

# Savannah River F-Area Seepage Basins

## Geochemical system description

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Table 1. Aqueous complexes proposed for the ASCEM-F-Area [Dong et al., 2012].

Reaction	$\log_{10} K$ (25° C)	Ref.
$\text{OH}^- \leftrightarrow \text{H}_2\text{O} - \text{H}^+$	14	(1)
$\text{AlOH}^{2+} \leftrightarrow \text{Al}^{3+} + \text{H}_2\text{O} - \text{H}^+$	5	(1)
$\text{Al}(\text{OH})_2^+ \leftrightarrow \text{Al}^{3+} + 2\text{H}_2\text{O} - 2\text{H}^+$	10.1	(1)
$\text{Al}(\text{OH})_3(\text{aq}) \leftrightarrow \text{Al}^{3+} + 3\text{H}_2\text{O} - 3\text{H}^+$	16.9	(1)
$\text{Al}(\text{OH})_4^- \leftrightarrow \text{Al}^{3+} + 4\text{H}_2\text{O} - 4\text{H}^+$	22.7	(1)
$\text{CaOH}^+ \leftrightarrow \text{Ca}^{2+} + \text{H}_2\text{O} - \text{H}^+$	12.78	(1)
$\text{CaHCO}_3^+ \leftrightarrow \text{HCO}_3^- + \text{Ca}^{2+}$	-1.106	(1)
$\text{CaCO}_3(\text{aq}) \leftrightarrow \text{HCO}_3^- + \text{Ca}^{2+} - \text{H}^+$	7.105	(1)
$\text{CO}_3^{2-} \leftrightarrow \text{HCO}_3^- - \text{H}^+$	10.329	(1)
$\text{CO}_2(\text{aq}) \leftrightarrow \text{HCO}_3^- - \text{H}_2\text{O} + \text{H}^+$	-6.352	(1)
$\text{NaCO}_3^- \leftrightarrow \text{Na}^+ + \text{HCO}_3^- - \text{H}^+$	10.579	(1)
$\text{NaHCO}_3(\text{aq}) \leftrightarrow \text{Na}^+ + \text{HCO}_3^-$	-0.1541	(1)
$\text{NaOH}(\text{aq}) \leftrightarrow \text{Na}^+ + \text{H}_2\text{O} - \text{H}^+$	14.18	(1)
$\text{MgCO}_3(\text{aq}) \leftrightarrow \text{HCO}_3^- + \text{Mg}^{2+} - \text{H}^+$	7.349	(1)
$\text{Mg}(\text{OH})^+ \leftrightarrow \text{Mg}^{+2} + \text{H}_2\text{O} - \text{H}^+$	11.44	(1)
$\text{MgHCO}_3^+ \leftrightarrow \text{Mg}^{+2} + \text{HCO}_3^-$	-1.07	(1)
$(\text{UO}_2)_2(\text{OH})_2^{2+} \leftrightarrow 2\text{UO}_2^{2+} + 2\text{H}_2\text{O} - 2\text{H}^+$	5.62	(1)
$(\text{UO}_2)_2\text{CO}_3(\text{OH})_3^- \leftrightarrow \text{HCO}_3^- - 4\text{H}^+ + 2\text{UO}_2^{2+} + 3\text{H}_2\text{O}$	11.184	(1)
$(\text{UO}_2)_2\text{OH}^{3+} \leftrightarrow 2\text{UO}_2^{2+} + \text{H}_2\text{O} - \text{H}^+$	2.7	(1)
$(\text{UO}_2)_3(\text{CO}_3)_6^{6-} \leftrightarrow 6\text{HCO}_3^- - 6\text{H}^+ + 3\text{UO}_2^{2+}$	7.973	(1)
$(\text{UO}_2)_3(\text{OH})_4^{2+} \leftrightarrow 3\text{UO}_2^{2+} + 4\text{H}_2\text{O} - 4\text{H}^+$	11.9	(1)
$\text{UO}_2(\text{OH})_4^{2-} \leftrightarrow \text{UO}_2^{2+} + 4\text{H}_2\text{O} - 4\text{H}^+$	32.4	(1)
$(\text{UO}_2)_3(\text{OH})_5^+ \leftrightarrow 3\text{UO}_2^{2+} + 5\text{H}_2\text{O} - 5\text{H}^+$	15.55	(1)

