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Distribution of U(IV), U(VI), Pu(III) and Nitric Acid between 30 vol.% Tributyl Phosphate in Dodecane and Aqueous Nitrate Solutions

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DISTRIBUTION OF U(IV), U(VI), Pu(III) AND NITRIC ACID BETWEEN 30 vol.% TRIBUTYL PHOSPHATE IN DODECANE AND AQUEOUS NITRATE SOLUTIONS

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Abstract: The distribution of U(IV), U(VI), Pu(III) and nitric acid between 30 vol.% TBP in dodecane and aqueous nitrate solutions, also containing hydrazine nitrate, was studied in batch experiments. Distribution ratios of the components were measured as functions of the concentrations of nitric acid and of uranium(IV), uranyl, plutonium(III) and hydrazine nitrates. Major part of the data was obtained at 25°C and temperature dependences of the distribution ratios of U(IV) and nitric acid were measured at 15 to 55°C.

VERTEILUNG VON U(IV), U(VI), Pu(III) UND SALPETERSÄURE ZWISCHEN 30 Vol.% TRIBUTYLPHOSPHAT IN DODEKAN UND WÄSSRIGEN NITRATLÖSUNGEN

Zusammenfassung: Die Verteilung von U(IV), U(VI), Pu(III) und Salpetersäure zwischen 30 vol.% TBP in Dodekan und wässrigen Nitratlösungen, die auch Hydrazinnitrat enthielten, wurde in statischen Versuchen untersucht. Verteilungskoeffizienten der genannten Komponenten wurden als Funktionen der Konzentrationen von Salpetersäure sowie von U(IV)-, U(VI)-, Pu(III)- und Hydrazinnitrat gemessen. Die Daten wurden zum größeren Teil bei 25°C gewonnen und die Temperaturabhängigkeit der Verteilungskoeffizienten von U(IV) und Salpetersäure wurde bei 15 bis 55°C verfolgt.

Knowledge of distribution ratios of actinides and nitric acid in the extraction with tributyl phosphate (henceforth TBP) is of importance in the modeling of extraction operations performed in the Purex process for the reprocessing of irradiated nuclear fuels. Detailed information is available on the extraction of uranyl and plutonium(IV) nitrates and of nitric acid (see [1], Part I), but rather little is known about distribution equilibria under the conditions of a reductive separation of plutonium from uranium. Especially of interest are data on the distribution of uranium(IV), uranium(VI) and plutonium(III) between 30 vol.% TBP in an aliphatic diluent and aqueous nitrate solutions, also containing hydrazine nitrate which is used as nitrite scavenger. Wherever possible, the distribution ratio of nitric acid, or at least the nitric acid concentration in the equilibrium aqueous phase, should be measured together with the distribution ratios of uranium(IV,VI) and/or plutonium(III). Only a part of data published before 1981, namely those obtained in this Institute and listed in Part II of [1], fulfilled these requirements. The data illustrate the effect of nitric acid on the distribution of uranium(IV) and the mutual influence of uranium(IV) and uranium(VI). More detailed information on these effects, and also information on the salting-out action of hydrazine nitrate and on the distribution of plutonium(III) is of great interest. Thus we continued our previous experimental work, gained such data, and present them numerically in this paper. The aim of this report is to give a complete list of distribution data obtained in this Institute in the investigation of sytems involving TBP in dodecane, uranium(IV) and hydrazine nitrate, and also to list results of distribution measurements made with uranium(VI) and nitric acid in the presence of hydrazine nitrate. An evaluation of the data will be published elsewhere.

EXPERIMENTAL

Reagents

Uranyl nitrate, nitric acid, ammonium oxalate, sodium fluoride, diethylenetriaminepentaacetic acid (henceforth DTPA), pyridine-2,6-dicarboxylic acid (henceforth PDCA), acetic acid and sodium acetate (all Merck, reagent grade), and n-dodecane (Fluka, olefine free), were used as received.

Stock solutions of uranium(IV) and plutonium(III) nitrates were prepared by electroreduction of uranium(VI) and plutonium(IV) respectively in solutions containing nitric acid and hydrazine nitrates. The stock solutions contained ~1M U(IV) and ~0.4M Pu(III) respectively, each together with ≤1M nitric acid and ~0.1M hydrazine nitrate. The content of U(VI) in the U(IV) solution was <2% and the oxidation of U(IV) did not exceed 2% during several weeks, if the solution was kept in a dark bottle at <10°C.

A stock solution of plutonium(IV) nitrate was provided by the reprocessing plant WAK at Karlsruhe, Germany. It contained 0.85M Pu(IV) and 6.5M nitric acid, and the fraction of Pu(IV) was ~2%.

A stock solution of hydrazine nitrate was prepared by careful neutralization of hydrazine hydrate with nitric acid to pH~4. The reaction mixture was cooled during the neutralization to <20°C. The content of hydrazinium ions was determined by alkalimetric titration.

Distribution measurements

The aqueous phase was mixed directly in a thermostated stirring vessel, in taking volumes of stock solutions needed for reaching desired starting concentrations. The starting volume of the aqueous phase was adjusted to a known volume (mostly 12 ml) by water. Plutonium(III) was added either directly to the starting aqueous phase, or produced by an addition of a small amount of plutonium(IV) in the presence of excess uranium(IV). A 30 vol.% solution of TBP in dodecane was added to a phase volume ratio of 1 and the phases were appropriately dispersed by a mechanical stirrer. A phase contact time of 10 min was sufficient for thermostating the system and for reaching the distribution equilibrium. The phases were then separated by gravitation, and samples were taken for analyses at the temperature of the experiment and cooled to the room temperature if necessary.

Analytical procedures

Solutions for complexometric titrations were prepared as follows: An aliquot of an aqueous phase was pipetted into a titration vessel containing 5 ml water or ≤0.4M nitric acid and the content of the vessel was diluted with water to a final volume of 50 - 60 ml. Nitric acid had to be present in the titration vessel only if an actinide(IV), especially Pu(IV), was to be determined. The amount of the acid

in the titration vessel was chosen so that a pH value suitable for the determination was reached by the dilution to the final volume, but pH~2 was never exceeded during the preparation of the titrated solution. Without this precaution taken the actinide(IV) was hydrolyzed, a subsequent addition of nitric acid did not dehydrolyse it fastly enough, and no reliable complexometric titration was possible. Nitric acid had not to be present in the titration vessel in the preparation of a solution for a titration of uranium(VI) and a pH value needed for the titration was adjusted after the dilution to the final volume by adding solutions of a buffer and, eventually, sodium hydroxide. An aliquot of an organic phase was pipetted into a titration vessel containing 5 - 10 ml methanol and, if an actinide(IV) was to be determined, 2 ml 2M nitric acid. The actinide to be determined was transferred from the aliquot into the methanol phase by a short, careful stirring and the content of the vessel was treated as described above.

