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Sandia National Laboratories
Operated for the U.S. Department of Energy by
Sandia Corporation

4100 National Parks Highway
Carlsbad, NM 88220

Phone: (575) 234-0056
Fax: (575) 234-0061
Internet: psdomsk@sandia.gov

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to: SNL WIPP Records Center
Defense Waste Management Programs

from: Paul S. Domski Shelly C. Nielsen for

subject: Total Actinide Concentrations as a function of Hydromagnesite5424 Log K

The dissolution of hydromagnesite5424 follows the reaction:

$$Mg_5(CO_3)_4(OH)_2.4H_2O + 6H^+ = 5Mg^{2+} + 3HCO_3^- + 5H_2O$$

The value of logK in DATA0.FM1 (Xiong 2011) for the dissolution of hydromagnesite5424 is 32.2529. Xiong (2010 and 2015) calculated the log K for hydromagnesite5424 to be 29.3328, the value which would be included in the DATA0.FM2 database (Domski and Xiong, 2015).

The CRA-2014 baseline solubility model (Brush and Domski, 2013) was executed in EQ3/6 using the DATA0.FM1 thermodynamic database. To examine the effect of changing the log K of hydromagnesite5424 a "one off" calculation was performed where the DATA0.FM1 database was copied to DATA0.HYD and the log K hydromagnesite5424 was changed to the new value of 29.3328. Using the DATA0.HYD database the 1x brine volume runs were executed for the ERDA-6 and GWB brines. The results of the CRA-2014 and the one-off calculation are shown in Table 1.

Table 1.

Database	Brine	FCO_2	TIC(M)	Am(M)	Th(M)	Np(M)
FM1	GWB	3.14E-06	3.79E-04	2.59E-06	6.05E-08	2.77E-07
HYD	GWB	5.83E-07	7.04E-05	2.59E-06	4.83E-08	1.14E-06
FM1	ERDA-6	3.14E-06	4.54E-04	1.48E-06	7.02E-08	8.76E-07
HYD	ERDA-6	5.83E-07	8.53E-05	1.66E-06	5.18E-08	3.04E-06

Table 1 shows that first order effect of decreasing hydromagnesite solubility by using the DATA0.HYD database is a decrease in total inorganic carbon (TIC), which is also reflected in the observed decrease in the fugacity of carbon dioxide for both brines. This decrease in TIC impacts both the thorium and neptunium solubility by changing the carbonate speciation of these elements. In the case of thorium, the total solubility decreases slightly due to the decrease in the primary thorium carbonate species, $Th(OH)_3(CO_3)^-$.

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The americium solubility is unchanged for the GWB brine, but is slightly increased in the one-off calculation for the ERDA-6 brine. The reason for this very slight increase is not clear, but is related to the increased activity of EDTA with decreasing carbonate, and the fact that magnesium is depleted in ERDA-6 which acts to slightly increase the activity of the $AmEDTA^-$ complex.

While for neptunium, its solubility increases slightly with decreasing carbonate due to NpO_2Acetate(aq) and NpO_2^+ becoming dominant over the carbonate complex, $NpO_2CO_3^-$.

In conclusion, the effect of decreasing the solubility of hydromagnesite5424 is a decrease in the total inorganic carbon of the system. Carbonate forms complexes with both thorium and neptunium and both of their solubilities were impacted, a slight decrease for thorium and a slight increase for neptunium. Americium solubility was only impacted for the ERDA-6 brine due to the interplay of the Mg-EDTA-carbonate system.

References

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