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Abstract

Own and published data on the extraction of Np(IV) and Np(VI) with tributyl phosphate (TBP) are gathered in the report. Purex process related data are solely considered. The scope of the gathering is limited to systems involving a 30 vol.% solution of TBP in an aliphatic diluent, and an aqueous nitrate solution. The concentration ranges in the equilibrium aqueous phase are 0.1 - 11 M HNO_3 , 0 - 210 g U(VI)/l und < 0.03 g Np(IV,VI)/l. The temperature range is 25 - 60°C. The distribution ratios of Np(IV) and Np(VI) were critically assessed and obviously erroneous values were indicated.

VERTEILUNGSDATEN VON NEPTUNIUM(IV,VI) IN BEZUG AUF DAS PUREX-VERFAHREN

Zusammenfassung

Der Bericht bietet eine Zusammenstellung von eigenen und veröffentlichten Daten über die Extraktion von Np(IV) and Np(VI) mit Tributylphosphat (TBP) unter den Bedingungen des Purex-Verfahrens. Die Daten sind auf Systeme beschränkt, die aus einer 30 vol.% Lösung von TBP in einem aliphatischen Verdünnungsmittel und aus einer wässrigen Nitratlösung bestehen. Die Konzentrationsbereiche in der wässrigen Phase im Verteilungsgleichgewicht sind 0.1 - 11 M HNO_3, 0 - 210 g U(VI)/l und < 0.03 g Np(IV,VI)/l. Der Temperaturbereich ist 25 - 60°C. Die Verteilungskoeffizienten von Np(IV) und Np(VI) wurden kritisch ausgewertet und diejenigen Werte wurden markiert, die als offensichtlich fehlerhaft erscheinen.

INTRODUCTION

Irradiated nuclear fuel from a ligh water cooled reactor can contain as much as 0.08 wt.% neptunium. After an appropriate cooling time it is exclusively present as the long-lived alpha nuclide ^{237}Np ($t_{½} = 2.2 \times 10^6$ y). The behaviour of neptunium in the Purex process can be rather complex [1,2], mainly due to its redox chemistry. Three valence states of neptunium, namely Np(IV,V,VI) are under appropriate conditions stable in nitric acid solutions. Two of them, namely Np(IV,VI) are extractable with tributyl phosphate (TBP). If the behaviour of neptunium in the Purex process is not appropriately controlled, it can distribute between several process streams. For two reasons it is highly desirable to prevent such a dispensation. First, both the uranium and plutonium products must be largely free of neptunium. Second, ^{237}Np is radiotoxic and waste management is facilitated if neptunium is directed to a single process stream, optimally to the high level waste.

Prediction and control of the neptunium behaviour in the Purex process require a mathematical model for the calculation of concentration profiles of neptunium in counter-current extractors. The model must be based on reliable distribution data of Np(IV,VI) in a system involving the Purex solvent, i.e. a 30 vol.% solution of TBP in a paraffinic diluent. A compilation of relevant distribution data has been given in [3], but the data are limited to room temperature with emphasis on aqueous nitric acid concentrations of 1 - 4 M. The validity range of the data corresponds to conventional conditions, under which U(VI) and Pu(IV) are simultaneously or individually extracted in the Purex process. A modification of the simultaneous extraction of U(VI) and Pu(IV) has recently been suggested [4], which prevents accumulation of plutonium in the extractor used. The extraction is performed at a nitric acid concentration of ~5 *M* and at temperature elevated to 50 - 55°C, contrary to conventional ~3 M nitric acid and 30 - 35°C. Neptunium can be effectively separated from U and Pu if it is electrolytically reduced to Np(IV,V) in the scrub section of the extractor. Np is transferred into the high level waste stream, in spite of partial reoxidation to Np(VI) [5]. Thus it became interesting to extend the knowledge of the extractability of Np(IV) and Np(VI) to higher nitric acid concentrations and to elevated temperature.

We obtained new distribution data on Np(IV,VI) under careful control of the valency state of neptunium. Together with published values they give a satisfactory picture of the Np(IV,VI) extractability, which has been presented in [6] in a generalized graphical form. In this report we give our new data in a numerical form, together with data from available literature. The general aim of this report is to provide a critical collection of distribution ratios of Np(IV,VI) valid under the conditions of the Purex process.