To prepare solutions for alkalimetric titrations, an aliquot of any phase was pipetted into a titration vessel containing 50 - 60 ml water and 50 mg of each ammonium oxalate and sodium fluride.

Uranium(IV) was determined by complexometric titration with 0.0025 - 0.01M DTPA at pH 1.5 - 2.0 in unbuffered solutions. Xylenol orange was taken as an indicator. Uranium(VI), even if present in excess, and plutonium(III) do not interfere.

Uranium(VI) was determined by complexometric titration with 0.01M PDCA at pH 3.6 (acetate buffer) with Arsenazo I used as an indicator [2].

Plutonium(III) was oxidized by sodium nitrite or air to plutonium(IV) and determined by titration with DTPA analogously to U(IV).

To determine a tetravalent actinide and uranium(VI) in one sample, first the actinide(IV) was titrated with DTPA as described above. Care was taken to avoid addition of any excess DTPA in this step of the procedure. Then acetate buffer was added to the solution, the pH value was adjusted to 3.6 with sodium hydroxide under checking with a glass electrode, and uranium(VI) was titrated with PDCA. It is most important to use particularly DTPA for the titration of the actinide(IV), because ethylenediaminetetraacetic acid tends to the formation of mixed actinide(IV)-uranium(VI) complexes and severely interferes with

the subsequent titration of U(VI) by PDCA. The determination of U(VI) is reliable if the initial ratio of actinide(IV) to U(VI) is ≤1 and the accuracy of the determination is remarkably lowered as the intital actinide(IV) to U(VI) ratio is increased above 1.

Alternatively, in early experiments (denoted by five-digits experiment numbers in Tables 1 and 2) U(IV) was determined by titration with DTPA, total uranium was determidend by the x-ray fluorescence method, and the concentration of U(VI) was calculated from the difference.

Nitric acid was determined by alkalimetric titration after complexing actinides by ammonium oxalate and sodium fluoride [3]. Due to favourable ratios of the concentrations of nitric acid and actinides in lower valence states, changes of the acid concentration eventually caused by the oxidation of U(IV) or Pu(III) during the titration could be expected to be negligible.

RESULTS

The data are gathered in Tables 1 to 5. Data obtained at 25°C are divided into four groups (Tables 1 to 4) according to the number of components present in the system. Table 5 gives results obtained at temperatures other than 25°C. To facilitate orientation in the Tables, we further grouped the data according to the concentrations of some components and, in Table 5, according to temperature. The subgroups are separated within the Tables by lines filled with asterisks. The Tables are computer-printed and for technical reasons the number of necessary decimal places had to be kept constant in each column. Thus last decimal places may in some cases have no physical significance. Results denoted by five-digits experiment numbers had been obtained in earlier work and are also given under the same numbers in Part II of [1].

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TABLE 1

Distribution ratios of uranium(IV) and nitric acid at 25°C. Initial organic phase: a 30 vol.% solution of TBP in dodecane. Initial aqueous phase: a solution of uranium(IV) nitrate, hydrazine nitrate and nitric acid.

Expt.no.	Equil. aq. molar concns.			Distribution ratios	
	U(IV)	HNO_3	$N_2H_5NO_3$	U(IV)	HNO_3
59	0.0220	0.100	0.0022	0.018	
72	0.0415	0.150	0.0041	0.068	
50131	0.0027	0.580	0.0006	0.204	
50101	0.0437	0.580	0.0104	0.348	
50132	0.0063	0.610	0.0014	0.254	
50135	0.0279	0.630	0.0063	0.287	
50134	0.0211	0.640	0.0047	0.256	
50133	0.0139	0.660	0.0030	0.223	
50136	0.0531	0.670	0.0131	0.407	
50137	0.0662	0.710	0.0164	0.409	
50138	0.0762	0.710	0.0193	0.444	
50102	0.0884	0.770	0.0242	0.555	
50122	0.0024	0.870	0.0006	0.288	
50123	0.0052	0.910	0.0013	0.385	
50127	0.0441	1.030	0.0125	0.617	
50124	0.0107	1.060	0.0029	0.561	
50126	0.0234	1.090	0.0064	0.564	
50125	0.0170	1.110	0.0047	0.559	
50128	0.0537	1.110	0.0155	0.639	
50129	0.0652	1.170	0.0188	0.644	
50108	0.0180	1.690	0.0068	1.161	
50105	0.0183	1.720	0.0069	1.137	
50118	0.0185	1.720	0.0067	1.076	
50114	0.0019	1.740	0.0007	1.105	
50117	0.0116	1.740	0.0043	1.129	
50120	0.0462	1.740	0.0166	1.050	
50115	0.0043	1.750	0.0015	1.023	
50116	0.0074	1.760	0.0028	1.149	
50119	0.0372	1.770	0.0135	1.070	
50104	0.0185	1.780	0.0069	1.130	
50121	0.0537	1.780	0.0193	1.047	
50106	0.0457	1.810	0.0169	1.101	
50107	0.0170	1.860	0.0063	1.124	
128	0.0880	1.960	0.0375	1.102	0.168
135	0.0730	2.260	0.0300	1.356	0.137
50130	0.0573	2.580	0.0165	0.635	
50112	0.0154	2.590	0.0069	1.545	
50150	0.0104	2.650	0.0050	1.731	
50153	0.0354	2.650	0.0163	1.624	

TABLE 1 (continued)

