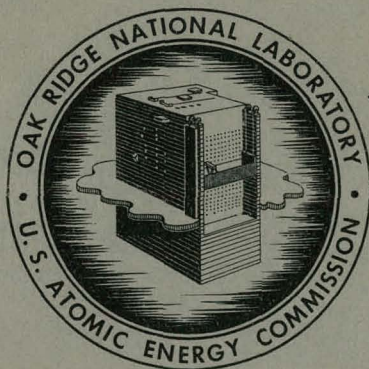


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SOLUBILITY OF ZIRCONIUM DIBUTYL PHOSPHATE
IN SOLVENT EXTRACTION SOLUTIONS

W. Davis, Jr.
H. H. Carmichael



OAK RIDGE NATIONAL LABORATORY
operated by
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CHEMICAL TECHNOLOGY DIVISION

Chemical Development Section B

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DATE ISSUED

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ABSTRACT

The solubility of zirconium dibutyl phosphate in aqueous uranyl nitrate and nitric acid solutions was found to vary from 0.35 to 159 mg of zirconium per liter, increasing with increasing uranium concentration. The solubility in 1.126 M TBP in Amsco 125-82 equilibrated with uranyl nitrate—nitric acid aqueous solutions was found to vary from 0.33 to 3.55 g of zirconium per liter. Particularly in the extractant solutions, the solubility of zirconium as zirconium dibutyl phosphate is well above the zirconium content of even extensively burned natural uranium fuels under Purex process conditions.

The zirconium dibutyl phosphate studied was prepared by direct synthesis in aqueous solution and found to have a variable composition. Precipitation from 2 M HNO_3 at 40°C yielded a product which had the approximate composition $\text{Zr}(\text{OH})(\text{NO}_3)(\text{DBP})_2$.

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1.0 INTRODUCTION

The work described in this report was performed to determine some of the properties of zirconium dibutyl phosphate in Purex solutions and to elucidate the role of dibutyl phosphoric acid (HDBP), a product of the degradation of tributyl phosphate (TBP), in the behavior of zirconium in the solvent extraction of nuclear fuels. Dibutyl phosphoric acid, which is formed by acid dealkylation¹ or radiolysis⁶ of TBP, has been shown^{2,8} to cause the extraction of zirconium into TBP-kerosene extractant solutions. Uranium represses this extraction very markedly, but Dukes² found a distribution coefficient, DC_a^0 , of zirconium from a 3.0 M HNO_3 solution into 30% TBP-dodecane of 0.44, at 70°C, with no dibutyl phosphoric acid initially present. Thus, dibutyl phosphoric acid is not the only factor involved in low decontamination of uranium and plutonium from zirconium.

The scope of this report includes measurements of the solubility of zirconium dibutyl phosphate, by use of Zr-95 tracer, in aqueous and 30% TBP-Amsco solutions containing uranyl nitrate and nitric acid in concentration ranges spanning those found in the Purex process. These measurements are probably accurate to a factor of about 3. In addition, qualitative observations on the behavior of solid zirconium dibutyl phosphate in contact with these and other solutions are described.

All chemical and radiochemical analyses described in this report were performed by members of the groups of W. R. Laing, O. Menis, P. F. Thomason, G. R. Wilson, and E. I. Wyatt of the ORNL Analytical Chemistry Division.

2.0 EXPERIMENTAL RESULTS

2.1 Solubility Data

Solid zirconium dibutyl phosphate, initially containing 3.71 mcurie of Zr-95 per gram of zirconium, was agitated with 16 aqueous solutions, of initial composition 0-3.96 M HNO_3 and 0-490 g of uranium per liter, and with 16 extractant solutions prepared by contacting portions of 1.126 M TBP in Amsco 125-82 with six successive portions of each of the aqueous solutions. This exhaustive contacting was performed to provide extractant solutions in equilibrium with the corresponding "as-prepared" aqueous solutions.

