OCT 4 - 1971

ORNL-4724
UC-10 – Chemical Separations Processes
for Plutonium and Uranium

FORMATION OF THIRD PHASES AND THE EFFECT OF TEMPERATURE ON THE DISTRIBUTION OF PLUTONIUM AND URANIUM IN EXTRACTIONS BY TRI-*n*-BUTYL PHOSPHATE

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OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

Printed in the United States of America. Available from
National Technical Information Service
U.S. Department of Commerce
5285 Port Royal Road, Springfield, Virginia 22151
Price: Printed Copy $3.00; Microfiche $0.95

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ORNL-4724

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

Chemical Development Section B

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SEPTEMBER 1971

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CONTENTS

		Page
Abstract		1
1.	Introduction	2
2.	Experimental Procedures	2
3.	Effect of Temperature on the Distribution of Plutonium	3
4.	Effect of Temperature on the Distribution of Uranium	10
5.	Formation of a Third Phase	16
6.	References	21

FORMATION OF THIRD PHASES AND THE EFFECT OF TEMPERATURE ON THE DISTRIBUTION OF PLUTONIUM AND URANIUM IN EXTRACTIONS BY TRI-n-BUTYL PHOSPHATE

D. E. Horner

ABSTRACT

The results obtained by various investigators in studies of the effect of temperature on the distribution of plutonium and uranium in extractions by tri-n-butyl phosphate (TBP) are summarized in this report. Data obtained in tests that were made recently at ORNL are also given. The results of all the tests of uranium extraction from 0.07-10 *M* HNO_3 solutions show that the extraction coefficients decrease as the temperature is increased. The results of all the tests of Pu(IV) extraction from nitric acid solutions with a concentration of 0.5 *M* or less show that the extraction coefficients for Pu(IV) also decrease as the temperature is increased; however, at higher nitric acid concentrations, there is some disagreement among data from different sources. The results of our recent tests with 1 and 4 *M* HNO_3 disagree with those obtained in previous investigations in that they show little or no effect of temperature on the plutonium extraction coefficients in the temperature range 30 to 60°C.

The maximum organic-phase plutonium concentration in TBP--n-dodecane that is obtainable without the formation of a third phase was shown to increase as the ionic strength of the aqueous phase in equilibrium with the organic phase was increased. For example, over the ionic strength range of 2.0 to 9.5, the allowable plutonium concentration increased from about 20 g/liter to 40 g/liter with 15% TBP, and increased from about 33 g/liter to 62 g/liter with 30% TBP, at room temperature (~ 23°C). In tests in which third phases were formed, the third phase disappeared in each case when the temperature was moderately increased. At 40°C, a loading of up to 68 g of plutonium per liter was obtained in 15% TBP, corresponding to about the theoretical maximum ratio of 0.5 mole of plutonium per mole of TBP.

1. INTRODUCTION

Adaptation of the tri-n-butyl phosphate (TBP) extraction process (Purex process) to the treatment of Liquid Metal Cooled Fast Breeder Reactor (LMFBR) fuels is being studied at ORNL. In the present conceptual flowsheet,[1] uranium and plutonium are coextracted and partitioned in the first cycle. The plutonium is then further purified and concentrated in two additional TBP cycles. Because of the high plutonium content of LMFBR fuels, the concentrations of plutonium in the extraction system are much higher than those encountered in past Purex experience. It is known that, at high plutonium concentrations, the solubility of the Pu(IV) nitrate--TBP complex in the diluent can be exceeded, resulting in formation of a second organic phase (third phase). The third phase, which is rich in both TBP and plutonium, represents both a process control problem and a potential criticality hazard. For these reasons, knowledge of the conditions under which a third phase can form is necessary prior to specifying flowsheet conditions.

A second important consideration in a Purex flowsheet is the effect of temperature on the distribution of the various components. In particular, the effect of temperature on the extraction of plutonium and uranium has an important bearing on the overall extraction and stripping efficiencies, while the effect of temperature on the extraction of fission products is important from the standpoint of decontamination efficiency.

This report summarizes conclusions from previous studies, made by other investigators, of the effect of temperature on plutonium and uranium in extractions by TBP and also describes recent results obtained from laboratory-scale tests that were made at ORNL in an effort to increase our understanding of both temperature effects and third-phase formation.

2. EXPERIMENTAL PROCEDURES

Technical-grade tri-n-butyl phosphate (TBP) purchased from the Commercial Solvents Corporation was purified by the usual sodium hydroxide treatment. The

desired TBP concentration (vol %) was obtained by diluting the purified material with practical-grade n-dodecane obtained from the Eastman Kodak Company. Analysis showed that a typical 15% solution used in this study contained 144.8 g of "dry TBP" per liter.

All aqueous feed solutions of plutonium were prepared by diluting, with acid, a stock solution of 6.6 *M* HNO_3 containing 246 g of Pu(IV) per liter. Each of the feed solutions was used as soon as possible after its preparation to minimize the effect of any possible valence changes.

In tests to determine the effect of temperature on extraction, the temperature of the aqueous and organic phases was adjusted to the desired level and then the phases were mixed vigorously in water-jacketed separatory funnels. A portion of the mixture was taken after 2 min of mixing, and subsequently the phases were allowed to separate; on separation, each was sampled for analysis.

In defining the regions of third-phase formation, the solvent was overloaded with plutonium by contact with feed solutions highly concentrated in plutonium. A small increment of fresh solvent was then added, and the phases were recontacted and centrifuged. Additional increments were added until no third phase was evident.

