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**Calculation of the Activity of Uranyl Nitrate and Nitric Acid
in the H₂O–HNO₃–UO₂(NO₃)₂–TBP–Diluent System
Using Zdanovskii’s Rule**

A. V. Ochkin*, A. O. Merkushkin, S. Yu. Nekhaevskii, and E. A. Tyupina

Mendeleev Russian University of Chemical Technology, Miusskaya pl. 9, Moscow, 125047 Russia

*e-mail: ochkin@rctu.ru

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Abstract—The description of the H₂O–HNO₃–UO₂(NO₃)₂–TBP–diluent system required calculation of the activity coefficients of uranyl nitrate and nitric acid in mixed solutions. Three methods for determining the activity coefficients were considered: from the experimental data, using Pitzer’s equation, and using Zdanovskii’s rule with Mikulin’s equations. All the three methods lead to similar results. The relative root-mean-square deviation is 7–10%, but it depends on correct evaluation of the error. The third method will allow calculation of the activity coefficients in multicomponent solutions.

Keywords: uranyl nitrate, extraction, spent nuclear fuel reprocessing, activity coefficients

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The spent fuel from nuclear reactors is reprocessed using PUREX process involving extraction with tri-*n*-butyl phosphate in a hydrocarbon diluent [1]. The aqueous phase contains fission products, and also Am and Cm, whereas uranium, plutonium, and neptunium nitrates are concentrated in the organic phase. The organic phase from the thermodynamic viewpoint can be considered as a quinary H₂O–HNO₃–UO₂(NO₃)₂–TBP–diluent system because of the low content of Pu and Np. The principles of description of a quinary system are presented in [1]. They involve subdivision of the main system into subsystems, each of which is described using mole fractions and rational activity coefficients. Mixed hydrocarbon diluents are commonly used; *n*-dodecane can be considered as their analog.

Previously we described the subsystems H₂O–HNO₃–TBP [2], H₂O–UO₂(NO₃)₂–TBP [3, 4], and H₂O–HNO₃–TBP–dodecane [5, 6]. We considered as the next step the description of the quaternary subsystem H₂O–UO₂(NO₃)₂–TBP–dodecane, but tabulated data for this system are lacking. Therefore, passing to the H₂O–HNO₃–UO₂(NO₃)₂–TBP–dodecane system is possible only through preliminary determination of the

activities in the mixed subsystem H₂O–HNO₃–UO₂(NO₃)₂ using the equations presented in [7, 8] or Zdanovskii’s rule [9]. Only after that it will be possible to pass to the description of the quinary system H₂O–HNO₃–UO₂(NO₃)₂–TBP–dodecane.

**DETERMINATION OF THE NITRIC ACID
ACTIVITY IN MIXTURES
WITH URANYL NITRATE**

Data on the vapor pressure of HNO₃ over mixed solutions of uranyl nitrate and acid are given in Table 1 [7]. The calculation was performed using the procedure described in [10]. It is based on the following equation:

$$(p/p^0) = (y_u c_u)/(y_u^0 c_u^0) = (y_s c_s)^2/(y_s^0 c_s^0)^2 = (y_{\pm} c_{\pm})^2/(y_{\pm}^0 c_{\pm}^0)^2, \quad (1)$$

where *p* is the HNO₃ vapor pressure; *c*, molar concentration; *y*, activity coefficient; subscripts u, s, and ± refer to the undissociated acid, total acid concentration, and acid in the ionic form, with *c*_s = *c*_u + *c*_±; superscript 0 refers to the vapor pressure and concentration of the acid in the pure form at 25°C. For pure HNO₃: density

Table 1. Acid activities in mixed solutions of uranyl nitrate and nitric acid [7]

