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A Method of Estimating Distribution Coefficients of Degradation Products from Organophosphorus Extractants

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A METHOD OF ESTIMATING DISTRIBUTION COEFFICIENTS OF DEGRADATION PRODUCTS FROM ORGANOPHOSPHORUS EXTRACTANTS

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A METHOD OF ESTIMATING DISTRIBUTION COEFFICIENTS OF DEGRADATION PRODUCTS FROM ORGANOPHOSPHORUS EXTRACTANTS

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ABSTRACT

The total phosphorus content of stripping solutions after their equilibration with organophosphorus extractants in successive contacts with fresh strip solution was employed to estimate the concentrations of phosphorus-containing hydrolysis (or degradation) products in the extractant phase and their distribution coefficients. Aqueous solutions and ethylene glycol were used as stripping agents. The procedure is especially useful for estimates of the concentrations of phosphorus-containing impurities that are originally present in the extractant or that are generated in separations processes. The utility of the method was demonstrated for separations processes that employ the following extractants: di-(2-ethylhexyl)-phosphoric acid, tributyl phosphate, and dihexyl-N,N-diethylcarbamylmethylene phosphonate.

1. INTRODUCTION

The distributions of organic extractants and their degradation products between the extractant phase and other process liquid phases are important parameters in separations processes based on solvent extraction. Extractant losses due to their solubility in other process liquid phases and their degradation by hydrolysis or radiolysis may be costly. Additionally, the degradation products may interfere with the desired separations. Phosphorus-containing degradation products are generally more soluble in

aqueous phases or in other liquid phases that contain hydrophilic molecules than is the parent extractant. This is true primarily because these degradation products are principally produced by removal of lipophile groups from the parent extractant. Purification methods for extractants are often based on this difference in solubility.

Analytical methods for estimation of distribution coefficients for the extractant and its degradation products between the extractant phase and the other phases with which it is contacted in chemical separations processes has often proved cumbersome. For a few mature processes such as Purex [1], such information has been obtained from cumulative effects in operating systems and from ^{32}P-tagged degradation products. Other methods have seen limited application: the titration procedure[2] lacks sensitivity, the radioactive tracer method[3] requires prior identification and preparation of each compound of interest, and the gas chromatographic method[4] demands prior identification of each compound of interest as well as a suitable analytical procedure for its measurement. Consequently, adequate knowledge of the effects of extractant losses and degradation is lacking for many proposed solvent extraction processes[5].

A general procedure is described for the estimation of distribution coefficient values of phosphorus-containing impurities which only requires determination of the phosphorus concentration of strip solutions that are used to preferentially strip the impurities. In this procedure, a known volume of impure extractant is contacted with known volumes of fresh stripping agent using several stages of batch, crosscurrent stripping. Samples of the stripping solutions from each of the equilibrium stages are analyzed for phosphorus by a suitable analytical method. The

distribution coefficient values of the impurities for the stripping process may be calculated using the equation which relates their phosphorus concentrations in the strip solution phase to the stage number. It is assumed that distribution coefficients for the extractable species in each stripping stage are the same. This procedure is applicable to cases having the following factors in common: (1) the impurities have a structural constituent such as phosphorus, sulfur, or nitrogen that is the same as the functional constitutent of the extractant; (2) the distribution coefficient values of the species are sufficiently different from each other and from that of the pure extractant; and (3) the stripping solutions employed do not initially contain any of the element used to analytically measure the concentrations of the impurities and extractant in the strip solution phase after its equilibration with the impure extractants.

The general procedure was applied to the estimation of distribution coefficient values for impurities derived from the extractants, di-(2-ethylhexyl) phosphoric acid (HDEHP), tributyl phosphate (TBP), and dihexyl-N,N-diethylcarbamylmethylene phosphonate (DHDECMP). The study was limited to the impurities that are the principal hydrolysis products of these extractants and are listed below:

Extractant	Impurity
HDEHP	Mono-(2-ethylhexyl)phosphoric acid (H_2MEHP)
TBP	Di-(n-butyl)phosphoric acid (DBP)
	Mono-(n-butyl)phosphoric acid (MBP)
DHDECMP	Unidentified