EXPERIMENTAL

Dodecane (Fluka, olefin free), and nitric acid and uranyl nitrate (Merck, reagent grade) were used as received. A 30 vol.% (1.096 M) solution of TBP in dodecane was used as a solvent. Before use, it was contacted with dilute solutions of sodium carbonate and nitric acid to remove dibutyl phosphate. ^{237}Np was purchased from the Isotope Division of Oak Ridge National Laboratory. Distribution ratios of neptunium were measured in batch experiments, in which equal volumes of the phases were contacted for ~30 min in a thermostated vessel. To keep the neptunium in the tetravalent or hexavalent states, 0.1 M ferrous sulfamate plus 0.125 M hydroxylammonium nitrate and 0.025 M ammonium hexanitratocerate(IV), respectively, were present in the initial aqueous phase. The phases were separated by gravity at temperature of the experiment, and aliquots were taken for analyses. The absolute chemical concentration of neptunium was < 0.001 M. Relative equilibrium concentrations of neptunium were determined by gamma spectrometry in measuring the intensity of the 86.5 keV line of ^{237}Np. Uranium was determined by an x-ray fluorescence method. Nitric acid was determined by alkalimetric titration after complexing U(VI) with a mixture of oxalic acid and potassium fluoride.

EVALUATION AND PRESENTATION OF THE DATA

Compilation of published data yielded 220 and 199 distribution ratios of Np(IV) ($D_{Np(IV)}$) and Np(VI) ($D_{Np(VI)}$), respectively. In our experimental work we gained 38 and 17 new values of $D_{Np(IV)}$ and $D_{Np(VI)}$ respectively. Some of the data scatter considerably, possibly due to the presence of Np(V) in experiments with Np(IV,VI), and a critical evaluation is necessary. To detect implausible values of $D_{Np(IV)}$ and $D_{Np(VI)}$, we assessed the distribution ratios as functions of the equilibrium concentrations of nitric acid and U(VI) in the aqueous phase and of temperature. A simple graphical assessment was not an appropriate method, because only a small part of the available distribution data was obtained in varying one single variable and keeping other variables constant. Consequently, at constant temperature only few distribution ratios could be plotted vs. the concentration of nitric acid at a constant U(VI) concentration and vice versa. Thus, the dependence of the $D_{Np(IV)}$ and $D_{Np(VI)}$ values on the above variables had to be described by suitable equations. Lack of information on activity coefficients prevented exact physico-chemical application of equilibrium constants of the respective extraction reactions. It was necessary to apply empirical model equations, which implicitly include variations of activity coefficients with concentration and temperature. Two types of equations were utilized. One of them is generally described in [7] and its application is illustrated in [8]. Equations of the another type are a part of an extensive mathematical model for calculation concentration profiles in counter-current extractors [9]. Parameters of the equations were optimized with available distribution ratios of Np(IV) and Np(VI). The fit beween experimental distribution ratios and values calculated from the equations with the optimized parameters was then

assessed. Those experimental distribution ratios were considered implausible which strongly differed from calculated values.

Since we compiled data from many sources, no strict criteria could be applied to the acceptability of the experimental distribution ratios. A distribution ratio of Np(VI) was taken as unreliable if the the experimental value differed as much as by $>40\%$ from the calculated value. Data on Np(IV) appeared to be of a worse quality, and a $>60\%$ difference between the experimental and calculated $D_{Np(IV)}$ values was taken as a criterion for reliability. Only those $D_{Np(IV)}$ and $D_{Np(VI)}$ values were considered unreliable, which exhibited the above deviations in the application of both types of model equations. Hence, 18 values of $D_{Np(IV)}$ and 5 values of $D_{Np(VI)}$ were suggested to be rejected, out of totally 281 values of $D_{Np(IV)}$ and 321 values of $D_{Np(VI)}$. Let us mention that model equations of the first type only (but the same criteria for reliability) were applied for the assessment of $D_{Np(IV)}$ and $D_{Np(IV)}$ values in [6]. This resulted in rejection of 23 $D_{Np(IV)}$ values.