Expt.no.	Equil. aq. molar concns.			Distribution ratios	
	U(IV)	HNO_3	$N_2H_5NO_3$	U(IV)	HNO_3
50148	0.0035	2.660	0.0017	1.771	
50151	0.0147	2.660	0.0070	1.714	
50149	0.0071	2.670	0.0034	1.746	
50147	0.0017	2.700	0.0009	1.882	
50152	0.0288	2.700	0.0136	1.681	
50154	0.0403	2.710	0.0188	1.650	
50110	0.0152	2.800	0.0073	1.717	
175	0.0235	2.900	0.0145	2.170	0.183
50109	0.0136	2.930	0.0069	1.875	
174	0.0534	2.980	0.0300	1.753	0.151
130	0.0490	3.130	0.0250	1.837	0.169
173	0.0710	3.260	0.0300	1.761	0.144
134	0.0440	3.400	0.0384	2.114	0.144
50111	0.0119	3.560	0.0072	2.429	
172	0.0956	3.590	0.0300	1.611	0.125
133	0.0380	4.070	0.0307	2.579	0.143
132	0.0360	4.150	0.0285	2.722	0.152
50139	0.0016	4.190	0.0011	2.750	
50140	0.0026	4.200	0.0018	2.962	
50141	0.0052	4.230	0.0034	2.731	
50142	0.0077	4.280	0.0051	2.792	
50143	0.0106	4.330	0.0070	2.783	
50144	0.0215	4.330	0.0135	2.577	
50145	0.0259	4.350	0.0166	2.645	
50146	0.0304	4.360	0.0191	2.572	
50103	0.0092	4.650	0.0076	3.696	
176	0.0320	4.960	0.0210	3.156	0.133
50113	0.0080	5.830	0.0073	4.177	
**********	**********	**********	**********	**********	**********
127	0.1310	1.450	0.0375	0.748	0.228
171	0.1510	3.310	0.0389	1.331	0.145
170	0.1320	4.000	0.0294	1.636	0.110
**********	**********	**********	**********	**********	**********
131	0.3060	0.400	0.0818	0.510	0.125
126	0.2570	0.640	0.0600	0.490	
177	0.5440	2.530	0.0830	0.585	0.087
179	0.5640	3.050	0.0810	0.652	0.079
178	0.6420	4.250	0.0860	1.218	0.073
**********	**********	**********	**********	**********	**********
14	0.0433	0.038	0.1000	0.079	
8	0.0640	0.038	0.1000	0.141	
1	0.0665	0.050	0.1000	0.132	
15	0.0433	0.051	0.1000	0.088	
21	0.0298	0.054	0.1000	0.048	
11	0.0677	0.056	0.1000	0.161	
78	0.0714	0.058	0.1000	0.085	

TABLE 1 (continued)

Expt.no.	Equil. aq. molar concns.			Distribution ratios	
	U(IV)	HNO_3	$N_2H_5NO_3$	U(IV)	HNO_3
24	0.0650	0.063	0.1000	0.155	
25	0.0683	0.065	0.1000	0.155	
26	0.0671	0.066	0.1000	0.155	
27	0.0677	0.066	0.1000	0.154	
63	0.0296	0.070	0.1000	0.010	
60	0.0195	0.071	0.1000	0.042	
61	0.0195	0.071	0.1000	0.042	
20	0.0305	0.071	0.1000	0.059	
16	0.0439	0.072	0.1000	0.093	
73	0.0523	0.075	0.1000	0.035	
79	0.0668	0.075	0.1000	0.067	
64	0.0365	0.077	0.1000	0.035	
12	0.0549	0.080	0.1000	0.133	
28	0.0542	0.081	0.1000	0.138	
29	0.0597	0.090	0.1000	0.152	
17	0.0427	0.091	0.1000	0.098	
22	0.0151	0.094	0.1000	0.042	
19	0.0292	0.095	0.1000	0.068	
74	0.0535	0.095	0.1000	0.048	
13	0.0555	0.098	0.1000	0.141	
5	0.0658	0.103	0.1000	0.148	
80	0.0688	0.104	0.1000	0.080	
4	0.0665	0.115	0.1000	0.154	
10	0.0677	0.121	0.1000	0.166	
3	0.0668	0.130	0.1000	0.160	
62	0.0200	0.140	0.1000	0.045	
18	0.0414	0.142	0.1000	0.128	
362	0.0827	0.148	0.0940	0.245	0.226
70	0.0390	0.160	0.1000	0.067	
65	0.0354	0.176	0.1000	0.069	
372	0.0896	0.178	0.1000	0.245	0.130
81	0.0683	0.192	0.1000	0.125	
56	0.0186	0.209	0.1000	0.037	
2	0.0650	0.211	0.1000	0.190	
75	0.0515	0.213	0.1000	0.092	
9	0.0659	0.220	0.1000	0.193	
66	0.0345	0.257	0.1000	0.105	
261	0.0826	0.320	0.1160	0.360	0.167
6	0.0610	0.335	0.1000	0.291	
23	0.0616	0.335	0.1000	0.292	
384	0.0981	0.336	0.1000	0.345	0.166
383	0.0894	0.340	0.1000	0.311	0.171
76	0.0482	0.383	0.1000	0.205	
71	0.0333	0.400	0.1000	0.204	
57	0.0170	0.419	0.1000	0.135	
67	0.0311	0.420	0.1000	0.240	

TABLE 1 (continued)

Expt.no.	Equil. aq. molar concns.			Distribution ratios	
	U(IV)	HNO_3	$N_2H_5NO_3$	U(IV)	HNO_3
68	0.0273	0.620	0.1000	0.380	
82	0.0525	0.709	0.1000	0.415	
77	0.0330	0.720	0.1000	0.420	
69	0.0266	0.740	0.1000	0.443	
58	0.0134	0.755	0.1000	0.388	
7	0.0239	0.798	0.1000	0.544	
294	0.0162	1.580	0.1000	1.150	0.224
295	0.0129	2.440	0.1000	1.700	0.209
296	0.0104	3.320	0.1000	2.320	0.205
297	0.0088	4.330	0.1000	2.970	0.180
363	0.1090	0.088	0.1000	0.286	0.407
376	0.1040	0.099	0.1000	0.266	0.111
377	0.1650	0.128	0.1000	0.449	
378	0.1900	0.153	0.1000	0.464	
373	0.1080	0.183	0.1000	0.290	0.121
375	0.1580	0.204	0.1000	0.404	0.239
379	0.1800	0.206	0.1000	0.496	0.065
374	0.1330	0.208	0.1000	0.370	0.102
382	0.1660	0.252	0.1000	0.437	0.118
364	0.1330	0.256	0.0980	0.405	0.134
369	0.1620	0.263	0.1000	0.425	0.114
368	0.1040	0.304	0.1000	0.312	0.154
381	0.1360	0.327	0.1000	0.406	0.146
365	0.1520	0.355	0.1000	0.443	0.155
380	0.1160	0.358	0.1000	0.347	0.158
370	0.2230	0.280	0.1000	0.473	0.059
371	0.3060	0.280	0.1000	0.455	0.036
366	0.2220	0.360	0.1000	0.491	0.172
367	0.2900	0.366	0.1000	0.510	0.086
260	0.0700	0.383	0.4920	0.652	0.177
292	0.0096	2.550	1.0000	2.430	0.209
293	0.0113	2.570	0.5000	2.010	0.214
291	0.0066	2.610	2.0000	3.670	0.194