$(\text{UO}_2)_3(\text{OH})_7^- \leftrightarrow 3\text{UO}_2^{2+} + 7\text{H}_2\text{O} - 7\text{H}^+$	32.2	(1)
$(\text{UO}_2)_3\text{O}(\text{OH})_2(\text{HCO}_3)^+ \leftrightarrow \text{HCO}_3^- - 4\text{H}^+ + 3\text{UO}_2^{2+} + 3\text{H}_2\text{O}$	9.68	(1)
$(\text{UO}_2)_4(\text{OH})_7^+ \leftrightarrow 4\text{UO}_2^{2+} + 7\text{H}_2\text{O} - 7\text{H}^+$	21.9	(1)
$\text{UO}_2\text{NO}_3^+ \leftrightarrow \text{UO}_2^{2+} + \text{NO}_3^-$	-0.3	(1)
$\text{UO}_2(\text{OH})^+ \leftrightarrow \text{UO}_2^{2+} + \text{H}_2\text{O}$	5.25	(1)
$\text{UO}_2(\text{OH})_2(\text{aq}) \leftrightarrow \text{UO}_2^{2+} + 2\text{H}_2\text{O} - 2\text{H}^+$	12.15	(1)
$\text{UO}_2(\text{OH})_3^- \leftrightarrow \text{UO}_2^{2+} + 3\text{H}_2\text{O} - 3\text{H}^+$	20.25	(1)
$\text{UO}_2\text{CO}_3(\text{aq}) \leftrightarrow \text{UO}_2^{2+} + \text{HCO}_3^- - \text{H}^+$	0.389	(1)
$\text{UO}_2(\text{CO}_3)_2^{2-} \leftrightarrow \text{UO}_2^{2+} + 2\text{HCO}_3^- - 2\text{H}^+$	4.048	(1)
$\text{UO}_2(\text{CO}_3)_3^{4-} \leftrightarrow \text{UO}_2^{2+} + 3\text{HCO}_3^- - 3\text{H}^+$	9.141	(1)
$\text{CaUO}_2(\text{CO}_3)_3^{2-} \leftrightarrow \text{Ca}^{+2} + \text{UO}_2^{2+} + 3\text{HCO}_3^- - 3\text{H}^+$	3.806	(1)
$\text{Ca}_2\text{UO}_2(\text{CO}_3)_3(\text{aq}) \leftrightarrow 2\text{Ca}^{+2} + \text{UO}_2^{2+} + 3\text{HCO}_3^- - 3\text{H}^+$	0.286	(1)
$\text{MgUO}_2(\text{CO}_3)_3^{2-} \leftrightarrow \text{Mg}^{+2} + \text{UO}_2^{2+} + 3\text{HCO}_3^- - 3\text{H}^+$	5.186	(1)
$\text{UO}_2\text{SiO}(\text{OH})_3^+ \leftrightarrow \text{SiO}_2(\text{aq}) + \text{UO}_2^{2+} + 2\text{H}_2\text{O} - \text{H}^+$	2.481	(1)

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<sup>(1)</sup>Dong et al. [2012].

Table 2. Surface complexation and cation-exchange reactions proposed for the ASCEM-F-Area.