Quantities of zirconium dibutyl phosphate dissolved in the aqueous solutions varied from 0.22 to 159 mg of zirconium per liter; corresponding quantities dissolved in the extractant solutions varied from 0.33 to 3.55 g of zirconium per liter (Table 1). Although there was excess solid in contact with all solutions at the time of sampling, the changes in zirconium content with agitation time noted in Table 1, and the observations of Moffat⁴ that several days to a week, or more, are required to achieve

Table 1. Zirconium Dibutyl Phosphate Dissolved in Aqueous and 30% TBP-Amsco Solutions^a

Sample No.	Aqueous Phase				Sample No.	30% TBP-Amsco Phase at Equilibrium			
	Initial Comp.		Agitation Time, hr	Zr Content, mg/liter		Initial Comp.		Agitation Time, hr	Zr Content, g/liter
	H ⁺ , M	U, M				H ⁺ , M	U, M		
1-A	0.08	1.21	96	67	1-0	0.000	0.405	48	1.85
			264	75				168	2.97
2-A	0.17	2.06	96	127	2-0	0.005	0.534	48	1.44
			264	159					
3-A	0.85	1.95	96	36	3-0	0.035	0.521	48	1.79
4-A	3.94	1.55	96	28	4-0	0.08	0.513	48	1.22
5-A	0.04	0.495	96	13	5-0	0.011	0.349	48	1.29
6-A	0.22	0.500	96	17	6-0	0.02	0.420	48	1.33
								168	2.45
7-A	0.94	0.508	96	19	7-0	0.04	0.462	48	1.70
								168	3.55
8-A	3.86	0.495	96	12	8-0	0.09	0.483	48	1.67
			264	41					
9-A	0.011	0.200	48	3.2	9-0	0.004	0.195	48	1.85
			360 ^b	2.7					
10-A	0.23	0.180	48	1.22	10-0	0.019	0.228	120	2.39
			360 ^b	0.78					
11-A	0.95	0.171	48	5.0	11-0	0.06	0.344	120	1.45
			360 ^b	6.1					
12-A	2.99	0.175	48	10.0	12-0	0.12	0.429	120	1.53
			360 ^b	11.6					
13-A	0.00	0	48	0.22	13-0	0.004	0	120	1.34
			360 ^b	0.35					
14-A	0.26	0	48	2.7	14-0	0.033	0	120	0.72
			360 ^b	3.0					
15-A	1.00	0	48	4.2	15-0	0.22	0	120	0.36
			360 ^b	4.7					
16-A	3.96	0	48	0.31	16-0	0.82	0	120	0.33
			360 ^b	0.42					

^a1.126 M TBP in Amsco before contacting with aqueous solutions.

^bAdditional time static except for occasional shaking.

solution saturation in similar studies, suggest that not all the values of zirconium content are true solubilities.

The reasons for the slow rate of dissolution of zirconium dibutyl phosphate in the aqueous and organic solutions used in the present study are not clear. It was observed that as much as two days were required to wet the solid in a few of the aqueous solutions. After such wetting, most of the solid formed a froth on the top of the aqueous solution and only a small quantity settled on standing. However, all organic solutions appeared to wet the zirconium dibutyl phosphate within a few hours.

Increasing the uranium concentration clearly increased the solubility of zirconium dibutyl phosphate in the aqueous phase. This effect of uranium may persist in the organic phase to a small extent, as suggested by the decreased zirconium content in samples 13-0, 14-0, 15-0, and 16-0, in which no uranium was present, below that in all other organic samples, in which uranium was present. It should be noted that the TBP in samples 1-0 through 8-0 was saturated with uranyl nitrate and nitric acid, primarily the former, and that samples 9-0 through 12-0 were nearly saturated with uranyl nitrate; thus, these solutions were so nearly of the same composition that only small changes in zirconium dibutyl phosphate solubility would be expected.

2.2 Behavior of Zirconium Dibutyl Phosphate

Air-dried zirconium dibutyl phosphate was wetted and dispersed by the TBP-Amsco solutions. When the compound was added to the aqueous solutions, some of it was wetted and settled, but the bulk formed a froth on top of the liquid.

Preliminary studies showed the zirconium dibutyl phosphate to be soluble to the order of 2% in methanol and slightly soluble in ethanol and carbon tetrachloride at 25°C. It was quite soluble in ethanol on warming but re-crystallized as a pasty mass which stuck to the glassware. A small amount of water added to a hot methanol solution caused precipitation, on cooling, of a material similar to the original precipitate. This, too, turned to a pasty mass on standing 48 hr.