TABLE 2

Distribution ratios of uranium(IV), uranium(VI) and nitric acid at 25°C. Initial organic phase: a 30 vol.% solution of TBP in dodecane. Initial aqueous phase: a solution of uranium(IV) nitrate, uranyl nitrate, hydrazine nitrate and nitric acid.

Expt.no.	Equil. aq. molar concns.[a]				Distribution ratios		
	U(VI)	U(IV)	HNO_3	$N_2H_5NO_3$	U(VI)	U(IV)	HNO_3
116	0.0097	0.053	0.0626	0.017		0.0560	
117	0.0097	0.052	0.1204	0.017		0.0790	
118	0.0097	0.037	0.8012	0.017		0.5260	0.195
119	0.0097	0.033	0.9148	0.017		0.6270	0.198
50161	0.0281	0.085	1.8400	0.021	6.758	0.4033	
50159	0.0357	0.078	1.8600	0.018	6.485	0.2905	
50185	0.0333	0.151	2.0900	0.036	6.667	0.3451	
50166	0.0266	0.072	3.0200	0.019	8.538	0.5160	
50170	0.0321	0.120	3.0900	0.032	6.866	0.5367	
50174	0.0288	0.112	4.3700	0.032	7.958	0.6299	
50201	0.0338	0.232	2.2200	0.056	5.473	0.3668	
50155	0.0478	0.087	1.1900	0.018	4.611	0.2092	
50182	0.0628	0.168	1.5100	0.036	4.299	0.2008	
50181	0.0436	0.158	1.5700	0.036	4.982	0.2924	
50160	0.0529	0.086	1.9500	0.018	5.231	0.2023	
50186	0.0548	0.163	1.9700	0.035	5.164	0.2254	
50167	0.0420	0.082	3.0900	0.019	7.017	0.3162	
50171	0.0479	0.136	3.1400	0.032	5.975	0.3417	
50168	0.0604	0.089	3.1500	0.019	5.674	0.2097	
50221	0.0504	0.150	4.6100	0.036	6.131	0.3780	
50189	0.0654	0.182	0.7500	0.037	3.425	0.1724	
50177	0.0550	0.204	1.0000	0.044	3.927	0.2190	
50193	0.0571	0.268	1.1800	0.057	4.249	0.1981	
50198	0.0534	0.261	1.5600	0.055	5	0.2048	
50197	0.0438	0.245	1.6100	0.056	4.893	0.2947	
50213	0.0623	0.307	1.7500	0.069	3.637	0.2837	
50202	0.0486	0.254	2.1000	0.055	5.632	0.2421	
50203	0.0674	0.276	2.1000	0.056	4.852	0.1551	
50156	0.0785	0.091	1.2300	0.018	3.619	0.0955	
50183	0.0835	0.179	1.4800	0.036	3.832	0.1349	
50187	0.0811	0.176	1.9500	0.036	4.059	0.1538	
50172	0.0726	0.150	3.2600	0.032	4.817	0.2060	
50222	0.0871	0.165	4.9600	0.036	4.202	0.2521	

TABLE 2 (continued)

Expt.no.	Equil. aq. molar concns.[a]				Distribution ratios		
	U(VI)	U(IV)	HNO_3	$N_2H_5NO_3$	U(VI)	U(IV)	HNO_3
50190	0.1020	0.188	0.7400	0.037	2.814	0.1101	
50205	0.0878	0.283	0.7800	0.056	3.200	0.1226	
50206	0.1156	0.296	0.7800	0.056	2.824	0.0823	
50209	0.0905	0.355	0.8400	0.070	3.381	0.1197	
50178	0.0760	0.213	0.9900	0.043	3.539	0.1533	
50218	0.1123	0.341	1.0100	0.067	2.917	0.1160	
50179	0.1020	0.223	1.0200	0.043	3.108	0.1081	
50217	0.0854	0.329	1.0500	0.066	3.361	0.1445	
50194	0.0795	0.282	1.0900	0.056	3.761	0.1309	
50195	0.1130	0.290	1.2100	0.056	3.004	0.0962	
50214	0.0823	0.324	1.4200	0.068	3.354	0.1970	
50215	0.1013	0.352	1.5600	0.071	3.248	0.1399	
50199	0.0787	0.265	1.5800	0.053	4.097	0.1309	
50175	0.0921	0.200	4.5500	0.042	4.017	0.1900	
50163	0.1628	0.104	0.7600	0.019	2.035	0.0548	
50157	0.1201	0.093	1.0700	0.017	2.732	0.0679	
50191	0.1420	0.191	0.7500	0.036	2.320	0.0763	
50207	0.1596	0.302	0.7800	0.056	2.337	0.0542	
50210	0.1216	0.366	0.8000	0.069	2.903	0.0738	
50211	0.1654	0.377	0.8100	0.069	2.366	0.0454	
50219	0.1465	0.345	1.0500	0.065	2.485	0.0803	
50200	0.1586	0.287	1.4600	0.053	2.491	0.0593	
50184	0.1593	0.194	1.5100	0.036	2.482	0.0612	
50216	0.1726	0.369	1.5700	0.069	2.370	0.0639	
50204	0.1460	0.295	2.0600	0.056	2.763	0.0708	
50188	0.1370	0.190	2.1000	0.036	2.927	0.0737	
50164	0.2832	0.103	0.7500	0.019	1.381	0.0261	
50224	0.3106	0.104	1.7400	0.019	1.442	0.0143	
50162	0.3016	0.102	1.8600	0.018	1.455	0.0217	
50173	0.2632	0.174	3.2600	0.033	1.660	0.0967	
50169	0.2377	0.111	3.5500	0.020	1.866	0.0533	
50192	0.2407	0.192	0.7800	0.035	1.635	0.0443	
50208	0.2850	0.307	0.7900	0.055	1.494	0.0229	
50212	0.2925	0.384	0.8400	0.069	1.497	0.0187	
50220	0.2504	0.338	1.0700	0.061	1.723	0.0284	
50196	0.2160	0.305	1.0900	0.056	1.921	0.0439	
50180	0.2590	0.249	1.3600	0.045	1.660	0.0297	
50223	0.2937	0.189	5.1700	0.036	1.530	0.0892	
50165	0.3970	0.103	0.7400	0.018	1.064	0.0165	
50158	0.3331	0.100	1.2400	0.018	1.308	0.0131	