Reaction	$\log_{10} K$ (25° C)	Ref.
<sup>(1)</sup> On Kaolinite		
$(> k - OH)_2 UO_2^+ \leftrightarrow 2 > k - OH^{-0.5} + UO_2^{2+}$	-5.3	(5)
$(> k - OH)_2 UO_2CO_3^- \leftrightarrow 2 > k - OH^{-0.5} + UO_2^{2+} + HCO_3^- - H^+$	-6.2	(5)
$> k - OH_2^{+0.5} \leftrightarrow > k - OH^{-0.5} + H^+$	-4.9	(5)
$> k - OHNa^{+0.5} \leftrightarrow > k - OH^{-0.5} + Na^+$	2.1	(5)
$> k - OH_2NO_3^{-0.5} \leftrightarrow > k - OH^{-0.5} + H^+ + NO_3^-$	-4.9	(5)
<sup>(3),(8)</sup> On Kaolinite		
$k_2UO_2 \leftrightarrow 2k^- + UO_2^{2+}$	-7.1	(5)
$kNa \leftrightarrow k^- + Na^+$	-2.9	(5)
$kH \leftrightarrow k^- + H^+$	-4.5	(5)
$k_2Ca \leftrightarrow 2k^- + Ca^{2+}$	-6.8	(5)
$k_3Al \leftrightarrow 3k^- + Al^{3+}$	-8	(5)
<sup>(3)</sup> On Goethite		
$(> Fe - OH)_2 UO_2^+ \leftrightarrow 2 > Fe - OH^{-0.5} + UO_2^{2+}$	-14.11	(5)
$(> Fe - OH)_2 UO_2CO_3^- \leftrightarrow 2 > Fe - OH^{-0.5} + UO_2^{2+} + HCO_3^- - H^+$	-4.35	(5)
$> Fe - OH_2^{+0.5} \leftrightarrow > Fe - OH^{-0.5} + H^+$	-9.18	(5)
$(> Fe - OH)_2 CO_2^- \leftrightarrow 2 > Fe - OH^{-0.5} + HCO_3^- - 2H_2O + H^+$	-12.23	(5)
$> Fe - OCO_2Na^{-0.5} \leftrightarrow > Fe - OH^{-0.5} + HCO_3^- - H_2O$	-3.28	(5)
<sup>(4)</sup> On Quartz		
$> qz - OH_2^+ \leftrightarrow > qz - OH + H^+$	1.1	(6)
$> qz - O^- \leftrightarrow > qz - OH - H^+$	8.1	(6)
$> qz - ONa \leftrightarrow > qz - OH - H^+ + Na^+$	6.8	(7)

<sup>(1)</sup>Diffuse-Layer, Gouy-Chapman, edge site density 2.3 [sites nm<sup>-2</sup>] [Heidmann et al., 2005; Dong et al., 2012]. Activity of the surface complexes are evaluated as molar fractions in the action mass law.

<sup>(2)</sup>Cation-exchange, Gaines-Thomas convention, exchange site density 0.28 [sites nm<sup>-2</sup>] [Heidmann et al., 2005; Dong et al., 2012].

<sup>(3)</sup>Diffuse-Layer, Gouy-Chapman, edge site density 3 [sites nm<sup>-2</sup>] [Sherman et al., 2008; Dong et al., 2012]. Activity of the surface complexes are evaluated as molar fractions in the action mass law.

<sup>(4)</sup>Diffuse-Layer, Gouy-Chapman, site density 10 [sites nm<sup>-2</sup>] [Landry et al., 2009].

<sup>(5)</sup>Dong et al. [2012].

<sup>(6)</sup>Sverjensky and Sahai [1996].

<sup>(7)</sup>Landry et al. [2009].

<sup>(8)</sup>Similar sorption behavior for U(VI) was obtained using a surface complexation model (SCM) without electrostatic correction and the activity of the surface complexes evaluated as molar fractions (in a similar way as was proposed by Heidmann et al., 2005).

Table 3. Mineral dissolution/precipitation reactions proposed for the ASCEM-F-Area.

