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Subject: Discussion on the influence of organic ligands on the solubility of U(VI)

This memorandum is written to demonstrate that the acetate, citrate, EDTA, and oxalate do not significantly impact the solubility of U(VI) under the conditions described by the FMT calculations. The present work shows calculations based on the input and the output data of the FMT Runs 12, 18, 22, and 28 (Brush and Xiong, 2003a).

The calculations presented in this memorandum are performed following the analysis plan AP-112 (Kirkes and Wagner, 2004) to answer part of the EPA question C-23-13 (EPA, 2004).

Description of the Organic Ligands in WIPP

Acetate, citrate, oxalate, and EDTA are capable of forming soluble complexes with the actinides, and have been identified in the WIPP inventory. The database describing the interaction of the organic ligands with the actinides was not complete at the time of the Compliance Certification Application (CCA) (DOE, 1996), but since 1996 both stability constants and Pitzer parameters have been determined, allowing inclusion of the organic ligands in the FMT speciation and solubility calculations.

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U(IV) is likely to be the most stable oxidation state of uranium under the mildly basic pH and chemically reducing conditions that will exist within the WIPP (DOE, Appendix PA, Attachment SOTERM, 2004). However, in order to address potential issues surrounding uncertainties of the prevailing oxidation states of this actinide, the WIPP PA models uranium as U(IV) in approximately half of the PA vectors, and as U(VI) in the other half. The solubility of U(VI) remains, as was the case for the CCA, based upon a literature survey carried out by Hobart and Moore (1996).

The concentrations of organic ligands are featured in Table 1, from Table SOTERM-4 of the Compliance Recertification Application (CRA). The values of "Molar Concentrations Used in FMT for the CRA-2004 PA" was determined by Brush and Xiong (2003b).

Table 1. Organic Ligand Concentrations used in WIPP Speciation and Solubility Calculations

Organic Ligand	Concentrations Based on Corrected CRA Inventory (M)	Concentrations Used in FMT for the CRA-2004 PA (M)	Concentrations Given in the CCA (M)
acetate	3.56×10^{-3}	5.05×10^{-3}	1.1×10^{-3}
citrate	2.71×10^{-4}	3.83×10^{-4}	7.4×10^{-3}
oxalate	2.73×10^{-6}	3.87×10^{-6}	4.2×10^{-6}
EDTA	1.53×10^{-2}	2.16×10^{-2}	4.7×10^{-4}

Effect of the Organic Ligands on U(VI)

The only actinide predicted to exist in the hexavalent oxidation state in the WIPP is U(VI). As stated in Attachment SOTERM of Appendix PA of the CRA, "the actinide(VI) speciation and solubility model for brines under basic conditions has not been developed sufficiently for use in FMT. The hydrolysis behavior of U(VI) is quite complicated and no satisfactory predictive models applicable to WIPP conditions are available." As a result, no database was developed to include An(VI) in the FMT calculations. The An(VI) solubility in systems containing no acetate, citrate, oxalate, or EDTA was estimated, based on literature data (Hobart and Moore, 1996). The solubility of U(VI) in the presence of these organic ligands was assumed to be equal to that in the absence of organic ligands.

Although no thermodynamic database is currently available for the uranyl species at high ionic strength, Mompean et al. (2003) published a revised database of the chemicals thermodynamics of actinides, which features data at zero ionic strength. Also, Choppin et al. (2001) published results of thermodynamic studies of uranyl complexation with organic ligands at a variety of ionic strengths, including at zero ionic strength. We will estimate the fraction of uranyl–organic complexes, using the zero-ionic-strength database and results from FMT calculations (Brush and Xiong, 2003a).

Estimation of the Uranyl Organic Ligands Fraction

The Gibbs free energy is defined as:

$$\Delta G = -RT \ln \underline{K} \tag{1}$$

where $R = 8.314$ $Jmol^{-1}K^{-1}$, $T = 298.15$, and K is the equilibrium constant.
The standard state Gibbs free energies of inorganic uranyl species are presented in Table 2.

