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SORPTION STUDY OF URANIUM (VI) IN ALANINE MEDIUM USING POLY [DIBENZO-18 CROWN-6] AND COLUMN CHROMATOGRAPHY

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ABSTRACT

A simple chromatographic separation method has been developed for quantitative sorption of Uranium(VI) from an aqueous solution of 1×10^{-3} M alanine using poly [dibenzo-18-crown-6] as stationary phase. The sorption of Uranium(VI) was quantitative 1×10^{-2}M to 1×10^{-5}M Alanine. The elution of Uranium(VI) was quantitative with 2.0–5.0 M HCl, 2.0-4.0M $HClO_4$. The capacity of poly [dibenzo-18-crown-6] for Uranium(VI) was found to be 1.092 ± 0.01 mmol/g of crown polymer. The effects of concentrations of Alanine, Uranium(VI) foreign ions and eluents have been studied. Uranium(VI) was separated from a number of cations in multicomponent mixtures. The applicability of the proposed method was ascertained for the determination of Uranium(VI) in real samples. The reliability of method was checked by comparison of the results with those obtained using flame photometer. The method is very simple, rapid and selective with good reproducibility (approximately±2%).

KEYWORDS: Sorption, Separation, Uranium (VI), Alanine, poly [dibenzo-18- crown-6], Chromatography.

INTRODUCTION

Separation methods based on extraction chromatography have become increasingly popular in radiochemical analysis. This is due to their simplicity, rapidity and the savings in reagent and waste disposal costs compared to the traditional separation methods based on e.g., anion exchange and liquid-liquid extraction. Uranium metal in its pure form is chemically active, anisotropic and has poor mechanical properties. On the other hand, uranium alloys are useful in diluting enriched uranium liquid fuel meant for nuclear reactors and pure uranium coated with silicon and canned in aluminium tubes are used in production reactors. However uranium and its compounds, like lead are highly toxic which cause progressive or irreversible renal injury and in acute cases may lead to kidney failure and death. The tolerable daily intake of uranium established by WHO based on Gilman's studies is 0.6 g/kg of body weight per day.[1-3] A considerable amount of work has been done in recent years on the separation of uranium from various other elements.[4-13]

According to literature survey reveals that there are no reports on the use of poly [dibenzo-18-rown-6] for the chromatographic separation of uranium (VI) in Alaninemedium. The aim of the present study is to optimize conditions on a laboratory scale for the chromatographic separation and recovery of uranium (VI) from other associated metal ions employing poly[dibenzo-18-crown-6] as the sorbent in Alanineas counter ion.

EXPERIMENTAL

Apparatus and reagents

A Ziess (German) Spectrophotometer, a digital pH meter (Model LI-120, ELICO, India) with glass and calomel electrodes and a digital Flame photometer (PI, Model No. 041, India) were used.

A stock solution of uranium(VI) was prepared by dissolving 2.166 g of uranlynitrate hexahydrate (Anala R grade, BDH, Poole, UK) in 100 mL of distilled deionized water and standardized gravimetrically.[14] A solution containing 100 µg/mL of uranium(VI) was prepared by appropriate dilution of standard stock solution. Alanine solution (1×10^{-1} M) was prepared by dissolving 2.22 g of Alanine in distilled demonized water and diluted to 250 mL. Poly[dibenzo-18-crown-6] (E Merck Darmstadt, Germany) was used after screening to 100-200 mesh. A total of 0.5 g of polymer was slurred with distilled deionised water and poured into a Pyrex glass chromatographic column (20×0.8 cm i.d.). The column was used after preconditioning with Alanine solution.

General procedure

50 µg of uranium (VI) was mixed with Alanine in the concentration range of 1×10^{-8} M to 1×10^{-1}M in a total volume of 10 mL. The solution was then passed through poly [dibenzo-18-crown-6] column, preconditioning with same concentration of Alanine as that of the sample solution at flow rate of 0.5 mL/min. The column was then washed with the same concentration of alanine. The sorbed uranium(VI) was then eluted with different eluting

agents (described later) at the flow rate of 0.5 mL/min. 5.0 mL fraction were collected and uranium(VI) in the aqueous phase was determined spectrophotometrically with Arsenazo(III) at 660 nm.[15-16] The concentration of uranium (VI) was calculated from a calibration graph.

