
Activity Coefficients of Nitrate Uranyl and Nitric Acid in Mixed Solutions

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Abstract: Aqueous solutions are frequently used in chemical technology. So numerous reference books contain basic data on major properties of binary electrolyte solutions. Usually they include densities, molar or molal concentrations, activity coefficients, water activities and sometimes osmotic coefficients. Unfortunately, most solutions are mixed ones. Therefore, it is necessary to use data of binary solutions in order to calculate thermodynamic properties of mixed solutions. For example, the aqueous phase in extraction reprocessing of nuclear reactor fuel can be considered as mixed solution of nitric acid and uranyl nitrate in first approximation. Thus, in order to calculate equilibrium during extraction of uranium with TBP it is necessary to create calculations algorithm of activity coefficients of uranyl nitrate and nitric acid with different ratios of their concentrations. Usually the integration of Gibbs-Duhem equation with some conditions is used. The first ones to offer this approach were Mc Kay and Perring in 1953. Practical implementation shows that it is recommended to integrate the equation under isopiestic conditions, meaning under constant osmotic coefficients. Zdanovskiy's rule states, that the sum of ratios of molal concentrations in mixed solution to molal concentrations in binary solutions is equal to 1. Mikulin's equations for the systems that abide by Zdanovskiy's rule allow calculations of molal coefficients of activity for such systems. Molal activity coefficients of nitrate uranyl (0.1-2.0 mole/kg) and nitric acid (0-5.0 mole/kg) in mixed solutions have been calculated using Zdanovskiy's rule and Mikulin's equations. Analytical equations were found in order to calculate these values under different concentrations. Literature data of activity coefficients for binary solutions can be combined with calculation results from equations.

Keywords: Uranyl Nitrate, Nitric Acid, Mixed Solutions, Activity Coefficients, Extraction, TBP

1. Introduction

PUREX process is the key technology used for recycling of used fuel from nuclear reactors. The process is carried out by using extraction process with TBP from carbonic solvent [1]. From the thermodynamic point of view the organic phase can be seen as a five-component system $H_2O-HNO_3-UO_2(NO_3)_2-TBP-solvent$ [2, 3]. In order to calculate an equilibrium in this system it is necessary to determine activity coefficients of uranyl nitrate and nitric acid in mixed solutions. The first ones to consider this problem were McKey and Perring in [4]. They offered to use the cross-differential equation

$$(\partial \ln a_e / \partial m_w)_{m_e} = (\partial \ln a_w / \partial m_e)_{m_w} \quad (1)$$

where a and m are an activity and molality of a component in the aqueous phase, subscripts "e" and "w" are related to electrolyte and water accordingly. The equation (1) has been integrated in different ways repeatedly.

Nitric acid activities in mixed solution have been found through acid steam pressure [5]. This data base can be interpreted in different ways. For example, Pitzer's equations [6] have been used in [7]. The average errors of calculated acid activities [7] from experimental ones [6] has been equal to 9.5% [8, 9]. Then it is necessary to consider the interpretation the experimental data through Zdanovskii's rule and Mikulin's equations. The results of the calculation were given in [8, 9] and the average errors of calculated acid activities [8, 9] from experimental ones [6] has been equal to 7.3%. It means that the second method should be preferred. Therefore this procedure has been

considered in details in [10].

The aim of this research is to calculate activity coefficients of uranyl nitrate and nitric acid in mixed solutions for different molalities and propose equations which can be used to calculate activity coefficients in mixed solutions with different concentrations of acid and salt.

2. Methodology and Discussion

The densities of mixed solutions were given in [5]. This data have been transferred in the equation [10]

$$d = 0.99704 + 0.31664c_U + 0.03377c_a - 0.0004 c_a^{2.07} \quad (2)$$

where 0,99704 is a density of water at 25°C, c_U and c_a are molar concentrations of uranyl nitrate and nitric acid, d is a density of mixed solution, g/cm^3 .

Later all molar concentrations c (mole/l) in mixed solutions were calculated using following equation:

$$c_a = m_a / (63,013 \cdot V) \quad (3)$$

$$c_U = m_U / (238,03 \cdot V) \quad (4)$$

where m_a и m_U are masses (g) of nitric acid and uranium and V is a volume of mixture in liters.

Following equation was used to calculate molal concentrations:

$$m_i = c_i / (d - 0,001 \cdot \sum c_j M_j) \quad (5)$$

where c_i is a molar concentration of component "i", c_j и M_j are molar concentrations and molecular masses of all mixture components, except for solvent.