3. EFFECT OF TEMPERATURE ON THE DISTRIBUTION OF PLUTONIUM

Previous studies of the effect of temperature on the distribution of plutonium included tests made with feed solutions containing plutonium or plutonium plus uranium at various nitric acid concentrations. In one series of tests[2] with solutions up to 5 *M* in HNO_3, the plutonium extraction coefficients decreased as the temperature was increased in the temperature range 20 to 70°C. This was attributed to a decrease in the equilibrium constant of the extraction reaction as the temperature was increased. At nitric acid concentrations higher than 5 *M*, an increase in temperature resulted in an increase of the coefficients, apparently because the activity coefficients of plutonium ions increased. With a feed containing uranyl nitrate up to a concentration

of 1.26 *M* and plutonium, the plutonium extraction coefficient increased over the entire range of acidity as the temperature was increased.

In another study[3] with both uranium- and non-uranium-bearing feed solutions, each of which was 3 *M* in HNO_3, the plutonium extraction coefficient increased by a factor of about 1.5 as the temperature was increased from 10°C to 40°C but decreased by a factor of 3 to 4 from 40°C to 80°C. Thus, the results of these two studies agree in the temperature range 40 to 70°C in nitric acid solutions with concentrations of 5 *M* or less but appear to be contradictory in the temperature range of 10 to 40°C. In the second study, the difference in results is ascribed to two factors: (1) a shift in equilibrium at higher temperatures, causing dissociation of the higher nitrate complexes of Pu(IV) to more extractable neutral species, and (2) the exothermic formation of the extracted complex, $Pu(NO_3)_4 \cdot 2TBP$. At acidities up to 1.7 *M* and temperatures up to 40°C, the first factor dominates; at higher temperatures, the second factor takes precedence. For extractions from nitric acid solutions having concentrations of 0.5 *M* or less, results of both studies show that increasing the temperature decreases the extraction coefficient. At this acid concentration, the inverse effect of temperature on the extraction coefficient is ascribed to the fact that the higher complexes are not present in significant concentrations and an increase in temperature results in greater dissociation of $Pu(NO_3)_4$ into lower complexes that are less extractable.

In our studies, batch tests were made with solutions initially containing 1 or ~ 20 g of plutonium per liter in 0.3, 1, and 4 *M* HNO_3. With 0.3 *M* HNO_3, the extraction coefficients decreased by a factor of about 3.5 as the temperature was increased from 30°C to 60°C (Table 1 and Fig. 1). These results with 0.3 *M* HNO_3 are in qualitative agreement with those obtained in the studies described above. At nitric acid concentrations of less than 0.3 *M*, it can be predicted that the same temperature effect will apply, although for different reasons. As the acidity is decreased, the disproportionation of Pu(IV) increases, resulting in formation of a larger amount of unextractable Pu(III). Also, at very low acidities, plutonium hydrolysis can occur, causing unextractable polymeric species to form. However, the formation of polymer

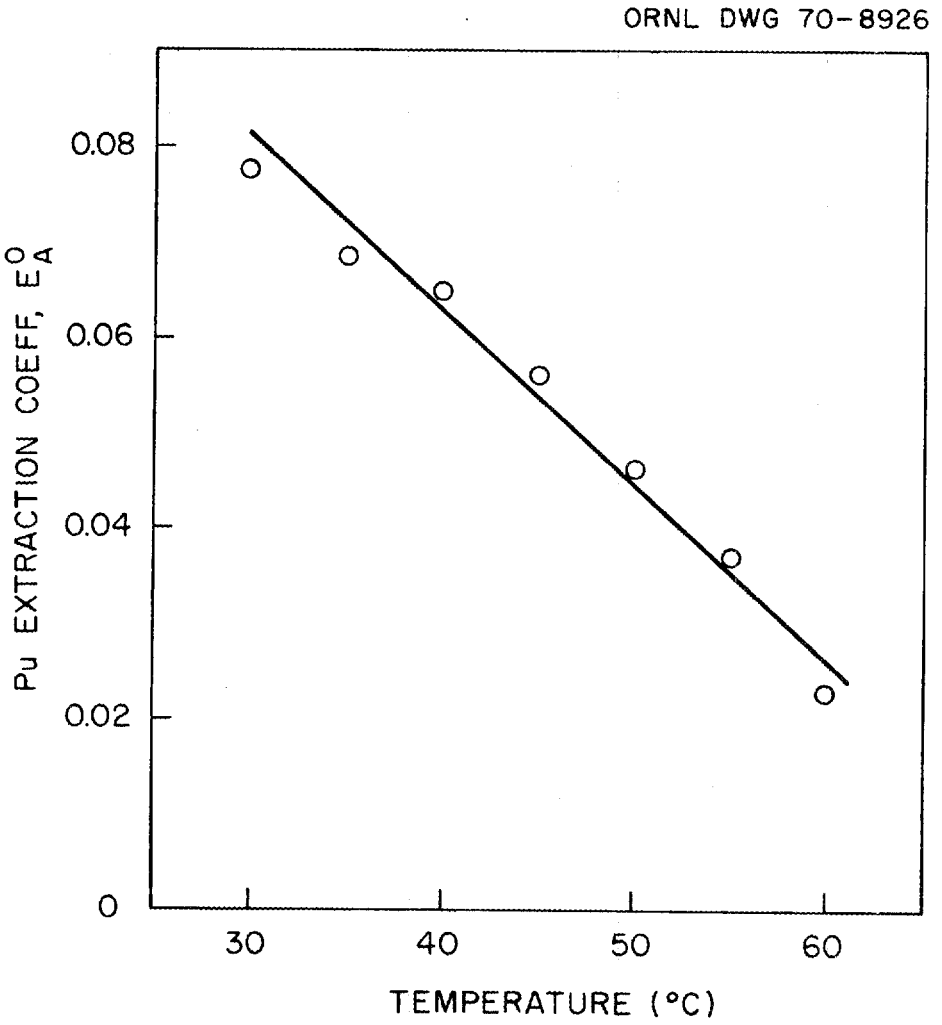

Fig. 1. Effect of Temperature on the Extraction of Pu(IV) from 0.3 *M* HNO_3 with 15% TBP--85% n-Dodecane.

