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TECHNICAL MATERIAL

Distribution Coefficients of U(VI), Nitric Acid and FP Elements in Extractions from Concentrated Aqueous Solutions of Nitrates by 30% Tri-*n*-butylphosphate Solution

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The distribution coefficients of uranium, nitric acid and 8 elements simulating fission products were obtained from the concentrated aqueous solutions of nitrates, whose nitrate ion concentrations were 4–15 mol dm⁻³. The relationships among the distribution coefficient of uranium, the nitrate ion concentration in the aqueous phase, and the concentration of tri-*n*-butylphosphate uncombined with nitrates in the organic phase were obtained from the data.

KEYWORDS: *distribution coefficients, uranium, FP elements, nitrate ion, tri-*n*-butylphosphate, nitric acid*

I. Introduction

A direct extraction method using supercritical carbon dioxide (SC-CO₂), Super-DIREX, has been developed as a new reprocessing process for spent nuclear fuels.^{1–7)} In this process, a complex of tri-*n*-butylphosphate (TBP) chemically complexed with nitric acid is used to dissolve uranium in the SC-CO₂ extraction media. Uranium oxides in the fuels are converted into a chemical complex of TBP and uranyl nitrate by the TBP-nitric acid complex, and directly dissolved in SC-CO₂.

The reaction of uranium oxide and the TBP-nitric acid complex in SC-CO₂ was considered as follows.⁸⁾ The complex forms reversed micelles in SC-CO₂. Uranium oxide in the fuel reacts with nitric acid contained in the water cores of the micelles and converts into uranyl nitrate in the water cores. The generated uranyl nitrate is complexed with TBP surrounding the cores. The acidity of the initial cores is expected to be greater than 12 mol dm⁻³. Considering that the PUREX process, which uses a bulk 3 mol dm⁻³ nitric acid solution as the aqueous phase, the nitrate ion concentration in the water cores in the Super-DIREX is expected to be greater than that in the aqueous phase employed in the PUREX.

For operation of the extraction processes, the distribution coefficient of each element contained in the spent fuels is one of the most important variables as a fundamental data. As for the uranium extraction systems with TBP and *n*-dodecane, a huge number of distribution coefficients for

PUREX were obtained under conditions in which the aqueous nitrate ion concentrations were lower than 5.4 mol dm⁻³, and their correlations were presented in the literature.^{9–13)} However, the extraction conditions of the Super-DIREX are different from those of the PUREX, especially the nitrate ion concentration is higher, as mentioned in Ref. 8). Moreover, the distribution coefficients of the fission product (FP) elements were not reported at the high nitrate ion concentration. In this study, we aimed to obtain the distribution coefficients of uranium, nitric acid and 8 elements simulating the FP elements when the nitrate ion concentrations in the aqueous phase were relatively high, *i.e.*, 4–15 mol dm⁻³. Neodymium, europium, cesium, *etc.* were employed as the FP elements, and *n*-dodecane was employed as the extraction medium instead of SC-CO₂. Although the distribution coefficients differ with the type of diluent, the distribution trends for the effects of the conditions, such as nitrate ion concentration and TBP concentration, are considered similar.¹⁴⁾ Among diluents, *n*-dodecane was chosen, and the trend of the uranium distribution coefficient obtained under high nitrate ion concentration was compared with that obtained in the PUREX. Most data were obtained at 333 K, which was the operating temperature of the Super-DIREX, and data at 313 and 323 K were also involved in this study.

II. Experimental

1. Chemicals

Uranyl nitrate was prepared by dissolving uranium dioxide powder in nitric acid, followed by crystallization. Eight reagents of neodymium nitrate hexahydrate, cerium(III) nitrate hexahydrate, europium nitrate hexahydrate, cesium

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nitrate, zirconium(IV) chloride, molybdenum(V) chloride, ruthenium nitrosyl nitrate, and rhenium(V) chloride were used to prepare the aqueous phase in this study. All the reagents were of analytical grade. The reagent of rhenium(V) chloride was purchased from Rare Metallic Co., Ltd., while the other reagents were purchased from Wako Pure Chemical Ind., Ltd. Nitric acid, TBP and *n*-dodecane were of special grade and purchased from Wako Pure Chemical Ind., Ltd.