Zirconium dibutyl phosphate is an effective carrier for niobium. Only 4% of the microscopic amount of niobium remained in the filtrate from the original precipitation, while 18.5% of the zirconium did not precipitate. From the amount of carrier zirconium, the loss to the filtrate, and the zirconium content of the precipitate, a theoretical yield of 14.5 g was calculated for the precipitation containing Zr-95. The 10% difference between this and the actual 13.0-g yield can probably be explained by the zirconium analysis being low. This sample did not dissolve in perchloric-sulfuric acid but required the addition of hydrofluoric acid. The original tracer had a niobium/zirconium radioactivity ratio of 1.7. The filtered solvent solutions had ratios varying from 0.37 to 106 with 14 of 46 samples having ratios below 2.0, the value calculated for the solid. Only two had ratios above 10.

Two organic solutions contacted with 4 M HNO_3 became discolored on equilibration with zirconium dibutyl phosphate.

The solubility of zirconium dibutyl phosphate has been reported by Moffat and Whitener⁴ as 0.0 and 3.6 g of zirconium per liter in water and 3% TBP in Amsco, respectively. The values found in the present work are 0.22 mg per liter of water and 1.34 g per liter of 30% TBP in Amsco. The value for the solubility in 3% TBP in Amsco was obtained with an equilibration of more than 1 week.

2.3 Preparation and Composition of Zirconium Dibutyl Phosphate

Four inactive and one Zr-95 labeled samples of zirconium dibutyl phosphate were made. The first precipitation, C-18-2, was performed by adding 1.3 ml of dibutyl phosphoric acid, HDBP, to 100 ml of a hot, 60-70°C, solution adjusted to 0.064 M zirconium in 3 M HNO_3 . This sample was filtered and dried under vacuum. The second precipitation, C-19-1, was performed by adding 100 ml of 13.7% solution of HDBP in ethanol to 100 ml of a hot, 65°C, 0.065 M zirconium in 3 M HNO_3 . The precipitate was filtered and dried under vacuum. The third precipitation, C-21-1, was performed by dropwise simultaneous addition of equal volumes of 0.130 M NaDBP and 0.065 M zirconium in 3 M HNO_3 to 2 M HNO_3 solution. During this addition vigorous stirring was used and the solution-precipitate was heated to 40°C. The precipitate was filtered, then redispersed in 200 ml of 2 M HNO_3 and stirred for 20 min, and finally refiltered and dried under vacuum. Conditions of the fourth precipitation, C-27-1, were the same as those of the third except that the solution was not heated.

The Zr-95 tracer used in this work was received as 10 mcuries of Zr-95 and 17 mcuries of Nb-95 in 6.9 ml of oxalate solution. This solution was diluted to 47 ml, the oxalate destroyed by addition of a small excess of potassium permanganate solution, and the excess of the latter just destroyed with hydroxylamine hydrochloride solution. Water and a stock solution of 5.89 M H^+ , 0.71 M Zr, 0.35 M Na, and 8.20 M NO_3^- were added to the Zr-95 solution to make 100 ml containing 0.25 M Zr and 2.80 M HNO_3 . This solution and 250 ml of 0.20 M NaDBP were added dropwise, with slow stirring, to 100 ml of 2 M HNO_3 . The zirconium was added much more rapidly than the NaDBP solution, which was added over a period of 30 min. After digesting for 30 min, the precipitate was filtered and washed with 100 ml of distilled water. The liquid was pulled out of the filter cake by vacuum filtration and the solid dried for 24 hr under a heat lamp. This material was labeled C-32-1.

Chemical and thermogravimetric analyses (Table 2 and Fig. 1) of the zirconium dibutyl phosphate preparations indicate that different compounds were formed under the various conditions of precipitation, neglecting differences in the amounts of water and nitric acid of solvolysis. Thus, the atomic ratio P/Zr, varying from 1 to 3, indicates a dibutyl phosphate anion to zirconium cation ratio of 1 to 3 since all phosphorus was added as the hydrogen or sodium dibutyl phosphate. Since the thermogravimetric weight loss at 200°C, a temperature at which sorbed solvent was removed (Fig. 1),

Table 2. Analyses of Zirconium Dibutyl
Phosphate Preparations

Sample	Chemical Analyses, wt %				Mole Ratios				Loss on Heating to 200°C, wt %
	Zr	P	C	NO ₃	P/Zr	C/Zr	NO ₃ /Zr	C/P	
C-18-1	6.38	7.36	27.51	--	3.38	32.7	--	9.7	8.5
C-19-1	5.25	4.13	14.63	21.25	2.32	21.2	5.9	9.2	41.3
C-21-1	14.72	10.82	28.18	14.30	2.16	14.6	1.4	6.8	15.8
C-27-1	20.10	6.77	30.54	11.47	0.98	11.5	0.84	11.6	4.4
C-32-1	12.81 ^a	--	--	--	--	--	--	--	--

^aThis value should be 14.3% on the basis of a Zr-95 material balance and the weight of the dried precipitate.