It is worth of noticing that high differences between experimental distribution ratios and those calculated from the model equations are rather seldom. With e.g. equations of the first type, as many as 180, 225, and 266 experimental $D_{Np(IV)}$ values fit the calculation within $\leq 10\%$, $\leq 20\%$, and $\leq 40\%$ respectively (out of totally 281 measurements). Still better, as many as 245, 294, and 314 experimental $D_{Np(VI)}$ values fit the calculation within $\leq 10\%$, $\leq 20\%$, and $\leq 30\%$ respectively (out of totally 321 measurements).

All available distribution ratios of Np(IV) and Np(VI) are gathered in the Table. Implausible values are denoted by a minus sign. The data are divided into two parts. The 1st part includes all data obtained in the absence of uranium(IV), and the data are ordered according to increasing concentration of nitric acid. Data obtained in the presence of uranium(VI) are given in the 2nd part. The data are further divided into groups, each of them being valid for a limited range of the nitric acid concentration. In each group the data are ordered according to increasing concentration of uranium(VI). All concentrations are related to the equilibrium aqueous phase.

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TABLE

Distribution ratios of Np(IV) and Np(VI) at varied concentrations of nitric acid and U(VI) in the equilibrium aqueous phase (25 - 60°C)

Organic phase: 30 vol.% TBP in an alkane dilent

Aqueous phase: Nitric acid, uranyl nitrate and >0.0001 M neptunium(IV) or neptunyl nitrate

Part 1: No uranium(VI) present, 0.1 - 11 M nitric acid (25 - 60°C).

HNO_3 conc. mol/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
0.100	25	-0.0019		[10]
0.150	25		0.446	own data
0.160	25		0.495	own data
0.190	25		0.587	own data
0.200	25	0.0039		[10]
0.200	25	0.0038		[10]
0.200	25		0.620	[10]
0.200	25		0.575	[10]
0.300	25	0.0150		[10]
0.370	25		1.523	own data
0.400	25	0.0390		[10]
0.400	25	0.0390		[10]
0.430	25	0.0487		own data
0.490	25	0.0837		own data
0.490	25	-0.150		[11]
0.490	25	0.113		[11]
0.500	25	-0.260		[10]
0.500	25	-0.212		[10]
0.500	25		2.06	[10]
0.500	25		1.74	[10]
0.500	25	-0.200		[12]
0.500	25		2.20	[12]
0.510	25	0.1264		own data
0.530	25		2.28	own data
0.600	25	-0.395		[10]
0.600	25	-0.386		[10]
0.620	25	0.289		own data

Part 1 (continued)

HNO_3 conc. mol/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
0.710	25		3.78	[13]
0.710	25	0.409		own data
0.850	25	0.251		[14]
0.860	25	0.633		own data
0.870	25		4.47	[14]
0.890	25		4.17	own data
0.920	25	0.620		[11]
0.920	25	0.620		[11]
1.00	25	0.899		[10]
1.00	25	0.863		[10]
1.00	25		4.79	[10]
1.00	25		4.43	[10]
1.00	25	1.10		[16]
1.00	25	0.807		[12]
1.00	25	0.750		[12]
1.00	25	-1.40		[14]
1.00	25	-1.50		[16]
1.00	25	-0.24		[15]
1.00	25		5.30	[12]
1.00	25		4.30	[16]
1.00	25		4.10	[16]
1.00	25		4.40	[16]
1.00	25		4.50	[15]
1.10	25		6.21	[13]
1.11	25	0.929		own data
1.30	25	1.114		own data
1.50	25	1.51		[10]
1.50	25	1.413		[10]
1.50	25	1.213		[10]
1.50	25	1.233		[10]
1.50	25		7.52	[10]
1.50	25		8.37	[10]
1.66	25	1.096		[14]
1.66	25		10.20	[14]
1.73	25	2.05		[11]
1.74	25	1.62		[11]

Part 1 (continued)

