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Extraction Behavior of Uranium(VI) with Polyurethane Foam

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Abstract

The extraction of uranium(VI) from aqueous solution with polyether-based polyurethane (PU) foam was studied. The effects of the kinds and concentrations of nitrate salts, uranium(VI) concentration, temperature, nitric acid concentration, pH, the content of poly(ethylene oxide) in the polyurethane foam, and the ratio of PU foam weight and solution volume on the extraction of uranium(VI) were investigated. The interferences of fluoride and carbonate ions on the extraction of uranium(VI) were also examined, and methods to overcome both interferences were suggested. It was found that no uranium was extracted in the absence of a nitrate salting-out agent, and the extraction behaviors of uranium(VI) with polyurethane foam could be explained in terms of an etherlike solvent extraction mechanism. In addition, the percentage extraction of a multiple stage was also estimated theoretically.

INTRODUCTION

Since Bowen reported that flexible polyurethane (PU) foams can be used as selective absorbents for a number of substances from dilute aqueous solution, the application of PU foam for the extraction and separation of various inorganic and organic species has attracted considerable attention (*1*, *2*). Because of the advantages of a high distribution coefficient, inexpensiveness, squeezability, and excellent hydrodynamic properties, PU foam has increasing importance in separation science.

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The mechanism for the extraction of inorganic and organic compounds with PU foam is not completely clear. Because of its large sorption capacity (0.5–1.5 mol/kg foam), it is now recognized that extraction is not a surface phenomenon and sorption into the bulk of PU must occur (*1*, *3*). Extraction phenomena with PU foam were generally interpreted by either a solvent-extraction mechanism (*1*, *3*) or a cation-chelation mechanism (*4*). Bowen (*1*) first found that most of the substances extracted by PU foam can be extracted from aqueous solutions by liquid diethyl ether. The view of an etherlike solvent extraction mechanism was also supported by Gesser et al. (*5*), Lo and Chow (*6*, *7*), Gesser and Gupta (*8*), Korkisch et al. (*9*), Abbas et al. (*10*), and Schumack and Chow (*3*).

The cation-chelation mechanism, proposed by Hamon et al. (*4*), demonstrated that PU foam acts predominantly as a long noncyclic polyether, and the metal ions are extracted in the form of anionic metal complexes which are solvated within the polymer matrix with the accompanying cations strongly solvated within crown-ether type structures within the polymer configuration. Accordingly, polyether-based PU foams which can form crown-ether type structures, particularly those containing poly(ethylene oxide), are superior to polyester-based PU foams for extraction, and the extraction efficiencies depend on the sizes of the countercations. The mechanism was also supported by Al-Bazi and Chow (*11*, *12*), Khan et al. (*13*), and Caletka et al. (*14*).

The application of PU foam for the extraction of uranium(VI) from nitrate solution has been studied by a few workers (*1*, *8*, *9*, *15*). Bowen (*1*) was the first to recover uranium(VI) from saturated aluminum nitrate solution with PU foam. Gesser et al. (*15*) reported that uranium(VI) can diffuse through a PU membrane in the presence of aluminum nitrate. From the effects of temperature and the concentrations of NH_4NO_3, $Ca(NO_3)_2$, and $Al(NO_3)_3$, Gesser and Gupta (*8*) and Korkisch et al. (*9*) found that the extraction of uranium(VI) with an open cell PU foam sponge is similar to and more efficient than that with liquid diethyl ether. Korkisch et al. (*9*) also investigated the possibility of the separation of uranium and thorium with PU foam.

In our present work the extraction of uranium(VI) from aqueous solution with polyether-based PU foam has been studied extensively. The effects of the kinds and concentrations of nitrate salts, the content of poly(ethylene oxide) (PEO) in PU foam, uranium(VI) concentration, temperature, nitric acid concentration, pH, the ratio of the weight of PU foam and solution volume, and the interferences of fluoride and carbonate ions on the extraction of uranium(VI) have been investigated. In addition, the extraction efficiency of a multiple stage has also been estimated theoretically.

