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Sandia National Laboratories
Waste Isolation Pilot Plant

Calculation of Organic-Ligand Concentrations for the WIPP Performance-Assessment Baseline Calculations

Work Carried Out under Task 1 of the Analysis Plan for the Calculation of Actinide Solubilities for the WIPP PABC, AP 120, Rev. 0
WIPP:1.4.2.2:PA:QA-L:539371

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1 ABBREVIATIONS, ACRONYMS, AND INITIALISMS

Table 1 defines the abbreviations, acronyms, and initialisms used in this report.

Table 1. Abbreviations, Acronyms, and Initialisms.

Abbreviation, Acronym, or Initialism	Definition
acetate	$CH_3CO_2^-$
acetic acid	CH_3CO_2H
AP	analysis plan
DOE	(U.S.) Department of Energy
C	carbon
CCA	(WIPP) Compliance Certification Application
citrate	$(CH_2CO_2H)_2C(OH)(CO_2)^-$
citric acid	$(CH_2CO_2H)_2C(OH)(CO_2H)$
CRA-2004	the first (WIPP) Compliance Recertification Application
DOE	(U.S.) Department of Energy
EDTA	ethylenediaminetetraacetate, $(CH_2CO_2H)_2N(CH_2)_2N(CH_2CO_2H)(CH_2CO_2)^-$; or ethylenediaminetetraacetic acid, $(CH_2CO_2H)_2N(CH_2)_2N(CH_2CO_2H)_2$
EPA	(U.S.) Environmental Protection Agency
g	gram(s)
H	hydrogen
kg	kilogram(s)
L	liter(s)
M	molar
m	meter(s) or molal
mol	moles
N	nitrogen
Na	sodium
Na acetate	CH_3CO_2Na
Na citrate	$(CH_2CO_2H)_2C(OH)(CO_2Na)$
Na EDTA	$(CH_2CO_2H)_2N(CH_2)_2N(CH_2CO_2H)(CH_2CO_2Na)$
Na oxalate	$(CO_2H)(CO_2Na)$
O	oxygen
oxalate	$(CO_2H)(CO_2)^-$
oxalic acid	$(CO_2H)_2$

Table 1. Abbreviations, Acronyms, and Initialisms.

Abbreviation, Acronym, or Initialism	Definition
PA	(WIPP) performance assessment
PABC	(WIPP) Performance Assessment Baseline Calculation(s)
WIPP	(U.S. DOE) Waste Isolation Pilot Plant
wt	weight

2 INTRODUCTION

This analysis report provides the current concentrations of acetate, citrate, ethylenediaminetetraacetate (EDTA), and oxalate (referred to hereafter as "organic ligands") in brines that could enter the U.S. Department of Energy's (DOE's) Waste Isolation Pilot Plant (WIPP) after closure of the waste panels. These concentrations will be used to calculate actinide solubilities for the WIPP Performance Assessment Baseline Calculations (PABC), required by the U.S. Environmental Protection Agency for the first recertification of the WIPP. This analysis described in this report was carried out under Task 1 of the analysis plan (AP) for the actinide-solubility calculations for the PABC (Brush and Xiong, 2005, Subsection 7.1).

Brush and Xiong (2003a) used revised estimates of the total masses of organic ligands to be emplaced in the WIPP (Crawford, 2003) to update the concentrations of these organic ligands calculated in 1996 for the WIPP Compliance Certification Application (CCA). (U.S. DOE, 1996a, Appendix B4, provided the masses of organic ligands used for the CCA; U.S. DOE, 1996b, Appendix SOTERM, Table SOTERM-4, column labeled "Organic Concentration (scaled)," provided the concentrations). However, Brush and Xiong (2003a) did not revise the volume of brine used to calculate organic-ligand concentrations. The updated concentrations (see Brush and Xiong, 2003a, Table 4, column labeled "CRA Concentration (M)"; and U.S. DOE, 2004, Appendix PA, Attachment SOTERM, Table SOTERM-4, column labeled "Concentrations Used in FMT for the CRA-2004 PA") were used to calculate actinide solubilities for the PA calculations for the first WIPP Compliance Recertification Application (CRA-2004 PA).