	Reaction	$\log_{10} K (25^\circ \text{C})$	Ref.
Primary minerals	Quartz $\leftrightarrow \text{SiO}_2(\text{aq})$	-3.7501	(1)
	Kaolinite $\leftrightarrow 2\text{Al}^{+3} + 2\text{SiO}_2(\text{aq}) + 5\text{H}_2\text{O} - 6\text{H}^+$	7.57	(2)
	Goethite $\leftrightarrow \text{Fe}^{+3} + 2\text{H}_2\text{O} - 3\text{H}^+$	0.1758	
Secondary minerals	Schoepite $\leftrightarrow \text{UO}_2^{+2} + 3\text{H}_2\text{O} - 2\text{H}^+$	4.8443	(1)
	Gibbsite $\leftrightarrow \text{Al}^{+3} + 3\text{H}_2\text{O} - 3\text{H}^+$	7.738	(3)
	Jurbanite $\leftrightarrow \text{Al}^{+3} + \text{SO}_4^{-2} + 6\text{H}_2\text{O} - \text{H}^+$	-3.8	(4)
	Basaluminite $\leftrightarrow 4\text{Al}^{+3} + \text{SO}_4^{-2} + 15\text{H}_2\text{O} - 10\text{H}^+$	22.251	(4)
	Opal $\leftrightarrow \text{SiO}_2(\text{aq})$	-3.005	(5)

<sup>(1)</sup>USDOE [2007].

<sup>(2)</sup>Yang and Steefel [2008].

<sup>(3)</sup>Pokrovskii and Helgeson [1995].

<sup>(4)</sup>Nordstrom [1982].

<sup>(5)</sup>Sonnenthal and Spycher [2000].

Table 4. Kinetic parameters proposed for the ASCEM-F-Area.

<sup>(1)</sup> Mechanism	Neutral	Acid	Base				
Mineral	kn <sub>i</sub>	ka <sub>i</sub>	p	kb <sub>i</sub>	q	η	θ
<sup>(2),(5)</sup> Quartz	10 <sup>-13.345</sup>	-	-	-	-	1	1
<sup>(3)</sup> Kaolinite (diss.)	10 <sup>-12.967</sup>	10 <sup>-11.098</sup>	0.777	10 <sup>-16.839</sup>	-0.472	1	0.5
<sup>(3)</sup> Kaolinite (prec.)	10 <sup>-14.126</sup>	10 <sup>-12.256</sup>	0.777	10 <sup>-17.996</sup>	-0.472	1	0.5
<sup>(2)</sup> Goethite	10 <sup>-7.94</sup>	-	-	-	-	1	1
Schoepite	Equilibrium						
<sup>(2)</sup> Gibbsite	10 <sup>-11.5</sup>	10 <sup>-7.65</sup>	0.992	10 <sup>-16.65</sup>	-0.784	1	1
<sup>(4)</sup> Jurbanite	10 <sup>-8</sup>	-	-	-	-	1	1
<sup>(4)</sup> Basaluminite	10 <sup>-8</sup>	-	-	-	-	1	1
<sup>(4)</sup> Opal (diss.)	10 <sup>-12.135</sup>	-	-	-	-	1	1
<sup>(4)</sup> Opal (prec.)	10 <sup>-9.135</sup>	-	-	-	-	1	1

<sup>(1)</sup>Computed as  $R_i = \left[ kn_i + ka_i a_{H^+}^p + kb_i a_{H^+}^q \right] (1 - \Omega^\eta)^\theta$  [mol m<sup>-2</sup> s<sup>-1</sup>].

<sup>(2)</sup>Palandri and Karaka [2004].

<sup>(3)</sup>Fitted from Yang and Steefel [2008].

<sup>(4)</sup>Estimated.

<sup>(5)</sup>Tester et al. [1994].

Table 5. Initial mineral volumetric fraction proposed for the ASCEM-F-Area.

Mineral	wt% [-]	vol. frac. [-]	surface area [m <sup>2</sup> g <sup>-1</sup> ]	density [g cm <sup>-3</sup> ]
Quartz	<sup>(1)</sup> 87	0.88	0.14	2.648
Kaolinite	<sup>(1)</sup> 10.48	0.11	<sup>(1)</sup> 20.71	2.594
Goethite	<sup>(1)</sup> 2.524	0.016	<sup>(1)</sup> 16.22	4.268
Schoepite	0	0	equilibrium	4.874
Gibbsite	0	0	120	2.44
Basaluminite	0	0	1	2.119
Opal	0	0	200	2.072
Jurbanite	0	0	1	1.789

<sup>(1)</sup>Dong et al. [2012].