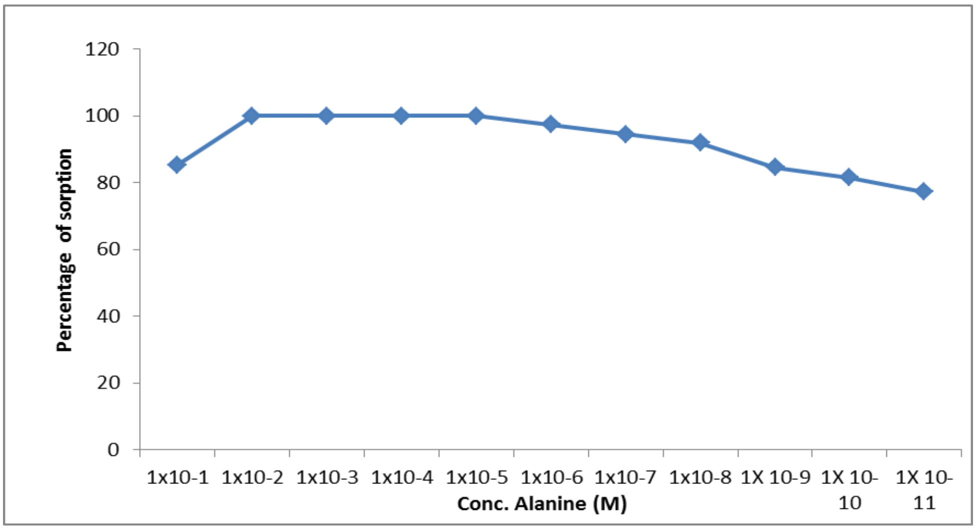
RESULTS AND DISCUSSION

Sorption of uranium (VI) on poly[dibenzo-18-crown-6] as a function of alanine concentration

Sorption studies of uranium (VI) were carried out from alanine medium. The concentration of alanine was varied from 1×10^{-8} M to 1×10^{-1}M. After sorption, the elution of uranium(VI) was carried out with 4.0 M hydrochloric acid. It was found that there was quantitative (100%) sorption of uranium (VI) from 1×10^{-5} M to 1×10^{-2} M alanine. The subsequent sorption studies of uranium(VI) were carried out with 1×10^{-3} M alanine.

Table: Sorption of uranium (VI) as a function of alanine concentration U(VI) = 50 µg/mL, Eluent = 4.0 M HCl.

Alanine concentration (M)	Sorption of U(VI) (%)
$1x10^{-1}$	85.4
$1x10^{-2}$	100
$1x10^{-3}$	100
$1x10^{-4}$	100
$1x10^{-5}$	100
$1x10^{-6}$	97.5
$1x10^{-7}$	94.6
$1x10^{-8}$	92



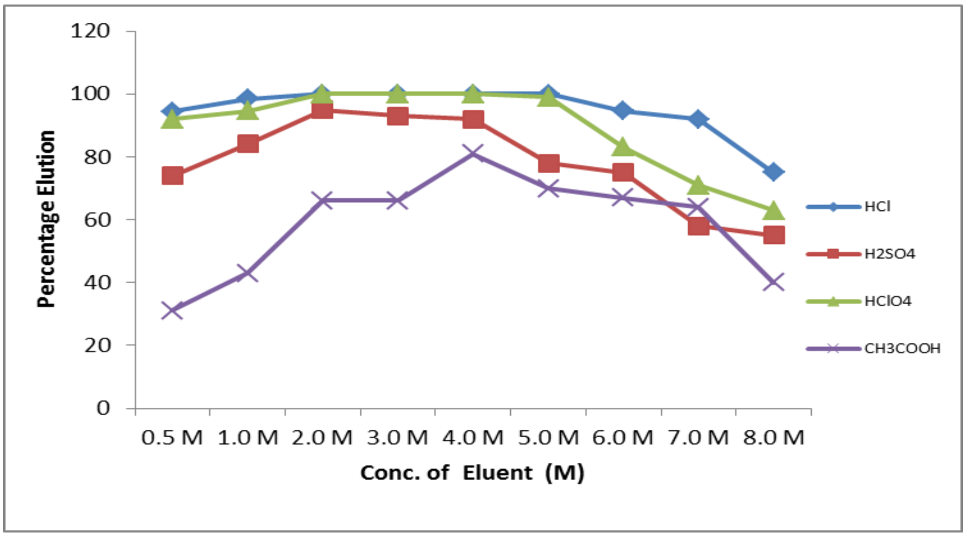
Sorption of Uranium (VI) as a function of alanine concentration

Elution study of uranium (VI) with various eluting agents

50µg/mL of Uranium (VI) was sorbed on the poly [dibenzo-18-crown-6] column at 1×10^{-3}M alanine concentration. After sorption, elution of Uranium (VI) was carried out using hydrochloric acid, sulphuric acid, perchloric acid and acetic acid. The concentration of eluting agents varied from 0.5 M to 8.0 M. The elution profile of uranium (VI) with various eluting agents is shown in figure. Showed that uranium (VI) was quantitatively eluted with 2.0 M to 5.0 M hydrochloric acid, 2.0 M to 4.0 M perchloric acid where as sulphuric acid and acetic acid were found to be inefficient eluents for uranium(VI). Further elution studies of uranium (VI) in this work was carried out with 4.0 M hydrochloric acid.