The distribution coefficients of interest were those relating to the partitioning of the impurities between the organophosphous extractant

phases and the other liquid phases that are contacted in certain chemical separations processes. Specifically, the distribution coefficient was determined for the partitioning of H_2MEHP between HDEHP-diluent phases and the various other liquid phases of a proposed Talspeak[5] process for the separation of lanthanides from trivalent actinides. These other process liquids were: (1) aqueous solutions of glycolic acid containing ethylenediaminepentacetic acid (DTPA), which were neutralized to a pH value of either 1.5 or 3; (2) oxalic acid solutions; and (3) ethylene glycol. The diluents employed with HDEHP were diethyl benzene (DEB), di-isopropylbenzene (DIPB), and normal dodecane (NDD) in which the HDEHP concentrations were 0.7 M and 0.5 M, respectively. Distribution coefficient values for the principal hydrolysis products of TBP and DHDECMP were determined between an aqueous nitric acid phase and phases of 1 M TBP in NDD and 1 M DHDECMP in DIPB, respectively.

2. RATIONALE OF METHOD

The procedure is based entirely on differences in distribution of the parent organophosphorus extractant and its impurities between the extractant-diluent phase and a phase which is more hydrophilic (e.g., an aqueous solution or ethylene glycol). By using successive contacts of the extractant-diluent phase with a more hydrophilic phase, the impurities which are more hydrophilic are preferentially removed. Since the extractant is present in a relatively large excess and also has a low solubility in the contacting phase, its concentration in aqueous solutions or ethylene glycol will be essentially constant after each stripping phase contact. The concentration of a hydrophilic impurity in the equilibrium stripping phase,

however, will diminish on repeated contacts of the extractant-diluent phase with the fresh stripping agents in crosscurrent stripping; the concentration approaches zero after this essentially complete removal from the extractant. After impurity removal, the total phosphorus analysis measures the solubility of the extractant in the stripping agent. Therefore, data regarding solubility of the extractant in the liquid phases of various tests may also be obtained.

If the distribution coefficient for a given species is constant, it may be estimated by the following equation that is readily derivable from the material-balance relationships in the crosscurrent stripping of an organic extractant with equal volumes of a stripping solution:

$$A_n = (1 + 1/K_d)^{-n} \cdot \frac{O_o}{K_d} \qquad (1)$$

where

n = number of equal volume stripping stages

A_n = concentration of impurity in the hydrophilic phase of the $\underline{n}^{th}$ stages

O_o = concentration of impurity in the original organic extractant (a constant)

K_d = concentration of impurity in the extractant phase divided by its concentration in the stripping phase.

Using the logarithmic form of eqn. 1, the slope of $[-\log(1 + 1/K_d)]$ and hence K_d can be evaluated from plots of A versus n,

$$\log A_n = [-\log (1 + \frac{1}{K_d})] \cdot n + \log \frac{O_o}{K_d} \qquad (2)$$

An equation similar to eqn. 2 has been proposed by Dean[6] for the

determination of K_d values of extractants and describes the change in concentration of the extractant phase as a function of n. In the case for unequal volumes of the organic and aqueous phase, eqn. 2 becomes:

$$\log A_n = [-\log (1 + \frac{1}{K_d} \cdot \frac{V_a}{V_o})]\ n + \log \frac{O_o}{K_d} \qquad (3)$$

where

V_o = volume of organic phase

V_a = volume of aqueous phase.

When only one impurity exists or is overwhelmingly predominant relative to other impurities, plots of phosphorus concentration of the stripping phase (corrected for solubility of the pure extractant) versus n according to eqn. 2 will yield a straight line. A curve comprised of composite straight lines is obtained when appreciable amounts of more than one phosphorus-containing degradation product is present. If K_d values for the impurities are sufficiently different, the composite curve may be resolved into its components in a manner similar to that used to resolve composite decay curves for radionculides[7]. The quantity of extractant present in the equilibrium strip liquid phase can be estimated either in separate experiments using pure extractants or by carrying out a sufficient number of stripping stages until all of the impurities are removed and the total phosphorus analysis of the subsequent equilibrium strip liquid phase is a measure of the soluble extractant.

The value of O_o, when unknown for a given impurity, may be estimated by the following method: (1) computation using the experimentally determined values of the slope and intercept from plots of log A versus n according to eqn. 2 or (2) summation of the phosphorus contents attribut-

able to that impurity in each stripping phase if the number of crosscurrent stages employed are sufficient to completely remove that impurity.