EXPERIMENTAL SECTION

Materials

Four types of polyether-based PU foams containing 0, 7, 10, and 12% PEO were kind gifts of Chiunglong Petrochemical Co., Ltd. These foams were pretreated with 1 mol/dm^3 HCl and acetone to remove possible inorganic and organic contaminants as described in the literature (*4*, *8*). Uranyl nitrate was purified with TBP extraction and then by recrystallization. All other chemicals were E.P. grade reagents. Deionized and distilled water was used for the preparation of solutions.

Procedure

The extraction of uranium(VI) with PU foam was conducted by a batch method. Generally, 10 cm^3 of a sample solution and 0.1–0.2 g of foam were placed in a stoppered test tube kept in a water bath at the desired temperature to achieve equilibrium. Preliminary experiments showed that equilibrium was achieved in about 1 day. To ensure the establishment of equilibrium, extraction was done for 2.5–3 days.

The pH value of the solution was adjusted by the addition of NH_4OH and HNO_3. Unless otherwise stated, the PU foam containing 12% PEO was used in this study, and the experiments were carried out in nitrate solutions containing 0.06 mol/dm^3 HNO_3 and 2.10 mmol/dm^3 uranium(VI) at 25°C.

The concentration of uranium(VI) in solution was determined by the thiocyanate method except that a 10% (w/v) solution of hydroxylamine hydrochloride was used instead of stannous chloride (*8*, *16*). In the presence of 0–0.5 mol/dm^3 fluoride and 0–1.0 mol/dm^3 carbonate ions, which were introduced in the form of sodium fluoride and sodium carbonate, respectively, appropriate amounts of $Al(NO_3)_3$ ($[Al^{3+}]/[F^-] > 1$) and HNO_3 solution (pH 1) were added to eliminate interferences during the analysis for the uranium(VI) concentration. The amount of uranium(VI) extracted in the PU foam was determined from the difference between the initial and final concentrations of uranium(VI) in solution.

The percentage extraction (E) and distribution coefficient (D) of uranium are defined as (*4*)

$$E = \frac{([U]_0 - [U]_{eq})}{[U]_0} \times 100\% \quad (1)$$

and

$$D = \frac{VE}{(100 - E)W} \quad (2)$$

where $[U]_0$ and $[U]_{eq}$ are the initial and equilibrium uranium(VI) concentrations in solution, V is the solution volume, and W is the weight of PU foam.

RESULTS AND DISCUSSION

Effect of Nitrate Salts

In the preliminary experiments it was found that no uranium was extracted in the absence of a nitrate salting-out agent, even if the content of PEO in the PU foam (0–12%), the uranium(VI) concentration (4.20×10^{-5} to 0.420 mol/dm^3), the temperature (15–45°C), the pH (1.3–3), and the nitric acid concentration (0.06–3.06 mol/dm^3) were varied.

The effects of the kinds and concentrations of nitrate salts on the extraction of uranium(VI) showed that the distribution coefficient of uranium between PU foam and aqueous nitrate solution is significantly enhanced with increasing nitrate concentration due to the salting-out effect, as shown in Fig. 1. It was also observed that the salting-out effects of different nitrate salts increase in the sequence $KNO_3 < NH_4NO_3 < NaNO_3 < Ca(NO_3)_2 < Mg(NO_3)_2 < Al(NO_3)_3$, which is the same as the order of the hydration of