HNO_3 conc. mol/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
2.00	25	2.00		[10]
2.00	25	1.96		[10]
2.00	25	1.92		[10]
2.00	25	2.384		[10]
2.00	25	2.46		[10]
2.00	25		9.64	[10]
2.00	25		9.12	[10]
2.00	25	2.00		[16]
2.00	25	2.04		[12]
2.00	25	2.10		[12]
2.00	25	2.26		[14]
2.00	25	2.04		[14]
2.00	25	2.30		[16]
2.00	25	-1.10		[15]
2.00	25		12.5	[12]
2.00	25		10.2	[15]
2.00	25		10.9	[13]
2.00	25		8.70	[16]
2.00	25		7.80	[16]
2.00	25		9.70	[16]
2.00	25		10.0	[15]
2.21	25		11.2	own data
3.00	25	3.33		[10]
2.54	25	2.14		[14]
2.54	25		15.1	[14]
2.57	25		13.3	[13]
3.00	25	3.00		[10]
3.00	25	3.09		[10]
3.00	25		14.2	[10]
3.00	25		14.35	[10]
3.00	25		14.7	[10]
3.00	25	3.10		[16]
3.00	25	3.42		[12]
3.00	25	3.50		[12]
3.00	25	2.95		[14]
3.00	25	3.50		[16]

Part 1 (continued)

HNO_3 conc. mol/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
3.00	25	2.20		[15]
3.00	25		17.0	[12]
3.00	25		14.7	[13]
3.00	25		13.3	[16]
3.00	25		10.9	[16]
3.00	25		13.5	[16]
3.00	25		15.0	[15]
3.19	25	3.41		own data
3.28	25	3.73		own data
3.39	25	3.55		[14]
3.39	25		14.8	own data
3.44	25	5.35		[11]
3.47	25		18.2	[14]
3.50	25	4.83		[12]
3.50	25		19.5	[12]
4.00	25	4.70		[16]
4.00	25	5.20		[12]
4.00	25	4.20		[14]
4.00	25	5.10		[16]
4.00	25	3.40		[15]
4.00	25		20.0	[12]
4.00	25		16.4	[13]
4.00	25		14.8	[16]
4.00	25		13.2	[16]
4.00	25		16.6	[16]
4.00	25		19.0	[15]
4.32	25	5.25		[14]
4.36	25		20.0	[14]
4.50	25		20.650	[16]
4.60	25		17.200	[13]
5.00	25	6.80		[16]
5.00	25	7.10		[12]
5.00	25	7.30		[16]
5.00	25	5.50		[15]
5.00	25		22.0	[12]
5.00	25		15.7	[16]

Part 1 (continued)

HNO_3 conc. mol/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
5.00	25		14.7	[16]
5.00	25		16.7	[16]
5.00	25		20.0	[15]
5.13	25	7.76		[14]
5.13	25		19.5	[14]
5.28	25	7.30		own data
5.36	25	7.37		own data
6.00	25	9.30		[16]
6.00	25	9.50		[12]
6.00	25	10.0		[16]
6.00	25	9.00		[15]
6.00	25		14.6	[16]
6.00	25		14.1	[16]
6.00	25		15.4	[16]
6.00	25		14.0	[15]
6.02	25		16.2	[14]
6.03	25	10.7		[14]
6.33	25	11.24		own data
7.00	25	12.6		[16]
7.00	25	12.0		[15]
7.08	25	13.5		[14]
7.94	25	13.33		[14]
7.94	25		10.0	[14]
8.00	25	14.5		[16]
8.00	25	15.0		[15]
9.00	25	14.0		[16]
9.00	25	14.0		[15]
9.02	25	11.22		[14]
9.12	25		8.13	[14]
10.0	25	11.0		[16]
10.0	25	13.0		[15]
10.0	25	9.77		[14]
10.0	25		5.62	[14]
10.96	25		5.13	[14]
11.0	25	7.76		[14]

Part 1 (continued)

HNO_3 conc. mol/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
1.02	45	0.800	3.60	[17]
1.95	45	2.340	7.91	[17]
2.98	45	4.0500	10.79	[17]
4.00	45	6.200	12.53	[17]
3.02	50	4.176		own data
3.28	50		7.46	own data
5.07	50	6.71		own data
6.54	50	8.24		own data
0.200	60		0.220	[10]
0.500	60		0.962	[10]
1.00	60		3.06	[10]
1.03	60	0.810	2.73	[17]
1.50	60		5.13	[10]
2.00	60		6.22	[10]
2.00	60	2.76	5.97	[17]
2.92	60	4.84	8.11	[17]
3.00	60		8.50	[10]
4.03	60	7.01	8.70	[17]

Part 2: Uranium(VI) present, 0.2 - 6.6 M nitric acid (25 - 60°C).