Run no.	Concentration, M		Density, g cm ⁻³	Vapor pressure, mm Hg		y_a	HNO ₃ activity, a_a
	UO ₂ (NO ₃) ₂ , c_U	HNO ₃ , c_a		H ₂ O	HNO ₃		
1	0.4660	0.976	1.1782	22.45	288×10^{-6}	1.102	1.158
2	0.4660	0.976	1.1782	22.51	288×10^{-6}	1.102	1.158
3	0.9020	0.951	1.3176	21.31	725×10^{-6}	1.795	2.914
4	0.9020	0.951	1.3176	21.44	868×10^{-6}	1.964	3.489
5	1.3140	0.920	1.4467	20.13	0.00159	2.748	6.391
6	1.3140	0.920	1.4467	20.40	0.00174	2.875	6.994
7	1.3140	0.920	1.4467	20.32	0.00175	2.883	7.034
8	0.4510	2.195	1.2163	20.87	0.00232	1.391	9.325
9	0.8540	2.108	1.3426	19.60	0.00332	1.733	13.344
10	0.8540	2.108	1.3426	19.43	0.00392	1.883	15.756
11	1.2370	2.026	1.4610	18.38	0.00724	2.663	29.100
12	1.2370	2.026	1.4610	18.75	0.00739	2.690	29.703
13	0.0860	4.290	1.1651	18.56	0.01038	1.506	41.721
14	0.2130	4.248	1.2027	18.62	0.01214	1.644	48.795
15	0.2130	4.248	1.2027	18.48	0.0122	1.648	49.036
16	0.4180	4.182	1.2646	17.73	0.0156	1.893	62.702
17	0.4180	4.182	1.2646	18.07	0.0155	1.887	62.300
18	0.4180	4.182	1.2646	17.92	0.0158	1.906	63.505
19	0.4180	4.182	1.2646	17.93	0.016	1.918	64.309
20	0.8030	3.923	1.3800	17.24	0.021	2.342	84.406
21	0.8030	3.923	1.3800	17.20	0.0214	2.364	86.014
22	0.8080	4.039	1.3804	17.00	0.0213	2.291	85.612
23	0.8080	4.039	1.3804	16.89	0.0204	2.242	81.994
24	1.1600	3.799	1.4850	16.46	0.0286	2.822	114.95
25	1.1600	3.799	1.4850	16.27	0.0269	2.737	108.12
26	1.1660	3.885	1.4812	15.86	0.0348	3.044	139.87
27	1.3760	3.716	1.5450	16.03	0.0379	3.321	152.33
28	1.3760	3.716	1.5450	16.50	0.0339	3.141	136.25
29	1.4280	3.818	1.5668	15.19	0.0384	3.254	154.34
30	1.4280	3.818	1.5668	15.68	0.0385	3.258	154.74
31	0.5331	4.981	1.3235	16.83	0.0358	2.408	143.9
32	0.3920	6.135	1.3136	15.37	0.0558	2.441	224.8
33	0.0984	7.462	1.2621	14.32	0.0924	2.583	371.4
34	0.2464	7.641	1.3148	13.40	0.1215	2.892	488.3
35	0.3740	7.135	1.3366	13.54	0.0953	2.743	383.0
36	0.3740	7.135	1.3366	13.19	0.0935	2.717	375.8
37	0.4871	7.674	1.4663	12.43	0.163	3.335	655.2
38	0.7437	7.776	1.4663	11.29	0.201	3.655	807.9
39	0.9843	7.654	1.5350	10.93	0.24	4.058	964.6
40	0.1015	8.054	1.2762	12.61	0.221	3.701	888.3
41	0.2469	8.070	1.3200	13.01	0.159	3.133	639.1
42	0.5232	8.227	1.3969	11.60	0.232	3.712	932.5
43	1.0030	8.129	1.5492	11.13	0.287	4.178	1153.5

$d = 1.5040 \text{ g cm}^{-3}$, vapor pressure $p^0 = 62.5 \text{ mm Hg}$ at 25°C , molar concentration $c_s^0 = 1000 \times 1.5040/63.015 = 23.867 \text{ M}$. The quantity y_s^* was taken equal to unity (the asterisk in [7] was used for denoting the activity at

infinite dilution), and for the pure acid $y_s^0 = 21.0 \pm 1.0$ was found. The key relationship is

