

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

July 28, 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:

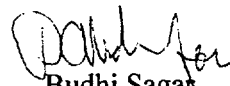
The enclosed abstract is being submitted for programmatic review. This abstract will be submitted for presentation at the Geological Society of America, to be held November 4–7, 2000, in Reno, Nevada. The title of this abstract is:

“Irreversible Coprecipitation in Source Term Analysis for Geologic Disposal of Nuclear Waste” by
W.M. Murphy

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

d:\gh&g\cfiscal 2000\letters\demarco\20.01402561\irreversible coprecipitation in source term analysis....

Enclosure

cc:	B. Meehan	D. Brooks	N. Coleman	T. Nagy (SwRI Contracts)
	E. Whitt	S. Wastler	K. Stablein	
	J. Linehan	T. Essig	W. Patrick	
	J. Greeves	J. Bradbury	CNWRA Directors	
	W. Reamer	B. Leslie	CNWRA Element Mgrs	



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

IRREVERSIBLE COPRECIPITATION IN SOURCE TERM ANALYSIS FOR GEOLOGIC DISPOSAL OF NUCLEAR WASTE

MURPHY, William M., Center for Nuclear Waste Regulatory Analyses, Southwest Research Institute, 6220 Culebra Rd., San Antonio, TX 78238

In geologic disposal of nuclear waste, radionuclides in minor concentrations, but of potentially severe radiological consequence, may be incorporated by coprecipitation with stable secondary uranyl minerals formed during irreversible oxidation of spent uranium dioxide fuel. Natural analog data demonstrate stability of secondary uranyl minerals containing trace Th. Experimental data suggest coprecipitation of Np with schoepite during irreversible spent fuel alteration. Crystallographic studies support compatibility of trace radionuclides in uranyl mineral structures. Calculations show beneficial effects of radionuclide coprecipitation on performance of the potential repository at Yucca Mountain due to diminished release of radionuclides from the source. The thermodynamic basis for coprecipitation as solid solution is well established. Configurational entropy of mixing of trace components in the carrier phase provides a thermodynamic potential for coprecipitation. Enthalpic effects lead to highly compatible or incompatible coprecipitation. However, strong kinetic effects on the distribution of coprecipitant species have been shown experimentally, and the theoretical basis for these effects remains speculative. Some experimental data for coprecipitation with calcite indicate maximum incorporation of compatible species at slow growth rates and maximum incorporation of incompatible species at high growth rates. This pattern is consistent with the Ostwald step rule of initial formation of less stable products. Irreversible thermodynamics dictates that the rate of entropy production during irreversible coprecipitation is minimized. Partial molar configurational entropy of mixing is maximal for minimal trace concentrations. Relative rates of trace and carrier species coprecipitation can be addressed using transition state theory by considering relative concentrations of activated complexes governing simultaneous reactions leading to coprecipitation of carrier and coprecipitant species. A theoretical understanding of irreversible coprecipitation may improve source term predictions for nuclear waste disposal and provide a basis for interpreting a variety of other geochemical phenomena. This work is sponsored by the Nuclear Regulatory Commission, but it is an independent product of the CNWRA and does not necessarily reflect the views or regulatory position of the NRC.