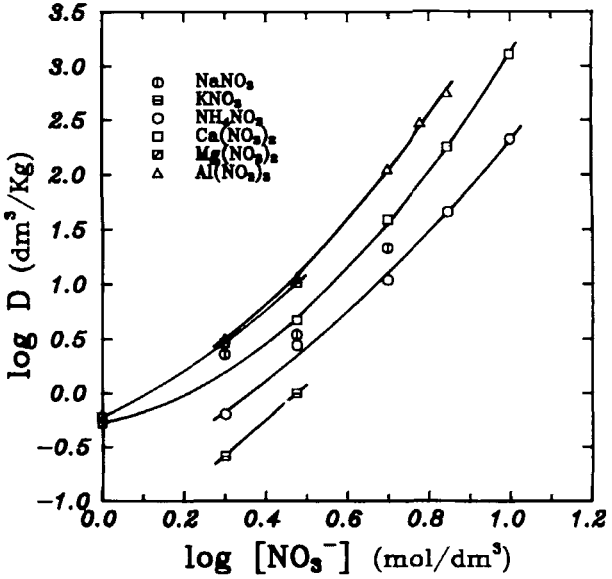

FIG. 1. Effects of the kinds and concentrations of nitrate salts on the distribution coefficient of uranium in 0.06 mol/dm^3 HNO_3 solution containing an initial uranium(VI) concentration of 2.10 mmol/dm^3 at 25°C.

their cations (*17*), but in disagreement with what is predicted by the cation-chelation mechanism (*4*, *11*, *12*, *14*). These phenomena are similar to those observed in the liquid–liquid extraction of uranium(VI) with diethyl ether (*18*), and they support the belief that the extraction of uranium(VI) with PU foam is characteristic of an etherlike solvent extraction mechanism.

Effect of PEO content in PU Foam

Four types of PU foam containing 0, 7, 10, and 12% PEO were tested. As shown in Table 1, the results indicated that the distribution coefficients of uranium increase with increasing PEO content in PU foam with different nitrates and nitrate concentrations.

According to the cation-chelation mechanism, this phenomenon can be explained by the fact that long-chain PEO may form a helical configuration and more effectively complex with uranium than can poly(propylene oxide) (PPO) due to the steric hindrance of the methyl group of PPO (*4*). However, the cation-chelation mechanism has been shown to be unsatisfactory for explaining the effect of nitrate salts. Since uranyl nitrate is generally extracted into ether in the form of a mixed solvate-hydrate, water molecules forming hydrogen bonds between the extractant and the extracted species play an important role in the extraction of uranium(VI) with ether (*19*). Therefore, the effect of the PEO content in PU foam may be attributed to the fact that increasing the content of PEO in PU foam makes the foam more hydrophilic and favors contact of solution and foam, which leads to an increase in the extraction of the solvated-hydrated species of uranium.

Effect of Uranium(VI) Concentration

The effects of equilibrium uranium(VI) concentration ($0.5–5 \times 10^{-5}$ mol/dm^3) in NH_4NO_3, $Ca(NO_3)_2$, and $Al(NO_3)_3$ solutions on the distri-

TABLE 1
Effect of the PEO Content in PU Foam on the Distribution Coefficient of Uranium in Various Nitrate Solutions Containing 0.06 mol/dm^3 HNO_3 at 25°C and an Initial Uranium(VI) Concentration of 2.10 mmol/dm^3

	D (dm^3/kg)				
% PEO	7 *N* NH_4NO_3	10 *N* NH_4NO_3	7 *N* $Ca(NO_3)_2$	10 *N* $Ca(NO_3)_2$	7 *N* $Al(NO_3)_3$
12	45.3	211	178	1286	566
10	44.0	191	118	1046	497
7	41.0	173	119	571	487
0	22.7	126	94.6	555	478