$$y_s = 21.0 \times 23.867(p/62.5)^{0.5}/c_s \quad (2)$$

Table 2. Acid activities in mixed solutions of uranyl nitrate and nitric acid

Run no.	Concentration, M		HNO ₃ vapor pressure, mm Hg [8]	y_a	HNO ₃ activity calculated using data from indicated source		$[(a_{a1} - a_a)/a_a]^2$
	UO ₂ (NO ₃) ₂ , c_U	HNO ₃ , c_a			[8], a_{a1}	[7], a_a	
1	0.4660	0.976	338×10^{-6}	1.194	1.359	1.158	0.0301
2	0.4660	0.976	338×10^{-6}	1.194	1.359	1.158	0.0301
3	0.9020	0.951	728×10^{-6}	1.799	2.926	2.914	0.0000
4	0.9020	0.951	728×10^{-6}	1.799	2.926	3.489	0.0260
5	1.3140	0.920	0.001397	2.576	5.615	6.391	0.0147
6	1.3140	0.920	0.001397	2.576	5.615	6.994	0.0389
7	1.3140	0.920	0.001397	2.576	5.615	7.034	0.0407
8	0.4510	2.195	0.002143	1.337	8.613	9.325	0.0058
9	0.8540	2.108	0.003663	1.820	14.723	13.344	0.0107
10	0.8540	2.108	0.003663	1.820	14.723	15.756	0.0043
11	1.2370	2.026	0.005921	2.408	23.798	29.100	0.0332
12	1.2370	2.026	0.005921	2.408	23.798	29.703	0.0395
13	0.0860	4.290	0.0105	1.514	42.203	41.721	0.0001
14	0.2130	4.248	0.01211	1.642	48.674	48.795	0.0000
15	0.2130	4.248	0.01211	1.642	48.674	49.036	0.0001
16	0.4180	4.182	0.01512	1.864	60.772	62.70	0.0009
17	0.4180	4.182	0.01512	1.864	60.772	62.30	0.0006
18	0.4180	4.182	0.01512	1.864	60.772	63.50	0.0019
19	0.4180	4.182	0.01512	1.864	60.772	64.31	0.0030
20	0.8030	3.923	0.01995	2.283	80.186	84.41	0.0025
21	0.8030	3.923	0.01995	2.283	80.186	86.01	0.0046
22	0.8080	4.039	0.01995	2.217	80.186	85.61	0.0040
23	0.8080	4.039	0.01995	2.217	80.186	81.99	0.0005
24	1.1600	3.799	0.02831	2.808	113.79	114.95	0.0001
25	1.1600	3.799	0.02831	2.808	113.79	108.12	0.0027
26	1.1660	3.885	0.03149	2.896	126.57	139.87	0.0090
27	1.3760	3.716	0.03477	3.181	139.75	152.33	0.0068
28	1.3760	3.716	0.03477	3.181	139.75	136.25	0.0007
29	1.4280	3.818	0.0392	3.288	157.56	154.34	0.0004
30	1.4280	3.818	0.0392	3.288	157.56	154.74	0.0003
31	0.5331	4.981	0.03229	2.287	129.78	143.9	0.0096
32	0.3920	6.135	0.0591	2.512	237.5	224.8	0.0035
33	0.0984	7.462	0.09525	2.622	382.8	371.4	0.0010
34	0.2464	7.641	0.1201	2.875	482.7	488.3	0.0001
35	0.3740	7.135	0.1042	2.868	418.8	383.0	0.0087
36	0.3740	7.135	0.1042	2.868	418.8	375.8	0.0131
37	0.4871	7.674	0.1551	3.254	623.4	655.2	0.0023
38	0.7437	7.776	0.1986	3.6334	798.2	807.9	0.0001
39	0.9843	7.654	0.2235	3.9159	898.3	964.6	0.0047
40	0.2469	8.070	0.1551	3.0939	623.4	639.1	0.0006
41	0.5232	8.227	0.2212	3.6243	889.1	932.5	0.0022
42	1.0030	8.129	0.2819	4.1408	1133.0	1153.5	0.0003
						\sum	0.3670
						σ	0.0946

In what follows, we will use subscript a for denoting the acid and subscript U for denoting uranium.

Davis et al. [7] estimate the uncertainty of their data at 4 rel. %. However, it is noted in [8] that the results

of run nos. 40 and 41 are inconsistent; therefore, data for run no. 40 were excluded from the analysis. To evaluate the accuracy of the experimental data, we compared the data of [7] and [8] (Table 2). As can be seen, the adequacy bias is 9.46%.