Concentrations of HNO_3 mol/l	U(VI) g/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
		0.200 - 0.214 M nitric acid			
0.200	1.35	25		0.610	[10]
0.200	2.50	25		0.623	[10]
0.201	6.10	25		0.703	[10]
0.202	10.8	25		0.757	[10]
0.204	21.4	25		0.796	[10]
0.208	41.7	25		0.749	[10]
0.211	69.2	25		0.637	[10]
0.212	99.2	25		0.524	[10]
0.214	137.0	25		0.414	[10]
0.200	3.60	60		0.261	[10]
0.200	7.84	60		0.287	[10]
0.202	14.3	60		0.329	[10]
0.204	27.7	60		0.331	[10]
0.208	50.8	60		0.327	[10]
0.211	78.3	60		0.368	[10]
0.212	109.0	60		0.288	[10]
0.214	143.0	60		0.285	[10]
		0.50 - 0.56 M nitric acid			
0.500	0.690	25		1.70	[10]
0.501	1.32	25	0.135		[10]
0.502	1.32	25		1.876	[10]
0.506	3.11	25	0.0625		[10]
0.506	3.11	25		1.81	[10]
0.510	6.44	25	0.0590		[10]
0.510	6.44	25		1.64	[10]
0.520	14.7	25	0.0330		[10]
0.520	14.7	25		1.37	[10]
0.535	34.0	25	0.0430		[10]
0.535	34.0	25		1.09	[10]

Part 2 (continued)

Concentrations of HNO_3 mol/l	U(VI) g/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
0.548	63.7	25	0.0350		[10]
0.548	63.7	25		0.666	[10]
0.548	63.7	25		0.732	[10]
0.555	96.7	25	0.0335		[10]
0.555	96.7	25		0.542	[10]
0.562	136.0	25	0.0295		[10]
0.562	136.0	25		0.400	[10]
0.502	2.18	60		0.951	[10]
0.505	5.17	60		0.857	[10]
0.508	9.06	60		0.845	[10]
0.518	19.7	60		0.829	[10]
0.532	41.1	60		0.675	[10]
0.544	69.9	60		0.535	[10]
0.553	99.3	60		0.378	[10]
0.560	138.0	60		0.299	[10]
		0.9 - 1.2 M nitric acid			
1.00	0.340	25		3.88	[10]
1.00	0.530	25		4.69	[10]
1.02	1.43	25		4.60	[10]
1.03	3.00	25		3.94	[10]
1.00	4.04	25	0.480		[12]
1.00	4.67	25		3.21	[12]
1.06	8.12	25		2.84	[10]
1.00	8.37	25	-0.376		[12]
1.00	8.41	25		2.62	[12]
0.900	8.50	25	0.190	2.45	[16]
0.900	8.50	25	0.190	2.41	[16]
1.11	24.0	25		1.70	[10]
0.930	25.5	25	0.100	1.60	[16]
0.930	25.5	25	0.0940	1.48	[16]
1.00	29.0	25	-0.200		[12]

Part 2 (continued)

Concentrations of HNO_3 mol/l	U(VI) g/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
1.00	30.5	25		1.26	[12]
1.00	42.8	25	-0.150		[12]
1.00	48.3	25		0.940	[12]
0.960	47.0	25	0.0580	1.07	[16]
0.960	47.0	25	0.0570	1.05	[16]
1.14	51.5	25		0.884	[10]
0.970	74.0	25	0.0460	0.710	[16]
0.970	74.0	25	0.0470	0.710	[16]
1.160	85.6	25		0.611	[10]
1.00	88.0	25	0.0650		[12]
1.00	96.8	25		0.540	[12]
0.980	108.0	25	0.0380	0.510	[16]
0.980	108.0	25	0.0380	0.500	[16]
1.00	121.2	25	0.0350		[12]
1.17	126.0	25		0.448	[10]
0.990	180.0	25	0.0350	0.270	[16]
0.990	180.0	25	0.0320	0.300	[16]
1.00	210.6	25		0.240	[12]
1.05	4.40	45	0.540	2.81	[17]
1.08	10.6	45	0.360	2.10	[17]
1.13	32.9	45	0.190	1.20	[17]
1.14	66.1	45	0.120	0.760	[17]
1.17	104.0	45	0.0870	0.520	[17]
1.18	162.0	45	0.0710	0.380	[17]
1.00	3.97	60		2.82	[10]
1.04	6.00	60	0.510	2.18	[17]
1.02	10.7	60		-2.82	[10]
1.07	14.0	60	0.400	1.68	[17]
1.03	18.1	60		-2.49	[10]
1.06	37.0	60		-1.85	[10]
1.11	37.8	60	0.260	1.05	[17]
1.10	65.3	60		-1.16	[10]
1.14	73.5	60	0.190	0.690	[17]