3. EXPERIMENTAL

3.1 Phosphorus Analysis

Phosphorus contents of aqueous phases were determined by phosphate ion analyses. Aqueous-phase samples were fumed with a 1:1 mixture of concentrated nitric and perchloric acids to convert all phosphorus compounds to the phosphate ion. The phosphate ion was measured either by the phosphovanadomolybdate or molybdenum blue method depending on the phosphate ion concentration.

The reliability of the method was established from a series of standard samples (Table 1). Good agreement was obtained between the actual and the analyzed values. Samples 2 through 7 of the standard series were prepared by making a dilution of Sample 1. Sample 1 was an aqueous 1 M glycolic acid - 0.05 M DTPA (pH = 3) solution that contained dissolved H_2MEHP and HDEHP. The value given for Sample 1 (Table 1) is the average of three determinations for total phosphorus 27.8, 27.6, and 27.5 mM.

3.2 The Contacting Phases

Successive stripping contacts were made at 25°C using equal volumes of extractant and stripping phases. An equal volume ratio was satisfactory in the present work. However, in some cases it may be helpful to use other ratios. For example if the impurity has a large value of K_d, it would be advantageous to increase the ratio of stripping volume to extractant volume to a value such that the impurity was completely removed by 5 to 10 succes-

Table 1. Comparison of values for the total phosphorus concentration of standard samples with those obtained by analysis

Standard sample number	Phosphate concentrations, mM	
	Actual	Analysis
1	27.6	27.7
2	13.8	14.2
3	6.92	7.04
4	3.47	3.37
5	1.75	1.82
6	0.88	0.86
7	0.45	0.47

sive contacts. For low K_d values, it would be desirable to decrease the ratio of stripping volume to extractant volume so that 5 to 10 experimental points are obtained. The K_d values of the degradation products in this study were sufficiently low to completely remove them from the extractant after 3 to 10 equal volume contacts. A 2-min contact time was sufficient to attain equilibrium. Aqueous phases were centrifuged to minimize the quantity of any entrained extractant phase. The commercial grade DHDECMP was obtained from the Carolina Chemical Company. After scrubbing out unidentified acidic compounds with caustic[8], the DHDECMP was purified by molecular distillation[9]. The DIPB was commercial grade. All chemicals employed to prepare stripping solutions from distilled water were reagent grade.

4. RESULTS AND DISCUSSION

The data showed that phosphorus content of the strip phase may be used to determine the presence of certain phosphorus-containing impurities in organophosphorus extractants. In some cases, it is possible to use the method for estimations of the distribution of these impurities throughout the various liquid streams employed in specific separations processes based on solvent extractions. Applicability of the method was demonstrated for the Talspeak process[5]. Limited studies also indicated the method may be useful for similar type estimations in solvent extraction processes[1,8] based on the use of TBP or DHDECMP.

4.1 HDEHP

The distribution of H_2MEHP between the immiscible liquid phases pre-

sent in a modified Talspeak process (Fig. 1) were not known. Of particular interest was the satisfactory purification of the HDEHP-diluent extractants for their recycle. Initially, the reproducibility of the experimental method and constancy of K_d values was determined (Table 2). In these four replicate tests, 10 ml of 0.7 M HDEHP-0.05 M H_2MEHP in DEB was stripped with 10-ml quantities of a 1 M glycolic acid - 0.05 M DTPA solution (pH 3) in six successive contacts of the organic phase with the fresh aqueous phase. Reproducibility of phosphorus concentrations of the equilibrated aqueous phases were quite good, as were the K_d values calculated for H_2MEHP. The K_d values were calculated by linear-regression analysis using eqn. 2. The concentrations of H_2MEHP employed in K_d calculations were obtained by subtracting the HDEHP concentrations (0.15 mM) from the total phosphorus concentration. Conformity of measured values to eqn. 2 is demonstrated by Fig. 2.

The values of K_d determined for other process steps of the Talspeak flowsheet are shown in Table 3. Values of K_d were observed to increase slightly as glycolic acid concentration of the aqueous phase was increased. The effect of pH was quite dramatic. The K_d values were decreased nearly an order of magnitude by increasing the pH of the glycolate solution from 1.5 to 3.0 by the addition of sodium hydroxide. This result suggests that increased formation of the sodium salt of H_2MEHP in the aqueous phase at higher pH values lessens the H_2MEHP extractability by HDEHP-DEB. Losses of HDEHP to the aqueous glycolic acid phases were well below 0.40 mM and were slightly less when NDD was the diluent rather than DEB. These concentrations of HDEHP were constant in all of the stripping stages as evidenced by the constancy of measured values of total phosphorus after all