2. Procedure

Nitrate solutions containing various amounts of uranyl nitrate, nitric acid, and the other nitrates are mixed well with 30 vol% TBP diluted with *n*-dodecane at constant temperatures. After extraction equilibrium was achieved, between the two phases, the organic and aqueous phases were separated using a centrifuge (Asone, C-12). The temperature of the solution was kept at constant at 313, 323 or 333 K until the phase separation.

Aliquots of each phase were taken for analyses of the density, nitric acid concentration, and concentrations of the metals. The densities were measured by a precision digital balance (A&D, GR-202) with an uncertainty of ± 0.0001 g and a calibrated micropipette. The concentrations of nitric acid were determined by an acid-base titration method with 0.01 mol dm^{-3} NaOH purchased from Wako Pure Chemical Ind., Ltd. Ammonium sulfate (10 wt% in water) was used for masking uranyl during titration so as to minimize the disturbance of uranyl ion on titration. The concentrations of metals in both phases were determined by inductively coupled plasma atomic emission spectrometry (ICP-AES, Shimadzu, ICPS-7000) and inductively coupled plasma mass spectrometry (ICP-MS, SII, SPQ9700). As for the organic phase, the water content was measured by a Karl Fischer titrator (Hiranuma, AQ-7).

III. Results and Discussion

The obtained distribution coefficients are shown along with the concentration of each element in the two phases in **Tables 1–17**. The definition of the distribution coefficient D is

$$D = \frac{C_{i,o}}{C_{i,a}}, \quad (1)$$

where C_i is the molar concentration of the i -th solute in mol dm^{-3} , and the subscript a or o represents the aqueous and organic phase, respectively. The densities and water content in the organic phase obtained in each experiment are summarized in **Table 18**. All the distribution coefficients of FP elements were smaller than that of uranium, as known in the PUREX in general.¹⁴⁾ The data in Tables 16 and 17 were obtained from the same solution with Run 7 with different temperatures of 313 and 323 K, respectively. The uranium distribution coefficient increased with decreasing temperature.

The extractions of nitric acid and uranyl nitrate by TBP are modeled by the following extraction equilibrium equations:^{11–13)}

Table 1 Distribution coefficients at 333 K obtained by Run 1

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm^{-3}	mol dm^{-3}	
HNO ₃	0.62	3.9	0.16
U	0.11	0.0095	12
Cs	0.000052	0.039	0.00013
Zr	0.000051	0.0021	0.024
Mo	N.D.*	0.00091	—
Re	0.000047	0.0014	0.034
Ru	0.00011	0.045	0.0024
Nd	0.000070	0.012	0.0058
Ce(III)	0.00017	0.036	0.0047
Eu	0.000062	0.0050	0.012

*N.D.: not detected

Table 2 Distribution coefficients at 333 K obtained by Run 2

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm^{-3}	mol dm^{-3}	
HNO ₃	0.71	4.5	0.16
U	0.12	0.010	12
Cs	0.00087	N.M.*	—
Zr	0.000025	0.00054	0.046
Mo	0.00020	0.0014	0.14
Re	0.000012	0.00048	0.025
Ru	0.00017	0.034	0.0050
Nd	0.00024	0.011	0.022
Ce(III)	0.0017	0.029	0.059
Eu	0.000071	0.0047	0.015

*N.M.: not measured

Table 3 Distribution coefficients at 333 K obtained by Run 3

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm^{-3}	mol dm^{-3}	
HNO ₃	0.78	6.7	0.12
U	0.12	0.013	9.2
Cs	0.0000017	0.037	0.000046
Zr	0.000040	0.00076	0.053
Mo	N.D.*	0.0016	—
Re	0.000011	0.00050	0.022
Ru	0.000023	0.040	0.00058
Nd	0.000039	0.013	0.0030
Ce(III)	0.00045	0.034	0.013
Eu	0.000044	0.0055	0.0080

*N.D.: not detected

Table 4 Distribution coefficients at 333 K obtained by Run 4

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm ⁻³	mol dm ⁻³	
HNO ₃	0.80	7.6	0.11
U	0.12	0.015	8.0
Cs	0.000030	0.041	0.000073
Zr	0.000046	0.00056	0.082
Mo	0.000023	0.0018	0.0013
Re	0.000012	0.00063	0.019
Ru	0.000024	0.046	0.00052
Nd	0.00013	0.015	0.0087
Ce(III)	0.00081	0.041	0.020
Eu	0.000055	0.0065	0.0085