TABLE 2 (continued)

Expt.no.	Equil. aq. molar concns.[a]				Distribution ratios		
	U(VI)	U(IV)	HNO_3	$N_2H_5NO_3$	U(VI)	U(IV)	HNO_3
50176	0.4230	0.173	4.7700	0.033	1.075	0.0924	
97	0.0097	0.051	0.0520	0.100		0.0990	
200	0.0003	0.006	0.0845	0.100		0.0410	
199	0.0018	0.038	0.0845	0.100		0.0800	0.144
198	0.0034	0.067	0.0883	0.100		0.1660	
206	0.0064	0.108	0.0902	0.100		0.3090	
205	0.0108	0.165	0.0912	0.100		0.4540	
197	0.0070	0.118	0.0984	0.100		0.3510	
93	0.0097	0.050	0.1406	0.100		0.1490	
94	0.0097	0.046	0.3544	0.100		0.2660	
236	0.0306	0.099	0.3610	0.116	3.830	0.2180	0.124
235	0.0130	0.093	0.3710	0.116	4.470	0.2970	0.150
95	0.0097	0.042	0.5441	0.100		0.3890	
302	0.0374	0.031	0.5780	0.105	4.030	0.2210	0.127
303	0.0300	0.074	0.5860	0.111	4.680	0.2580	0.126
304	0.0320	0.118	0.5860	0.116	4.860	0.2720	0.124
305	0.0352	0.159	0.6240	0.122	4.660	0.2720	0.103
96	0.0097	0.037	0.7781	0.100		0.5470	
188	0.0032	0.044	0.8260	0.100		0.6360	0.195
186	0.0051	0.071	0.8260	0.100		0.6360	0.189
184	0.0067	0.092	0.8260	0.100		0.6450	0.186
185	0.0087	0.120	0.8350	0.100		0.6500	0.187
183	0.0044	0.060	0.8400	0.100		0.6410	0.194
189	0.0124	0.170	0.8740	0.100		0.6600	0.152
187	0.0099	0.136	0.8830	0.100		0.6590	0.164
308	0.0206	0.037	0.9460	0.105	5.940	0.3760	0.146
309	0.0294	0.154	0.9960	0.122	5.720	0.3410	0.116
204	0.0151	0.221	0.1032	0.100		0.5260	
203	0.0220	0.315	0.1150	0.100		0.5520	
202	0.0169	0.249	0.1272	0.100		0.5350	
196	0.0173	0.257	0.1490	0.100		0.5340	
201	0.0239	0.355	0.1610	0.100		0.5310	
195	0.0230	0.340	0.1704	0.100		0.5330	
306	0.0357	0.207	0.6200	0.127	4.620	0.2790	0.107
307	0.0360	0.279	0.6380	0.133	4.900	0.2600	0.107
237	0.0516	0.101	0.3690	0.116	3.140	0.1630	0.100
254	0.0646	0.106	2.3500	0.116	5.480	0.1250	0.063
238	0.0726	0.106	0.3760	0.116	2.920	0.1180	0.089
252	0.1110	0.108	0.8340	0.116	2.830	0.0695	0.065
253	0.0951	0.110	1.3000	0.116	3.470	0.0855	0.062

TABLE 2 (continued)

Expt.no.	Equil. aq. molar concns.[a]				Distribution ratios		
	U(VI)	U(IV)	HNO_3	$N_2H_5NO_3$	U(VI)	U(IV)	HNO_3
250	0.1540	0.107	0.1780	0.116	1.810	0.0560	0.061
239	0.1340	0.113	0.3760	0.116	2.200	0.0567	0.064
251	0.1240	0.110	0.6100	0.116	2.470	0.0691	0.063
240	0.2180	0.116	0.3930	0.116	1.590	0.0301	0.048
335	0.2460	0.150	1.4600	0.159	1.840	0.0047	0.045
337	0.2300	0.148	2.6600	0.159	2.060	0.0130	0.037
241	0.4380	0.119	0.4180	0.116	0.976	0.0092	0.026
336	0.3420	0.137	2.0100	0.145	1.400	0.0080	0.031
99	0.0097	0.047	0.0453	0.200		0.1820	
98	0.0097	0.047	0.0896	0.200		0.1830	0.172
100	0.0097	0.047	0.1743	0.200		0.2130	
181	0.0020	0.038	0.2640	0.200		0.2670	0.162
101	0.0097	0.043	0.3823	0.200		0.3390	
310	0.0327	0.029	0.4710	0.205	4.050	0.2000	0.124
311	0.0345	0.159	0.4900	0.222	4.660	0.2760	0.106
102	0.0097	0.039	0.5662	0.200		0.4580	
103	0.0097	0.036	0.7752	0.200		0.6190	
180	0.0020	0.020	1.7950	0.200		1.4400	0.211
182	0.0025	0.019	3.0000	0.200		2.1300	0.198
347	0.1670	0.298	0.7810	0.212	2.480	0.0440	0.078
350	0.1580	0.298	1.0810	0.212	2.688	0.0525	0.060
344	0.1620	0.289	1.2240	0.212	2.710	0.0510	0.057
346	0.1490	0.298	1.5760	0.212	2.870	0.0460	0.055
349	0.1450	0.295	1.8250	0.212	3.020	0.0534	0.052
342	0.1490	0.295	1.9600	0.212	3.030	0.0330	0.050
348	0.1420	0.286	2.2150	0.212	3.070	0.0484	0.055
345	0.1420	0.291	2.7230	0.212	3.086	0.0617	0.049
386	0.1430	0.292	3.0000	0.212	3	0.0770	0.048
343	0.1440	0.286	3.5600	0.212	3.163	0.0970	0.045
385	0.1460	0.229	4.9000	0.212	2.840	0.1440	0.043
340	0.2570	0.150	0.8730	0.212	1.680	0.0120	0.085
339	0.2580	0.143	1.2500	0.212	1.750	0.0106	0.068
389	0.2260	0.311	0.5930	0.212	1.812	0.0305	0.061
390	0.2110	0.317	0.7410	0.212	2.060	0.0315	0.065
388	0.2050	0.332	0.8880	0.212	2	0.0367	0.056
387	0.1910	0.330	0.9020	0.212	2.230	0.0382	0.052
104	0.0097	0.037	0.0414	0.500		0.4890	
105	0.0097	0.039	0.1059	0.500		0.4480	