CALCULATION OF THE ACTIVITIES
OF NITRIC ACID AND URANYL NITRATE
USING ZDANOVSKII'S RULE
AND MIKULIN'S EQUATIONS

In 1936, Zdanovskii [11] discovered a relationship that was later termed Zdanovskii's rule:

$$m_1/m_1^* + m_2/m_2^* = 1, \quad (3)$$

where m_1 and m_2 are the molal concentrations of electrolytes 1 and 2 in a mixed solution with the water activity a_w , and m_1^* and m_2^* are the molal concentrations of electrolytes 1 and 2 in their individual isopiestic solutions with the same water activity a_w . Mikulin [9] suggested equations for calculating the activity coefficients in such solutions:

$$\gamma_1 = v_1 \gamma_1^* m_1^* / (v_1 m_1 + v_2 m_2), \quad (4a)$$

$$\gamma_2 = v_2 \gamma_2^* m_2^* / (v_1 m_1 + v_2 m_2), \quad (4b)$$

where γ_1 and γ_2 are the activity coefficients of electrolytes 1 and 2 in a mixed solution with the water activity a_w , and γ_1^* and γ_2^* are the activity coefficients of the same electrolytes in the isopiestic solutions with the same water activity a_w . Equations (3), (4a), and (4b) were generalized by Mikulin for the case of more than two components. Because analysis of the applicability of Zdanovskii's rule revealed cases of deviations from Eqs. (4a) and (4b) [9], it was necessary to compare the acid activities calculated from data in Table 2 and using Mikulin's equations.

The activity of uranyl nitrate a_U in binary aqueous solutions was calculated by the common equation

$$a_U = 4m_U^3 \gamma_{\pm}^3, \quad (5)$$

where m_U is the molal concentration of uranyl nitrate in aqueous solution, and γ_{\pm} is the mean ionic activity coefficient. The values of γ_{\pm} are given in Table 3 [12]. The activities a_U calculated from these data in the interval of uranyl nitrate concentrations from 0.04 to 2.0 M were approximated by the following equation [3, 4]:

$$a_U = 0.1879m_U^{2.5} + 0.752m_U^{4.18} + 0.2881m_U^{7.22}. \quad (6)$$

However, in what follows we will also perform calculations for higher concentrations; therefore, it was necessary to use the whole volume of data in Table 3.

Table 3. Activity coefficients of uranyl nitrate and water activities

$m_U, \text{mol kg}^{-1}$ [12]	γ_U [12]	a_w	a_w [14]
0.00	1.0000	1.0000	1.000000
0.10	0.5430	0.9952	0.995283
0.20	0.5120	0.9903	0.990289
0.30	0.5100	0.9850	0.984962
0.40	0.5180	0.9794	0.979313
0.50	0.5340	0.9734	0.973336
0.60	0.5550	0.9670	0.967018
0.70	0.5780	0.9605	0.960347
0.80	0.6080	0.9533	0.953316
0.90	0.6410	0.9459	0.945928
1.00	0.6790	0.9380	0.938188
1.20	0.7610	0.9217	—
1.40	0.8550	0.9043	—
1.60	0.9622	0.8861	—
1.80	1.0830	0.8671	—
2.00	1.2180	0.8475	0.846830
2.50	1.6020	0.7980	0.798237
3.00	2.0000	0.7516	0.752624
3.50	2.3700	0.7102	0.711968
4.00	2.6400	0.6764	0.676758
4.50	2.8500	0.6470	0.646336
5.00	3.0100	0.6210	0.619310
5.50	3.2000	0.5940	0.593972

In addition, for calculations using Eqs. (4a) and (4b) it is necessary to know the water activities a_w , which were interpolated using data from [13]. Alternative values of a_w are given in [14]. The differences are small, and we used the values from [13].

A grapho-analytical method for calculating m^* is described in [9]. First, from the experimental data for binary solutions it is necessary to determine the dependence of m_1^* and m_2^* on the water activity a_w . This is followed by construction of the dependence in the coordinates m_2^*/m_1^* , where m_1^* and m_2^* are the molal concentrations at equal water activities, i.e., in isopiestic solutions. In so doing, it should be taken into account that, at $m_2 \rightarrow 0$, $m_2^*/m_1^* = v_1/v_2$. To determine m_2^* in the ternary solution with concentrations m_1 and m_2 , the quantity m_2/m_1 is plotted (lower point on the ordinate). Then, the line is drawn from m_2/m_1 to m_2 on the abscissa and then to the line describing the dependence $m_2^*/m_1^* - m_2^*$. The perpendicular drawn from the interception point gives the sought-for value of m_2^* . It can be readily shown that such procedure corresponds to