Table 1. Effect of Temperature on the Extraction of Plutonium from 0.3 M HNO_3

Organic phase: 15% TBP--85% n-dodecane

Aqueous phase: 0.3 M HNO_3 containing 1.06 g of Pu per liter

Contact: 2 min at phase ratio of 1/1

Temp. (°C)	Plutonium Concentration (g/liter) In the Organic Phase	In the Aqueous Phase	Pu Extraction Coefficient, E_a^o
30	0.075	0.96	0.078
35	0.065	0.95	0.069
40	0.063	0.98	0.065
45	0.055	0.98	0.056
50	0.046	1.0	0.046
55	0.038	1.0	0.038
60	0.023	1.0	0.023

was shown not to be the reason for the effect obtained in our tests with 0.3 M HNO_3.

Since conditions that decrease the extraction coefficient have the opposite effect on the stripping coefficient, the results discussed above indicate that increasing the temperature should improve the efficiency of plutonium stripping with 0.3 M HNO_3. This was confirmed by results of batch tests made over a temperature range 25 to 60°C (Table 2). In these tests, the percentage of Pu(III) in the final aqueous phase increased from 0.7 to 32% of the total aqueous-phase plutonium as the result of an increase in disproportionation of Pu(IV) at the higher temperatures. Comparison of results of the plutonium stripping tests with the results of extraction tests from 0.3 M HNO_3 (Fig. 1) shows an apparent much smaller temperature effect in the former case. This is attributed to the fact that the equilibrium acid concentration

Table 2. Effect of Temperature on the Stripping of Plutonium with 0.3 M HNO_3

Organic phase:	15% TBP--85% n-dodecane, scrubbed with Na_2CO_3 solution and containing 5.2 g of plutonium per liter
Aqueous phase:	0.3 M HNO_3
Contact:	1 min at an A/O phase ratio of 2/1

Temp. (°C)	Pu Concentration (g/liter) In the Organic Phase	In the Aqueous Phase	Fraction (%) of Total Aqueous Pu as Pu(III)	Stripping Coefficient, S_o^a
25	0.50	2.2	0.69	4.4
30	0.49	2.2	1.1	4.6
35	0.50	2.1	1.6	4.3
40	0.44	2.2	>10	5.0
45	0.41	2.2	14	5.2
50	0.38	2.2	20	5.8
55	0.34	2.2	26	6.6
60	0.30	2.2	32	7.2

in the aqueous phase in the stripping tests was 0.5 M, including the acid stripped from the organic feed; and the amount of Pu(IV) disproportionation, therefore, was considerably lower than in the extraction tests. Since the stripping efficiency is higher and the tendency for third-phase formation is lower at elevated temperatures, we have specified a temperature of 40°C for the stripping step in both the second and third extraction cycles of a proposed flowsheet[1] for processing LMFBR fuels. Results of a batch countercurrent test of the stripping system are described in Sect. 5.

The data from tests carried out at ORNL to determine the effect of increasing the temperature on the extraction of plutonium from 1 and 4 M HNO_3 (Tables 3 and 4) show no significant trend over the range of 23 to 60°C. Thus our results are in disagreement with those reported by other investigators (discussed above).

Table 3. Effect of Temperature on the Extraction of Plutonium from 1 *M* HNO_3

Organic phase: 15% TBP--85% n-dodecane

Aqueous phase: 1 *M* HNO_3 containing 0.96 or 20 g of Pu per liter

Contact: 1 min at phase ratio of 1/1

Temp. (°C)	Plutonium Concentration (g/liter)			Pu Extraction Coefficient, E^o_a
	In the Initial Aqueous Phase	In the Organic Phase	In the Final Aqueous Phase	
23	20	11	11	1.1
40	20	12	10	1.1
60	20	13	10	1.3
30	0.96	0.49	0.59	0.83
35	0.96	0.49	0.56	0.87
40	0.96	0.47	0.52	0.91
45	0.96	0.47	0.54	0.88
50	0.96	0.47	0.57	0.83
55	0.96	0.44	0.57	0.77
60	0.96	0.46	0.46	0.98

Table 4. Effect of Temperature on the Extraction of Plutonium from 4 *M* HNO_3

Organic phase: 15% TBP--85% n-dodecane

Aqueous phase: 4 *M* HNO_3 containing 1.0 or 22 g of Pu per liter

Contact: 1 min at phase ratio of 1/1

Temp. (°C)	Plutonium Concentration (g/liter)			Plutonium Extraction Coefficient, E^o_a
	In the Initial Aqueous Phase	In the Organic Phase	In the Final Aqueous Phase	
30	22	15	4.1	3.7
35	22	16	4.2	3.8
40	22	16	4.1	3.8
45	22	16	4.1	3.8
50	22	16	4.1	3.9
55	22	16	4.0	4.1
60	22	16	4.1	4.0
30	1.0	0.96	0.18	5.3
35	1.0	0.96	0.20	4.8
40	1.0	0.94	0.22	4.3
45	1.0	0.94	0.20	4.8
50	1.0	0.97	0.21	4.6
55	1.0	0.96	0.20	4.8
60	1.0	0.96	0.18	5.3