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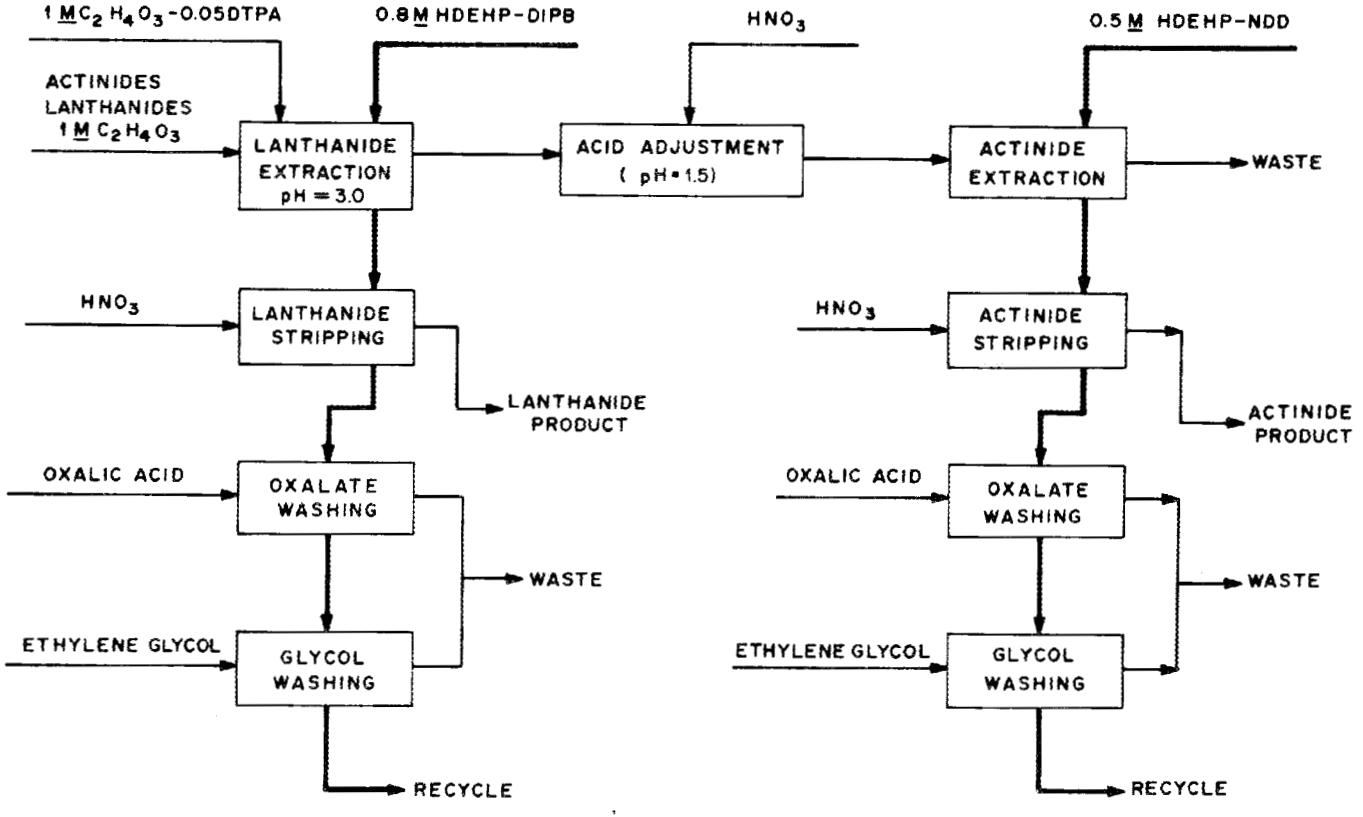

Fig. 1. Chemical flowsheet for separation of americium and curium from the lanthanide elements. The feed and the 1 M $C_2H_4O_3$- 0.05 M DTPA scrubbing solution are adjusted to pH = 3 with NaOH for lanthanide extraction.

