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June 13, 2001 Contract No. NRC-02-97-009 Account No. 20-01402-871

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 Materials Research Society Annual meeting, to be held November 26–29, 2001, in Boston, Massachusetts. The title of this abstract is:

"Radioisotope Fractionation and Secular Disequilibrium in Performance Assessment" William M. Murphy and <u>David A. Pickett</u>

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

D. Pickett

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

Sincerely,

Budhi Sagar

Technical Director

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RADIOISOTOPE FRACTIONATION AND SECULAR DISEQUILIBRIUM IN PERFORMANCE ASSESSMENT

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Radioisotopes of actinides and their decay products are fractionated in natural geochemical systems and in chemical processes associated with geologic disposal of nuclear waste. Two potential applications of isotope fractionation and decay-series disequilibrium in performance assessment for geologic repositories for nuclear waste are preferential radionuclide release and characterization of system closure. Systems that are closed on time scales that are long relative to the half-lives of decay chain nuclides achieve secular equilibrium, characterized by unit activity ratios among nuclides. Natural systems are commonly out of secular equilibrium. For example, U-234/U-238 activity ratios in groundwaters reach values of 5 to 10. A primary mechanism is selective release and mobility of alpha decay products because of nuclear recoil effects. Preferential release of radioisotopes from nuclear waste forms or solubility limiting solid phases could affect repository performance; however, consequences of differential radioisotope release have not been regarded previously in performance assessments. For example, in DOE TSPA-SR performance assessment calculations for the proposed repository at Yucca Mountain, Pu-239 is a major dose contributor at times greater than 50,000 years. If Pu-239 were released preferentially to other Pu isotopes, dose could be affected. Another possible application arises from the usefulness of decay-series disequilibrium to characterize open-system behavior in natural systems. For geologic disposal of nuclear waste, a criterion focused on chemical system closure could capture the essential performance feature of the natural system with respect to radionuclide transport and could be based quantitatively on uranium and thorium decay series isotope equilibria/disequilibria.

This work is supported in part by the U.S. Nuclear Regulatory Commission (NRC) under contract number NRC-02-97-009. This is an independent product and does not necessarily represent the views or regulatory position of the NRC.