Small adjustments can be made in equation (1) for solution density in order to adjust them with concentrations of uranyl nitrate and other components of water solution according to Mikulin's equation [11] and Colon with colleagues article [12] in which use of molal volumes for calculation of concentrations in water phase is discussed.

Coefficients of activity of uranyl nitrate γ_U in binary solution are presented in table 1 in accordance with works [13-15]. The scale of molal concentrations was used in this table. In addition, activities of water a_w are presented for the aim of comparison. It is clearly shown in table 1, that the results of two works practically are identical. Data acquired by Vosnesenskaya [13] will be used this work in order to make use of software which allows use Mikulin's equations. Results of calculations are presented in row 8.

Table 1. Activity coefficients on uranyl nitrate γ_U (calculation).

N	m_U , mol/kg	γ_{U1} [13]	a_w [14]	γ_{U2} [15]	a_w [15]	$10^7 \cdot [(\gamma_{U1} - \gamma_{U2}) / \gamma_{U1}]^2$	Calculations results γ_U
1	0.100	0.543	0.9952	0.5363	0.99528	1523	0.5429
2	0.200	0.512	0.9903	0.5088	0.99029	391	0.5122
3	0.300	0.510	0.9850	0.5079	0.98496	170	0.5097
4	0.400	0.518	0.9794	0.5171	0.97931	30	0.5180
5	0.500	0.534	0.9734	0.5325	0.97334	79	0.5340
6	0.600	0.555	0.9670	0.5527	0.96702	172	0.5550
7	0.700	0.578	0.9605	0.5771	0.96035	24	0.5774
8	0.800	0.608	0.9533	0.6054	0.95332	183	0.6083
9	0.900	0.641	0.9459	0.6375	0.94593	298	0.6409
10	1.000	0.679	0.9380	0.6733	0.93819	705	0.6799
11	2.000	1.218	0.8475	1.2231	0.84683	175	1.2181
					Σ	3749	
					δ	0.58%	

Equation (4) was used to calculate intermediate concentrations:

$$\gamma_{Uc} = 1 - 1.20570 \cdot m_U^{0,288745} + 0,875093 \cdot m_U^{0,7603} + 0.0012527 \cdot m_U^{-0,55601} \quad (6)$$

where γ_{Uc} are calculated values of activity coefficients, m_U is a molal concentration of nitrate uranyl.

In paper [10] a calculation of activity coefficients of nitric acid and nitrate uranyl for 4 concentrations of the salt can be found. The calculation was performed using Zdanovskiy's rule [16]:

$$m_1 / m_1^* + m_2 / m_2^* = 1 \quad (7)$$

where m_1 and m_2 – molality of electrolytes 1 and 2 in mixed solution with water activity a_w , m_1^* and m_2^* – molality of

electrolytes 1 and 2 in their respective individual isopiestic solutions with the same water activity a_w . Mikulin's equations [11] were used to calculate coefficients of activity in previously mentioned solutions.

$$\gamma_1 = v_1 \cdot \gamma_1^* \cdot m_1^* / (v_1 \cdot m_1 + v_2 \cdot m_2) \quad (8)$$

$$\gamma_2 = v_2 \cdot \gamma_2^* \cdot m_2^* / (v_1 \cdot m_1 + v_2 \cdot m_2) \quad (9)$$

where γ_1 and γ_2 are coefficients of activity of electrolytes 1 and 2 in mixed solution with water activity a_w , γ_1^* and γ_2^* are their coefficients of activity in isopiestic solutions with the same water activity a_w .

Values of density of solutions, molality coefficients and water activity for mixed solutions of nitric acid and nitrate uranyl are given in table 2.

Table 2. Molality coefficients of activity of nitrate unranlyl γ_U , nitric acid γ_a solution density d and water activity a_w .