4. EFFECT OF TEMPERATURE ON THE DISTRIBUTION OF URANIUM

Results of previous studies, made by other investigators,[4-6] of the effect of increasing the temperature on the distribution of uranium in nitric acid feed solutions containing uranium show that the uranium extraction coefficient decreases as the temperature is increased; less change is found to occur at higher solvent loadings. Results of similar tests, made recently at ORNL, are in agreement with these. The latter tests were made with aqueous feeds containing 10.8 to 11.7 g of uranium per liter in 0.3, 2, and 3 *M* HNO_3, at temperatures ranging between 25 and 60°C (Tables 5-7). A plot of the logarithm of the extraction coefficients vs the reciprocals of the absolute temperatures for each acidity resulted in parallel straight lines, with very little scatter in the points (Fig. 2). From these data, the following equation, which expresses the extraction coefficient, E_{T_2}, at any temperature between 25 and 60°C as a function of the coefficient, E_{T_1}, at room temperature (25°C), was obtained by computer fit:

$$E_{T_2} = E_{T_1} \times e^{2220(1/T_2 - 1/T_1)} ,$$

where T is the absolute temperature.

Use of this equation should be restricted to data obtained at relatively low solvent loadings (≤ 18% of the theoretical maximum) since the slope of the curve decreases as the solvent loading is increased and approaches zero as a limit. This was shown by results of another series of tests using an aqueous feed containing 93.6 g of uranium per liter, in which the maximum loading was 60% of theoretical. The slope of the straight line obtained by plotting these data is less than that determined in previous tests made at lower loadings (Fig. 3 and Table 7).

In tests with a feed containing both plutonium and uranium in 3 *M* HNO_3, the extraction of plutonium increased, while that of uranium decreased, as the temperature was increased. These results are in agreement with those obtained previously. For example, in batch tests with a feed containing 7 g of plutonium and 58 g of uranium per liter in 3 *M* HNO_3, the plutonium coefficient increased from 0.5 to 1.2 and the

Table 5. Effect of Temperature on the Extraction of Uranium from 0.3 M HNO_3

Organic phase: 15% TBP--85% n-dodecane

Aqueous phase: 0.3 M HNO_3 containing 10.8 g of U per liter

Contact: 1 min at phase ratio of 1/1

Temp. (°C)	Uranium Concentration (g/liter)		Uranium Extraction Coefficient, E^o_a
	In the Organic Phase	In the Aqueous Phase	
25	3.4	7.8	0.44
30	3.1	7.3	0.43
35	2.7	8.0	0.34
40	2.5	8.1	0.31
45	2.3	8.3	0.28
50	2.2	8.6	0.25
55	1.9	8.6	0.23
60	1.9	9.1	0.21

Table 6. Effect of Temperature on the Extraction of Uranium from 2 *M* HNO_3

Organic phase: 15% TBP--85% n-dodecane

Aqueous phase: 2 *M* HNO_3 containing 11.7 g of U per liter

Contact: 1 min at phase ratio of 1/1

Temp. (°C)	Uranium Concentration (g/liter)		Uranium Extraction Coefficient, E^o_a
	In the Organic Phase	In the Aqueous Phase	
25	9.8	1.2	8.1
30	9.8	1.9	5.3
35	9.5	2.0	4.8
40	9.1	2.2	4.2
45	8.9	2.4	3.7
50	8.8	2.5	3.5
55	8.6	2.8	3.1
60	8.4	3.1	2.8

Table 7. Effect of Temperature on the Extraction of Uranium from 3 *M* HNO_3

Organic phase: 15% TBP--85% n-dodecane

Aqueous phase: 3 *M* HNO_3 containing 11 or 94 g of U per liter

Contact: 1 min at phase ratio of 1/1

Temp. (°C)	Uranium Concentration (g/liter) In the Organic Phase	In the Aqueous Phase	Uranium Extraction Coefficient, E_a^o
25	11	1.1	11
30	12	1.1	11
35	12	1.4	8.7
40	12	1.5	7.8
45	11	1.6	7.1
50	12	1.8	6.7
55	11	2.0	5.6
60	11	2.2	5.1
25	44	51	0.86
30	45	52	0.87
35	44	52	0.85
40	43	54	0.80
45	42	54	0.78
50	42	55	0.77
55	41	55	0.75
60	41	58	0.70