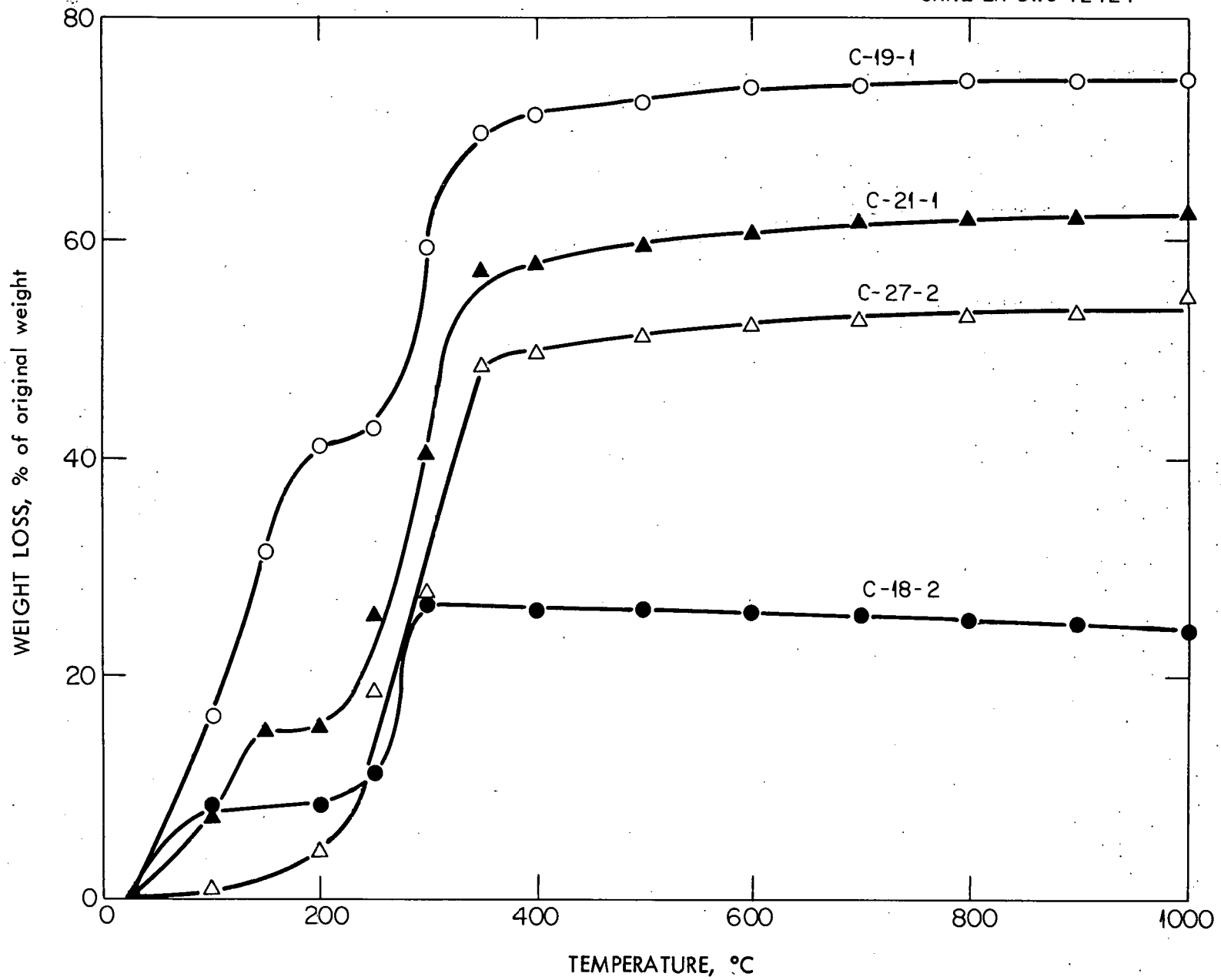


Fig. 1. Thermogravimetric curves for zirconium dibutyl phosphates.