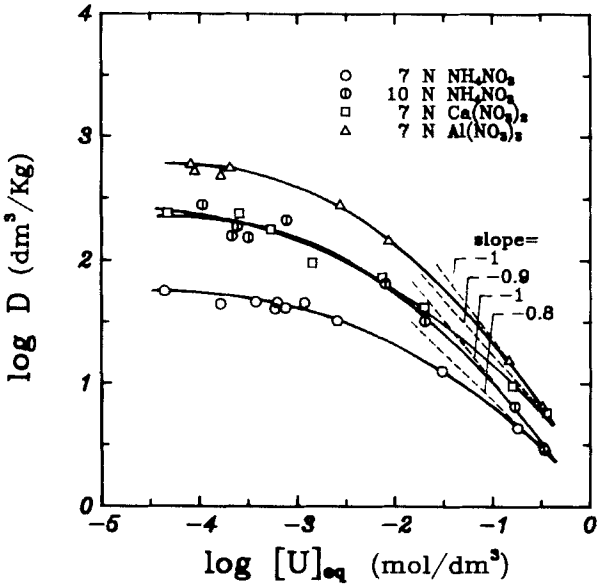

FIG. 2. A log-log plot of the distribution coefficient of uranium and equilibrium uranium(VI) concentration for various nitrate systems containing 0.06 mol/dm^3 HNO_3 at 25°C.

bution coefficients of uranium are shown in Fig. 2, where the effects of different nitrates and the nitrate concentration are also illustrated. It can be seen that the distribution coefficient of uranium decreases with an increase in the equilibrium uranium(VI) concentration in solution, particularly within the higher uranium(VI) concentration range. The slopes of the curve of log D vs log $[U]_{eq}$ at high uranium(VI) concentrations were found to approach -1. This result is similar to that observed by Gesser and Gupta (*8*), and could be interpreted by the fact that the foam has been saturated with uranium(VI).

From Eqs. (1) and (2), the following equation can be derived:

$$\log D = \log (([U]_0 - [U]_{eq})V/W) - \log [U]_{eq} \qquad (3)$$

When the foam is saturated with uranium(VI), the value of $([U]_0 - [U]_{eq})V/W$ is constant, and hence the slope is equal to -1. The larger slopes for the cases of 7 *N* NH_4NO_3 (-0.8) and 7 *N* $Ca(NO_3)_2$ (-0.9) may be due to the fact that the foams have not been saturated with uranium(VI).

The data in Fig. 2 were recalculated and illustrated in a log-log plot of the extraction capacity of PU foam ($([U]_0 - [U]_{eq})V/W$) and $[U]_{eq}$ as shown

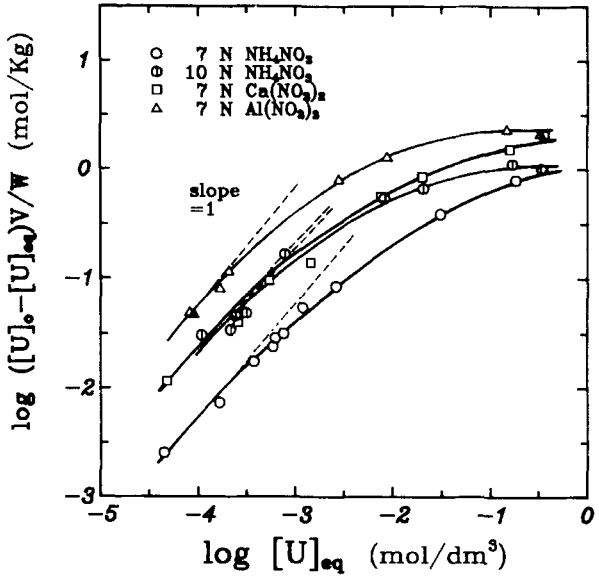

FIG. 3. A log-log plot of the extraction capacity of PU foam and equilibrium uranium(VI) concentration for various nitrate systems containing 0.06 mol/dm^3 HNO_3 at 25°C.

in Fig. 3. At low equilibrium uranium(VI) concentrations, all the slopes of the curves are approximately equal to 1. This implies that uranium(VI) should be extracted into PU foam in a mononuclear type at low uranium(VI) concentrations.