After the revised organic-ligand concentrations of Brush and Xiong (2003a) had been used for the actinide-solubility calculations for the CRA-2004 PA, Crawford and Leigh (2003) and Leigh (2003) corrected Crawford's (2003) estimates of the total masses of organic ligands to be emplaced in the WIPP, and Brush and Xiong (2003b) corrected the concentrations of these organic ligands. The corrected concentrations appear in Brush and Xiong (2003b, Table 4, column labeled "Corrected Concentration for a 10-Panel, Homogeneous Repository (M), This Report") and U.S. DOE (2004, Appendix PA, Attachment SOTERM, Table SOTERM-4, column labeled "Concentrations Based on Corrected CRA Inventory") provided these corrected organic-ligand concentrations.

However, the actinide solubilites calculated for the CRA-2004 PA were not corrected because Crawford and Leigh (2003) and Leigh (2003) decreased the masses; and Brush and Xiong (2003b) decreased the concentrations of organic ligands in the WIPP. Therefore, the solubilities calculated for the CRA-2004 PA were slightly higher - and thus slightly conservative (i.e., resulted in higher releases of radioelements from the repository) - relative to those that would have been obtained had they been recalculated.

The analysis plan (AP) for the actinide-solubility calculations for PABC specifies that both the masses of organic ligands to be emplaced in the WIPP and the brine volume used to

calculate organic-ligand concentrations be re-evaluated and - if necessary - revised prior to the actinide solubility calculations (see Brush and Xiong, 2005, Subsection 7.1).

Leigh (2005a, 2005b) reviewed the most recent information on the WIPP inventory and concluded that the masses of organic ligands from Crawford and Leigh (2003) and Leigh (2003) should be used to calculate organic-ligand concentrations for the PABC.

However, Stein (2005) concluded that the minimum brine volume required for release of dissolved and colloidal radioelements - established by Larson (1996) and used by the U.S. DOE (1996b, Appendix SOTERM, Section SOTERM.5), Brush and Xiong (2003a, 2003b), and the U.S. DOE (2004, Appendix PA, Attachment SOTERM, Section SOTERM-5.0) - should be revised. Therefore, this analysis report describes the calculation of organic-ligand concentrations based on previous estimates of the masses of these organic ligands to be emplaced in the WIPP (Crawford and Leigh, 2003; Leigh, 2003) and Stein's (2005) revised estimate of the minimum brine volume.

The new organic-ligand concentrations reported herein (see Subsection 5 below) replace those used for the actinide-solubility calculations for the CRA-2004 PA (see Brush and Xiong, 2003a, Table 4, column labeled "CRA Concentration (M)"; U.S. DOE, 2004, Appendix PA, Attachment SOTERM, Table SOTERM-4, column labeled "Concentrations Used in FMT for the CRA-2004 PA"). The current organic-ligand concentrations also replace those calculated after the CRA-2004 PA (Brush and Xiong, 2003b, Table 4, column labeled "Corrected Concentration for a 10-Panel, Homogeneous Repository (M), This Report"; and U.S. DOE, 2004, Appendix PA Attachment SOTERM, Table SOTERM-4, column labeled "Concentrations Based on Corrected CRA Inventory").

3 MASSES OF ORGANIC LIGANDS TO BE EMPLACED IN THE WIPP

Leigh (2005a, 2005b) reviewed the most recent information on the WIPP inventory and concluded that we should continue to use the total masses of acetic acid, Na acetate, citric acid, Na citrate, Na EDTA, oxalic acid, and Na oxalate estimated by Crawford and Leigh (2003) and Leigh (2003) to calculate the concentrations of acetate, citrate, EDTA, and oxalate in WIPP brines for the PABC. These estimated total masses are in Crawford and Leigh (2003, Table 4, column labeled "Total Mass"). These masses are the same as those used by Brush and Xiong (2003b) to correct the organic-ligand concentrations used for the actinide-solubility calculations for the CRA-2004 PA (see Section 1 above).

However, it must be noted that - although Crawford (2003) reported that all of the EDTA in the WIPP inventory comprises Na EDTA - Crawford and Leigh (2003) and Leigh (2005a) erroneously reported that it comprises EDTA. Therefore, Leigh (2005b) corrected Crawford and Leigh (2003) and Leigh (2005a) by stating that all of the EDTA in the WIPP inventory is actually Na EDTA.

4 CALCULATION OF MOLECULAR WEIGHTS OF ORGANIC LIGANDS

We used the following atomic weights to calculate the molecular weights of acetic acid, Na acetate, citric acid, Na citrate, Na EDTA, oxalic acid, and Na oxalate: H, 1.00794 g/mol; C, 12.0107 g/mol; N, 14.00674 g/mol; O, 15.9994 g/mol, and Na, 22.989770 g/mol (Lide, 2002). Table 2 provides the formulas for these compounds.