does not account for all the nitrate anion, each preparation contained at least one nitrate ion. In view of the ratio $(\text{DBP}^- + \text{NO}_3^-)/\text{Zr}$ being between 2 and 4, it is evident that the precipitates did not contain the zirconyl (ZrO^{++}) grouping but contained zirconium as the Zr^{4+} cation. The varying compositions can be explained by a general formula $\text{Zr}(\text{OH})_x(\text{NO}_3)_y(\text{DBP})_z$ in which the only restriction is that $x + y + z = 4$. The chemical analyses and thermogravimetric weight loss of preparation C-21-1 correspond very closely to the formula $\text{Zr}(\text{OH})(\text{NO}_3)(\text{DBP})_2 \cdot 1/2 \text{H}_2\text{O} \cdot 1/2 \text{HNO}_3$. The water and nitric acid may be combined as solvates, or, more probably, may be merely sorbed on and in the solid. In the latter case it is fortuitous that excess water and nitric acid are equal and add up to 1 mole per mole of zirconium dibutyl phosphate.

The present interpretation of analyses of zirconium dibutyl phosphate preparations is consistent with that of Moffat,³ who has prepared, by adding an acetone solution of HDBP to a 2 M HNO_3 solution of zirconium, a compound containing 14.4% Zr, 18.0% NO_3 , 29.4% C, and 5.0% H, which are very close to the theoretical values for $\text{Zr}(\text{NO}_3)_2(\text{DBP})_2$.

Only zirconium in the Zr-95 labeled zirconium dibutyl phosphoric acid preparation was determined. Since this material, C-32-1, was prepared in a manner very nearly like that used in preparing materials C-21-1 and C-27-1, except that C-32-1 was dried with an infrared lamp for 6 hr, it is believed that its composition corresponds to $\text{Zr}(\text{OH})(\text{NO}_3)(\text{DBP})_2$. The theoretical zirconium content of this compound is 15.50 wt % as compared with the analytical value of 12.81 wt % zirconium in preparation C-32-1. However, the latter value is too low on the basis of the Zr-95 material balance.

3.0 EXPERIMENTAL PROCEDURES

Commercial dibutyl phosphoric acid from Victor Chemical Company, containing 87.7% HDBP (dibutyl phosphoric acid) and 1.8% H_2MBP (monobutyl phosphoric acid) with most of the remainder being water, was purified as follows. Forty-seven milliliters of HDBP was heated with 10 g of activated charcoal. This was diluted with 10 ml of carbon tetrachloride and filtered. After diluting the filtered solution with 250 ml of carbon tetrachloride, the HDBP was scrubbed four times with 50-ml portions of distilled water to remove the H_2MBP . The HDBP was then contacted with three 70-ml volumes of 1.0 N NaOH, each of which converted one third of the original acid to the water-soluble sodium salt. The third extraction yielded a yellowish solution with brown interfacial solids. The first two extracts, both acidic to pH paper, were combined and scrubbed four times with 32-ml portions of ether to remove organic-soluble components. Ninety percent of the equivalent amount of nitric acid and 32 ml of ether were added and mixed. The organic phase was separated and the ether removed under vacuum. Similarly prepared materials had equivalent weights of 233.5, water contents of 10.8 to 16.3%, and less than 0.2% H_2MBP .

A zirconium stock solution was prepared by dissolving 40 g of A. D. McKay "pure zirconium hydroxide" in 50 ml of hot concentrated

nitric acid. After adding another 50 ml of acid and refluxing, the solids were allowed to settle. The supernatant was filtered and diluted to 200 ml. Less than 1 g of undissolved solids remained. This solution contained 5.89 M H^+ , 0.71 M Zr, 0.35 M Na^+ , and 8.20 M NO_3^- . The sodium, as well as small quantities of aluminum, boron, cadmium, copper, iron, and lead, was introduced from the zirconium hydroxide.

The solutions (Table 3) in which the solubility of zirconium dibutyl phosphate was to be measured were prepared from a stock solution, containing 408 g of uranium per liter and 1.02 M HNO_3 , c.p. nitric acid, drum grade Amsco 125-82, and sodium carbonate-washed tributyl phosphate (TBP). Since some of the solutions were to have higher uranium/acid mole ratios than were available in the stock solution, UO_3 was prepared by thermal decomposition of UO_4 . The UO_3 was added to portions of the stock solution to decrease the acid concentration or to raise the uranium concentration. The solutions with very low acidities were prepared from recrystallized uranyl nitrate hexahydrate. Thirty-six milliliters of 1.126 M TBP in Amsco (300 g of TBP per liter) was equilibrated with five successive 20-ml batches of each of the 16 aqueous solutions. After a sixth equilibration with 35 ml of aqueous solution, the phases were separated and filtered to remove entrained material.