Effect of Temperature

The temperature dependence of the distribution coefficient of uranium was investigated in NH_4NO_3, $Ca(NO_3)_2$, and $Al(NO_3)_3$ solutions within the temperature range of 15 to 45°C. Plots of log D vs $1/T$, as shown in Fig. 4, indicate that higher distribution coefficients of uranium can be obtained at a lower temperature, which is similar to extraction with diethyl ether (*19*).

Since the distribution coefficient D can be expressed as (*8*)

$$\log D = \frac{-\Delta H}{2.303RT} + \frac{\Delta S}{2.303R} \tag{4}$$

where ΔH and ΔS are the changes of enthalpy and entropy, respectively, and R is the gas constant.

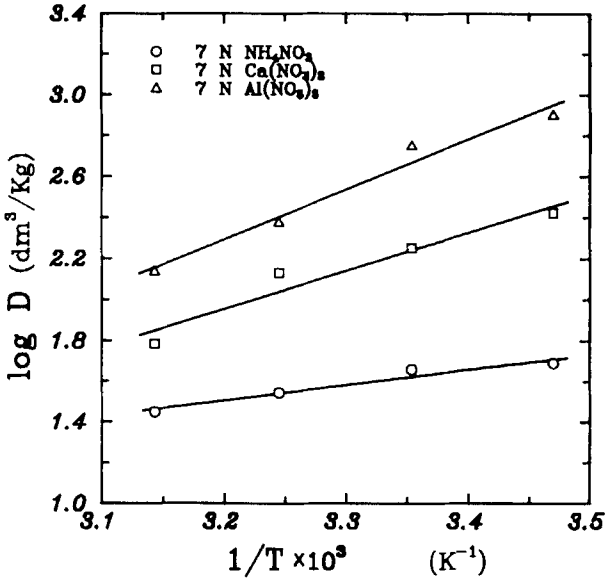

FIG. 4. A plot of log D vs $1/T$ for various nitrate systems containing 0.06 mol/dm^3 HNO_3 and an initial uranium(VI) concentration of 2.10 mmol/dm^3.

The values of ΔH and ΔS for the three cases illustrated in Fig. 4 were calculated and are listed in Table 2. They indicate that the changes of enthalpy and entropy for different nitrate salts follow the order of the salting-out effect: $NH_4NO_3 < Ca(NO_3)_2 < Al(NO_3)_3$.

Effect of Nitric Acid Concentration

The effects of nitric acid concentration in various nitrate solutions were studied at a constant total nitrate ion concentration of 7.06 N. The results are shown in Table 3, where the data obtained at various concentrations

TABLE 2
The Values of ΔH and ΔS in Various Nitrate Solutions Containing 0.06 mol/dm^3 HNO_3 at an Initial Uranium(VI) Concentration of 2.10 mmol/dm^3

Nitrate	ΔH (kJ/mol)	ΔS (J/mol·K)
7 N NH_4NO_3	−14.6	−18.0
7 N $Ca(NO_3)_2$	−35.6	−76.5
7 N $Al(NO_3)_3$	−46.6	−105.0

TABLE 3
Effect of Nitric Acid Concentration on the Distribution Coefficient of Uranium at a Total Nitrate Ion Concentration of 7.06 mol/dm^3 with an Initial Uranium(VI) Concentration of 2.10 mmol/dm^3 at 25°C

$[HNO_3]$ (mol/dm^3)	Nitrate salt (N)	NH_4NO_3 D/D^a (dm^3/kg)	$Ca(NO_3)_2$ D/D^a (dm^3/kg)	$Al(NO_3)_3$ D/D^a (dm^3/kg)
0.06	7.00	45.3/45.3	178/178	566/566
1.06	6.00	7.5/25.1	13.5/100	59.6/297
2.06	5.00	5.5/10.8	8.4/38.4	21.7/113
3.06	4.00	0.3/5.6	5.2/15.9	9.5/45.7

[a]Obtained under the same conditions except that $[HNO_3]$ was fixed at 0.06 mol/dm^3.

of nitrate salts and a constant nitric acid of 0.06 mol/dm^3 are also listed for comparison. The distribution coefficients of uranium were found to decrease with increasing concentration of nitric acid for all cases, which is in agreement with what was observed by Korkisch et al. (*9*). This indicates that the acidity of the solution can lower the extraction efficiency.