Table 6. Chemical composition for the background and seepage solution proposed for the ASCEM-F-Area.

Component	Solution 1	<sup>(1)</sup> Solution 2	Solution 3	Units
pH	5.4	2.05	5.4	[-]
Na	2.78x10 <sup>-4</sup>	6.82x10 <sup>-5</sup>	2.78x10 <sup>-4</sup>	[mol kgw <sup>-1</sup> ]
Cl	9.98x10 <sup>-3</sup>	3.39x10 <sup>-5</sup>	9.98x10 <sup>-3</sup>	[mol kgw <sup>-1</sup> ]
<sup>(3)</sup> TIC	1.23x10 <sup>-5</sup>	1.09x10 <sup>-5</sup>	1.23x10 <sup>-5</sup>	[mol kgw <sup>-1</sup> ]
Al	3.09x10 <sup>-8</sup>	10 <sup>-8</sup>	3.09x10 <sup>-8</sup>	[mol kgw <sup>-1</sup> ]
Fe(III)	2.92x10 <sup>-16</sup>	2.75x10 <sup>-6</sup>	2.92x10 <sup>-16</sup>	[mol kgw <sup>-1</sup> ]
K	3.32x10 <sup>-5</sup>	1.72x10 <sup>-6</sup>	3.32x10 <sup>-5</sup>	[mol kgw <sup>-1</sup> ]
Ca	10 <sup>-5</sup>	10 <sup>-5</sup>	10 <sup>-5</sup>	[mol kgw <sup>-1</sup> ]
Mg	5.35x10 <sup>-3</sup>	2.47x10 <sup>-6</sup>	5.35x10 <sup>-3</sup>	[mol kgw <sup>-1</sup> ]
U(VI)	<sup>(2)</sup> 1.25x10 <sup>-10</sup>	3.01x10 <sup>-5</sup>	<sup>(2)</sup> 1.25x10 <sup>-10</sup>	[mol kgw <sup>-1</sup> ]
Nitrates	10 <sup>-3</sup>	10 <sup>-2</sup>	10 <sup>-3</sup>	[mol kgw <sup>-1</sup> ]
SO <sub>4</sub>	2.25x10 <sup>-5</sup>	4.8x10 <sup>-5</sup>	2.25x10 <sup>-5</sup>	[mol kgw <sup>-1</sup> ]
SiO <sub>2</sub> (aq)	1.77x10 <sup>-4</sup>	1.18x10 <sup>-4</sup>	1.77x10 <sup>-4</sup>	[mol kgw <sup>-1</sup> ]
<sup>3</sup> H	10 <sup>-15</sup>	2.17x10 <sup>-9</sup>	10 <sup>-15</sup>	[mol kgw <sup>-1</sup> ]
Ionic strength	1.64x10 <sup>-2</sup>	1.02x10 <sup>-2</sup>	1.64x10 <sup>-2</sup>	[mol kgw <sup>-1</sup> ]
P <sub>CO2(g)</sub>	10 <sup>-3.5</sup>	10 <sup>-3.5</sup>	10 <sup>-3.5</sup>	[atm]
Mineral saturation indices				
SI <sub>quartz</sub>	0	-0.17	0	[-]
SI <sub>kaolinite</sub>	0	-18.42	0	[-]
SI <sub>goethite</sub>	0	0	0	[-]
SI <sub>schoepite</sub>	-4.5	-5.46	-4.5	[-]
SI <sub>basaluminite</sub>	-7.63	-35.89	-7.63	[-]
SI <sub>gibbsite</sub>	-0.17	-9	-0.17	[-]
SI <sub>jurbanite</sub>	-4.29	-6.05	-4.29	[-]
SI <sub>opal</sub>	-0.74	-0.92	-0.74	[-]

<sup>(1)</sup>Based on Killian et al. [1987].

<sup>(2)</sup>Calculated for an U(VI) in the solid sediment of about 0.1 [μg g<sup>-1</sup>].

<sup>(3)</sup>Total Inorganic Carbon.

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