Part 2 (continued)

Concentrations of HNO_3 mol/l	U(VI) g/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
1.13	84.0	60		0.692	[10]
1.15	98.4	60		0.517	[10]
1.16	107.0	60		0.383	[10]
1.16	113.0	60	0.160	0.500	[17]
1.16	160.5	60	0.150	0.390	[17]
		1.5 - 1.8 M nitric acid			
1.50	0.160	25		8.04	[10]
1.50	0.350	25	1.193		[10]
1.50	0.350	25		7.41	[10]
1.53	0.940	25	1.08		[10]
1.63	0.940	25		6.72	[10]
1.55	2.00	25	0.933		[10]
1.55	2.00	25		5.83	[10]
1.78	4.20	25	0.560	4.26	[16]
1.78	4.20	25	0.550	3.81	[16]
1.60	5.16	25	0.617		[10]
1.60	5.16	25		4.46	[10]
1.68	16.8	25	0.225		[10]
1.68	16.8	25		2.444	[10]
1.75	42.2	25	0.106		[10]
1.75	42.2	25	0.112		[10]
1.75	42.2	25		1.27	[10]
1.75	42.2	25		1.045	[10]
1.78	76.6	25	0.0670		[10]
1.78	76.6	25		0.723	[10]
1.79	118.0	25	0.0480		[10]
1.79	118.0	25		0.495	[10]
1.51	0.540	60		4.79	[10]
1.53	1.26	60		4.24	[10]
1.55	2.90	60		3.97	[10]
1.60	7.60	60		2.565	[10]

Part 2 (continued)

Concentrations of HNO_3 mol/l	U(VI) g/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
1.67	22.9	60		1.746	[10]
1.74	49.8	60		0.915	[10]
1.77	86.7	60		0.570	[10]
1.78	125.0	60		0.414	[10]
		1.9 - 2.4 M nitric acid			
2.02	0.170	25		8.94	[10]
2.04	0.290	25	2.174		[10]
2.04	0.290	25		9.00	[10]
2.04	0.770	25	1.805		[10]
2.04	0.770	25		8.66	[10]
2.07	1.49	25	1.48		[10]
2.07	1.49	25		7.71	[10]
2.00	1.83	25		7.03	[12]
2.00	1.88	25	1.36		[12]
2.00	3.13	25	1.24		[12]
2.00	3.62	25		5.90	[12]
2.15	4.17	25	0.938		[10]
2.15	4.17	25		5.46	[10]
2.00	12.12	25	0.453		[12]
2.00	12.27	25		3.21	[12]
1.90	15.7	25	0.220	2.49	[16]
1.90	15.7	25	0.220	2.22	[16]
2.26	16.8	25	0.316		[10]
2.26	16.8	25		2.41	[10]
2.00	23.4	25	0.193		[12]
2.00	28.2	25		1.51	[12]
1.96	33.7	25	0.110	1.28	[16]
1.96	33.7	25	0.100	1.20	[16]
2.31	43.8	25	0.121		[10]
2.31	43.8	25		1.08	[10]
1.98	45.0	25	0.0880	1.07	[16]
1.98	45.0	25	0.0900	0.940	[16]

Part 2 (continued)

