OFFICE OF RADIATION AND INDOOR AIR
WASHINGTON, D.C. 20460

April 24, 2024

Mr. Michael Gerle, Director
Environmental Regulatory Compliance Division
Carlsbad Field Office
U.S. Department of Energy
P.O. Box 3090
Carlsbad, New Mexico 88221-3090

Re: Second set of questions on the Replacement Panels Planned Change Request (RPPCR)

Dear Mr. Gerle:

The U.S. Environmental Protection Agency (EPA) is continuing its review of the U.S. Department of Energy's (DOE) submittal of the RPPCR. This letter transmits a set of Agency technical questions and comments (see enclosure). EPA would appreciate a timely response to these questions to help expedite its review.

If you have any questions concerning this request, please contact Jay Santillan at (202) 343-9343 or at Santillan.Jay@epa.gov.

Sincerely,

Tom Peake

Digitally signed by Tom Peake
Date: 2024.04.24 16:19:45 -04'00'

Tom Peake
Director
Center for Waste Management and Regulations

ENCLOSURE
1. Second set of technical questions on the RPPCR

cc: Anderson Ward, DOE CBFO
Justin Marble, DOE EM
Lee Veal, EPA
Ray Lee, EPA

Winifred Okoye, EPA
EPA Docket

Enclosure 1: Second set of technical questions on the RPPCR

RPPCR2-GENERAL-1: Dimensions of replaced area and new panels

How much unused total repository area for waste disposal in square feet and square meters has been lost during WIPP's operational history, including the ground control and contamination issues from the 2014 incidents?

a. Approximately how much disposal area was lost in the repository due to radiological contamination and ground control issues stemming from the 2014 incidents?
    i. Approximately how much disposal area was lost in Panel 7 of the repository due to contamination and ground control issues?
    ii. What is the disposal area of Panels 9 and 10, individually and combined, that DOE is no longer planning to use?
b. Approximately how much disposal area has been lost in Panel 1 and any other panels in repository due to unplanned delays and ground control issues prior to the 2014 incidents?
c. What is the combined total disposal area of replacement Panels 11 and 12?

EPA is interested in verifying quantitatively how much lost repository space Panels 11 and 12 will recover as well as the amount of additional space to the repository they will provide.

RPPCR2-DATA0.FM6-1: $Am(OH)_3(am)$ verification calculations at low ionic strength

Please compare $Am(OH)_3(am)$ solubilities calculated with DATA0.FM6 to the low-ionic-strength solubility data from the studies cited by Guillaumont et al. (2003) for their log K value, including:

a. Rai et al. (1983) solubility data from pH 7 to 11,
b. Edelstein et al. (1983) solubility data at approximately pH 8, 9 and 10, and
c. Nitsche and Edelstein (1985) solubility data at pH 7.

Edelstein, N., J. Bucher, R. Silva, and H. Nitsche. 1983. Thermodynamic Properties of Chemical Species in Nuclear Waste. ONWI-399, Lawrence Berkeley Laboratory, Berkeley, California.

Guillaumont, R., T. Fanghänel, V. Neck, J. Fuger, D.A. Palmer, I. Grenthe, and M.H. Rand. 2003. Update on the Chemical Thermodynamics of Uranium, Neptunium, Plutonium, Americium and Technetium. OECD Nuclear Energy Agency Data Bank, Eds., OECD Publications, Issy-les-Moulineaux, France.

Nitsche, H., and N.M. Edelstein. 1985. Determination of the Solubilities and Complexation of Waste Radionuclides Pertinent to Geologic Disposal at the Nevada Tuff Site. Topical Report, Solubilities and Speciation of Actinide Ions in Near-Neutral Solution. LBL-18900, Lawrence Berkeley Laboratory, Berkeley, California.

Rai, D., R.G. Strickert, D.A. Moore, and J.L. Ryan. Am(III) hydrolysis constants and solubility of Am(III) hydroxide. Radiochimica Acta 33:201-206.

