimates provide a lower bound to rates of episodic release from the uranium ore body. Using the approach of Murphy and Percy (1992), Murphy and Codell (1999) estimated an upper bound to the mass of uranium removed from the Nopal I deposit [$U(r)$, moles] based on a water infiltration rate estimated from meteoric precipitation rates, uranium concentration in water from solubility limits, and duration time of removal from ages of oxidized uranium minerals. The amount of uranium trapped in carbonates and opals [$U(T)$] can be compared with $U(r)$ to help evaluate the reasonableness of each estimate. The estimated rate of uranium redeposition can be compared to the long-term estimate [$U(r)/\text{time}$]. Another long-term rate estimate--greater than $U(r)/\text{time}$ --was

considered by Murphy and Codell (1999) using the total mass of uranium oxidized. This rate was used in a U.S. Nuclear Regulatory Commission (NRC) performance assessment model for Yucca Mountain; the calculated dose from repository radionuclide release was lower than those using laboratory-derived release parameters from spent nuclear fuel tests. If release is episodic, short-term rates may differ from these long-term averages.

U.S. Department of Energy plans include study of natural analogs for building confidence in dissolution models for predicting spent fuel degradation rates at Yucca Mountain. It is in this context that release rates from Nopal I may prove useful. Radionuclide release rate is a function of both the degradation rate and the flux of water. Rates from episodic release and transport at Nopal I may further test the assumptions used to predict radionuclide release from Yucca Mountain.

This work, funded by the NRC under contract NRC-02-97-009, does not necessarily reflect the views or regulatory position of the NRC.

References

- Murphy, W.M., and R.C. Codell. 1999. Alternate source term models for Yucca Mountain performance assessment based on natural analog data and secondary mineral solubility. *Scientific Basis for Nuclear Waste Management XXII*. D.J. Wronkiewicz, and J.H. Lee, eds. Warrendale, PA: Materials Research Society: 551-558.
- Murphy, W.M., and E.C. Percy. 1992. Source-term constraints for the proposed repository at Yucca Mountain, Nevada, derived from the natural analog at Peña Blanca, Mexico. *Scientific Basis for Nuclear Waste Management XV*. C.G. Sombret, ed. Pittsburgh, PA: Materials Research Society: 521-527.
- Percy, E.C., J.D. Prikryl, W.M. Murphy, and B.W. Leslie. 1994. Alteration of uraninite from the Nopal I deposit, Peña Blanca District, Chihuahua, Mexico, compared to degradation of spent nuclear fuel in the proposed U.S. high-level nuclear waste repository at Yucca Mountain, Nevada. *Applied Geochemistry* 9: 713-732.
- Pickett, D.A., and W.M. Murphy. 1999a. Uranium chemistry and isotopy in waters and rocks at Peña Blanca. *Eighth EC Natural Analogue Working Group Meeting Proceedings*. Luxembourg: European Commission, in press.
- Pickett, D.A., and W.M. Murphy. 1999b. Unsaturated zone waters from the Nopal I natural analog, Chihuahua, Mexico - Implications for radionuclide mobility at Yucca Mountain. *Scientific Basis for Nuclear Waste Management XXII*. D.J. Wronkiewicz, and J.H. Lee, eds. Warrendale, PA: Materials Research Society: 809-816.
- Prikryl, J.D., D.A. Pickett, W.M. Murphy, and E.C. Percy. 1997. Migration behavior of naturally-occurring radionuclides at the Nopal I uranium deposit, Chihuahua, Mexico. *Journal of Contaminant Hydrology* 26: 61-69.