Table 2. Total phosphorus concentrations and the K_d values of H_2MEHP for the crosscurrent stripping of 0.7 M HDEHP-0.05 M H_2MEHP in DEB with an aqueous, 1 M glycolic acid-0.05 DTPA solution (pH = 3)

Replicate series	A	B	C	D
Total phosphorus concentration of aqueous phase, mM				
Stage 1	27.7	27.8	27.6	27.5
2	12.7	12.8	12.4	12.6
3	5.32	5.22	5.46	5.48
4	2.23	2.31	2.18	2.37
5	1.00	1.07	1.02	
6	0.45	0.52	0.48	0.52
K_d	0.682	0.727	0.702	0.729

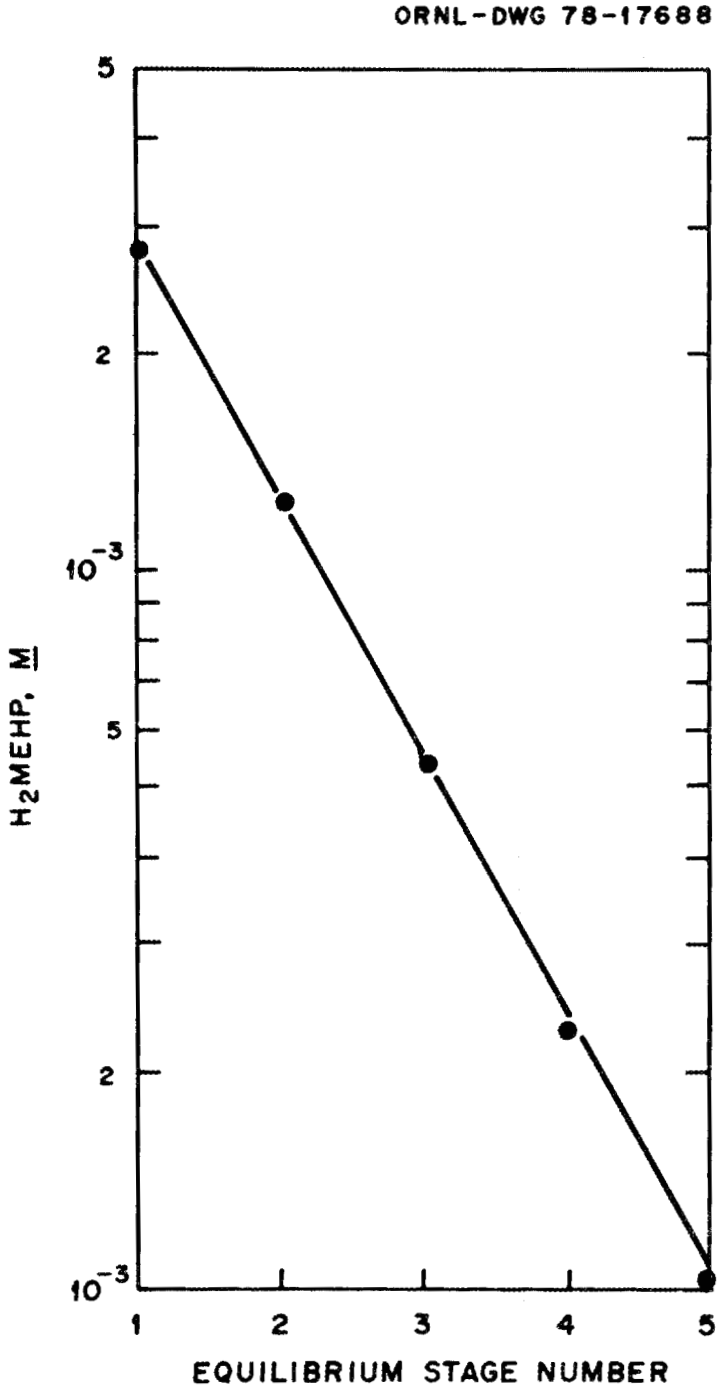

Fig. 2. Stripping of H_2MEHP from 0.7 M HDEHP - 0.05 M H_2MEHP - DEB. Aqueous phase: 1 M $C_2H_4O_3$ - 0.05 M DTPA ($p\overline{H}$ = 3).