Table 2: Gibbs Energy of Uranyl–Inorganic Species [a]

Species	ΔG^0 (kJ/mol)
UO_2^{2+}	-952.551
UO_2OH^+	-1159.724
$UO_2(OH)_2^0(aq)$	-1357.479
$UO_2(OH)_2(s)$	-1398.683
$UO_2(CO_3)^0$	-1537.188
$UO_2(CO_3)(cr)$	-1564.701
$UO_2(CO_3)_2^{2-}$	-2103.161
$UO_2(CO_3)_3^{4-}$	-2660.914
UO_2Cl^+	-1084.738
$UO_2Cl_2(aq)$	-1208.707
$UO_2SO_4(aq)$	-1714.535
$UO_2(SO_4)_2^{2-}$	-2464.190
$UO_2(SO_4)_3^{4-}$	-3201.801

[a] Data are taken from Mompean et al. (2003)

FMT uses the normalized standard chemical potential μ^0/RT, defined as:

$$\mu^0\big/RT = \Delta G^0\big/RT \qquad (2)$$

Values of μ^0/RT are calculated from the values of ΔG^0 given in Table 2. The values of μ^0/RT are presented in Table 3.

Table 3: Normalized Standard Chemical Potentials for Uranyl–Inorganic Species

Species	μ^0/RT
UO_2^{2+}	-384.276
UO_2OH^+	-467.853
$UO_2(OH)_2^0(aq)$	-547.631
$UO_2(OH)_2(s)$	-564.254
$UO_2(CO_3)^0(aq)$	-620.129
$UO_2(CO_3)(cr)$	-631.228
$UO_2(CO_3)_2^{2-}$	-848.453
$UO_2(CO_3)_3^{4-}$	-1073.460
UO_2Cl^+	-437.603
$UO_2Cl_2(aq)$	-487.614
$UO_2SO_4(aq)$	-691.674
$UO_2(SO_4)_2^{2-}$	-994.098
$UO_2(SO_4)_3^{4-}$	-1291.664

Additional standard chemical potentials, available in the FMT database are presented in Table 4.

Table 4: Standard Chemical Potentials of the Additional Species

Species	μ^0/RT
HCO_3^-	-236.7510
CO_3^{2-}	-212.9440
CO_2 (aq)	-155.6800
H_2O	-95.6635
H^+	0.0000
OH^-	-63.4350
Cl^-	-52.9550
SO_4^{2-}	-300.3860

Choppin et al. (2001) determined the standard chemical potentials of the uranyl–organic ligand complexes; the data are summarized in Table 5.

Table 5: Normalized Standard Chemical Potentials of the Uranyl–Organic Complexes

Species	μ^0/RT
UO_2Ac^+	-538.585
UO_2Cit^-	-371.429
UO_2EDTA^{2-}	-361.555
UO_2Ox(aq)	-387.779

Given the following chemical equilibrium,

$$A + B \rightleftharpoons C + D \tag{3}$$

The equilibrium constant is defined as:

$$K = \frac{[C][D]\gamma_C\gamma_D}{[A][B]\gamma_A\gamma_B} \tag{4}$$

At zero ionic strength, the ionic activity coefficient terms, $\gamma = 1$. Substituting definitions from equations (1) and (2) gives logK as:

$$-\log K = \frac{\left(\mu_0/RT\right)_C + \left(\mu_0/RT\right)_D - \left(\mu_0/RT\right)_A - \left(\mu_0/RT\right)_B}{\ln 10} \tag{5}$$

The equilibrium constants for each uranyl-organic and -inorganic complex formation are presented in Table 6.

Table 6: Equilibrium Constants for Uranyl-Inorganic and -Organic Species, at zero ionic strength