TABLE 2 (continued)

Expt.no.	Equil. aq. molar concns.[a]				Distribution ratios		
	U(VI)	U(IV)	HNO_3	$N_2H_5NO_3$	U(VI)	U(IV)	HNO_3
106	0.0097	0.036	0.1974	0.500		0.4660	
107	0.0097	0.036	0.3948	0.500		0.5850	
242	0.0082	0.082	0.4170	0.492	8.540	0.4450	0.147
243	0.0340	0.088	0.4220	0.492	3.765	0.3350	0.117
108	0.0097	0.031	0.7800	0.500		0.8390	
109	0.0097	0.026	1.0920	0.500		1.0100	
244	0.0532	0.103	0.4430	0.492	4.340	0.1560	0.082
327	0.1720	0.291	0.7900	0.584	2.396	0.0476	0.067
331	0.1656	0.296	1.0810	0.584	2.540	0.0553	0.047
324	0.1539	0.299	1.1940	0.584	2.713	0.0531	0.052
330	0.1530	0.295	1.3770	0.584	2.825	0.0659	0.060
326	0.1543	0.296	1.5240	0.584	2.763	0.0541	0.055
334	0.1476	0.298	1.6210	0.584	2.870	0.0596	0.054
322	0.1547	0.296	1.7730	0.584	2.733	0.0682	0.047
329	0.1623	0.296	1.7890	0.584	2.622	0.0598	0.054
332	0.1438	0.296	1.9190	0.584	2.942	0.0651	0.051
328	0.1430	0.270	2.2300	0.581	3.097	0.0710	0.048
325	0.1403	0.255	2.6260	0.577	3.069	0.0796	0.046
333	0.1284	0.239	3.0000	0.574	3.311	0.0899	0.046
323	0.1254	0.217	3.2500	0.571	3.332	0.0952	0.060
245	0.1990	0.116	0.4690	0.492	1.850	0.0302	0.039
246	0.4320	0.118	0.4850	0.492	1.024	0.0074	0.019
110	0.0097	0.026	0.0832	1.000		1.2000	
111	0.0097	0.027	0.1194	1.000		1.0400	
112	0.0097	0.028	0.2196	1.000		0.9580	
113	0.0097	0.029	0.3688	1.000		1.0000	
247	0.0050	0.079	0.3760	0.967	27.200	0.4750	0.105
248	0.0322	0.098	0.4070	0.967	7.380	0.2090	0.069
114	0.0097	0.026	0.6760	1.000		1.1800	
115	0.0097	0.025	0.9726	1.000		1.3300	
259	0.0556	0.096	1.8180	0.967	6.250	0.1510	0.057
255	0.1040	0.106	0.2360	0.967	3.170	0.0752	0.044
256	0.0941	0.110	0.4330	0.967	3.480	0.0819	0.048
257	0.0890	0.109	0.6590	0.967	3.820	0.0826	0.052
258	0.0828	0.107	0.8810	0.967	4.090	0.0939	0.052
249	0.1770	0.112	0.4230	0.967	2.140	0.0313	0.032

TABLE 2 (continued)

Expt.no.	Equil. aq. molar concns.[a]				Distribution ratios		
	U(VI)	U(IV)	HNO_3	$N_2H_5NO_3$	U(VI)	U(IV)	HNO_3
120	0.0097	0.013	0.1021	2.000		3.2700	
121	0.0097	0.014	0.1319	2.000		2.9700	
122	0.0097	0.016	0.2225	2.000.		2.4400	0.152
123	0.0097	0.016	0.4160	2.000		2.3100	0.188
124	0.0097	0.017	0.7049	2.000		2.3500	0.190
125	0.0097	0.015	1.1074	2.000		2.5500	0.194

[a] The U(VI) concentration is an intitial value, where no distribution ratio is given for U(VI) in the same line

TABLE 3

Distribution ratios of uranium(IV), uranium(VI), plutonium(III) and nitric acid at 25°C. Initial organic phase: a 30 vol.% solution of TBP in dodecane. Initial aqueous phase: a solution of uranium(IV) nitrate, plutonium(III) nitrate, hydrazine nitrate, nitric acid and, in some cases, uranyl nitrate.