Table 2. Formulas and Molecular Weights of Two Forms of Four Ligands That Could Be Emplaced in the WIPP.

Compound	Formula	Mol Wt (g)
Acetic acid	CH_3CO_2H	60.0520
Na acetate	CH_3CO_2Na	82.0338
Citric acid	$(CH_2CO_2H)_2C(OH)(CO_2H)$	192.1235
Na citrate	$(CH_2CO_2H)_2C(OH)(CO_2Na)$	214.1054
EDTA	$(CH_2CO_2H)_2N(CH_2)_2N(CH_2CO_2H)_2$	292.2427
Na EDTA	$(CH_2CO_2H)_2N(CH_2)_2N(CH_2CO_2H)(CH_2CO_2Na)$	314.2246
Oxalic acid	$(CO_2H)_2$	90.0349
Na oxalate	$(CO_2H)(CO_2Na)$	112.0167

Acetic acid, citric acid, EDTA, and oxalic acid contain one, three, four, and two acidic hydrogen atoms, respectively, that can be substituted with Na (or other alkali or alkaline-earth metals). Crawford and Leigh (2003), and Leigh (2003, 2005) did not specify how many of the acidic hydrogens in Na citrate, Na EDTA, or Na oxalate were replaced by Na. Therefore, we assumed that only one of the acidic hydrogens was substituted with Na to calculate the molecular weights of Na citrate, Na EDTA, and Na oxalate. This assumption is conservative (i.e., it results in the highest molar quantities of the Na-bearing forms of these ligands.

We calculated the molecular weights of acetic acid, Na acetate, citric acid, Na citrate, EDTA (ethylenediaminetetraacetic acid, in this case), Na-EDTA, oxalic acid, and Na oxalate as follows:

- Acetic acid: (4 × atomic mass H) + (2 × atomic mass C) + (2 × atomic mass O) = (4 × 1.00794 g/mol) + (2 × 12.0107 g/mol) + 2 × 15.9994 g/mol) = (4.0318 + 24.0214 + 31.9988) g/mol = 60.0520 g/mol.

- Na acetate: (3 × atomic mass H) + (2 × atomic mass C) + (2 × atomic mass O) + (1 × atomic mass Na) = (3 × 1.00794 g/mol) + (2 × 12.0107 g/mol) + 2 × 15.9994 g/mol) + (1 × 22.989770 g/mol) = (3.0238 + 24.0214 + 31.9988 + 22.989770) g/mol = 82.0338 g/mol.

- Citric acid: (8 × atomic mass H) + (6 × atomic mass C) + (7 × atomic mass O) = (8 × 1.00794 g/mol) + (6 × 12.0107 g/mol) + (7 × 15.9994 g/mol) = (8.0635 + 72.0642 + 111.9958) g/mol = 192.1235 g/mol.

- Na citrate: (7 × atomic mass H) + (6 × atomic mass C) + (7 × atomic mass O) + (1 × atomic mass Na) = (7 × 1.00794 g/mol) + (6 × 12.0107 g/mol) + (7 × 15.9994 g/mol) + (1 × 22.989770 g/mol) = (7.0556 + 72.0642 + 111.9958 + 22.989770) g/mol = 214.1054 g/mol.

- EDTA: (16 × atomic mass H) + (10 × atomic mass C) + (2 × atomic mass of N) + (8 × atomic mass O) = (16 × 1.00794 g/mol) + (10 × 12.0107 g/mol) + (2 × 14.00674 g/mol) + (8 × 15.9994 g/mol) = (16.1270 + 120.1070 + 28.0135 + 127.9952) g/mol = 292.2427 g/mol.

- Na EDTA: (15 × atomic mass H) + (10 × atomic mass C) + (2 × of N) + (8 × atomic mass O) + (1 × atomic mass Na)= (15 × 1.00794 g/mol) + (10 × 12.0107 g/mol) + (2 × 14.00674 g/mol) + (8 × 15.9994 g/mol) + (1 × 22.989770 g/mol) = (15.1191 + 120.1070 + 28.0135 + 127.9952 + 22.989770) g/mol = 314.2246 g/mol.

- Oxalic acid: (2 × atomic mass H) + (2 × atomic mass C) + (4 × atomic mass O) = (2 × 1.00794 g/mol) + (2 × 12.0107 g/mol) + 4 × 15.9994 g/mol) = (2.0159 + 24.0214 + 63.9976) g/mol = 90.0349 g/mol.