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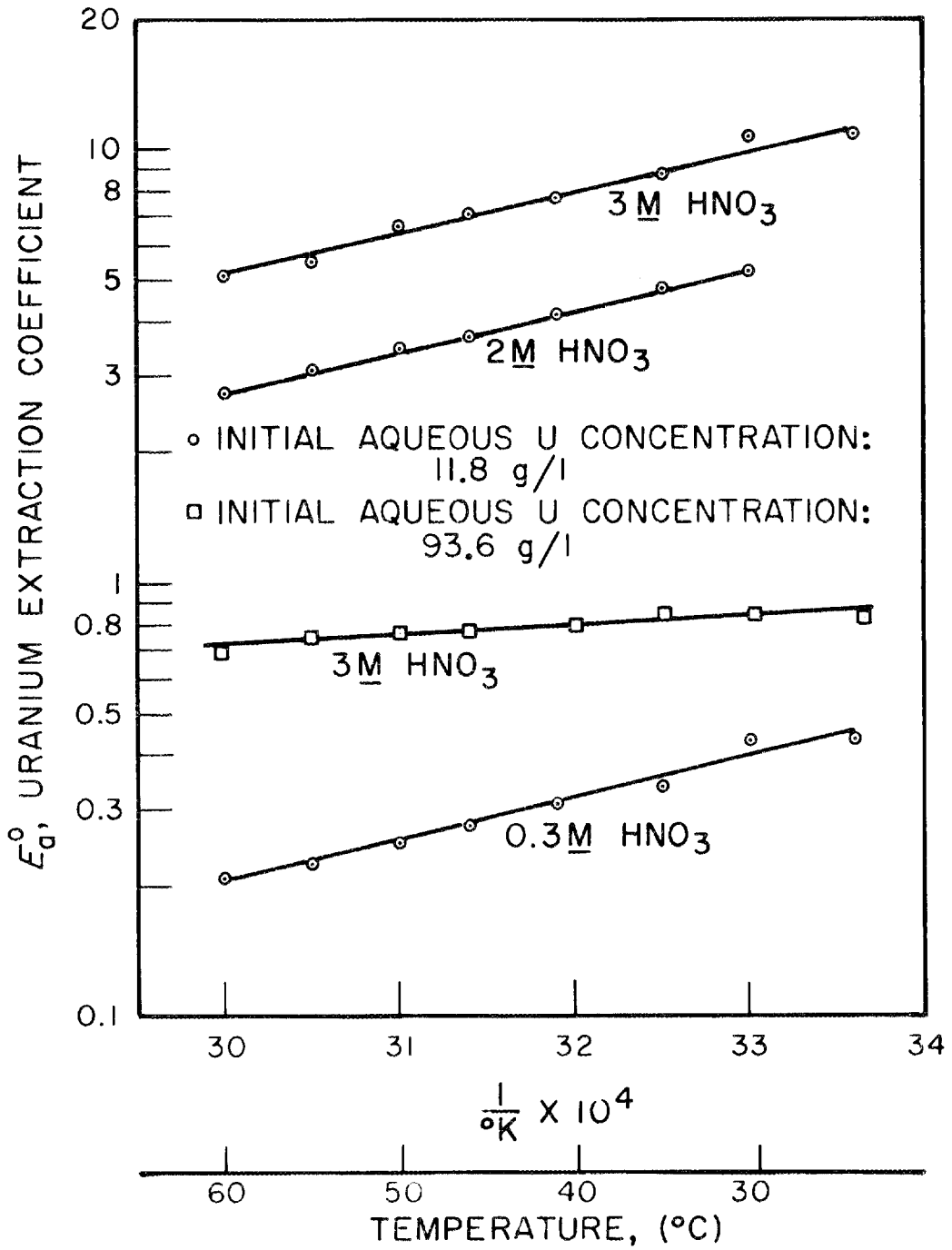

Fig. 2. Effect of Temperature on the Extraction of Uranium from Nitric Acid with 15% TBP--85% n-Dodecane. The initial uranium concentration was 10.8-11.7 g/liter; the phase ratio was 1/1.