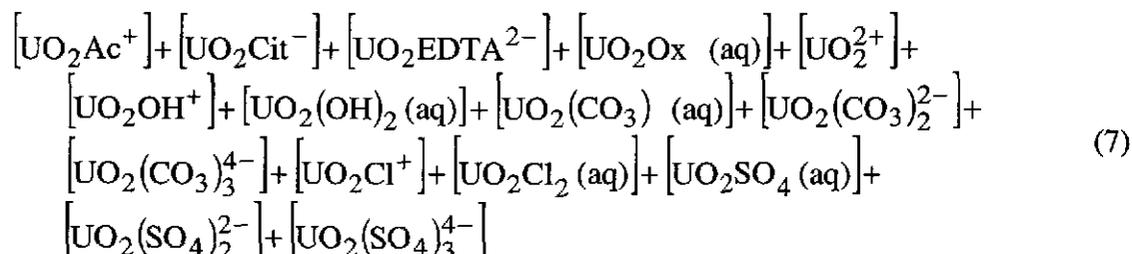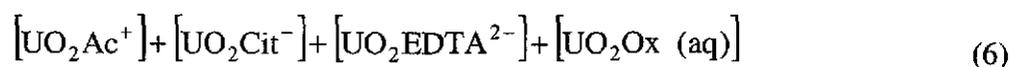| Equilibrium	log K^0
$UO_2^{2+} + OH^- \rightleftharpoons UO_2(OH)^+$	8.75
$UO_2^{2+} + 2\ OH^- \rightleftharpoons UO_2(OH)_2^0(aq)$	15.85
$UO_2^{2+} + 2\ OH^- \rightleftharpoons UO_2(OH)_2(solid)$	23.06
$UO_2^{2+} + CO_3^{2-} \rightleftharpoons UO_2(CO_3)^0$	9.95
$UO_2^{2+} + CO_3^{2-} \rightleftharpoons UO_2(CO_3)(cr)$	14.77
$UO_2^{2+} + 2\ CO_3^{2-} \rightleftharpoons UO_2(CO_3)_2^{2-}$	16.63
$UO_2^{2+} + 3\ CO_3^{2-} \rightleftharpoons UO_2(CO_3)_3^{4-}$	21.87
$UO_2^{2+} + Cl^+ \rightleftharpoons UO_2Cl^+$	0.16
$UO_2^{2+} + 2\ Cl^+ \rightleftharpoons UO_2Cl_2(aq)$	-1.12
$UO_2^{2+} + SO_4^{2-} \rightleftharpoons UO_2(SO_4)(aq)$	3.05
$UO_2^{2+} + 2\ SO_4^{2-} \rightleftharpoons UO_2(SO_4)_2^{2-}$	3.93
$UO_2^{2+} + 3\ SO_4^{2-} \rightleftharpoons UO_2(SO_4)_3^{4-}$	2.71
$UO_2^{2+} + Ac^- \rightleftharpoons UO_2(Ac)^+$	3.03
$UO_2^{2+} + Cit^{3-} \rightleftharpoons UO_2(Cit)^+$	8.94
$UO_2^{2+} + EDTA^{4-} \rightleftharpoons UO_2(EDTA)^{2-}$	13.18
$UO_2^{2+} + Ox^{2-} \rightleftharpoons UO_2(Ox)^0\ (aq)$	7.18

We calculate the fraction of uranyl-organic complexes in a system containing acetate, citrate, EDTA, and oxalate in concentrations equal to the ones determined from the FMT calculations for the solubility of An(III, IV, and V) (Brush and Xiong, 2003a). The calculations were carried out with the same pH and carbonate concentrations given by the FMT output. The FMT calculations were performed to model systems at high ionic strength, whereas the stability constant values for uranyl with organic ligands used in this report are for low ionic strength. However, the goal of this report is to demonstrate that organic ligand complexation of U(VI) under WIPP conditions will render a negligible impact upon U(VI) speciation and solubility, and the results of this analysis should be regarded as an approximation. The FMT Run 12 was performed to calculate species concentrations in a system featuring the brine GWB in absence of microbial activity, containing the organic ligands described above, and in which CO_2 fugacity is maintained by the brucite-calcite carbonation reaction (Brush and Xiong, 2003a). The FMT Run 18 was performed to calculate species concentrations in a system featuring the brine GWB in presence of microbial activity, containing the organic ligands described above, and which the CO_2 fugacity is maintained by the brucite-hydromagnesite buffer (Brush and Xiong, 2003a). The FMT Run 22 was performed to calculate species concentrations in a system featuring the brine ERDA–6 in absence of microbial activity, containing the organic ligands described above, and which the CO_2 fugacity is maintained by brucite-calcite buffer (Brush and Xiong, 2003a). The FMT Run 28 was performed to calculate species concentrations in a system featuring the brine ERDA–6 in presence of microbial activity, containing the organic ligands described above, and which the CO_2 fugacity is maintained by brucite- hydromagnesite buffer (Brush and Xiong, 2003a). The free species concentrations obtained in the output of the FMT Runs 12, 18, 22 and 28 are presented in Table 7.