Concentrations of HNO_3 mol/l	U(VI) g/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
1.91	45.6	25		1.13	[10]
2.00	59.5	25		0.796	[12]
2.00	60.0	25	0.0730	0.830	[16]
2.00	60.0	25	0.0760	0.730	[16]
2.00	82.8	25	0.0630		[12]
2.35	83.9	25	0.0710		[10]
2.35	83.9	25		0.605	[10]
2.02	115.0	25	0.0580	0.560	[16]
2.02	115.0	25	0.0550	0.490	[16]
2.37	129.0	25	0.0500		[10]
2.37	129.0	25		0.418	[10]
2.00	178.3	25	0.0340		[12]
2.08	2.64	45	1.52	5.64	[17]
2.16	7.65	45	0.890	3.64	[17]
2.27	32.0	45	0.340	1.42	[17]
2.34	77.9	45	0.170	0.700	[17]
2.35	130.0	45	0.130	0.450	[17]
2.39	184.5	45	0.110	0.330	[17]
2.00	0.430	60		5.66	[10]
2.04	1.11	60		5.09	[10]
2.07	2.41	60		4.585	[10]
2.05	3.60	60	1.87	4.41	[17]
2.14	7.00	60		3.22	[10]
2.13	9.60	60	1.18	2.89	[17]
2.23	22.8	60		1.44	[10]
2.24	36.3	60	0.520	1.24	[17]
2.29	52.0	60		0.777	[10]
2.29	80.8	60	0.290	0.680	[17]
2.33	90.0	60		0.545	[10]
2.37	133.0	60		0.318	[10]

Part 2 (continued)

Concentrations of HNO_3 mol/l	U(VI) g/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
2.30	134.5	60	0.220	0.450	[17]
2.33	186.0	60	0.200	0.330	[17]
		2.7 - 3.1 M nitric acid			
3.01	0.100	25		13.4	[10]
3.02	0.180	25	2.92		[10]
3.02	0.160	25		13.34	[10]
3.02	0.180	25		13.75	[10]
3.05	0.430	25	2.70		[10]
3.05	0.480	25		12.25	[10]
3.05	0.430	25		13.25	[10]
3.10	0.940	25	2.23		[10]
3.10	0.940	25		11.5	[10]
3.10	1.10	25		10.7	[10]
3.00	1.19	25		11.6	[12]
3.00	1.19	25	2.995		[12]
3.00	2.36	25		8.03	[12]
3.00	2.85	25	-2.33		[12]
2.70	3.40	25	1.05	6.67	[16]
2.70	3.40	25	1.10	7.14	[16]
3.00	4.41	25	1.405		[12]
3.00	7.92	25		4.09	[12]
3.00	8.51	25	0.963		[12]
2.87	10.0	25	0.390	3.39	[16]
2.87	10.0	25	0.440	3.68	[16]
3.27	11.9	25		3.36	own data
3.00	16.4	25	-0.677		[12]
3.00	17.7	25		2.48	[12]
2.98	29.2	25	0.170	1.52	[16]
2.98	29.2	25	0.190	1.32	[16]
3.01	40.5	25	0.130	1.18	[16]
3.01	40.5	25	0.140	1.08	[16]
3.00	46.4	25	-0.373		[12]

Part 2 (continued)

Concentrations of HNO_3 mol/l	U(VI) g/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
3.00	48.4	25		0.977	[12]
3.03	62.0	25	0.100	0.830	[16]
3.03	62.0	25	0.100	0.730	[16]
3.00	72.7	25	0.138		[12]
3.05	101.2	25	0.0760	0.480	[16]
3.05	101.2	25	0.0880	0.420	[16]
3.00	182.0	25	0.0390		[12]
3.02	0.340	60		7.90	[10]
3.05	0.790	60		7.28	[10]
3.10	1.66	60		5.51	[10]
3.09	2.50	60	3.27	5.62	[17]
	3.2 - 3.8 M nitric acid				
3.50	0.940	25	3.93		[12]
3.50	1.04	25		12.1	[12]
3.52	2.08	25	2.01	9.47	[16]
3.50	2.14	25		9.08	[12]
3.50	2.21	25	-3.41		[12]
3.21	2.54	25	1.47		[10]
3.21	2.54	25		8.20	[10]
3.16	2.55	25		8.64	own data
3.21	2.69	25		7.65	[10]
3.38	4.00	25	1.74		own data
3.38	4.00	25	1.74		own data
3.73	4.50	25	1.33	6.63	[16]
3.50	7.21	25		5.24	[12]
3.50	7.92	25	1.20		[12]
3.38	11.2	25		3.37	[10]
3.38	11.2	25	0.559		[10]
3.38	13.4	25		2.82	[10]
3.50	13.8	25		2.90	[12]
3.50	14.5	25	0.568		[12]