m_U , mol/kg	c_U , mol/L	c_a , mol/L	d , g/sm ³	a_w	γ_U	γ_a
			$m_a = 0.500$ mol/kg			
0.1	0.09752	0.488	1.0443	0.9779	0.5141	0.7184
0.2	0.1936	0.484	1.0746	0.9722	0.5246	0.7274
0.3	0.2882	0.480	1.1044	0.9662	0.5403	0.7427
0.4	0.3814	0.477	1.1338	0.9599	0.5590	0.7620
0.5	0.4732	0.473	1.1628	0.9531	0.5863	0.7846
0.6	0.5638	0.470	1.1913	0.9461	0.6153	0.8105
0.7	0.6530	0.466	1.2195	0.9387	0.6501	0.8394
0.8	0.7409	0.463	1.2472	0.9309	0.6869	0.8703
0.9	0.8275	0.460	1.2745	0.9229	0.7240	0.9021
1.0	0.9130	0.456	1.3015	0.9146	0.7651	0.9357
1.25	1.1213	0.449	1.3672	0.8930	0.8879	1.0264
1.50	1.3226	0.441	1.4306	0.8699	1.0279	1.1288
1.75	1.5170	0.433	1.4920	0.8456	1.1896	1.2416
2.00	1.7051	0.426	1.5513	0.8205	1.3716	1.3619
			$m_a = 1.000$ mol/kg			
0.1	0.0961	0.961	1.0596	0.9594	0.5414	0.7374
0.2	0.1908	0.954	1.0893	0.9530	0.5658	0.7569
0.3	0.2841	0.947	1.1186	0.9463	0.5918	0.7796
0.4	0.3760	0.940	1.1475	0.9392	0.6229	0.8055
0.5	0.4666	0.933	1.1760	0.9318	0.6568	0.8337
0.6	0.5559	0.927	1.2040	0.9241	0.6909	0.8631
0.7	0.6440	0.920	1.2317	0.9162	0.7286	0.8943
0.8	0.7308	0.913	1.2589	0.9080	0.7716	0.9276
0.9	0.8163	0.907	1.2858	0.8995	0.8194	0.9627
1.0	0.9007	0.901	1.3123	0.8908	0.8691	0.9994
1.25	1.1065	0.885	1.3770	0.8680	1.0059	1.1004
1.50	1.3054	0.870	1.4395	0.8440	1.1629	1.2098
1.75	1.4977	0.856	1.4999	0.8193	1.3410	1.3284
2.00	1.6837	0.842	1.5583	0.7953	1.5333	1.4472
			$m_a = 1.500$ mol/kg			
0.1	0.0947	1.421	1.0742	0.9397	0.5979	0.7744
0.2	0.1881	1.411	1.1034	0.9326	0.6292	0.8000
0.3	0.2801	1.400	1.1322	0.9253	0.6607	0.8272
0.4	0.3708	1.390	1.1606	0.9176	0.6952	0.8562
0.5	0.4601	1.380	1.1886	0.9097	0.7349	0.8875
0.6	0.5483	1.371	1.2162	0.9015	0.7798	0.9209
0.7	0.6351	1.361	1.2434	0.8930	0.8272	0.9562
0.8	0.7208	1.351	1.2702	0.8842	0.8771	0.9934
0.9	0.8053	1.342	1.2966	0.8753	0.9298	1.0325
1.0	0.8886	1.333	1.3227	0.8662	0.9852	1.0738
1.25	1.0919	1.310	1.3863	0.8425	1.1379	1.1801
1.50	1.2885	1.288	1.4479	0.8180	1.3123	1.2969
1.75	1.4786	1.267	1.5074	0.7942	1.5021	1.4144
2.00	1.6625	1.247	1.5649	0.7711	1.6952	1.5141
			$m_a = 2.000$ mol/kg			
0.1	0.0934	1.867	1.0882	0.9189	0.6647	0.8211
0.2	0.1854	1.854	1.1169	0.9113	0.7012	0.8504
0.3	0.2761	1.841	1.1452	0.9033	0.7432	0.8821
0.4	0.3656	1.828	1.1731	0.8951	0.7882	0.9159
0.5	0.4537	1.815	1.2006	0.8865	0.8357	0.9516
0.6	0.5407	1.802	1.2278	0.8778	0.8859	0.9891
0.7	0.6264	1.790	1.2545	0.8689	0.9386	1.0285
0.8	0.7110	1.777	1.2809	0.8598	0.9938	1.0688
0.9	0.7944	1.765	1.3069	0.8505	1.0522	1.1096
1.0	0.8766	1.753	1.3326	0.8411	1.1143	1.1521
1.25	1.0775	1.724	1.3952	0.8168	1.2854	1.2673
1.50	1.2718	1.696	1.4558	0.7931	1.4727	1.3834
1.75	1.4597	1.668	1.5144	0.7701	1.6634	1.4827
2.00	1.6417	1.642	1.5712	0.7479	1.8548	1.5881
			$m_a = 2.500$ mol/kg			
0.1	0.0920	2.301	1.1016	0.8971	0.7520	0.8781
0.2	0.1828	2.285	1.1299	0.8887	0.7972	0.9124
0.3	0.2722	2.269	1.1577	0.8802	0.8450	0.9485
0.4	0.3604	2.253	1.1851	0.8715	0.8951	0.9862
0.5	0.4474	2.237	1.2122	0.8626	0.9474	1.0251