Table 7: Molarity of Free Species in the Output of the FMT Runs 22 and 12 (Brush and Xiong, 2003a)

Ligand	Concentration of the Free Species (M)			
	Run 12 (GWB) [a]	Run 18 (GWB) [a]	Run 22 (ERDA–6) [a]	Run 28 (ERDA–6) [a]
Acetate	2.71059×10^{-3}	2.71058×10^{-3}	3.50750×10^{-3}	3.62315×10^{-3}
Citrate	8.49384×10^{-6}	8.49141×10^{-6}	9.90235×10^{-6}	1.07159×10^{-5}
EDTA	4.04495×10^{-11}	4.04395×10^{-11}	8.09661×10^{-12}	8.28602×10^{-12}
Oxalate	2.08478×10^{-4}	2.08444×10^{-4}	5.39563×10^{-4}	5.99387×10^{-4}
OH^-	7.09877×10^{-6}	7.09852×10^{-6}	1.23390×10^{-5}	1.30662×10^{-5}
CO_3^{2-}	2.27525×10^{-5}	2.16432×10^{-5}	1.38787×10^{-5}	6.88136×10^{-5}
Cl^-	5.43128	5.43090	5.24055	5.24592
SO_4^{2-}	0.182010	0.182363	0.176016	0.168900

[a] The number of significant figures, far in excess of representing a realistic error estimate, is reproduced here in their entirety to avoid rounding errors in further calculations.

The fraction of uranyl-organic complexes can be calculated as the ratio of terms that respectively describe the organic-ligand-bound U(VI) (equation 6), and the total U(VI) in solution (equation 7), as illustrated:

$$[UO_2Ac^+] + [UO_2Cit^-] + [UO_2EDTA^{2-}] + [UO_2Ox \ (aq)] \tag{6}$$

$$\begin{aligned}
&[UO_2Ac^+] + [UO_2Cit^-] + [UO_2EDTA^{2-}] + [UO_2Ox \ (aq)] + [UO_2^{2+}] + \\
&[UO_2OH^+] + [UO_2(OH)_2 \ (aq)] + [UO_2(CO_3) \ (aq)] + [UO_2(CO_3)_2^{2-}] + \\
&[UO_2(CO_3)_3^{4-}] + [UO_2Cl^+] + [UO_2Cl_2 \ (aq)] + [UO_2SO_4 \ (aq)] + \\
&[UO_2(SO_4)_2^{2-}] + [UO_2(SO_4)_3^{4-}]
\end{aligned} \tag{7}$$

Substituting the definitions for the terms described in equation 4 into equations 6 and 7, and rearranging yields a format that describes the organic-ligand bound U(VI) (equation 8) and the total U(VI) concentration (equation 9):

$$[UO_2^{2+}] \times \left(\begin{array}{l} K_{UO_2Ac^+} \times [Ac^-] + K_{UO_2Cit^-} \times [Cit^{3-}] + K_{UO_2EDTA^{2-}} \times [EDTA^{4-}] + \\ K_{UO_2Ox(aq)} \times [Ox^{2-}] \end{array} \right) \tag{8}$$

$$[\text{UO}_2^{2+}] \times \begin{pmatrix} 1 + \text{K}_{\text{UO}_2\text{Ac}^+} \times [\text{Ac}^-] + \text{K}_{\text{UO}_2\text{Cit}^-} \times [\text{Cit}^{3-}] + \text{K}_{\text{UO}_2\text{EDTA}^{2-}} \times [\text{EDTA}^{4-}] + \\ \text{K}_{\text{UO}_2\text{Ox(aq)}} \times [\text{Ox}^{2-}] + \text{K}_{\text{UO}_2\text{OH}^+} \times [\text{OH}^-] + \text{K}_{\text{UO}_2(\text{OH})_2(\text{aq})} \times [\text{OH}^-]^2 + \\ \text{K}_{\text{UO}_2\text{CO}_3(\text{aq})} \times [\text{CO}_3^{2-}] + \text{K}_{\text{UO}_2(\text{CO}_3)_2^{2-}} \times [\text{CO}_3^{2-}]^2 + \text{K}_{\text{UO}_2(\text{CO}_3)_3^{4-}} \times [\text{CO}_3^{2-}]^3 + \\ \text{K}_{\text{UO}_2\text{Cl}^+} \times [\text{Cl}^-] + \text{K}_{\text{UO}_2(\text{Cl})_2(\text{aq})} \times [\text{Cl}^-]^2 + \text{K}_{\text{UO}_2\text{SO}_4(\text{aq})} \times [\text{SO}_4^{2-}] + \\ \text{K}_{\text{UO}_2(\text{SO}_4)_2^{2-}} \times [\text{SO}_4^{2-}]^2 + \text{K}_{\text{UO}_2(\text{SO}_4)_3^{4-}} \times [\text{SO}_4^{2-}]^3 \end{pmatrix} \quad (9)$$