Table Elution of uranium (VI) with different eluting agents U(VI) = 50 µg/mL, alanine 1×10^{-3}M.

Conc. **Acid**	**0.5**	**1.0**	**2.0**	**3.0**	**4.0**	**5.0**	**6.0**	**7.0**	**8.0**
	Percentage Elution								
HCl	94.4	98.5	100	100	100	100	94.5	92	75
H_2SO_4	74	84	95	93	92.0	78	75	58	55
$HClO_4$	92	94.5	100	100	100	99	83	71	63
CH_3COOH	31	43	66	66	81	70	67	64	40



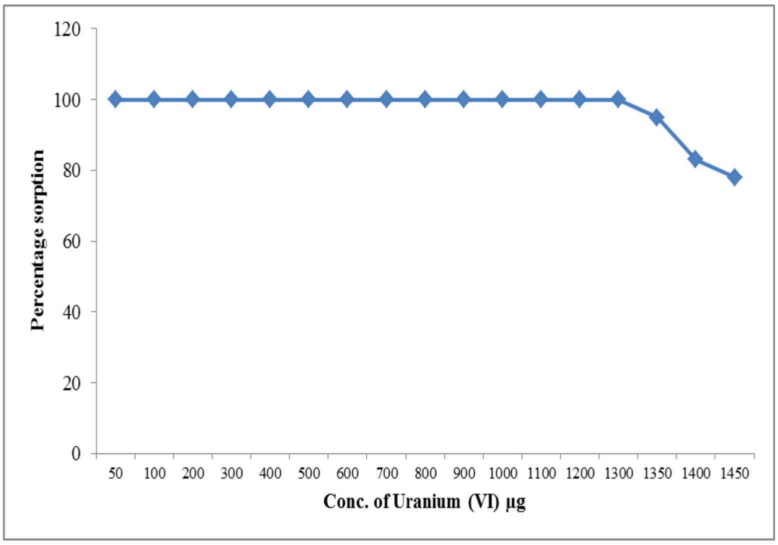
Elution of uranium (VI) with various eluting agents.

Effect of varying concentration of uranium (VI)

In order to find out the capacity of poly [dibenzo-18-crown-6] for the uranium(VI), the concentration of uranium(VI) was varied from 100-1000 µg/10 mL in alanine and 4.0 M hydrochloric acid as eluent. The results showed that the sorption of uranium(VI) was quantitative (100%) up to 1300µg. With increase in concentration of uranium(VI) there was decrease in the percentage sorption of uranium(VI). From this study it was found that the capacity of poly [dibenzo-18-crown-6] for uranium(VI) was found to be 1.092 ± 0.01 mmol/g of crown polymer.

Table: Effect of varying concentration of uranium(VI) alanine 0.001 M, eluent 1.0 M HCl.

Uranium (VI) (μg)	Percentage sorption (%)
50	100
100	100
200	100
300	100
400	100
500	100
600	100
700	100
800	100
900	100
1000	100
1100	100
1200	100
1300	100
1350	95
1400	83
1450	78



Effect of varying concentration of uranium (VI).

Separation of uranium (VI) from binary mixtures

An aliquot of solution containing 50µg of uranium (VI) was mixed with foreign ions and alanine was added so that its concentration was 1×10^{-3} M in total volume of 10 mL. The tolerance limit was set as the amount of foreign ions required to cause ±2% deviation in the recovery of uranium (VI). The solution was passed through a poly[dibenzo-18-crown-6] column, preconditioned with 1×10^{-3} M alanine at a flow rate of 0.5 mL/min. Subsequently the column was washed with 15 mL of 1×10^{-3} M alanine to remove unsorbed metal ions. Various foreign ions were not sorbed and hence passed through the column. The effluent was collected and analyzed for foreign ion content. The tolerance limit of various foreign ions is shown in Table. The most of the alkali metals show high tolerance limit. In case of alkaline metal lithium(I), sodium(I), potassium (I), rubidium(I) and cesium(I) are strongly tolerates. Most of the p-block and d-block elements were sorbed and shows low tolerance limit. Amongst the inner transition elements, as compared lanthanum(III), cerium(III), thallium(III) show low tolerance limit. The anion of inorganic and organic acids showed high tolerance limit.

