URANINITE SOLUBILITY IN HCl AQUEOUS SOLUTIONS AT 500°C, 10^2 MPa

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Solubility measurements are done by capsula technique under Ni/NiO buffer hydrogen fugacity. Uraninite crystals have been synthesized from U_3O_8 [1]. To obtain uraninite parameter a CAD4 Enraf-Nonius and STAD I STOE apparatus are used. The synthesized uraninite composition is $UO_{2,008}$.

Uraninite crystals and 1 ml of 0.001-2 m HCl solution have been placed into gold capsulas. They have been welded, weighted and put into stainless bomb filled with pure water. The bomb is heated up to 500°C. Calculated pressure is 10^5 PA. After 4-5 months heating the bomb is quenched in cooled water. Quenched capsulas are taken off, weighted and unsealed. One ml of concentrated HCl is added in each capsula, and solution is filtered. Concentrations of U(IV) and U(total) are determined by arsenaso-3 in 6 N HCl solution using КФК-3 photometer, 50 mm cuvee at λ=662,5 nm. Analytical procedure and standard curve are done according to [2]. ICP-MS Element-XR analysis id done to determine total uranium concentration. Both methods results are matched.

Obtained results of uraninite solubility are in accordance with published data at hydrothermal parameters [4-6]. Data on chemical element speciation in supercritical water solution [7] and the dependence of uraninite solubility on HCl^0 concentration shown on tab. and fig. below evidence on the following reactions: UO_2(uraninite) + HCl^0 = $U(OH)_3Cl^0$, UO_2(uraninite) + $2HCl^0$ = $U(OH)_2Cl_2^0$ and UO_2(uraninite) + $3HCl^0$ = $UOHCl_3^0$ + H_2O.

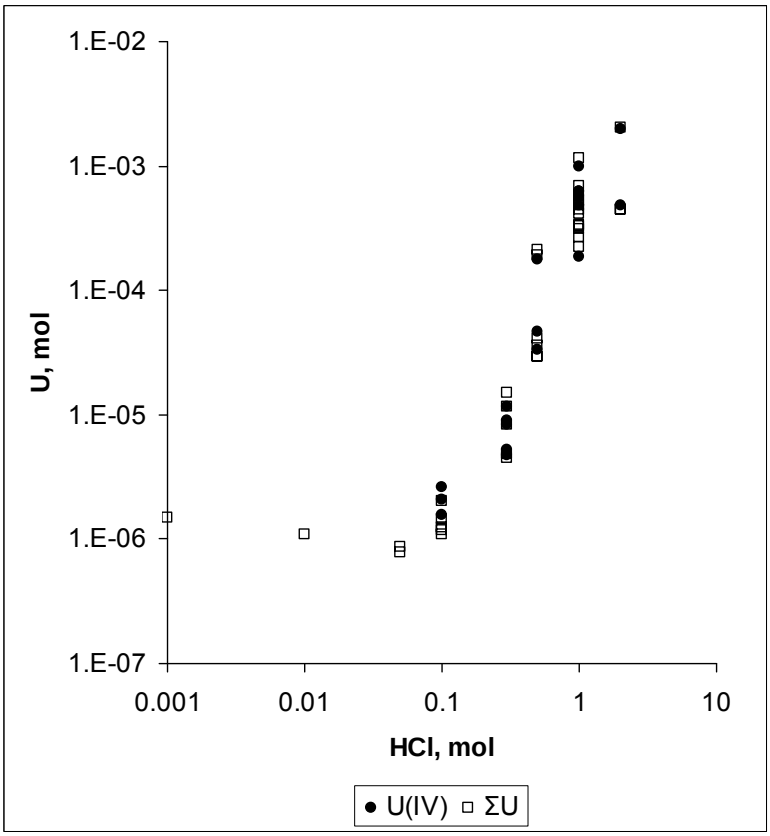

Fig. Solubility UO_2 in HCl aqueous solutions

Table

Solubility of UO_2(uraninite) in HCl solutions at 500°C, 1 kb.

№	Initial concentration of HCl, m	Photometric determination of uranium concentration, $m\times10^{-4}$		ICP-MS determination of uranium concentration, $m\times10^{-4}$
		U(IV)	U_Σ	U_Σ
3/13	0.001			0.0147
3/14	0.01			0.0108
2/1	0.05			0.0078
2/2	0.05			0.0086
1/7	0.1	0.021		
2/13+2/14	0.1	0.0156		
2/1+2/4	0.1	0.026		
2/3	0.1			0.0117
2/7	0.1			0.0123
2/8	0.1			0.011
2/13	0.1			0.0144
2/14	0.1			0.0204
1/2*	0.3		0.151	
1/14*	0.3	0.084		
2/5	0.3	0.052		0.0481
2/6	0.3	0.047		0.0451
2/9	0.3	0.09		0.0836
2/15	0.3	0.116		0.115
1/15*	0.5	1.75	2.1	1.91
2/10	0.5	0.465	0.413	0.295
2/16	0.5	0.33	0.36	0.295
3/16	0.5	0.292	0.255	0.235
3/19*	0.5	0.289	0.352	0.308
1/4*	1.0		4.80	
1/5*	1.0		6.99	
1/10*	1.0	5.70	5.25	3.28
1/11*	1.0	5.76	4.1	11.43
1/16	1.0	6.30	3.11	3.36
1/17	1.0	4.83	4.51	5.78
2/11*	1.0	6.23	52.4	58.7
2/12	1.0	1.85	2.67	2.21
2/17*	1.0	10	34	
13/17	1.0	1.08	1.27	1.14
1/18*	2.0	4.83	4.51	4.50
2/18*	2.0	20	20.5	
3/18	2.0	6.2	7.23	7.50

* Initial phase is U_3O_8(cr).

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Analysis of experimental solids by X-Ray study shows there is no new phases except uraninite. Investigation of uraninite crystals by binocular shows there are deposited gold scales on uraninite crystal surface and among uraninite crystals. We think the deposition of capsule gold on uraninite crystal surface results from following processes: gold oxidation by uranium $UO_{2+x}(cr) + 2Au(cr) + 2H^+ = UO_2(cr) + 2Au^+(aq) + H_2O$ and $Au^+(aq)$ reduction by hydrogen of Ni/NiO buffer $2Au^+ + H_2(g) = 2Au(cr) + 2H^+$.

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