Results

The fractions of uranyl bound to the organic ligands acetate, citrate, EDTA, and oxalate in the conditions described by the FMT Runs 12, 18, 22, and 28 are presented in Table 8. The fraction of urany-organic complexes varies from 0.00 % in the case of Run 28 to 0.06 % in the case of Run 22. These results show that the impact of acetate, citrate, EDTA, and oxalate on the solubility of uranyl is insignificant under WIPP-relevant conditions.

Table 8: Uranyl bound to organic ligands in systems described by FMT Runs 12, 18, 22, and 28

	UO_2^{2+} bound to organic ligands (%)
Run 12: GWB, nonmicrobial vector, f_{CO_2} buffer = brucite-calcite	0.01
Run 18: GWB, microbial vector, f_{CO_2} buffer = brucite-hydromagnesite	0.01
Run 22: ERDA–6, nonmicrobial vector, f_{CO_2} buffer = brucite-calcite	0.06
Run 28: ERDA–6, microbial vector, f_{CO_2} buffer = brucite-hydromagnesite	0.00

Note on the Validity of our Calculations

Appendix SOTERM in the CCA stated that Ni^{2+} ions that would be released to the brines due to corrosion and dissolution of steels would be capable of binding 99.8% of the EDTA in the WIPP inventory; therefore, it was not necessary to consider the potential impact of organic ligands upon the actinide speciation and solubility. However, actinide ions are fully capable of competing with Ni^{2+}, and other ions for binding sites on organic ligands, and the high charge density on the actinide ions can allow them to act as very effective competitors for binding sites on organic ligands, effectively replacing other ions in the organic ligand complexes. Attachment SOTERM in the CRA considers the effect of organic ligands on the solubility of the +III, +IV, and +V actinide ions.

The assessment of the impact of organic ligands upon An(VI) speciation in this analysis uses the free ligand concentrations that result from FMT modeling calculations that were carried out to determine the speciation of the +III, +IV, and +V actinides. In essence, the use of free ligand concentrations in the analysis of impact upon An(VI) speciation parallels the argument put forth in Appendix SOTERM of the CCA, namely that only free ligand concentrations need to be considered, as opposed to total ligand

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concentrations. It is the goal of this section to demonstrate that the generalization is appropriate for U(VI) under the conditions expected to exist within the WIPP.

In order for the use for free ligand concentrations to be appropriate in the analysis presented in this paper, it is necessary to demonstrate that the An(VI) ions cannot effectively compete for ligand binding sites with the metal ions that account for the bulk of bound ligand concentration, i.e. Mg^{2+} in the present case. This can be shown by demonstrating that the ratio of An(VI)–ligand complex concentration to Mg^{2+}–ligand concentration is a very small number, i.e. the ability of An(VI) to displace Mg^{2+} is very small.

The following illustration uses UO_2^{2+} as the representative for the +VI actinides. The calculations will be carried out using stability constant values for zero ionic strength. The stability constants of formation of only 1:1 complexes of a ligand, L, with either UO_2^{2+} or Mg^{2+} are defined as:

$$\beta_{UO_2^{2+}} = \frac{[UO_2 L^{2-n}]}{[UO_2^{2+}][L^{n-}]} \tag{10}$$

$$\beta_{Mg^{2+}} = \frac{[MgL^{2-n}]}{[Mg^{2+}][L^{n-}]} \tag{11}$$

Dividing the $\beta_{UO_2^{2+}}$ by $\beta_{Mg^{2+}}$ and rearranging gives the following ratio:

$$\frac{\beta_{UO_2^{2+}}[UO_2^{2+}]}{\beta_{Mg^{2+}}[Mg^{2+}]} = \frac{[UO_2 L^{2-n}]}{[MgL^{2-n}]} \tag{12}$$

The values for the stability constants of Mg^{2+} with acetate, citrate, oxalate, and EDTA are given in Table 8. These data were calculated using the FMT database (Brush and Xiong, 2003a) and equation (5)