Table 4. Activities of nitric acid, a_a , and uranyl nitrate, a_U , calculated using Zdanovskii's rule

Run no.	c_a	c_U	a_w	m_a^*	γ_a	a_a	m_U^*	γ_{\pm}	a_U
1	0.976	0.466	0.930	1.914	0.8957	1.4939	1.101	0.7086	0.1453
2	0.976	0.466	0.930	1.914	0.8957	1.4939	1.101	0.7086	0.1453
3	0.951	0.902	0.888	2.877	1.1125	3.2429	1.578	0.9734	2.7315
4	0.951	0.902	0.888	2.877	1.1125	3.2429	1.578	0.9734	2.7315
5	0.920	1.314	0.840	3.902	1.4002	6.3994	2.069	1.3565	22.854
6	0.920	1.314	0.840	3.902	1.4002	6.3994	2.069	1.3565	22.854
7	0.920	1.314	0.840	3.902	1.4002	6.3994	2.069	1.3565	22.854
8	2.195	0.451	0.866	3.367	1.1254	8.6103	1.816	1.0340	0.4093
9	2.108	0.854	0.822	4.273	1.3855	15.441	2.248	1.3913	6.7702
10	2.108	0.854	0.822	4.273	1.3855	15.441	2.248	1.3913	6.7702
11	2.026	1.237	0.778	5.196	1.6788	25.696	2.713	1.8523	48.543
12	2.026	1.237	0.778	5.196	1.6788	25.696	2.713	1.8523	48.543
13	4.290	0.086	0.779	5.175	1.4650	41.080	2.702	1.6124	0.01076
14	4.248	0.213	0.765	5.471	1.5508	47.750	2.851	1.7579	0.2118
15	4.248	0.213	0.765	5.471	1.5508	47.750	2.851	1.7579	0.2118
16	4.182	0.418	0.743	5.960	1.7037	60.913	3.103	2.0087	2.3888
17	4.182	0.418	0.743	5.960	1.7037	60.913	3.103	2.0087	2.3888
18	4.182	0.418	0.743	5.960	1.7037	60.913	3.103	2.0087	2.3888
19	4.182	0.418	0.743	5.960	1.7037	60.913	3.103	2.0087	2.3888
20	3.923	0.803	0.711	6.685	1.9396	81.602	3.499	2.4174	29.518
21	3.923	0.803	0.711	6.685	1.9396	81.602	3.499	2.4174	29.518
22	4.039	0.808	0.701	6.907	2.0010	91.454	3.628	2.5320	34.558
23	4.039	0.808	0.701	6.907	2.0010	91.454	3.628	2.5320	34.558
24	3.799	1.160	0.673	7.569	2.2145	114.00	4.047	2.9164	156.26
25	3.799	1.160	0.673	7.569	2.2145	114.00	4.047	2.9164	156.26
26	3.885	1.166	0.664	7.785	2.2847	126.08	4.195	3.0424	180.17
27	3.716	1.376	0.651	8.115	2.3909	137.39	4.428	3.2326	355.16
28	3.716	1.376	0.651	8.115	2.3909	137.39	4.428	3.2326	355.16
29	3.818	1.428	0.640	8.391	2.4550	153.57	4.628	3.3574	444.73
30	3.818	1.428	0.640	8.391	2.4550	153.57	4.628	3.3574	444.73
31	4.981	0.5331	0.677	7.480	2.0797	130.28	3.988	2.7257	12.381
32	6.135	0.392	0.623	8.846	2.3798	240.39	4.959	3.3073	8.794

Eq. (3). Indeed, the equality $(m_2^*/m_1^*)/(m_2^* - m_2) = (m_2/m_1)/m_2$ is observed.

The results of calculating the activities of the acid and uranyl nitrate are given in Table 4. In so doing, the same molar concentrations as in [7] were used. However, the calculation was limited to available values of a_w from Table 3.

COMPARISON OF THE TWO METHODS FOR CALCULATING THE ACID ACTIVITY

The results of calculating the acid activity from the experimental data [7] and using Zdanovskii's rule (Table 4) are compared in Table 5.