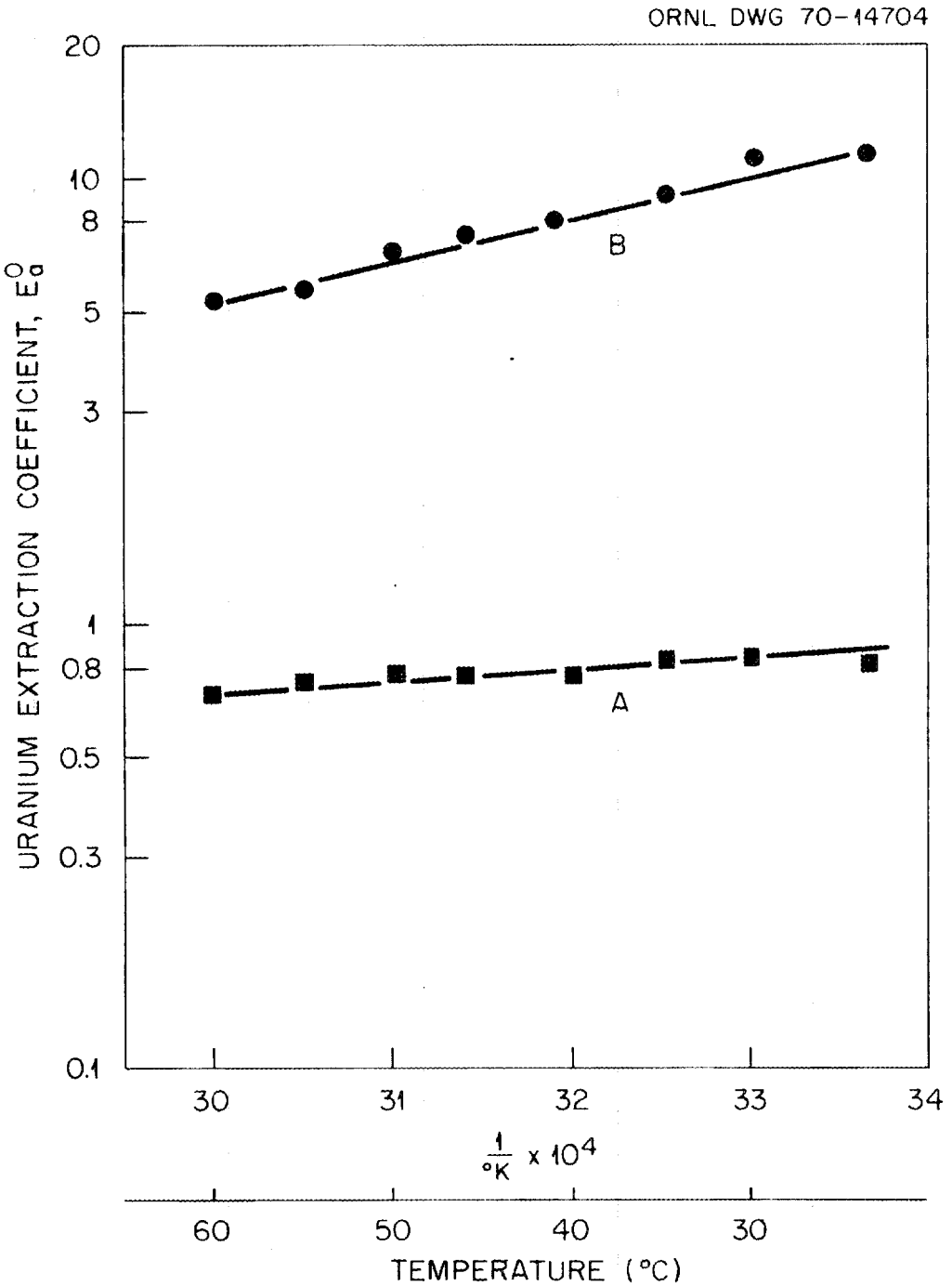

Fig. 3. Effects of Temperature and Solvent Loading on the Extraction of Uranium with 15% TBP--85% n-Dodecane. Aqueous phase: A - 3 *M* HNO_3 containing 94 g of U per liter; B - 3 *M* HNO_3 containing 12 g of U per liter. Phase ratio: 1/1.

uranium coefficient decreased from 1.7 to 1.0 as the temperature was increased from 23°C to 60°C (Fig. 4 and Table 8). Since the extraction of plutonium in nitric acid solutions having a concentration of 1 *M* or greater was previously shown in our tests to be essentially independent of temperature, the improved plutonium extraction in this case is thought to be due primarily to a decrease in competition by uranium for the solvent as the temperature is increased. For this reason, operation at an elevated temperature (e.g., 40°C) rather than at 25°C in the first-cycle coextraction step of a Purex flowsheet should result in more efficient extraction of plutonium and less efficient extraction of uranium. This, in turn, would cause plutonium reflux to be decreased at high solvent loadings. Another important potential advantage of elevating the temperature is that it ensures that the formation of a third phase will not occur, even under conditions of maloperation (see Sect. 5). It is known that an increase in the temperature increases the extraction of ^{95}Zr-Nb[3,5,6] and decreases the extraction of ^{106}Ru. Also, increasing the temperature increases the rate at which TBP and the diluent are degraded. All of these factors must be carefully considered when decisions concerning the optimum operating temperatures for the extraction and stripping cycles are made.

5. FORMATION OF A THIRD PHASE

The formation of a third phase occurs under certain conditions in some solvent extraction systems. This must be avoided since it interferes with process operation and control. In addition, in extractions of plutonium with TBP, the third phase can represent a criticality hazard since it is highly concentrated in plutonium. (We have obtained plutonium concentrations in the third phase as high as 187 g/liter.) To avoid the formation of a third phase, the concentrations of the extracted components in the solvent phase must frequently be limited by appropriate control of the extraction conditions. Operation in this manner suffers the disadvantages of decreasing the fission product decontamination efficiencies and the throughput capacity of the system. Reference 7 provides a thorough discussion of the chemistry