The concentration of zirconium dissolved in each of the test solutions was calculated from experimental gamma counting data⁵ and the known specific activity of Zr-95 in preparation C-32-1.

Equilibration of the Zr-95 labeled zirconium dibutyl phosphate with the various solutions was carried out as follows. About 65 mg of the dried solid was added, as a mixture of powder and small lumps, to 20-ml portions of each aqueous or organic solution, which was contained in a 25-ml glass-stoppered graduated cylinder. The solids dispersed well in the organic solutions but not in the aqueous solutions. An additional 130 mg was added to each of the organic solutions after they had stood for 72 hr. The solutions were then tumbled at room temperature (20-26°C) and 20 rpm for 48 to 96 hr. Samples of the tumbled solutions were pipetted and filtered through Whatman No. 1 filter paper and analyzed. Samples 1A and 2A were returned to the tumbler after sampling and tumbled for an additional 168 hr. Additional solid was added to samples 8A, 1-0, 6-0, and 7-0 after sampling, and they, too, were returned to the tumbler.

4.0. PROCESS APPLICATIONS

The present data and those of Moffat and Whitener⁴ on the solubility of zirconium dibutyl phosphate in uranium nitrate-nitric acid aqueous or TBP-hydrocarbon solutions have an immediate use in predicting the behavior of zirconium in radiochemical reprocessing of nuclear fuels. For example, after extended burnup, the ratio of total atoms of zirconium to atoms of U-235 fissioned for natural uranium fuel reaches a limiting value⁷ of 0.32, corresponding to 29 mg of total zirconium per mole of natural uranium. If this

Table 3. Compositions of Aqueous and Organic Solutions Prior to Contacting with Zirconium Dibutyl Phosphate

Aqueous Phase				30% TBP-Amsco Phase at Equilibrium				
Sample No.	H ⁺	U	Density, g/l	Sample No.	H ⁺	U	H ₂ O	Density, g/l
	Conc, M	Conc, g/l			Conc, M	Conc, g/l	Conc, g/l	
1-A	0.08	287	1.3793	1-0	0.000	96.4	13.17	0.9478
2-A	0.17	490	1.6634	2-0	0.005	127	24.39	0.9839
3-A	0.85	464	1.6441	3-0	0.035	124	22.61	0.9852
4-A	3.94	368	1.6079	4-0	0.08	122	17.38	0.9823
5-A	0.04	118	1.1579	5-0	0.011	83.1	13.92	0.9075
6-A	0.22	119	1.1644	6-0	0.02	100	14.20	0.9514
7-A	0.94	121	1.1889	7-0	0.04	110	20.93	0.9648
8-A	3.86	118	1.2789	8-0	0.09	115	10.28	0.9749
9-A	0.011	47.7	1.0623	9-0	0.004	46.4	21.03	0.8796
10-A	0.23	42.9	1.0577	10-0	0.019	54.4	18.69	0.8928
11-A	0.95	40.7	1.0848	11-0	0.06	81.7	12.15	0.9322
12-A	2.99	41.6	1.1556	12-0	0.12	102	13.08	0.9592
13-A	0.000	0	0.9947	13-0	0.004	0	12.80	0.8193
14-A	0.26	0	1.0557	14-0	0.033	0	15.15	0.8185
15-A	1.00	0	1.0302	15-0	0.22	0	22.52	0.8248
16-A	3.96	0	1.1264	16-0	0.82	0	22.34	0.8424

uranium is reprocessed in the Purex process, the uranium content of the IAF stream is about 1.5 M; the associated zirconium content would be, at most, 44 mg of zirconium per liter. Since the ratio of solvent to feed flow rate is about 5, the zirconium content would be equivalent to 8.8 mg of zirconium per liter of solvent. Thus, the 44 mg of zirconium per liter, maximum, in the feed would be close to the solubility limit in the aqueous phase (Table 1) but the 8.8 mg per liter of solvent would be well below the solubility value of 1 to 5 g/liter. Hence, zirconium dibutyl phosphate would not precipitate from the extractant; it might precipitate at the feed plate of a solvent extraction unit if the uranium concentration is decreased sufficiently by scrub solution.

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