As follows from Table 5, the difference in the acid activities a_a and a_{ac} is approximately the same as the difference between a_a and a_{a1} in Table 2. Then, when comparing Tables 1, 2, and 5, we can make the following conclusions:

(1) The error of the experimental data is not 4 rel %, as indicated in [7], but appreciably larger.

(2) The acid vapor pressures in run nos. 1 and 2 in Table 1 are probably underestimated, which follows from Tables 2 and 5.

(3) Comparison of Tables 1, 2, and 5 leads to the estimated error of the acid activities of approximately 7 rel %.

Table 5. Comparison of the acid activities determined from the experimental data (a_a) and calculated using Zdanovskii's rule (a_{ac})

Run no.	c_a	c_U	a_a	a_w	y_a	a_{ac}	$[(a_a - a_{ac})/a_a]^2$
1	0.976	0.466	1.158	0.930	0.8957	1.494	0.084427
2	0.976	0.466	1.158	0.930	0.8957	1.494	0.084427
3	0.951	0.902	2.914	0.888	1.1125	3.243	0.012734
4	0.951	0.902	3.489	0.888	1.1125	3.243	0.004969
5	0.920	1.314	6.391	0.840	1.4002	6.399	1.86×10^{-6}
6	0.920	1.314	6.994	0.840	1.4002	6.399	0.007219
7	0.920	1.314	7.034	0.840	1.4002	6.399	0.008134
8	2.195	0.451	9.325	0.866	1.1254	8.610	0.005872
9	2.108	0.854	13.344	0.822	1.3855	15.441	0.02468
10	2.108	0.854	15.756	0.822	1.3855	15.441	0.0004
11	2.026	1.237	29.100	0.778	1.6788	25.696	0.013682
12	2.026	1.237	29.703	0.778	1.6788	25.696	0.018196
13	4.29	0.086	41.721	0.779	1.4650	41.080	0.000235
14	4.248	0.213	48.795	0.765	1.5508	47.750	0.000458
15	4.248	0.213	49.036	0.765	1.5508	47.750	0.000687
16	4.182	0.418	62.70	0.743	1.7037	60.91	0.000814
17	4.182	0.418	62.30	0.743	1.7037	60.91	0.000496
18	4.182	0.418	63.51	0.743	1.7037	60.91	0.001667
19	4.182	0.418	64.31	0.743	1.7037	60.91	0.00279
20	3.923	0.803	84.41	0.711	1.9396	81.60	0.001103
21	3.923	0.803	86.01	0.711	1.9396	81.60	0.002631
22	4.039	0.808	85.61	0.701	2.0010	91.45	0.004657
23	4.039	0.808	81.99	0.701	2.0010	91.45	0.013311
24	3.799	1.160	114.95	0.673	2.2145	114.00	6.85×10^{-5}
25	3.799	1.160	108.12	0.673	2.2145	114.00	0.002959
26	3.885	1.166	139.88	0.664	2.2847	126.08	0.009731
27	3.716	1.376	152.33	0.651	2.3909	137.39	0.009619
28	3.716	1.376	136.26	0.651	2.3909	137.39	6.96×10^{-5}
29	3.818	1.428	154.34	0.640	2.4550	153.57	2.5×10^{-5}
30	3.818	1.428	154.74	0.640	2.4550	153.57	5.75×10^{-5}
31	4.981	0.5331	143.89	0.677	2.0797	130.28	0.008943
32	6.135	0.392	224.28	0.623	2.3798	240.39	0.005163
$\Sigma(1-32)$							0.330228
$\sigma(1-32)$							0.103
$\Sigma(3-32)$							0.161374
$\sigma(3-32)$							0.0733

(4) Zdanovskii's rule and Mikulin's equations allow reliable calculation of the activities of the acid and uranyl nitrate.

Thus, the use of Zdanovskii's rule and Mikulin's equations for calculating the activities of nitric acid

and uranyl nitrate opens prospects for calculating the equilibrium in the quinary system $H_2O-HNO_3-UO_2(NO_3)_2-TBP$ -dodecane and hence in the extraction system on which the PUREX process is based. In so doing, it is probably possible to take into account the

effect of fission products on the water activity in the mixed solutions and hence on the activity of the acid and uranyl nitrate.

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