RPPCR2-DATA0.FM6-2: $Am(OH)_3(am)$ verification calculations at high ionic strength

Please provide the following verification calculations for the $Am(OH)_3(am)$ log K value included in DATA0.FM6, including pcH vs. concentration graphs comparing calculated and experimentally measured values and spreadsheets containing the experimental pcH data, experimental solubility data, and calculated solubility results:

a. $Am(OH)_3(am)$ solubilities from pcH 8 to 11.2 compared to data from Neck et al. (2009) in:

i. 2.5 M NaCl
ii. 5 M NaCl
iii. 0.25 M $MgCl_2$
iv. 1.0 M $MgCl_2$

b. $Am(OH)_3$(am) solubilities from pcH 8 to 11.2 compared to data from Herm et al. (2015) in:
v. 2.64 m NaCl
vi. 2.64 m NaCl-$NaNO_3$ with 0.54 m $NaNO_3$
vii. 5.61 m NaCl
viii. 5.61 m NaCl-$NaNO_3$ with 1.15 m $NaNO_3$

c. $Am(OH)_3$(am) solubilities compared to data from Hinz et al. (2015) in:
ix. 5.0 M NaCl plus 0.004 M total borate from pcH 8 to 11.2
x. 5.0 M NaCl plus 0.04 M total borate from pcH 9.5 to 11.2
xi. 5.0 M NaCl plus 0.16 M total borate from pcH 9.5 to 11.2
xii. 0.25 M $MgCl_2$ plus 0.004 M total borate from pcH 8 to 11.2
xiii. 0.25 M $MgCl_2$ plus 0.04 M total borate from pcH 8 to 11.2
xiv. 0.25 M $MgCl_2$ plus 0.16 M total borate from pcH 9.5 to 11.2
xv. 1.0 M $MgCl_2$ plus 0.004 M total borate from pcH 8 to 11.2
xvi. 1.0 M $MgCl_2$ plus 0.04 M total borate from pcH 9.5 to 11.2
xvii. 1.0 M $MgCl_2$ plus 0.16 M total borate from pcH 9.5 to 11.2

The $Am(OH)_3$(am) log K from Guillaumont et al. (2003) was added to DATA0.FM6 because it was better able to simulate the $Nd(OH)_3$(s) – $MgCl_2$ – H_2O system (Miller et al. 2023). Although verification calculations per AP-200 (Domski 2022) were performed in $MgCl_2$ solutions close to 2 M (Domski 2023), DOE has not provided verification calculations in high ionic strength NaCl solutions typical of WIPP brines. The D values calculated for $Nd(OH)_3$(s) solubilities in concentrated NaCl solutions range from -2.76 to -1.88 (Domski 2024), and these negative D values indicate that calculations using the $Am(OH)_3$(am) log K consistently overpredict these solubilities. The nitrate concentrations selected above from Herm et al. (2015) are consistent with the concentrations that would occur in WIPP brine if the entire RPPCR nitrate inventory (Van Soest 2022) dissolved in the RPPCR minimum brine volume (King 2021). The borate concentrations selected from Hinz et al. are consistent with borate concentrations in WIPP brines.

Domski, P. 2022. Analysis Plan: Procedure(s) for the Creation of Thermodynamic Databases for WIPP Compliance Calculations. Sandia National Laboratories, Carlsbad, NM.

Domski, P. 2023. An Update to the WIPP EQ3/6 Database Data0.FM1 with the Creation of Data0.FM6. Sandia National Laboratories, Carlsbad, NM, ERMS 579370.

Domski, P. 2024. Uncertainty Analysis of Actinide Solubilities for the Replacement Panels Planned Change Request (RPPCR) Performance Assessment and the 2024 Compliance Recertification Application (CRA-2024). Unofficial Draft Sandia National Laboratories, Carlsbad, NM, ERMS XXXXX.

Guillaumont, R., T. Fanghänel, V. Neck, J. Fuger, D.A. Palmer, I. Grenthe, and M.H. Rand. 2003. Update on the Chemical Thermodynamics of Uranium, Neptunium, Plutonium, Americium and Technetium. OECD Nuclear Energy Agency Data Bank, Eds., OECD Publications, Issy-les-Moulineaux, France.

Herm, M., X. Gaona, T. Rabung, D. Fellhauer, C. Crepin, K. Dardenne, M. Altmaier, and H. Geckeis. 2015. Solubility and spectroscopic study of An^{III}/Ln^{III} in dilute to concentrated Na-Mg-Ca-Cl-NO_3 solutions. Pure and Applied Chemistry 87:487-502.