Part 2 (continued)

Concentrations of HNO_3 mol/l	U(VI) g/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
3.50	35.9	25	0.220		[12]
3.50	38.0	25	0.198		[10]
3.50	38.0	25	0.186		[10]
3.50	38.0	25		1.22	[10]
3.50	38.0	25		1.136	[10]
3.50	41.5	25		1.09	[10]
3.50	50.3	25		1.05	[12]
3.50	70.2	25	0.0830		[12]
3.52	78.5	25		0.616	[10]
3.52	78.5	25	0.114		[10]
3.52	81.6	25		0.580	[10]
3.75	102.0	25	0.103		own data
3.70	104.0	25	0.107		own data
3.54	111.0	25	0.0910		[10]
3.54	111.0	25		0.400	[10]
3.54	123.0	25		0.400	[10]
3.36	124.0	25		0.410	own data
3.50	139.0	25		-0.193	[12]
3.39	191.0	25		0.235	own data
3.13	1.83	45	2.65	7.52	[17]
3.27	6.08	45	1.61	4.64	[17]
3.48	27.5	45	0.550	1.51	[17]
3.54	73.4	45	0.250	0.670	[17]
3.59	128.0	45	0.190	0.430	[17]
3.60	185.0	45	0.160	0.300	[17]
3.29	3.54	50	2.30		own data
3.16	3.92	50		4.67	own data
3.31	16.3	50		2.17	own data
3.51	92.6	50	0.292		own data
3.33	124.0	50		0.406	own data
3.42	191.0	50		0.246	own data

Part 2 (continued)

Concentrations of HNO_3 mol/l	U(VI) g/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
3.21	4.68	60		4.41	[10]
3.22	7.42	60	2.04	3.49	[17]
3.36	18.3	60		2.06	[10]
3.42	32.9	60	0.810	1.27	[17]
3.44	47.5	60		0.905	[10]
3.51	81.0	60	0.420	0.650	[17]
3.49	85.1	60		0.515	[10]
3.53	130.0	60		0.354	[10]
3.54	139.7	60	0.310	0.410	[17]
3.56	183.5	60	0.280	0.310	[17]
	3.9 - 5.3 M nitric acid				
5.28	1.71	25	3.22		own data
3.88	8.50	25	0.840	4.24	[16]
5.24	9.88	25	1.0175		own data
3.90	16.0	25	0.510	2.42	[16]
3.95	26.5	25	0.320	1.48	[16]
4.87	34.3	25	0.386		own data
3.99	53.0	25	0.230	1.01	[16]
5.07	71.2	25	0.236		own data
5.19	143.0	25	0.156		own data
4.79	189.0	25	0.1294		own data
4.09	1.64	45	4.12	8.38	[17]
4.26	5.21	45	2.48	4.99	[17]
4.53	30.6	45	0.790	1.36	[17]
4.70	78.6	45	0.400	0.640	[17]
4.73	129.0	45	0.290	0.400	[17]
4.74	193.0	45	0.260	0.290	[17]
4.99	3.51	50	3.56		own data
4.95	16.1	50	1.417		own data
4.83	43.3	50	0.697		own data

Part 2 (continued)

Concentrations of HNO_3 mol/l	U(VI) g/l	Temp. °C	Distr. ratio of Np(IV)	Np(VI)	Ref.
4.95	78.2	50	0.488		own data
5.07	154.0	50	0.381		own data
4.75	203	50	0.329		own data
4.15	2.49	60	4.71	5.87	[17]
4.22	7.35	60	2.96	3.34	[17]
4.51	35.2	60	1.12	1.16	[17]
4.59	82.0	60	0.620	0.580	[17]
4.63	135.5	60	0.480	0.380	[17]
4.65	192.0	60	0.410	0.300	[17]
	6.3 - 6.7 M nitric acid				
6.54	9.91	25	2.03		own data
6.70	35.2	25	0.888		own data
6.54	76.8	25	0.515		own data
6.41	122.0	25	0.342		own data
6.62	19.3	50	2.22		own data
6.54	49.7	50	1.113		own data
6.49	88.7	50	0.761		own data
6.33	136.0	50	0.520		own data