Available online at www.sciencedirect.com

ScienceDirect

Procedia Chemistry 21 (2016) 87 – 92

Procedia Chemistry

5th International ATALANTE Conference on Nuclear Chemistry for Sustainable Fuel Cycles

Activity Coefficients of Uranyl Nitrate and Nitric Acid in Aqueous Mixtures

Alexander Ochkin *, Dmitry Gladilov, Sergey Nekhaevskiy, Alexey Merkushkin

D. Mendeleev University of Chemical Technology of Russia, 9 Miusskay Sqr., Moscow, 125047, Russia

Abstract

The activity coefficients of nitric acid and uranyl nitrate in in aqueous mixtures are necessary to model the system H_2O-HNO_3-$UO_2(NO_3)_2$-TBP-diluent. Three methods have been compared in this work to determine activity coefficients based on experimental data, Pitzer's equation or Zdanovskiy's rule. Acid activities have been calculated from the data of two first methods. These results were compared with the data of third method. Errors were about 3.3.%. Activity coefficients of uranyl nitrate γ_U as a function of concentration of uranyl nitrate and nitric acid were determined in 76 mixed solutions. The equation to calculate γ_U was proposed.

Peer-review under responsibility of the organizing committee of ATALANTE 2016

Keywords: PUREX; extraction; mathematical modeling;

1. Introduction

The nuclear fuel reprocessing is based on PUREX process. The organic solvent is usually composed of 30% tributyl phosphate (TBP) in a hydrocarbon diluents. Then the composition of the organic phase can be expressed as H_2O-HNO_3-$UO_2(NO_3)_2$-$Pu(NO_3)_4$-TBP-diluent. But because the concentration of plutonium is low the system can be considered a five-component one [1,2]. Activities of uranyl nitrate and nitric acid in mixtures should be determined to model this extracting system. The methods of the derivation of this data will be considered here.

* Ochkin A.. Tel.: +7-495-496-4557; fax: +7-495-944-1987.
E-mail address: ochkin@rctu.ru

Peer-review under responsibility of the organizing committee of ATALANTE 2016
doi:10.1016/j.proche.2016.10.013

2. Determination of activities from the experiments and Pitzer parameters

Table 1. Acid activities in mixed solutions of acid and uranyl nitrate

N	Concentrations[3], mole/L		Density[3], g/cm^3	Vapor pressure of HNO_3[3], mm Hg	HNO_3 activity				
	$UO_2(NO_3)_2$ c_U	HNO_3 c_a			a_{a3}[3]	a_{a4}[4]	a_{a5}[5]	Average a_{am}	
1	0.466	0.976	1.1782	$288\cdot10^{-6}$	**1.158**	1.359	1.543	1.353	1.451*
2	0.466	0.976	1.1782	$288\cdot10^{-6}$	**1.158**	1.359	1.543	1.353	1.451*
3	0.902	0.951	1.3176	$725\cdot10^{-6}$	2.914	2.926	3.433	3.091	
4	0.902	0.951	1.3176	$868\cdot10^{-6}$	3.489	2.926	3.433	3.283	
5	1.314	0.92	1.4467	0.00159	6.391	5.615	6.592	6.199	
6	1.314	0.92	1.4467	0.00174	6.994	5.615	6.592	6.400	
7	1.314	0.92	1.4467	0.00175	7.034	5.615	6.592	6.414	
8	0.451	2.195	1.2163	0.00232	9.325	8.613	8.963	8.967	
9	0.854	2.108	1.3426	0.00332	13.344	14.723	15.796	14.621	
10	0.854	2.108	1.3426	0.00392	15.756	14.723	15.796	15.425	
11	1.237	2.026	1.461	0.00724	29.100	23.798	25.804	26.234	
12	1.237	2.026	1.461	0.00739	29.703	23.798	25.804	26.435	
13	0.086	4.29	1.1651	0.01038	41.721	42.203	40.595	41.506	
14	0.213	4.248	1.2027	0.01214	48.795	48.674	47.026	48.165	
15	0.213	4.248	1.2027	0.0122	49.036	48.674	47.026	48.245	
16	0.418	4.182	1.2646	0.0156	62.702	60.772	59.486	60.987	
17	0.418	4.182	1.2646	0.0155	62.300	60.772	59.486	60.853	
18	0.418	4.182	1.2646	0.0158	63.505	60.772	59.486	61.254	
19	0.418	4.182	1.2646	0.016	64.309	60.772	59.486	61.522	
20	0.803	3.923	1.38	0.021	84.406	80.186	80.387	81.660	
21	0.803	3.923	1.38	0.0214	86.014	80.186	80.387	82.196	
22	0.808	4.039	1.3804	0.0213	85.612	80.186	90.435	85.411	
23	0.808	4.039	1.3804	0.0204	81.994	80.186	90.435	84.205	
24	1.16	3.799	1.485	0.0286	114.95	113.79	116.56	115.10	
25	1.16	3.799	1.485	0.0269	108.12	113.79	116.56	112.82	
26	1.166	3.885	1.4812	0.0348	139.87	126.57	129.42	131.95	
27	1.376	3.716	1.545	0.0379	152.33	139.75	143.89	145.32	
28	1.376	3.716	1.545	0.0339	136.25	139.75	143.89	139.96	
29	1.428	3.818	1.5668	0.0384	154.34	157.56	162.38	158.09	
30	1.428	3.818	1.5668	0.0385	154.74	157.56	162.38	158.23	
31	0.5331	4.981	1.3235	0.0358	143.9	129.78	127.81	133.83	
32	0.392	6.135	1.3136	0.0558	224.8	237.5	235.9	232.7	
33	0.0984	7.462	1.2621	0.0924	371.4	382.8	395.5	383.2	
34	0.2464	7.641	1.3148	0.1215	488.3	482.7	490.4	487.1	
35	0.374	7.135	1.3366	0.0953	383.0	418.8	422	407.9	
36	0.374	7.135	1.3366	0.0935	375.8	418.8	422	405.5	
37	0.4871	7.674	1.4663	0.163	655.2	623.4	619	632.5	
38	0.7437	7.776	1.4663	0.201	807.9	798.2	783.8	796.6	
39	0.9843	7.654	1.535	0.24	964.6	898.3	924.4	929.1	
40	0.2469	8.07	1.32	0.159	639.1	623.4	635.1	632.5	
41	0.5232	8.227	1.3969	0.232	932.5	889.1	858.2	893.3	
42	1.003	8.129	1.5492	0.287	1153.5	1133.0	1213.8	1166.8	
	δ				6.0	4.8	4.8		
	δ*				5.1	4.9	3.8		