Effect of pH

The effect of pH on the distribution coefficient of uranium was examined in 10 N NH_4NO_3. A plot of log D vs pH indicates that the optimum pH is around 1, as shown in Fig. 5. In the more acidic range, the D value will decrease due to the acidity of the solution, as observed in the last section. The lower distribution coefficient found when pH > 3 could be due to the hydrolysis of uranyl ion into a nonextractable species (*20*, *21*). When pH > 4, the precipitation of uranium(VI) is significantly observed. This is probably due to the formation of ammonium diuranate (*19*).

Interferences of Fluoride and Carbonate Ions

The fluoride and carbonate complexes of uranyl ion are frequently present in solution in the uranium refining process, and more stable than uranyl nitrate (*19*). It was found from preliminary experiments that they cannot be extracted with PU foam. The interferences of fluoride and carbonate ions on the extraction of uranium(VI) in 10 N NH_4NO_3 solution are shown in Fig. 6. It was observed that the interference of fluoride ion is more significant than that of carbonate ion. Because of the strong complex of fluoride and uranyl ions, almost no uranium was extracted when the concentration of fluoride ion was larger than 0.25 mol/dm^3. The smaller in-

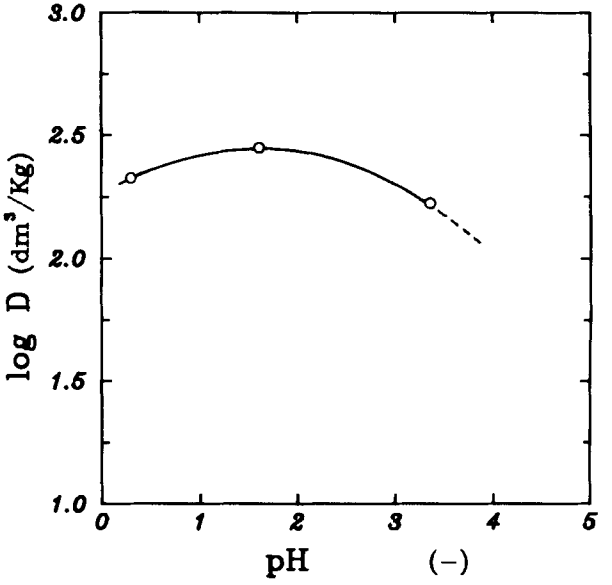

FIG. 5. Effect of pH on the distribution coefficient of uranium in 10 N NH_4NO_3 solution containing an initial uranium(VI) concentration of 2.10 mmol/dm³ at 25°C.

terference of the carbonate ion may be attributed to the fact that the complex of uranyl with carbonate ions is weaker than that with fluoride ion and that the acidity of the solution partially converts carbonate ions to carbon dioxide gas.

When the experiments were repeated with 10 N NH_4NO_3 replaced by 7 N $Al(NO_3)_3$, no interferences by fluoride and carbonate ions were observed. The elimination of the interference of fluoride ion could be due to the much stronger complex of Al^{3+} and F^- (*19*). Further experimental results showed that the interference of fluoride ion is eliminated almost completely when the concentration of Al^{3+} is larger than that of F^-. This is consistent with the results for the extraction of uranium(VI) with diethyl ether (*19*). The elimination of interference by carbonate ion may be due to the fact that the acidity of $Al(NO_3)_3$ is higher than that of NH_4NO_3. In fact, it has been confirmed by additional experiments that interference by carbonate ion can be eliminated by adding an appropriate amount of acid to reduce the solution pH to about 1. Therefore, it can be concluded that aluminum nitrate is a better nitrate salting-out agent for the extraction of uranium(VI) with PU foam because it eliminates interference by fluoride and carbonate ions.