Expt. no.	Equil. aq. molar concns.[a]					Distribution ratios			
	U(VI)	U(IV)	Pu(III)	HNO_3	$N_2H_5NO_3$	U(VI)	U(IV)	Pu(III)	HNO_3
34	0.0008	0.0300	0.0015	0.095	0.100		0.065		
33	0.0008	0.0365	0.0015	0.100	0.100		0.084		
30	0.0008	0.0348	0.0015	0.136	0.100		0.078		
83	nil	0.0169	0.0010	0.157	0.100		0.042		
89	nil	0.0382	0.0010	0.177	0.100		0.068		
86	nil	0.0295	0.0010	0.190	0.100		0.071		
31	0.0008	0.0341	0.0015	0.265	0.100		0.135		
84	nil	0.0160	0.0010	0.355	0.100		0.120		
32	0.0008	0.0324	0.0015	0.380	0.100		0.246		
90	nil	0.0350	0.0010	0.383	0.100		0.160		
87	nil	0.0280	0.0010	0.400	0.100		0.158		
49	0.0075	0.0170	0.0150	0.413	0.100		0.283		
35	0.0008	0.0292	0.0015	0.503	0.100		0.348		
85	nil	0.0152	0.0010	0.600	0.100		0.320		
88	nil	0.0302	0.0010	0.615	0.100		0.315		
46	0.0075	0.0323	0.0150	0.688	0.100		0.514		
312	0.0025	0.0319	0.0018	0.699	0.100	7.120	0.539	0.0854	0.205
54	0.0005	0.0227	0.0010	0.793	0.100		0.542		
45	0.0075	0.0260	0.0150	0.793	0.100		0.616		
44	0.0075	0.0400	0.0150	0.982	0.100		0.622		
55	0.0005	0.0292	0.0010	1.112	0.100		0.740		
47	0.0075	0.0360	0.0150	1.225	0.100		0.840		
51	0.0075	0.0299	0.0150	1.860	0.100		1.190		
36	0.0008	0.0660	0.0015	0.070	0.100		0.154		
39	0.0008	0.0660	0.0015	0.122	0.100		0.175		
91	nil	0.0540	0.0010	0.163	0.100		0.111		
40	0.0008	0.0586	0.0015	0.193	0.100		0.188		
37	0.0008	0.0641	0.0015	0.204	0.100		0.210		
43	0.0075	0.0562	0.0150	0.265	0.100		0.256		
42	0.0075	0.0536	0.0150	0.312	0.100		0.291		
38	0.0008	0.0588	0.0015	0.388	0.100		0.318		
92	nil	0.0490	0.0010	0.390	0.100		0.222		
48	0.0075	0.0485	0.0150	0.470	0.100		0.372		
41	0.0075	0.0475	0.0150	0.581	0.100		0.457		
50	0.0075	0.0445	0.0150	0.737	0.100		0.558		

TABLE 3 (continued)

Expt. no.	Equil. aq. molar concns.[a]					Distribution ratios			
	U(VI)	U(IV)	Pu(III)	HNO_3	$N_2H_5NO_3$	U(VI)	U(IV)	Pu(III)	HNO_3
315	0.0050	0.0561	0.0251	1.190	0.100	7.480	0.727	0.0714	0.205
317	0.0064	0.0516	0.0085	1.890	0.100	4.030	1.065	0.0229	0.185
318	0.0050	0.0464	0.0241	1.970	0.100	7.140	1.060	0.0318	0.202
279	0.0163	0.0962	0.0387	1.020	0.112	7.120	0.509	0.0284	0.152
313	0.0166	0.0547	0.0087	1.040	0.100	1.700	0.684	0.0314	0.180
314	0.0114	0.1161	0.0066	1.160	0.100	6.290	0.567	0.0218	0.160
316	0.0110	0.1174	0.0218	1.220	0.100	5.250	0.629	0.0192	0.181
320	0.0164	0.1174	0.0991	1.250	0.100	9.620	0.511	0.0208	0.126
191	0.0110	0.0891	0.0047	1.294	0.100		0.857		0.172
192	0.0140	0.0863	0.0117	1.333	0.100		0.904		0.170
282	0.0157	0.0918	0.0393	1.350	0.112	7.520	0.599	0.0255	0.147
53	0.0225	0.0348	0.0450	0.700	0.100		0.592		
52	0.0225	0.0332	0.0450	0.857	0.100		0.693		
193	0.0190	0.1052	0.0223	0.500	0.100		0.483		0.178
283	0.0237	0.0975	0.0392	0.622	0.112	5.170	0.357	0.0140	0.153
281	0.0220	0.0925	0.0387	0.767	0.112	5.850	0.384	0.0129	0.155
194	0.0190	0.0970	0.0223	0.842	0.100		0.655		0.185
280	0.0230	0.0887	0.0355	0.860	0.112	5.540	0.436	0.0211	0.144
285	0.0255	0.1190	0.0732	0.937	0.127	6.550	0.411	0.0170	0.173
288	0.0235	0.1230	0.0775	1.040	0.127	6.730	0.482	0.0097	0.154
319	0.0216	0.1084	0.0955	1.180	0.100	7.560	0.523	0.0480	0.122
289	0.0197	0.1240	0.0750	1.230	0.127	7.840	0.550	0.0200	0.128
190	0.0190	0.0814	0.0223	1.333	0.100		0.835		0.170
321	0.0206	0.1174	0.0973	1.390	0.100	8.730	0.499	0.0175	0.146
286	0.0187	0.1210	0.0750	1.390	0.127	8.360	0.600	0.0147	0.139
287	0.0313	0.1230	0.0763	0.759	0.127	5.370	0.392	0.0197	0.105
284	0.0312	0.1075	0.0392	1.260	0.112	6.850	0.360	0.0158	0.122
290	0.0363	0.1175	0.1525	1.530	0.127	7.880	0.243	0.0184	0.105

[a] The U(VI) concentration is an intitial value, where no distribution ratio is given for U(VI) in the same line

TABLE 4

Distribution ratios of uranium(VI) and nitric acid at 25°C. Initial organic phase: a 30 vol.% solution of TBP in dodecane. Initial aqueous phase: a solution of uranyl nitrate, hydrazine nitrate, nitric acid and, in two cases, uranium(IV) nitrate.

Expt.no.	Equil. aq. molar concns.				Distribution ratios	
	U(VI)	U(IV)	HNO_3	$N_2H_5NO_3$	U(VI)	HNO_3
299	0.0273	nil	0.556	0.100	3.700	0.1290
298	0.0278	nil	0.589	0.100	3.450	0.1310
300	0.0189	nil	0.961	0.100	5.890	0.1540
233	nil	nil	0.082	0.200		0.0970
234	nil	nil	0.910	0.200		0.2100
341	0.3920	0.1430	0.263	0.212	1.130	0.0380
338	0.7620	0.1050	0.473	0.206	0.648	
301	0.0286	nil	0.483	0.200	3.540	0.1230
270	0.0372	nil	0.081	0.507	2.823	0.0940
271	0.0332	nil	0.174	0.507	3.223	0.1080
278	0.0015	nil	0.411	0.507	8.133	0.1940
277	0.0099	nil	0.416	0.507	6.182	0.1630
276	0.0250	nil	0.431	0.507	4.680	0.1290
275	0.1360	nil	0.465	0.507	2.118	0.0532
274	0.5510	nil	0.480	0.507	0.793	0.0185
272	0.0143	nil	0.866	0.507	8.601	0.1450
273	0.0094	nil	1.740	0.507	13.936	0.1450
269	0.0260	nil	0.066	1.010	4.500	0.1420
351	0.0031	nil	0.068	1.062	8.355	0.2330
352	0.0031	nil	0.144	1.062	8.580	0.2330
268	0.0232	nil	0.160	1.010	5.129	0.1330
266	0.0006	nil	0.391	1.010	21.333	0.2170
265	0.0044	nil	0.411	1.010	14.591	0.1770
264	0.0163	nil	0.416	1.010	7.730	0.1310
263	0.1120	nil	0.455	1.010	2.714	0.0414
262	0.3030	nil	0.470	1.010	1.287	0.0226
353	0.0021	nil	0.473	1.062	13.250	0.2060
267	0.0138	nil	0.851	1.010	9.130	0.1500
354	0.0016	nil	1.043	1.062	17.530	0.2050
355	0.0009	nil	2.000	1.062	30.400	0.2020
356	0.0007	nil	2.932	1.062	39.100	0.1960

