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TITLE

AMINE EXTRACTION OF PLUTONIUM FROM NITRIC ACID SOLUTIONS

LOADING AND STRIPPING EXPERIMENTS

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AMINE EXTRACTION OF PLUTONIUM FROM NITRIC ACID SOLUTIONS

LOADING AND STRIPPING EXPERIMENTS

by

A. S. Wilson

Chemical Research
Chemical Research and Development
HANFORD LABORATORIES

January 19, 1961

HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND, WASHINGTON

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The information in this document has been extracted from the monthly reports of the author during a search (1957-1958) for a suitable amine processing system for plutonium nitrate.[1] These experiments with concentrated plutonium nitrate solutions show that trilaurylamine (TLA) plus an aromatic diluent, xylene, was the solvent system which did not form a second organic phase. Experiments are also reported with tri-n-octylamine (TnOA)-xylene and TLA-Amsco, 2 volume percent normal octyl alcohol. Second organic phases appear in both of these systems at high plutonium nitrate concentrations.

Distribution experiments with plutonium(VI) nitric acid - perchloric acid mixtures indicate that perchloric acid suppresses the extraction of plutonium.

Loading Experiments

These experiments were conducted by contacting equal volumes of the aqueous and organic solutions of interest for three to five minutes. The phases were allowed to separate and then were sampled and the plutonium determined by alpha counting. The organic solutions were pre-equilibrated with an aqueous phase containing all constituents of interest except the plutonium. The results are presented in Tables I, II, III, and IV.

Stripping Experiments

The stripping of plutonium from the amine solutions has generally been accomplished by reducing the plutonium(IV) to plutonium(III). Other methods would include a dilution of the amine solution by the addition of diluent and the use of an acid or anion which would react more strongly with the alkyl ammonium ion than does the plutonium species. The former procedure may be satisfactory for analytical procedures, but would probably not be acceptable for process work. If a suitable acid or anion could be found, the latter procedure would be acceptable. In this regard the extraction of plutonium from nitric-perchloric acid mixtures was examined. The results are in Table V. It is seen that perchloric acid profoundly effects the extraction of plutonium. Other anions which have a high affinity for the alkyl ammonium ions would also be expected to be effective.

(1) A. S. Wilson, "Tertiary Amine Extraction of Plutonium From Nitric Acid Solutions," Proceedings of the Second International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1958, P/544, Vol. 17, p. 348 United Nations (1959).

TABLE I

EFFECT OF PLUTONIUM CONCENTRATION ON ITS EXTRACTION BY TLA-AMSCO

Organic Phase: 10 vol.% TLA, 2 vol.% n-octylalcohol-Amsco (0.15 M TLA) pre-equilibrated with 2 M HNO_3, 0.03 M $NaNO_2$.

Aqueous Phase: 2 M HNO_3, 0.03 M $NaNO_2$, Pu(IV) as shown.

#	Pu gm/l		E_A^o
	Aq. Phase	Org. Phase	
1	4.88×10^{-3}	0.455	93
2	1.02×10^{-2}	0.890	87
3	4.23×10^{-2}	2.12	50
4	2.58×10^{-1}	4.24	16
5	6.21	25.7 H	1.1*
		1.85 L	
		6.65 A	
6	14.8	34.8 H	0.53*
		1.21 L	
		7.85 A	

* E_A^o was calculated using the average plutonium concentration in the organic phase

H - The heavier of the two organic phases which formed

L - The lighter organic phase

TABLE II

EFFECT OF PLUTONIUM CONCENTRATION ON ITS EXTRACTION BY TnOA-XYLENE

Organic Phase: 10 vol.% TnOA-xylene (0.22 M TnOA)

Aqueous Phase: 2 M HNO , 0.03 M NaNO , Pu(IV) as shown.

#	Pu gm/l		E_A^o
	Aq. Phase	Org. Phase	
1	6.65×10^{-4}	0.482	725
2	2.54×10^{-3}	0.920	362
3	1.08×10^{-2}	2.24	207
4	4.08×10^{-2}	5.60	137
5	0.157	8.90	57
6	0.635	29.0 H	20*
		2.42 L	
		13.0 A	
7	7.25	47.8 H	1.8*
		1.78 L	
		13.3 A	

* E_A^o was calculated using the average plutonium concentration in the organic phase

H - The heavier of the two organic phases which formed

L - The lighter organic phase

TABLE III

EFFECT OF PLUTONIUM CONCENTRATION ON ITS EXTRACTION BY TLA-XYLENE

Organic Phase: 10 vol.% TLA-xylene (0.15 M TLA)

Aqueous Phase: 2 M HNO_3, 0.03 M $NaNO_2$, Pu(IV) as shown.

#	Pu gm/l		E_A^o	M HNO_3(aq.)
	Aq. Phase	Org. Phase		
1	8.5×10^{-3}	0.47	55	1.81
2	1.9×10^{-2}	0.92	49	1.81
3	7.5×10^{02}	2.08	28	1.81
4	0.36	3.96	12	1.81
5	6.54	7.20	1.1	2.03
6	14.7	7.86	0.54	2.08
7	27.2	9.39	0.34	2.51

TABLE IV

EXTRACTION OF PLUTONIUM BY TLA-XYLENE AT HIGH AQUEOUS PLUTONIUM CONCENTRATIONS

Organic Phase: 20 vol.% TLA-xylene

Aqueous Phase: HNO_3 as shown, Pu as shown.

M HNO_3 Aq.	Pu gm/l		E_A^o	M TLA/M Pu
	Aq. Phase	Org. Phase		
2	38	22	0.58	3.3
4	32	25	0.78	2.9
5.5	27	30	1.1	2.4
6.7	24	30	1.26	2.4

Organic Phase: 10 vol.% TLA-xylene

Aqueous Phase: HNO_3 as shown, Pu as shown.

M HNO_3 Aq.	Pu gm/l		E_A^o	M TLA/M Pu
	Aq. Phase	Org. Phase		
2	43	10	0.23	3.6
4	38	12	0.32	3.0
5.3	32	13	0.40	2.8
6.6	28	13	0.46	2.8

TABLE V

EXTRACTION OF PLUTONIUM FROM NITRIC-PERCHLORIC ACID MIXTURES

Organic Phase: 10 vol.% TnOA-xylene pre-equilibrated with the proper acid solution.

Aqueous Phase: Nitric acid and perchloric acid concentration as shown; 0.03 M $NaNO_2$, tracer Pu(IV).

M HNO_3	M $HClO_4$	E_A^o
2.0	0.0	2.8×10^2
1.5	0.5	0.7
1.0	1.0	0.04
0.8	0.2	0.04