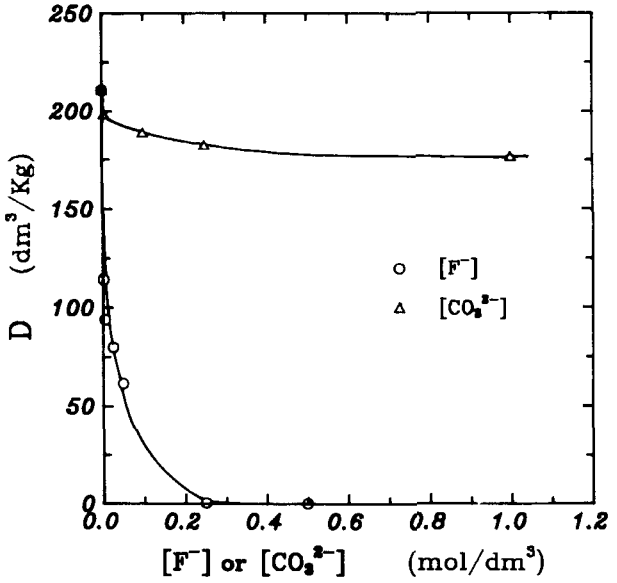

FIG. 6. Effects of fluoride and carbonate ion concentrations on the distribution coefficient of uranium in 10 *N* NH_4NO_3 solution containing 0.06 mol/dm^3 HNO_3 and an initial uranium(VI) concentration of 2.10 mmol/dm^3 at 25°C.

Effect of Foam Weight and Solution Volume Ratio

The effects of the ratio of the weight of PU foam and solution volume W/V (0.0025–0.0325 kg/dm^3) on the distribution coefficient of uranium and on the extraction capacity of PU foam were investigated in 10 *N* NH_4NO_3 solution. As can be seen from Fig. 7, W/V does not have a significant influence on the distribution coefficient of uranium. However, the extraction capacity of PU foam decreases with an increase in the value of W/V.

Extraction Efficiency of Multiple Stage

Multiple stage extraction is usually considered more efficient than single stage operation. The percentage extraction of a multiple stage can be expressed as

$$E_n\,(\%) = \left(1 - \frac{1}{(DW/nV + 1)^n}\right) \tag{5}$$

where n is the extraction stage and E_n is the total percentage extraction of an n stage extraction. Figure 8 shows the relationship of E_n and n at different

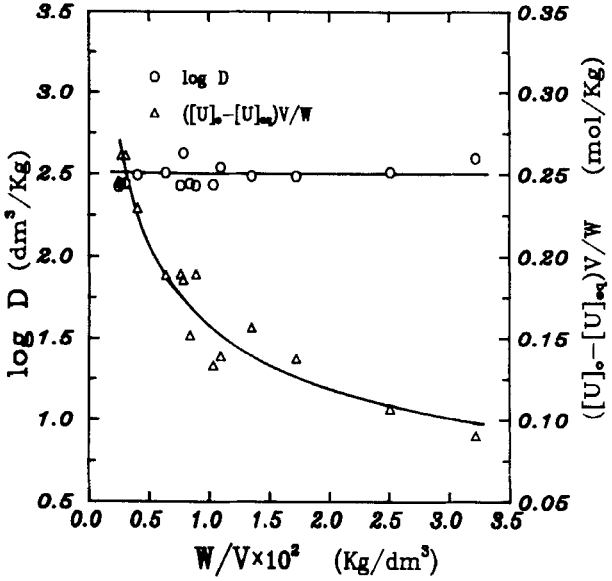

FIG. 7. Effects of W/V value on the distribution coefficient of uranium and the extraction capacity of PU foam in 10 N NH_4NO_3 solution containing 0.06 mol/dm^3 and an initial uranium(VI) concentration of 2.10 mmol/dm^3 at 25°C.