Hinz, K., M. Altmaier, X. Gaona, T. Rabung, D. Schild, M. Richmann, D. Reed, E. Alekseev, and H. Geckeis. 2015. Interaction of Nd(III) and Cm(III) with borate in dilute to concentrated alkaline NaCl, $MgCl_2$ and $CaCl_2$ solutions: solubility and TRLFS studies. New Journal of Chemistry 39:849-859.

King, S. 2021. Repository Volume, DRZ Volume, and Minimum Brine Volume for a Direct Brine Release for a Repository with New Panels. Sandia National Laboratories, Carlsbad, New Mexico, ERMS 576475.

Miller, C., P. Domski, J. Jang, I. Foli. 2023. Development of a WIPP Thermodynamic Database for RPPCR and CRA-2019 Compliance Calculations AP-200 Revision 0. Sandia National Laboratories, Carlsbad, New Mexico.

Neck, V., M. Altmaier, T. Rabung, J. Lützenkirchen, and T. Fanghänel. 2009. Thermodynamics of trivalent actinides and neodymium in NaCl, $MgCl_2$, and $CaCl_2$ solutions: solubility, hydrolysis, and ternary Ca-M(III)-OH complexes. Pure and Applied Chemistry 81:1555-1568.

Van Soest, G.D. 2022. Performance Assessment Inventory Report – 2022. Los Alamos National Laboratory Carlsbad Operations INV-PA-22, Revision 0, December 7, 2022.

RPPCR2-DATA0.FM6-3: $AmOHCO_3$(c) verification calculations

Please provide verification calculations for the $AmOHCO_3$(c) log K value included in DATA0.FM6, including pcH vs. concentration graphs comparing calculated and experimentally measured values and spreadsheets containing the experimental pH data, experimental solubility data, and calculated solubility results for $P_{CO2} = 10^{-3}$ atm from Felmy et al. (1990) from pcH 8 to 9.5.

DOE has not provided verification calculations for the $AmOHCO_3$(c) log K value included in DATA0.FM6. The solubility of this solid is important because it is predicted to control Am(III) solubilities in WIPP brines for the RPPCR PA.

Felmy, A.R., D. Rai and R.W. Fulton, 1990. The solubility of $AmOHCO_3$(c) and the aqueous thermodynamics of the system Na^+ - Am^{3+} - HCO_3^- - OH^- - H_2O. Radiochimica Acta 50:193-204.

RPPCR2-DATA0.FM6-4: Omitted Pitzer interaction parameters

The Pitzer interaction parameters from Moog et al. (2004) listed in the following table were previously included in the DATA0.FM4 database reviewed by EPA (2022). Please explain the reasons for omitting these parameters from DATA0.FM6. Please provide calculations that show whether omission of these parameters would affect predicted dissolved iron concentrations in WIPP brines under repository conditions.

i	j	k	Θ_{ij}	ψ_{ijk}
K^+	Fe^{2+}	--	0.0274	--
Ca^{2+}	Fe^{2+}	--	0.0811	--
Fe^{2+}	K^+	Cl^-	--	-0.0252

Fe^{2+}	Ca^{2+}	Cl^-	--	-0.016[a]

RPPCR2-12PanelAnalyses:

EPA is interested in the possibility of using the individual panel releases shown in Figure 3 of ERMS 580656 Estimation of Releases from a 12-Panel Repository by Hansen et al. (2023) to estimate releases from a 12-panel repository. We have the following two questions:

1. Please provide a detailed explanation of how the individual panel releases shown in Figure 3 of ERMS 580656 were calculated, with accompanying conceptual descriptions and justifications.
2. Please provide an explanation of how the individual panel releases shown in Figure 3 of ERMS 580656 accumulate to yield the combined releases of all 19 panels shown in Figure 4-43 of ERMS 581044 (the RPPCR PA).

Hansen, C., Brunell, S., King, S. (2023). Estimation of Releases from a 12-Panel Repository, Revision 0. Sandia National Laboratories, Carlsbad, NM. ERMS 580656.