- Na oxalate: (1 × atomic mass H) + (2 × atomic mass C) + (4 × atomic mass O) + (1 × atomic mass Na) = (1 × 1.00794 g/mol) + (2 × 12.0107 g/mol) + 4 × 15.9994 g/mol) + (1 × 22.989770 g/mol) = (1.00794 + 24.0214 + 63.9976 + 22.989770) g/mol = 112.0167 g/mol.

Table 2 provides the molecular weights of these compounds (see preceding page).

5 CALCULATION OF ORGANIC-LIGAND CONCENTRATIONS

We used 10,011 m^3 of brine, "a reasonable minimum volume of brine in the repository required for a brine release" (Stein, 2005), to calculate the dissolved concentrations of acetate, citrate, EDTA, and oxalate in a homogeneous, 10-panel PA repository. This volume replaces the previously used value of 29,841 m^3 of brine, "the smallest quantity of brine required to be in the repository [for] transport away from the repository" (Larson, 1996; U.S. DOE, 1996b). A volume of 29,841 m^3 of brine was used by the U.S. DOE (1996b, Appendix SOTERM, Section SOTERM.5), Brush and Xiong (2003a, 2003b), and the U.S. DOE (2004, Appendix PA, Attachment SOTERM, Section SOTERM-5.0) to calculate organic-ligand concentrations for the CCA and the CRA-2004 PA (see Section 1 above). A volume of 10,011 m^3 of brine is conservative because any volume greater than 10,011 m^3 would result in lower organic-ligand concentrations. A volume of 10,011 m^3 of brine is equal to 10,011,000 L of brine.

We calculated the concentrations of acetic acid, Na acetate, citric acid, Na citrate, Na EDTA, oxalic acid, and Na oxalate by multiplying the total masses of these compounds in kg from Crawford and Leigh (2003, Table 4, column labeled "Total Mass") by 1000 g/kg to convert Crawford and Leigh's estimates to total masses in grams. Next, we divided these masses by the molecular weights of these compounds from Table 2 of this report, column labeled "Mol Wt (g)," which yielded the total quantities of these compounds to be emplaced in moles. We then divided these quantities by 10,011,000 L to obtain the concentrations of these compounds in units of mol/L (M):

- Acetic acid: $((1.42 \times 10^2 \text{ kg}) \times (1000 \text{ g/kg}) \div (60.0520 \text{ g/mol})) \div 1.0011 \times 10^7 \text{ L} = 2.36 \times 10^{-4}$ M.
- Na acetate: $((8.51 \times 10^3 \text{ kg}) \times (1000 \text{ g/kg}) \div (82.0338 \text{ g/mol})) \div 1.0011 \times 10^7 \text{ L} = 1.04 \times 10^{-2}$ M.
- Citric acid: $((1.19 \times 10^3 \text{ kg}) \times (1000 \text{ g/kg}) \div (192.1235 \text{ g/mol})) \div 1.0011 \times 10^7 \text{ L} = 6.19 \times 10^{-4}$ M.
- Na citrate: $((4.00 \times 10^2 \text{ kg}) \times (1000 \text{ g/kg}) \div (214.1054 \text{ g/mol})) \div 1.0011 \times 10^7 \text{ L} = 1.87 \times 10^{-4}$ M.
- Na EDTA: $((2.56 \times 10^1 \text{ kg}) \times (1000 \text{ g/kg}) \div (314.2246 \text{ g/mol})) \div 1.0011 \times 10^7 \text{ L} = 8.14 \times 10^{-6}$ M.
- Oxalic acid: $((1.38 \times 10^4 \text{ kg}) \times (1000 \text{ g/kg}) \div (90.0349 \text{ g/mol})) \div 1.0011 \times 10^7 \text{ L} = 1.53 \times 10^{-2}$ M.
- Na oxalate: $((3.39 \times 10^4 \text{ kg}) \times (1000 \text{ g/kg}) \div (112.0167 \text{ g/mol})) \div 1.0011 \times 10^7 \text{ L} = 3.02 \times 10^{-2}$ M.

Table 3 summarizes these calculations and provides the results (see page 19).

Table 3. Dissolved Concentrations of One or Two Forms of Four Ligands for a Homogeneous, 10-Panel Repository.