Table 5 Distribution coefficients at 333 K obtained by Run 5

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm ⁻³	mol dm ⁻³	
HNO ₃	0.87	8.7	0.10
U	0.11	0.022	5.0
Cs	0.00011	0.031	0.0035
Zr	N.D.*	0.00054	—
Mo	N.D.*	0.0016	—
Re	0.00014	0.00053	0.26
Ru	0.00021	0.039	0.0054
Nd	0.000098	0.013	0.0075
Ce(III)	0.00078	0.035	0.022
Eu	0.00072	0.0055	0.13

*N.D.: not detected

Table 6 Distribution coefficients at 333 K obtained by Run 6

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm ⁻³	mol dm ⁻³	
HNO ₃	0.79	8.4	0.094
U	0.13	0.030	4.3
Cs	0.000046	0.028	0.0016
Zr	0.0013	0.00080	1.6
Mo	0.000048	0.0030	0.016
Re	0.000013	0.00062	0.021
Ru	0.000091	0.054	0.0017
Nd	0.00019	0.016	0.012
Ce(III)	0.000064	0.043	0.0015
Eu	0.000040	0.015	0.0027

Table 7 Distribution coefficients at 333 K obtained by Run 7

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm ⁻³	mol dm ⁻³	
HNO ₃	0.72	8.5	0.085
U	0.14	0.046	3.0
Cs	0.000057	0.034	0.0017
Zr	0.00061	0.00089	0.69
Mo	0.00034	0.0036	0.094
Re	0.000019	0.00072	0.026
Ru	0.0018	0.064	0.028
Nd	0.00031	0.019	0.016
Ce(III)	0.000062	0.053	0.0012
Eu	0.000031	0.0096	0.0032

Table 8 Distribution coefficients at 333 K obtained by Run 8

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm ⁻³	mol dm ⁻³	
HNO ₃	0.87	9.5	0.092
U	0.13	0.036	3.6
Cs	0.000020	0.028	0.00071
Zr	0.00024	0.00080	0.30
Mo	0.00011	0.0027	0.041
Re	0.000017	0.00062	0.027
Ru	0.000041	0.049	0.00084
Nd	0.000043	0.015	0.0029
Ce(III)	0.000064	0.041	0.0016
Eu	0.000049	0.0074	0.0066

Table 9 Distribution coefficients at 333 K obtained by Run 9

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm ⁻³	mol dm ⁻³	
HNO ₃	0.52	7.8	0.067
U	0.21	0.052	4.0
Cs	—	0.044	—
Zr	0.00084	0.0026	0.32
Mo	0.0000073	0.0050	0.0015
Re	0.000036	0.0012	0.030
Ru	0.0000085	0.082	0.00010
Nd	0.000019	0.025	0.00076
Ce(III)	0.000019	0.068	0.00028
Eu	0.000020	0.013	0.0015

Table 10 Distribution coefficients at 333 K obtained by Run 10

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm ⁻³	mol dm ⁻³	
HNO ₃	0.38	6.5	0.058
U	0.27	0.11	2.5
Cs	0.000032	0.071	0.00045
Zr	0.0027	0.0038	0.71
Mo	0.000050	0.0069	0.0072
Re	0.00031	0.0017	0.18
Ru	0.000038	0.11	0.00035
Nd	0.000084	0.035	0.0024
Ce(III)	0.000080	0.095	0.00084
Eu	0.000065	0.018	0.0036

Table 11 Distribution coefficients at 333 K obtained by Run 11

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm ⁻³	mol dm ⁻³	
HNO ₃	0.17	3.3	0.052
U	0.40	0.085	4.7
Zr	0.0022	0.080	0.028
Mo	0.00023	0.10	0.0023
Re	0.00095	0.0091	0.10
Ru	0.00032	0.88	0.00036
Nd	0.00038	0.47	0.00081
Ce(III)	0.00022	0.47	0.00047
Eu	0.00091	0.58	0.0016

Table 12 Distribution coefficients at 333 K obtained by Run 12

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm ⁻³	mol dm ⁻³	
HNO ₃	0.17	4.2	0.040
U	0.43	0.078	5.5
Zr	0.0044	0.15	0.029
Mo	0.00038	0.13	0.0029
Re	0.0015	0.012	0.13
Ru	0.00021	1.1	0.00019
Nd	0.00031	0.54	0.00057
Ce(III)	N.D.*	0.57	—
Eu	0.00084	0.67	0.0013