Table 8. Stability Constants of Mg^{2+} with Organic Ligands

Equilibrium	Log K^0
$Mg^{2+} + Ac^- \rightleftharpoons MgAc^+$	1.11
$Mg^{2+} + Cit^{3-} \rightleftharpoons MgCit^-$	5.30
$Mg^{2+} + EDTA^{4-} \rightleftharpoons MgEDTA^{2-}$	10.13
$Mg^{2+} + Ox^{2-} \rightleftharpoons UO_2Ox(aq)$	3.79

For simplicity, it is assumed that $UO_2(OH)_2(s)$ is the solubility-limiting phase. The concentration of UO_2^{2+} at pH = 9 is calculated from the solubility product constant for the reaction

$$UO_2(OH)_2(s) \rightleftharpoons UO_2^{2+} + 2OH^- \tag{13}$$

The value of log K_{SP} = -23.06 at 25 °C and zero ionic strength (see Table 6) was used to calculate the concentration of free uranyl ion. At pH = 9 the concentration of UO_2^{2+} is about 10^{-13} M. The values for

[Mg^{2+}] are taken from FMT Runs 12, 18, 22, and 28 (Brush and Xiong, 2003a), and are provided in Table 9.

Table 9. Concentrations of Mg^{2+} and Mg Complexes

Species	Run 12 Concentration (M)	Run 18 Concentration (M)	Run 22 Concentration (M)	Run 28 Concentration (M)
Mg^{2+}	0.49	0.49	0.11	0.097

Using the data in Tables 6 and 9 and in equation (12) one obtains the ratios given in Table 10.

Table 10. Ratios of UO_2^{2+}–Organic Ligand Complexes to Mg^+–Organic Ligand Complexes

Ligand	[UO$_2$L]/[MgL] Ratio Run 12	[UO$_2$L]/[MgL] Ratio Run 18	[UO$_2$L]/[MgL] Ratio Run 22	[UO$_2$L]/[MgL] Ratio Run 28
acetate	1.7×10^{-11}	1.7×10^{-11}	7.4×10^{-11}	8.5×10^{-11}
citrate	8.9×10^{-10}	8.9×10^{-10}	3.9×10^{-9}	4.5×10^{-9}
oxalate	5.0×10^{-10}	5.0×10^{-10}	2.2×10^{-9}	2.5×10^{-9}
EDTA	2.3×10^{-10}	2.3×10^{-10}	1.0×10^{-9}	1.2×10^{-9}

The small values for the ratios of UO_2^{2+} complex concentrations to Mg^{2+} complex concentrations demonstrate that under the conditions of the model, UO_2^{2+} will not be able to displace an appreciable amount of Mg from any of the complexes. This result is primarily driven by the confluence of two conditions, i.e. the abundance of Mg^{2+} and the low concentration of free UO_2^{2+}, which arises from the hydrolysis and precipitation of UO_2^{2+} at pH = 9. It should be emphasized that the ratios given in Table 10 are defined for a specific set of conditions. A change in pH or Mg^{2+} concentration would lead to a change in the ratio, and consequently a change in the conclusions drawn from the model. Additionally, the present illustration can be regarded only as an approximation since the calculations were carried out for a system at zero ionic strength, yet the concentrations of Mg were drawn from FMT Runs meant to model a high-ionic-strength solution. Nevertheless, the approximation makes a clear point that UO_2^{2+} will not effectively compete with Mg^{2+} for organic ligand binding sites under the conditions of this example. Consequently, using the free ligand concentrations rather than total ligand concentrations makes an acceptable approximation for the purpose of illustrating that acetate, citrate, oxalate, and EDTA will not significantly increase the solubility of U(VI) in the WIPP.

Conclusions

Organic ligands do not significantly impact solubility of U(VI) under WIPP-relevant conditions, for the following reasons:

- There is extensive hydrolysis of U(VI) at pH = 9
- acetate, citrate, oxalate and EDTA are unable to compete with OH⁻ under the conditions described in the FMT Runs
- U(VI) does not displace Mg^{2+} from organic ligand complexes to an appreciable extent under the defined conditions

It is important to note that the above arguments apply only to U(VI) under the conditions defined by the FMT calculations. For example, Np(V) does not undergo extensive hydrolysis at pH = 9, and will exhibit behavior quite different than that of U(VI).

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