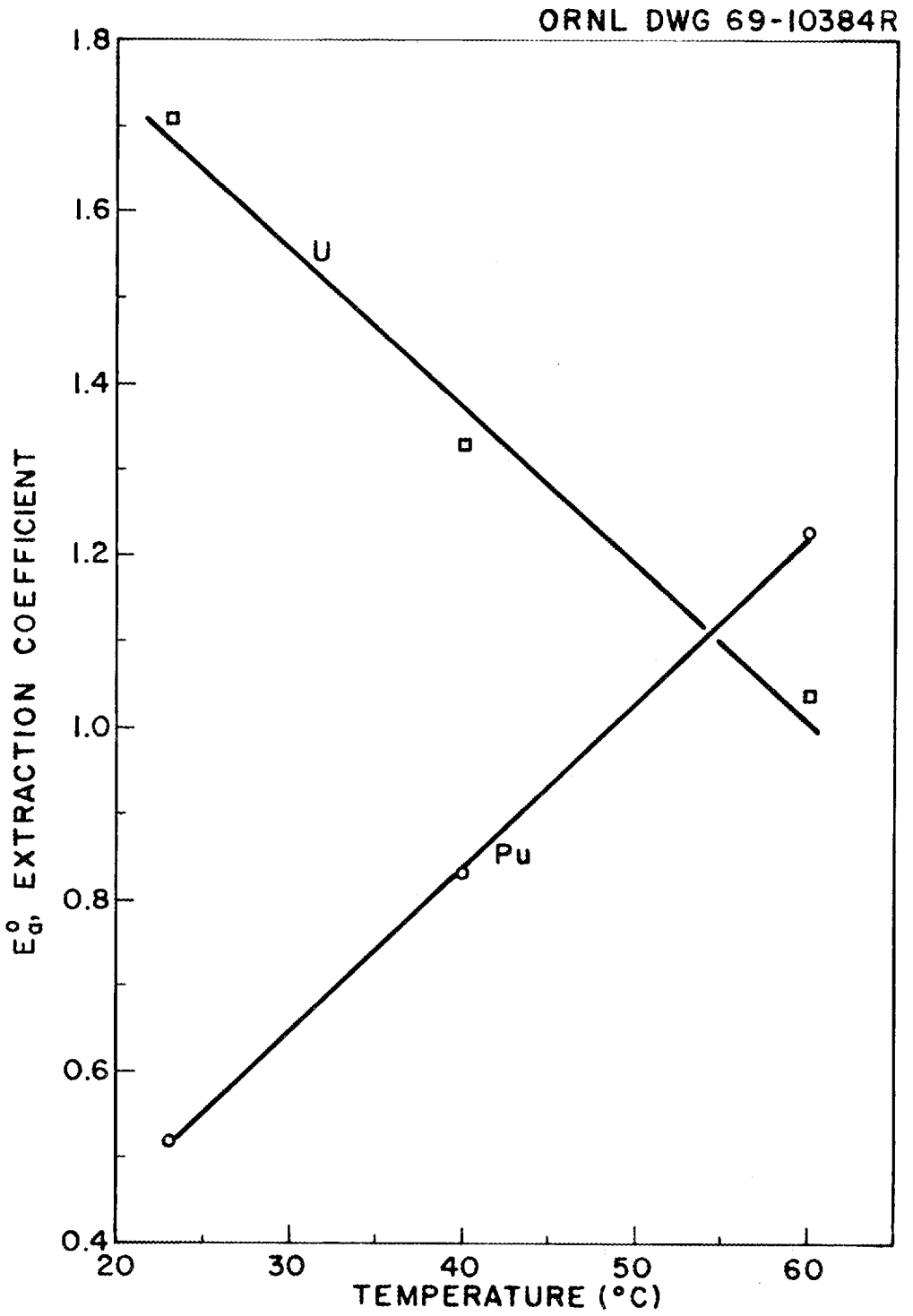

Fig. 4. Effect of Temperature on the Extraction of Plutonium and Uranium with 15% TBP--85% n-Dodecane. Aqueous phase: 3 *M* HNO_3 containing 7 g of plutonium and 58 g of uranium per liter. Phase ratio: 1/1.

Table 8. Effect of Temperature on the Extraction of Uranium and Plutonium from Mixed Feed

Organic phase: 15% TBP--85% n-dodecane

Aqueous phase: 3 M HNO_3 containing, in g/liter, 7 Pu and 58 U

Contact: 1 min at phase ratio of 1/1

Temp. (°C)	Concentration						Extraction Coefficient, E_a^o	
	In the Organic Phase			In the Aqueous Phase				
	U (g/liter)	Pu (g/liter)	HNO_3 (M)	U (g/liter)	Pu (g/liter)	HNO_3 (M)	U	Pu
23	42	2.1	0.26	24	4.1	2.5	1.7	0.52
40	38	3.0	0.26	29	3.6	2.5	1.3	0.84
60	34	3.7	0.27	33	3.0	2.5	1.0	1.2

of this phenomenon as it occurs with three major extraction reagents: (1) neutral organophosphorus compounds, (2) acidic organophosphorus compounds, and (3) long-chain aliphatic amines. Our studies to define the conditions that result in the formation of a third phase in the nitric acid--plutonium--TBP--n-dodecane system included a series of batch tests in which the maximum plutonium concentrations obtainable under a given set of conditions without formation of a third phase were determined. The procedure used is described in Sect. 2. At room temperature (~ 23°C), the maximum organic-phase plutonium concentration was shown to increase as the acidity, total nitrate concentration, or ionic strength of the aqueous phase was increased. For this system the ionic strength, μ, is defined as:

$$1/2\ \Sigma c_i z_i^2 = [H^+] + 10[Pu^{4+}] \quad , *$$

where c_i is the ionic concentration in molarity and z_i is the ionic charge. Over an ionic strength range of 2.0 to 9.5, the maximum plutonium concentration attainable without third-phase formation increases from about 20 g/liter to 40 g/liter with 15% TBP and from about 33 g/liter to 62 g/liter with 30% TBP (Fig. 5).

Mills and Logan[9] have reported results of third-phase studies with 20% TBP--80% odorless kerosene and Pu(IV); Th(IV), U(VI), and Pu(VI) were also studied. In their work, the organic-phase Pu(IV) concentration at the point of third-phase formation at 20°C and 25°C was shown to increase as the aqueous-phase acidity was increased from 3 to 7 *M* HNO_3 and then decrease as the acidity was further increased to 10 *M*. The increase over the acid concentration range up to 7 *M* HNO_3 agrees qualitatively with results of our tests. However, our tests did not include acidities greater than 8 *M* HNO_3. Interpolation of the values given by Mills and Logan to 23°C for 20% TBP--80% kerosene resulted in plutonium concentrations slightly greater than our values for 30% TBP--70% n-dodecane at acidities of less than 7 *M*. Presumably the difference between their results and ours is due to differences in the solubilities of the extracted plutonium complex in n-dodecane as compared with those in kerosene.

*More exactly, the quantity on the left side of this equation is one-half the ional concentration[8] since the ionic concentration is expressed in molarity instead of molality, but for the purpose of this report it is referred to as the ionic strength.