m_U , mol/kg	c_U , mol/L	c_n , mol/L	d , g/sm ³	a_w	γ_U	γ_a
0.6	0.5332	2.222	1.2388	0.8536	1.0027	1.0642
0.7	0.6178	2.207	1.2651	0.8443	1.0613	1.1048
0.8	0.7013	2.192	1.2911	0.8348	1.1241	1.1475
0.9	0.7837	2.177	1.3167	0.8252	1.1906	1.1926
1.0	0.8649	2.162	1.3420	0.8156	1.2601	1.2395
1.25	1.0633	2.127	1.4036	0.7920	1.4450	1.3542
1.50	1.2553	2.092	1.4633	0.7691	1.6332	1.4529
1.75	1.4412	2.059	1.5211	0.7470	1.8229	1.5583
2.00	1.6212	2.026	1.5771	0.7259	2.0119	1.6566
$m_a = 3.000$ mol/kg						
0.1	0.0907	2.722	1.1145	0.8740	0.8546	0.9465
0.2	0.1802	2.703	1.1422	0.8653	0.9043	0.9839
0.3	0.2684	2.684	1.1696	0.8565	0.9565	1.0216
0.4	0.3554	2.666	1.1966	0.8475	1.0119	1.0603
0.5	0.4412	2.647	1.2232	0.8382	1.0713	1.1012
0.6	0.5259	2.629	1.2494	0.8287	1.1348	1.1445
0.7	0.6094	2.612	1.2753	0.8192	1.2016	1.1900
0.8	0.6918	2.594	1.3008	0.8096	1.2718	1.2366
0.9	0.7731	2.577	1.3260	0.8003	1.3445	1.2826
1.0	0.8533	2.560	1.3509	0.7910	1.4188	1.3265
1.25	1.0493	2.518	1.4116	0.7681	1.6045	1.4247
1.50	1.2391	2.478	1.4705	0.7461	1.7926	1.5300
1.75	1.4229	2.439	1.5274	0.7250	1.9809	1.6285
2.00	1.6009	2.401	1.5826	0.7051	2.1590	1.7169
$m_a = 3.500$ mol/kg						
0.1	0.0894	3.130	1.1268	0.8505	0.9659	1.0186
0.2	0.1776	3.109	1.1541	0.8415	1.0219	1.0575
0.3	0.2646	3.088	1.1810	0.8322	1.0823	1.0989
0.4	0.3505	3.067	1.2075	0.8227	1.1463	1.1428
0.5	0.4351	3.046	1.2337	0.8132	1.2138	1.1885
0.6	0.5187	3.026	1.2595	0.8038	1.2844	1.2344
0.7	0.6011	3.006	1.2850	0.7945	1.3573	1.2790
0.8	0.6825	2.986	1.3101	0.7853	1.4308	1.3207
0.9	0.7627	2.966	1.3349	0.7762	1.5040	1.3590
1.0	0.8420	2.947	1.3594	0.7672	1.5773	1.3979
1.25	1.0356	2.900	1.4193	0.7452	1.7638	1.5030
1.50	1.2231	2.854	1.4772	0.7241	1.9514	1.6017
1.75	1.4048	2.810	1.5334	0.7041	2.1296	1.6910
2.00	1.5810	2.767	1.5878	0.6855	2.2917	1.7696
$m_a = 4.000$ mol/kg						
0.1	0.0882	3.527	1.1386	0.8262	1.0980	1.0980
0.2	0.1751	3.503	1.1654	0.8167	1.1588	1.1424
0.3	0.2609	3.479	1.1919	0.8074	1.2271	1.1878
0.4	0.3456	3.456	1.2180	0.7981	1.2981	1.2327
0.5	0.4291	3.433	1.2437	0.7889	1.3706	1.2754
0.6	0.5116	3.411	1.2691	0.7798	1.4430	1.3148
0.7	0.5929	3.388	1.2942	0.7707	1.5151	1.3523
0.8	0.6733	3.366	1.3190	0.7618	1.5878	1.3933
0.9	0.7525	3.345	1.3434	0.7530	1.6616	1.4355
1.0	0.8308	3.323	1.3675	0.7443	1.7363	1.4773
1.25	1.0221	3.271	1.4265	0.7232	1.9232	1.5761
1.50	1.2074	3.220	1.4836	0.7031	2.1017	1.6663
1.75	1.3871	3.170	1.5390	0.6844	2.2654	1.7466
2.00	1.5613	3.123	1.5926	0.6670	2.4131	1.8180
$m_a = 4.500$ mol/kg						
0.1	0.0869	3.911	1.1499	0.8016	1.2414	1.1877
0.2	0.1727	3.885	1.1763	0.7924	1.3125	1.2310
0.3	0.2573	3.860	1.2023	0.7833	1.3841	1.2715
0.4	0.3408	3.834	1.2280	0.7742	1.4552	1.3087
0.5	0.4233	3.809	1.2533	0.7653	1.5266	1.3482
0.6	0.5046	3.785	1.2783	0.7564	1.5992	1.3900
0.7	0.5849	3.760	1.3030	0.7477	1.6729	1.4320
0.8	0.6642	3.736	1.3274	0.7391	1.7474	1.4731
0.9	0.7425	3.712	1.3515	0.7307	1.8222	1.5130
1.0	0.8198	3.689	1.3752	0.7223	1.8964	1.5517
1.25	1.0088	3.632	1.4333	0.7022	2.0751	1.6427
1.50	1.1919	3.576	1.4896	0.6833	2.2405	1.7247