TABLE 4 (continued)

Expt.no.	Equil. aq. molar concns.				Distribution ratios	
	U(VI)	U(IV)	HNO_3	$N_2H_5NO_3$	U(VI)	HNO_3
357	0.0011	nil	0.449	2.124	29.200	0.1250
358	0.0010	nil	0.524	2.124	34.200	0.2170
359	0.0009	nil	1.006	2.124	35.900	0.2030
360	0.0006	nil	1.784	2.124	47.200	0.1950
361	0.0005	nil	2.532	2.124	57.600	0.1900

TABLE 5

Distribution ratios of uranium(IV) and nitric acid at temperatures (t, °C) other than 25°C. Initial organic phase: a 30 vol.% solution of TBP in dodecane. Initial aqueous phase: a solution of uranium(IV) nitrate, hydrazine nitrate, nitric acid and, in some cases, uranyl nitrate.

Expt. no.	Eqil. aq. molar concns.[a]				Distribution ratios		t
	U(VI)	U(IV)	HNO_3	$N_2H_5NO_3$	U(IV)	HNO_3	
207	nil	nil	0.083	0.200		0.119	15
214	nil	nil	0.905	0.200		0.225	15
210	0.0136	0.044	0.099	0.200	0.2490	0.000	15
225	0.0136	0.036	0.254	0.200	0.3380	0.184	15
218	0.0136	0.030	0.920	0.200	0.8210	0.208	15
222	0.0019	0.019	1.833	0.200	1.5100	0.209	15
229	0.0025	0.018	3.000	0.200	2.2400	0.199	15
136	nil	0.312	0.440	0.075	0.5080		28
143	nil	0.314	0.450	0.075	0.4586		28
144	nil	0.233	0.830	0.050	0.5622	0.157	28
137	nil	0.137	1.540	0.037	0.8613	0.149	28
138	nil	0.089	2.120	0.035	1.2081	0.160	28
141	nil	0.060	2.980	0.030	1.5401	0.161	28
142	nil	0.051	3.540	0.021	1.9804	0.155	28
140	nil	0.042	4.270	0.030	2.5178	0.148	28
129	nil	0.032	4.960	0.026	3.1563	0.133	26
139	nil	0.037	4.970	0.021	2.9973	0.135	28
145	nil	0.320	0.470	0.075	0.4969		31
146	nil	0.243	0.870	0.050	0.5597	0.138	31
147	nil	0.142	1.560	0.037	0.8592	0.167	31
149	nil	0.065	3.130	0.032	1.6923	0.157	31
148	nil	0.058	3.660	0.023	1.8621	0.134	31
150	nil	0.042	5.140	0.023	2.8266	0.130	31
215	nil	0.000	0.915	0.200		0.196	35
211	0.0136	0.047	0.100	0.200	0.1280		35
226	0.0136	0.039	0.261	0.200	0.2160	0.144	35
155	nil	0.349	0.460	0.079	0.4413	0.152	34
154	nil	0.255	0.860	0.052	0.5608	0.174	34
219	0.0136	0.033	0.943	0.200	0.6630	0.191	35
157	nil	0.156	1.760	0.037	0.8782	0.159	37
153	nil	0.132	1.860	0.032	0.9545	0.167	34
158	nil	0.097	2.730	0.028	1.3031	0.150	37

TABLE 5 (continued)

Expt. no.	Eqil. aq. molar concns.[a]				Distribution ratios		t
	U(VI)	U(IV)	HNO_3	$N_2H_5NO_3$	U(IV)	HNO_3	
151	nil	0.082	2.830	0.027	1.4095	0.152	34
230	0.0025	0.018	3.000	0.200	2.0900	0.202	35
160	nil	0.066	4.330	0.028	2.0575	0.132	37
156	nil	0.054	4.510	0.032	2.1852	0.133	34
159	nil	0.059	5.080	0.028	2.3946	0.120	37
152	nil	0.048	5.190	0.025	2.7292	0.129	34
164	nil	0.299	0.780	0.047	0.5017	0.141	40
163	nil	0.111	2.750	0.032	1.1802	0.142	40
165	nil	0.096	3.070	0.037	1.3613	0.137	40
161	nil	0.073	4.180	0.028	1.9456	0.134	40
162	nil	0.071	4.960	0.021	2.1530	0.115	40
208	nil	0.000	0.086	0.200		0.071	45
216	nil	0.000	0.931	0.200		0.188	45
212	0.0136	0.049	0.100	0.200	0.1010		45
227	0.0136	0.041	0.268	0.200	0.1770	0.132	45
220	0.0136	0.034	0.936	0.200	0.6220	0.179	45
169	nil	0.178	1.740	0.037	0.8034	0.132	43
223	0.0019	0.020	1.823	0.200	1.3200	0.205	45
166	nil	0.125	2.590	0.027	1.0960	0.131	43
231	0.0025	0.018	3.010	0.200	2.1100	0.195	45
167	nil	0.102	3.310	0.027	1.4608	0.139	43
168	nil	0.086	4.120	0.033	1.8203	0.126	43
209	nil	0.000	0.084	0.200		0.063	55
217	nil	0.000	0.939	0.200		0.181	55
213	0.0136	0.049	0.104	0.200	0.0780		55
228	0.0136	0.041	0.272	0.200	0.1540	0.124	55
221	0.0136	0.033	0.954	0.200	0.6170	0.175	55
224	0.0019	0.019	1.794	0.200	1.3900	0.211	55
232	0.0025	0.018	3.000	0.200	2.1400	0.198	55

a) The U(VI) concentrations are initial values