Compound	Total Mass (kg)	Total Mass (g)	Total Quantity (mol)	Concentration (M)
Acetic acid	1.42×10^{2} [A]	1.42×10^{5}	2.36×10^{3}	2.36×10^{-4}
Na acetate	8.51×10^{3} [A]	8.51×10^{6}	1.04×10^{5}	1.04×10^{-2}
Citric acid	1.19×10^{3} [A]	1.19×10^{6}	6.20×10^{3}	6.19×10^{-4}
Na citrate	4.00×10^{2} [A]	4.00×10^{5}	1.87×10^{3}	1.87×10^{-4}
EDTA	None reported[B]	None reported[B]	None reported[B]	None reported[B]
Na EDTA	2.56×10^{1} [B]	2.56×10^{4}	8.15×10^{1}	8.14×10^{-6}
Oxalic acid	1.38×10^{4} [A]	1.38×10^{7}	1.53×10^{5}	1.53×10^{-2}
Na oxalate	3.39×10^{4} [A]	3.39×10^{7}	3.03×10^{5}	3.02×10^{-2}

A. From Crawford and Leigh (2003, Table 4, column labeled "Total Mass"), and Leigh (2005a, 2005b, column labeled "Total").

B. Crawford (2003) reported that all of the EDTA in the WIPP inventory is Na EDTA. Crawford and Leigh (2003) and Leigh (2005a) misreported that it comprises EDTA. Therefore, Leigh (2005b, Table 1, column labeled "Total") corrected Crawford and Leigh (2003) and Leigh (2005a) by stating that all of the EDTA in the WIPP inventory is actually Na EDTA.

We calculated the total dissolved concentrations of acetate, citrate, and oxalate by adding the concentrations of acetic acid and Na acetate, citric acid and Na citrate, and oxalic acid and Na oxalate (see Table 3, column labeled "Concentration (M)" above). The concentration of Na EDTA is equal to the total concentration of EDTA (see Section 3; and Table 3, Note B, above). Table 4 (see next page) provides the new total dissolved concentrations of acetate, citrate, EDTA, and oxalate for a homogeneous, 10-panel repository and compares them to the concentrations calculated by U.S. DOE (1996b, Appendix SOTERM, Table SOTERM-4, column labeled "Organic Concentration (scaled),") for the CCA, Brush and Xiong (2003a) for the actinide-solubility calculations for the CRA-2004 PA, and the corrected concentrations of Brush and Xiong (2003b).

Table 4. Concentrations of Four Organic Ligands for a Homogeneous, 10-Panel Repository.

Organic Ligand	CCA[A] (m)	CRA-2004 PA[B] (M)	CRA-2004 PA, Corrected[C] (M)	PABC[D] (M)
Acetate	1.1×10^{-3}	5.05×10^{-3}	3.56×10^{-3}	1.06×10^{-2}
Citrate	7.4×10^{-3}	3.83×10^{-4}	2.71×10^{-4}	8.06×10^{-4}
EDTA	4.2×10^{-6}	3.87×10^{-6}	2.73×10^{-6}	8.14×10^{-6}
Oxalate	4.7×10^{-4}	2.16×10^{-2}	1.53×10^{-2}	4.55×10^{-2}

A. U.S. DOE (1996b, Appendix SOTERM, Table SOTERM-4, column labeled "Organic Concentration (scaled)."
B. Brush and Xiong (2003a, Table 4, column labeled "CRA Concentration (M)").
C. Brush and Xiong (2003b, Table 4, column labeled "Corrected Concentration for a 10-Panel, Homogeneous Repository (M), This Report").
D. This analysis report.

6 CONCLUSIONS

This analysis yielded the following organic-ligand concentrations for the actinide-solubility calculations for the PABC: (1) acetate: 1.06×10^{-2} M, (2) citrate: 8.06×10^{-4} M; (3) EDTA: 8.14×10^{-6} M; (4) oxalate: 4.55×10^{-2} M (see Table 4 above). We will use these organic-ligand concentrations to calculate actinide solubilities for the PABC.

The corrected masses of organic ligands estimated by Crawford and Leigh (2003), the masses used to calculate organic-ligand concentrations for the PABC actinide-solubility calculations, are 70.5% of the scaled masses estimated by Crawford (2003, Table 2, column labeled "Scaled Mass (kg)"), the masses used by Brush and Xiong (2003a) to calculate these concentrations for the CRA-2004 PA solubility calculations.

However, the minimum volume of brine in the repository required for a brine release established by Stein (2005), the volume used to calculate organic-ligand concentrations for the PABC, is about 33.5% of that estimated by Larson (1996), the volume used by Brush and Xiong (2003a, 2003b) for the CRA-2004 PA.

Therefore, the organic-ligand concentrations to be used for the PABC solubility calculations exceed those used for the CRA-2004 PA solubility calculations by about a factor of two.

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