m_U , mol/kg	c_U , mol/L	c_a , mol/L	d , g/sm ³	a_w	γ_U	γ_a
1.75	1.3696	3.522	1.5442	0.6657	2.3910	1.7985
2.00	1.5420	3.469	1.5972	0.6493	2.5265	1.8651
$m_a = 5.000$ mol/kg						
0.1	0.0857	4.285	1.1607	0.7778	1.3974	1.2672
0.2	0.1703	4.257	1.1867	0.7688	1.4676	1.3048
0.3	0.2537	4.229	1.2123	0.7599	1.5388	1.3458
0.4	0.3361	4.202	1.2376	0.7512	1.6114	1.3877
0.5	0.4175	4.175	1.2625	0.7425	1.6851	1.4292
0.6	0.4978	4.148	1.2871	0.7340	1.7595	1.4697
0.7	0.5770	4.122	1.3114	0.7256	1.8339	1.5091
0.8	0.6553	4.096	1.3355	0.7173	1.9073	1.5473
0.9	0.7326	4.070	1.3592	0.7092	1.9793	1.5844
1.0	0.8089	4.045	1.3826	0.7012	2.0497	1.6202
1.25	0.9957	3.983	1.4398	0.6821	2.2170	1.7039
1.50	1.1767	3.922	1.4953	0.6644	2.3703	1.7801
1.75	1.3524	3.864	1.5492	0.6478	2.5089	1.8492
2.00	1.5229	3.807	1.6015	0.6321	2.6349	1.9118

Following equations were used to calculate intermediate values

$$\gamma_{Uc} = 1 - 1.2057 \cdot m_U^{0.288745} + 0.875093 \cdot m_U^{0.7603} + 0.0012527 \cdot m_U^{-0.55601} + 0.022544 \cdot m_a^{1.939} + 0.1965 \cdot m_U^{0.36492} \cdot m_a^{1.0269} - 0.05434 \cdot m_U^{0.01938} \cdot m_a^{0.85829} + 0.0008968 \cdot m_a^{2.863} - 1.395 \cdot 10^{-6} \cdot m_a^{6.335} \quad (10)$$

$$\gamma_a = 0.51835 + 0.28785 \cdot m_U^{0.42072} + 0.0029304 \cdot m_U^{-1.72029} + 0.114566 \cdot m_U^{1.13785} \cdot m_a^{0.48261} + 0.12686 \cdot m_a^{1.1158} - 0.08377 \cdot m_a^{0.5694} \quad (11)$$

Mikulin's equations can be used to calculate greater number of components γ_{Uc} [17]:

$$\gamma_1 = v_1 \cdot \gamma_1^* \cdot m_1^* / (v_1 \cdot m_1 + v_2 \cdot m_2 + v_3 \cdot m_3) \quad (12)$$

$$\gamma_2 = v_2 \cdot \gamma_2^* \cdot m_2^* / (v_1 \cdot m_1 + v_2 \cdot m_2 + v_3 \cdot m_3) \quad (13)$$

where molal concentrations of sodium nitrate and rare elements can be as a third and later as a fourth component. But in order to do so we need to properly adjust equation for calculating density of water solution (1).

3. Conclusion

It was shown earlier [8-10] that the calculation using Zdanovskii's rule and Mikulin's equations resulted in good agreement between experimental values and calculated ones. Now the equation (10) and (11) have been used to calculate molal activity coefficients of nitric acid and uranyl nitrate in mixed solutions in order to increase upper limit of molal concentrations of uranyl nitrate to 2.0 mole/kg. Thus the problem of calculating equilibrium in extraction of nitrate uranyl during the reprocessing of spent fuel with the use of TBP extraction can be solved.

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