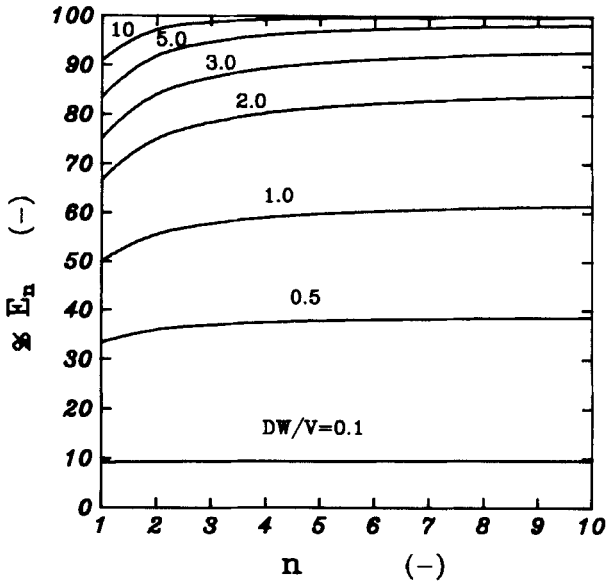

FIG. 8. Relationship of percentage extraction and the extraction stage at various DW/V values.

DW/V values ranging from 0.1 to 10, which covers most practical conditions. It is noteworthy that increasing DW/V enhances the extraction efficiency more significantly than does increasing the extraction stage. Since PU foam is quite inexpensive, it is better to operate the extraction process at a higher W/V ratio.

CONCLUSIONS

The extraction of uranium(VI) from aqueous solution with polyether-based PU foam was studied extensively. It was found that no uranium was extracted in the absence of a nitrate salting-out agent. The extraction efficiency increased significantly with increasing nitrate concentration and hydration of the cation of the nitrate salt. Increasing the PEO content in PU foam favored the extraction of uranium(VI). It was observed that the distribution coefficient of uranium decreases with increasing equilibrium uranium(VI) concentration in solution, and uranium(VI) should be extracted into PU foam of the mononuclear type at low uranium(VI) concentration. The temperature dependence showed that low temperature favors the extraction of uranium(VI), from which changes of enthalpies and entropies were determined. The acidity of the solution lowered the extraction efficiency, and the optimum pH value for the extraction of uranium(VI) with PU foam was found to be around 1. Beyond pH 4, a precipitate of uranium(VI) was observed, probably due to the formation of ammonium diuranate. It was also found that fluoride ions interfere significantly with the extraction of uranium(VI), but the interference can be overcome by the addition of aluminum nitrate. The interference of carbonate ion was less evident than that of fluoride ion and could be overcome by adjusting the pH value of the solution to about 1.

In addition, the distribution coefficient of uranium was not obviously affected by increasing the ratio of the weight of PU foam and solution volume, while the extraction capacity of the PU foam decreased. According to a theoretical estimation of the extraction of efficiency for a multiple stage with varying values of DW/V, increasing the W/V ratio may be more practicable than increasing the stage of extraction.

Acknowledgments

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NOMENCLATURE

D	distribution coefficient (dm^3/kg)
E	percentage extraction

E_n	percentage extraction of n stage
ΔH	enthalpy change (kJ/mol)
n	extraction stage
R	gas constant (J/mol·K)
ΔS	entropy change (J/mol·K)
T	temperature (K)
$[U]_0$	initial uranium concentration (mol/dm^3)
$[U]_{eq}$	equilibrium uranium concentration (mol/dm^3)
V	volume of solution (dm^3)
W	weight of PU foam (kg)
[ ]	concentration of the species in the bracket

Subscripts

eq	equilibrium value
n	extraction stage
0	initial value

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