*N.D.: not detected

Table 13 Distribution coefficients at 333 K obtained by Run 13

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm ⁻³	mol dm ⁻³	
HNO ₃	0.078	4.3	0.018
U	0.39	0.049	8.0
Zr	0.0018	0.076	0.024
Mo	0.00019	0.092	0.0021
Re	0.0014	0.010	0.14
Ru	0.00048	0.83	0.00058
Nd	0.00074	0.36	0.0021
Ce(III)	0.00034	0.39	0.00087
Eu	0.0017	0.40	0.0043

Table 14 Distribution coefficients at 333 K obtained by Run 14

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm ⁻³	mol dm ⁻³	
HNO ₃	0.08	3.6	0.02
U	0.39	0.057	6.8
Zr	0.0011	0.076	0.014
Mo	0.00012	0.084	0.0014
Re	0.0012	0.010	0.12
Ru	0.0010	0.82	0.0012
Nd	0.0016	0.40	0.0040
Ce(III)	0.00070	0.39	0.0018
Eu	0.0040	0.43	0.0093

Table 15 Distribution coefficients at 333 K obtained by Run 15

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm ⁻³	mol dm ⁻³	
HNO ₃	0.17	3.1	0.055
U	0.39	0.10	3.9
Zr	0.00066	0.076	0.0087
Mo	0.000089	0.092	0.00097
Re	0.00074	0.010	0.074
Ru	0.00010	0.45	0.00022
Nd	0.00017	0.19	0.00090
Ce(III)	0.00019	0.43	0.00044
Eu	0.00007	0.046	0.0015

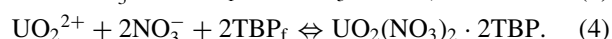
Table 16 Distribution coefficients at 313 K obtained by Run 16

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm ⁻³	mol dm ⁻³	
HNO ₃	0.80	10.7	0.075
U	0.14	0.030	4.7
Cs	N.D.*	0.033	—
Zr	0.00065	0.0012	0.54
Mo	0.000070	0.0034	0.021
Re	0.000017	0.00082	0.021
Ru	0.000020	0.057	0.00035
Nd	0.000058	0.017	0.0034
Ce(III)	0.000056	0.048	0.0012
Eu	0.000065	0.0086	0.0076

*N.D.: not detected

Table 17 Distribution coefficients at 323 K obtained by Run 17

	Concentration		Distribution coefficient
	Organic phase	Aqueous phase	
	mol dm ⁻³	mol dm ⁻³	
HNO ₃	0.83	10.4	0.080
U	0.12	0.031	3.9
Cs	0.0000050	0.030	0.00017
Zr	0.0013	0.00093	1.4
Mo	0.000047	0.0029	0.016
Re	0.000013	0.00072	0.018
Ru	0.000090	0.050	0.0018
Nd	0.00019	0.014	0.014
Ce(III)	0.000063	0.040	0.0016
Eu	0.000039	0.0074	0.0053



The equilibrium constants for Eqs. (2)–(4) can be represented as

$$K_{H1} = \frac{[\text{HNO}_3 \cdot \text{TBP}]}{[\text{H}^+][\text{NO}_3^-]T_f}, \quad (5)$$

$$K_{H2} = \frac{[\text{HNO}_3 \cdot 2\text{TBP}]}{[\text{H}^+][\text{NO}_3^-]T_f^2}, \quad (6)$$

$$K_U = \frac{[\text{UO}_2(\text{NO}_3)_2 \cdot 2\text{TBP}]}{[\text{UO}_2^{2+}][\text{NO}_3^-]^2T_f^2}, \quad (7)$$

where T_f is the concentration of the uncombined TBP. From Eq. (7), the distribution coefficient of uranium is found to be affected by two parameters, the nitrate ion concentration in the aqueous phase and the concentration of TBP uncombined with nitrates in the organic phase as

Table 18 Densities and water content in organic phase

Run No.	Density		H ₂ O content in organic phase
	Organic phase	Aqueous phase	
	g cm ⁻³	g cm ⁻³	
1	0.822	1.096	0.107
2	0.870	1.00	0.156
3	0.886	1.21	0.211
4	0.8786	1.35	0.298
5	0.843	1.27	0.204
6	0.842	1.27	0.224
7	0.848	1.28	0.150
8	0.847	1.27	0.190
9	0.839	1.24	0.144
10	0.868	1.27	0.152
11	0.93	1.5	0.13
12	0.94	1.6	0.13
13	0.92	1.6	0.1
14	0.93	1.5	0.1
15	0.9	1.4	0.14
16	0.845	1.35	0.268
17	0.850	1.30	0.347