Table 3. Calculated values of K_d for H_2MEHP using eqn. 2 and the measured solubility of HDEHP in the aqueous phase

Extractant	Stripping liquid	K_d	HDEHP solubility in strip phase, mM
0.7 M HDEHP-DEB	Aqueous, 1 M $C_2H_4O_3$-0.05 M DTPA (pH = 3)	0.66	0.15
0.7 M HDEHP-DEB	Aqueous, 2 M $C_2H_4O_3$-0.05 M DTPA (pH = 3)	1.00	0.40
0.7 M HDEHP-DEB	Aqueous, 1 M $C_2H_4O_3$-0.05 M DTPA (pH = 1.5)	5.78	0.10
0.5 M HDEHP-NDD	Aqueous, 1 M $C_2H_4O_3$-0.05 M DTPA (pH = 1.5)	2.70	0.25
0.5 M HDEHP-NDD	Aqueous, 2 M $C_2H_4O_3$-0.05 M DTPA (pH = 1.5)	3.12	0.20
0.7 M HDEHP-DEB	Ethylene glycol	0.41	4.0
0.5 M HDEHP-DEB	Ethylene glycol	0.82	3.0
0.7 M HDEHP-DEB	10 wt % oxalic acid	1.11	0.80

of the H_2MEHP had been stripped from the HDEHP phase. The K_d values for H_2MEHP were about a factor of 2 higher when NDD was the diluent rather than DEB. Losses of extractant using ethylene glycol to remove H_2MEHP would be expected to be moderate. The measured concentration of HDEHP in the ethylene glycol was 4 m$\underline{M}$. Hydrogen bonding would appear to be a logical explanation of the marginally favorable K_d for stripping H_2MEHP from HDEHP-DEB with oxalic acid. The oxalic acid scrub is primarily employed to remove any fission product zirconium impurity that may be present in the actinide-lanthanide feed. Any H_2MEHP formed by hydrolysis in the lanthanide or actinide stripping steps (see Fig. 1) would not be removed by the nitric acid. The total phosphorus analyses indicated its aqueous solubility was much too low (<5 m$\underline{M}$).

The presence of trivalent elements in the extractant does not severely hamper the removal of H_2MEHP from the extractant when it is stripped with 1 $\underline{M}$ glycolic acid - 0.05 $\underline{M}$ DTPA (pH = 3). Tests conducted with commercial HDEHP (0.8 $\underline{M}$ in DIPB) containing 0.04 $\underline{M}$ europium and 0.012 $\underline{M}$ H_2MEHP showed the H_2MEHP was stripped in few stages (see Fig. 3). The K_d values for Eu were constant throughout the stripping stages within experimental error (18.5 $\pm$ 1.0).

The knowledge of K_d values for impurities in the various steps of chemical separations processes are of considerable import. For example, in the lanthanide extraction step (see Fig. 1), the system is "self-cleaning" with respect to the extractant when impurities are generated by radiation and/or hydrolysis within the system. The measured K_d values indicate its concentrations could not build up to appreciable values in the extractant because appreciable quantities of H_2MEHP are rejected to the trivalent