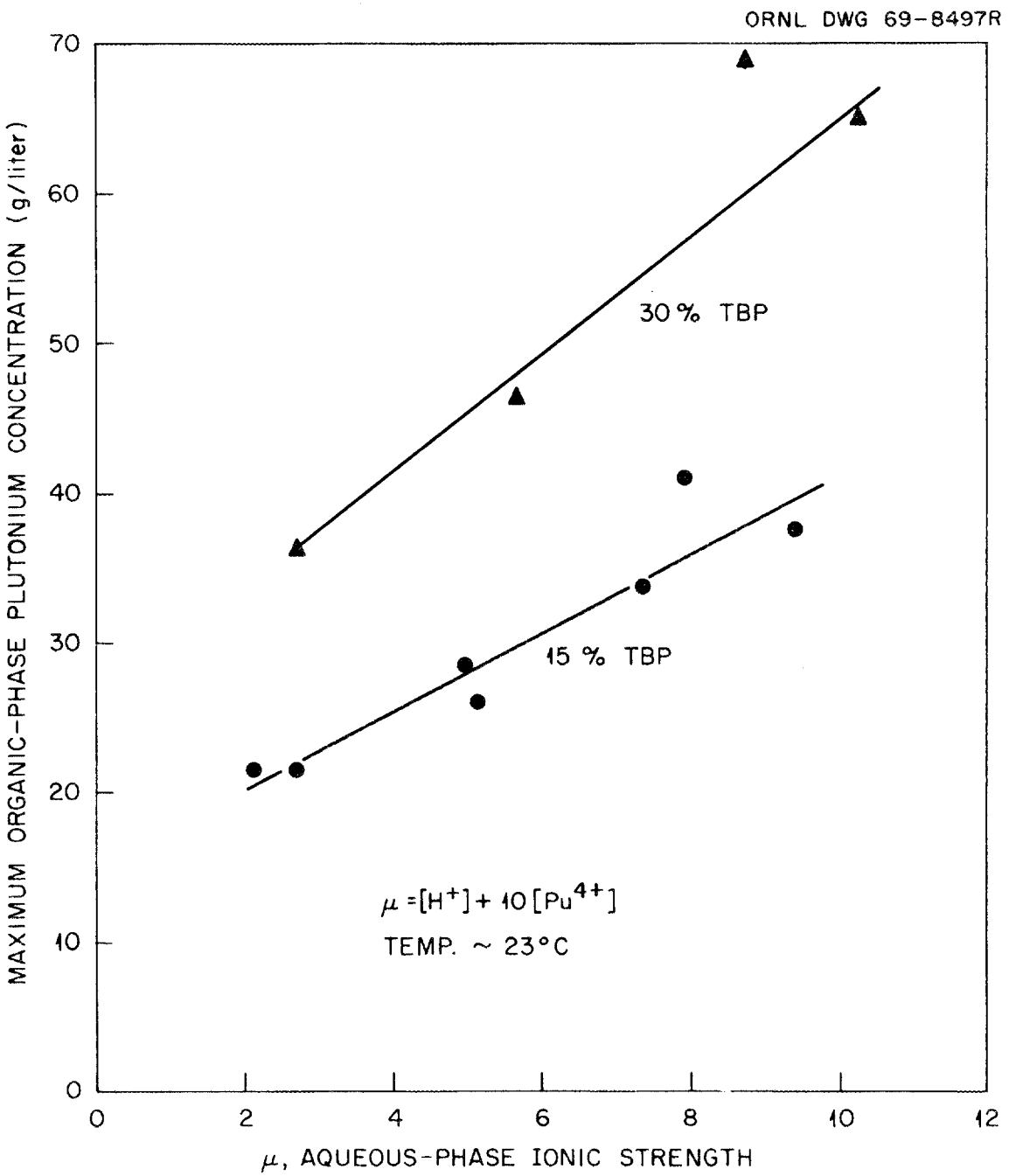

Fig. 5. Maximum Plutonium Loading of TBP--n-Dodecane Solvent Without Third-Phase Formation. Aqueous phase: $Pu(NO_3)_4^-$--HNO_3.

At elevated temperatures, much higher plutonium concentrations in the solvent phase can be obtained without the formation of a third phase. At 40°C, we have not observed third-phase formation with a plutonium concentration of 68 g per liter of 15% TBP--85% n-dodecane, which is close to the theoretical maximum loading (assuming 0.5 mole of plutonium per mole of TBP). These results lead to the conclusion that no third phase will form in process applications with 15% TBP--85% n-dodecane, provided the minimum temperature is not less than 40°C.

The higher solvent loading possible at elevated temperatures was confirmed in a batch countercurrent test at 40 to 45°C, which simulated the third plutonium purification cycle (Table 9). Five stages were used for extraction, one for scrubbing, and five for stripping with relative organic/feed/scrub/strip flow ratios of 30/30/4/15. More than 99.5% and 99.8% of the plutonium was recovered in the extraction step and in the stripping test, respectively, to give an overall recovery greater than 99.2%. Actually, the recovery was probably much higher than this but could not be accurately measured because of interference from americium in the raffinate. The organic phase in the feed stage was loaded to 43.6 g/liter (about 66% of theoretical maximum loading). (In comparison, a loading of only about 25 g/liter can be obtained without third-phase formation at a temperature of about 23°C.)

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Table 9. Data for Batch Countercurrent Third-Cycle Extraction and Stripping at Elevated Temperature

Organic phase: 15% TBP--85% n-dodecane

Feed: 4 *M* in HNO_3, 0.04 *M* in Fe^{3+}, and containing 41.3 g of Pu per liter

Scrub: 2 *M* HNO_3

Strip: 0.3 *M* HNO_3--0.02 *M* hydroxylamine nitrate--0.01 *M* hydrazine

Organic/feed/scrub/strip: 30/30/4/15

Contact time: 2 min

Temperature: 40-45°C

	Organic-Phase Concentrations		Aqueous-Phase Concentrations	
Stage No.	Pu (g/liter)	HNO_3 (*M*)	Pu (g/liter)	HNO_3 (*M*)
Strip-5	0.11	0.019	8.9	0.40
Strip-4	2.9	0.028	26	0.37
Strip-3	12	0.043	44	0.41
Strip-2	23	0.025	63	0.59
Strip-1	34	0.039	77	0.64
Scrub	41	0.079	30	2.4
Ext.-1	44	0.11	24	3.8
Ext.-2	24	0.25	6.0	3.9
Ext.-3	4.9	0.39	0.69	4.1
Ext.-4	0.89	0.42	<0.2	4.0
Ext.-5	0.10	0.41	< 0.2	3.7

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