$$\frac{D}{T_f^2} = f([\text{NO}_3^-]). \quad (8)$$

The two parameters are estimated using

$$[\text{NO}_3^-] = \sum_i f_i C_{i,a}, \quad (9)$$

$$T_f = T_0 - 2(K_{H2}C_{\text{HNO}_3,a} + K_U C_{U,a}) \cdot T_f^2 - K_{H1}C_{\text{HNO}_3,a}T_f, \quad (10)$$

where f_i is the molar ratio of the i -th solute to the nitrate ion, for example, unity for the nitric acid and two for the uranyl nitrate, and T_0 is the total TBP concentration in mol dm⁻³. According to the previous data compiled for PUREX, the following estimations were presented for equilibrium constants:¹³⁾

$$K_{H1} = (0.1416[\text{NO}_3^-]^{0.6724} + 0.006058[\text{NO}_3^-]^{3.418})(1 - 0.54e^{-15F}), \quad (11)$$

$$K_{H2} = K_{H1}, \quad (12)$$

$$K_U = (5.284[\text{NO}_3^-]^{1.582} + 1.557[\text{NO}_3^-]^{3.843} + 0.01267[\text{NO}_3^-]^{7.447})(4F^{-0.17} - 3), \quad (13)$$

where F is the volume fraction of TBP. The total TBP concentration is calculated as

$$T_0 = 3.6538F. \quad (14)$$

The concentrations of the nitrate ion and uncombined TBP calculated using the above equations are summarized in **Table 19**.

Table 19 Concentrations of nitrate ion and uncombined TBP

Run No.	[NO ₃ ⁻] _{aq}	T _f
	mol dm ⁻³	mol dm ⁻³
1	4.22	0.0978
2	4.76	0.0671
3	6.98	0.0167
4	7.98	0.00982
5	9.08	0.00534
6	8.86	0.00537
7	9.03	0.00429
8	9.89	0.00328
9	8.54	0.00511
10	7.62	0.00586
11	11.6	0.0016
12	15.0	0.00065
13	12.4	0.0016
14	12.2	0.0016
15	7.5	0.0073
16	11.2	0.00217
17	10.8	0.00243

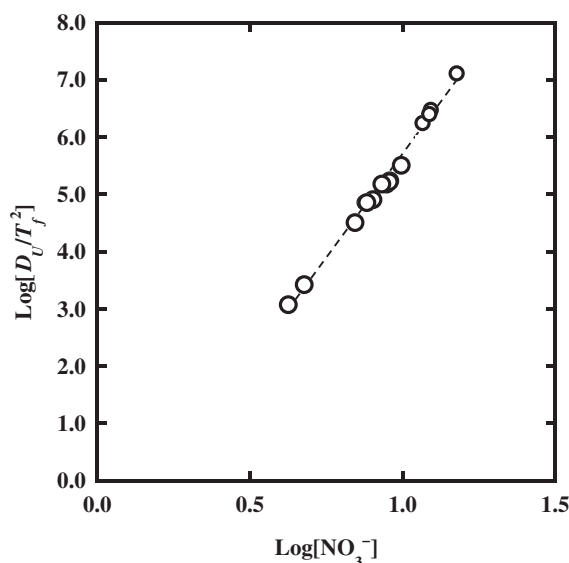
**Fig. 1** Uranium distribution coefficient at 333 K

Figure 1 shows a double logarithmic plot of the uranium distribution coefficient at 333 K with the nitrate ion and uncombined TBP concentrations according to Eq. (2). Even for the high nitrate ion concentration conditions, a linear relationship was confirmed. This means that the extraction characteristics under high nitrate ion concentration conditions are considered to be similar to those under low nitrate ion concentration conditions. Moreover, the effect of the presence of FP elements on the uranium distribution was negligible under the present condition because of their small distribution coefficients.

IV. Conclusion

The distribution coefficient data for uranium, nitric acid and 8 elements, neodymium, cerium(III), europium, cesium, zirconium(IV), molybdenum(V), ruthenium and rhenium(V), which simulate the FP elements in the spent fuels, were obtained from the concentrated aqueous solutions of nitrates, whose nitrate ion concentrations were 4–15 mol dm⁻³. The relationships among the distribution coefficient of uranium, the nitrate ion concentration in the aqueous phase, and the concentration of TBP uncombined with nitrates in the organic phase were obtained from the data.

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