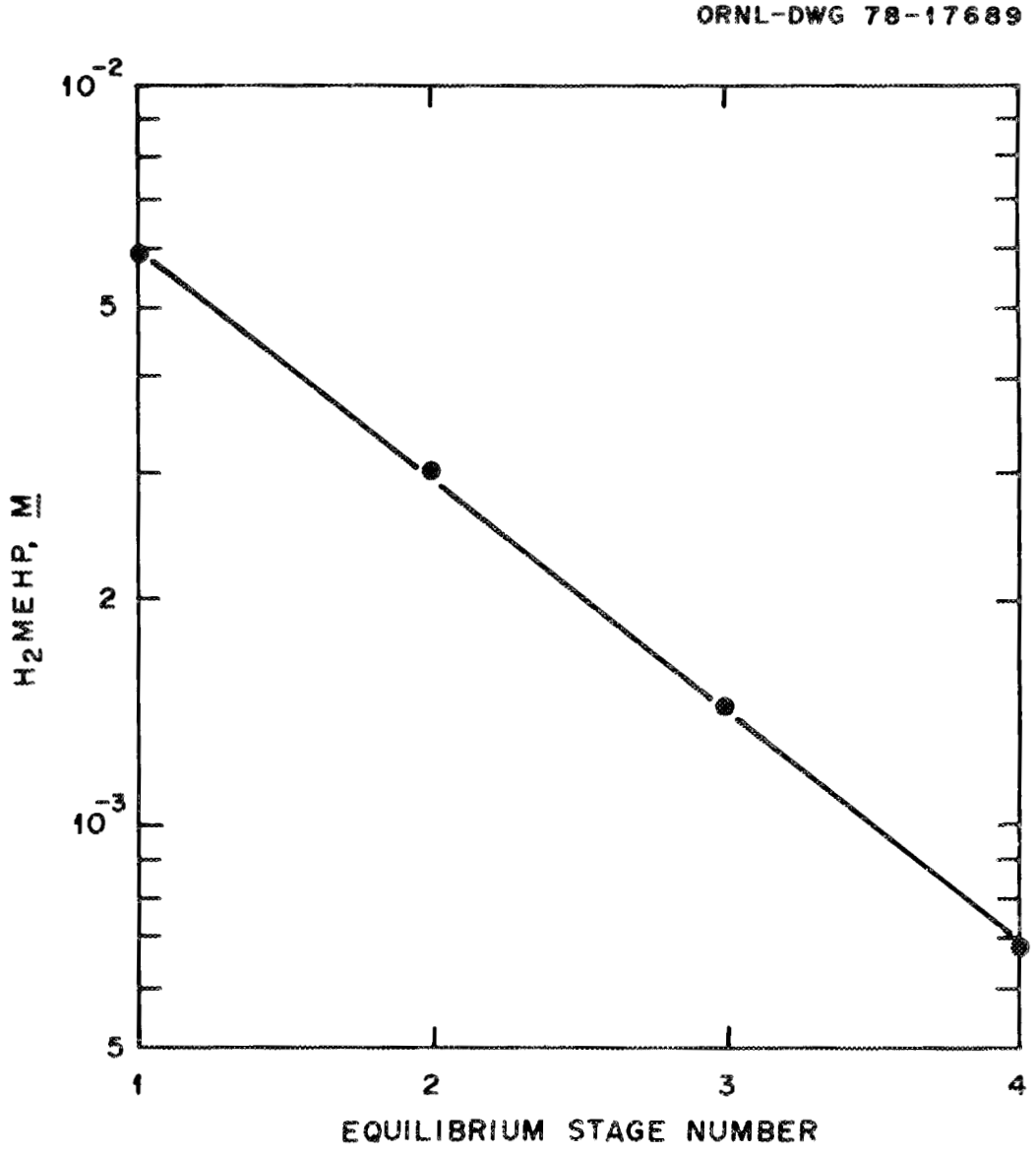

Fig. 3. Stripping of H_2MEHP with the presence of europium in the organic phase. Organic phase: 0.8 M HDEHP - 0.01 M H_2MEHP - 0.04 M Eu in DIPB; aqueous phase: 1 M $C_2H_4O_3$ - 0.5 M $DTPA^{-}$ (pH = 3).

actinide product. However, in the actinide extraction step where lower pH values are employed, the K_d values indicate that appreciable buildup would be expected unless the extractant were purified by glycolic acid extraction prior to its recycle.

4.2 TBP

Distribution coefficients were determined for DBP and MBP between 1 M TBP-NDD and 3 M HNO_3 employing a 40 mM concentration for each of these impurities in the extractant. The K_d values were 0.79 and 6.2 for MBP and DBP, respectively. The K_d values were calculated from a determination of the phosphorus concentrations of the aqueous phases from five stages of crosscurrent stripping. In order to determine if the total phosphorus method was suitable for the estimation of rates of TBP hydrolysis, the following experiment was performed. The organic phase remaining from the above experiment was aged for 29 days at room temperature (∿25°C) and was again stripped with 3 M HNO_3. The total phosphorus concentration of the combined aqueous phases was 0.8 mM, which is about a factor of 2 higher than the 0.42 mM value obtained using pure extractant. Thus the total phosphorus content is satisfactory for detecting the occurrence of such low rates of hydrolysis.

4.3 DHDECMP

A process based on the use of this extractant for the extraction of lanthanides and trivalent actinides from nitric acid solutions is currently under development[8]. Although the hydrolysis products have not been identified, stability of the extractant and the distribution of these im-

purities between the process liquid phases are of interest. Impurities for the present work were generated by equilibrating 1 M DHDECMP-DIPB with 3 M HNO_3, separating the phases, and allowing the organic phase to age for 29 days. Subsequent stripping tests were carried out in a manner identical to those with the aged 1 M TBP phase that was discussed previously. Total phosphorus determination showed that the rate of hydrolysis of DHDECMP was somewhat higher than that for TBP. The observed concentrations of phosphorus in the combined 3 M HNO_3 strip solutions were ~1.4 mM, whereas 0.65 mM would have been expected using unaged extractant. The principal hydrolysis products, though unidentified, were partially stripped by 3 M HNO_3. The observed decrease in phosphorus concentration of the aqueous phase as additional contacts were made indicated a K_d value that was probably <0.2.

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