Table: Separation of uranium(VI) from binary mixtures U(VI)- 50 µg, sorption- 1×10^{-3} M alanine, eluent- 2.0 M HCl.

Ion	Added as	Tol. Limit (mg)	Ion	Added as	Tol. Limit
Li^+	LiCl	14	Tl^{3+}	$Tl(NO_3)_3.3H_2O$	0.1
Na^+	NaCl	11	La^{3+}	$La(NO_3)_3.6H_2O$	0.1
K^+	KCl	7	Ce^{3+}	$CeCl_3.6H_2O$	0.05
Rb^+	RbCl	9	V^{4+}	$VOSO_4.4H_2O$	0.5
Cs^+	CsCl	12	Th^{4+}	$Th(NO_3)_4$	Co-extran
NH_4^+	NH_4Cl	18	Cr^{6+}	$K_2Cr_2O_7$	0.2
Be^{2+}	$BeSO_4.4H_2O$	Co-extran	Mo^{6+}	$(NH_4)_6Mo_7O_{24}.4H_2O$	14
Mg^{2+}	$MgCl_2.6H_2O$	0.5	W^{6+}	$Na_2WO_4O.4H_2O$	6
Ca^{2+}	$CaCl_2$	0.5	Cl^-	HCl	1
Sr^{2+}	$Sr(NO_3)_2$	0.2	Br-	HBr	12
Ba^{2+}	$Ba(NO_3)_2$	0.8	SCN^-	NaSCN	5
Co^{2+}	$CoCl_2.6H_2O$	Co-extran	ClO_4^-	$HClO_4$	4
Ni^{2+}	$NiCl_2.6H_2O$	Co-extran	CH_3COO^-	CH_3COOH	3
Zn^{2+}	$ZnCl_2$	0.1	SO4 $_2{}^-$	H_2SO_4	1
U^{6+}	$UO_2(NO_3)_2.6H_2O$	1	$BO_3{}^{3-}$	H_3BO_3	5
Pb^{2+}	$Pb(NO_3)_2$	6	Tartrate	Tartaric acid	15
Cr^{2+}	$Cr(NO_3)_3.9H_2O$	0.1	EDTA	EDTA	6.5
Fe^{3+}	$FeCl_3.6H_2O$	0.5	Ascorbate	Ascorbic acid	3
Al^{3+}	$Al(NO_3)_3.9H_2O$	0.1			

Separation of uranium (VI) from Multicomponent mixtures

Separation of uranium (VI) was carried out from number of associated elements in multicomponent mixture. The mixture containing lithium(I), uranium(VI), thorium(IV), potassium(I), sodium(I), strontium(II), lead(II) was passed through the poly[dibenzo-18-crown-6] column at 1×10^{-3} M alanine concentration, lithium(I), potassium (I), sodium(I) was not sorbed and hence passed through the column. The uranium(VI), thorium(IV), lead(II), strontium(II) were sorbed. The sorbed stronsium(II) was first eluted with 25 mL of 0.1 M acetic acid. After that uranium(VI) were eluted with 3.0 M hydrochloric acid then lead(II) was eluted with 5.0 M sulphuric acid and finally the thorium (VI) was eluted with 3.0.M hydrochloric acid and effluents are analyzed spectrophotometrically. Using this method, separation of lithium (I), uranium(VI), thorium(IV), potassium(I)), lead(II), strontium(II), sodium(I) mixtures was achieved. The results are shown in table.

Table: Separation of uranium (VI) from Multicomponent Mixtures.

No	Mixture	Taken µg	Found µg	Recovery %	Sorption Condition	Eluent
1	Li(I)	100	100	100	$1\text{x}10^{-3}$M Alanine	NSPC
	U(VI)	50	50	100		3.0MCH_3COOH
	Th(VI)	30	30	100		3.0 M HCl
2	K(I)	100	99	99	$1\text{x}10^{-3}$ M Alanine	NSPC
	U(VI)	50	50	100		3.0MCH_3COOH
	Th(VI)	30	30	100		3.0 M HCl
3	Na(I)	100	100	100	$1\text{x}10^{-3}$M Alanine	NSPC
	U(VI)	50	50	100		3.0MCH_3COOH
	Th(VI)	30	30	100		3.0 M HCl
4	Li(I)	100	100	100	$1\text{x}10^{-3}$M Alanine	NSPC
	Sr(II)	50	49	98		3.0MCH_3COOH
	U(VI)	50	50	100		3.0 M HCl
5	Li(I)	100	100	100	$1\text{x}10^{-3}$ M Alanine	NSPC
	Sr(II)	50	49	98		0.1MCH_3COOH
	U(VI)	50	50	100		3.0MCH_3COOH
	Th(VI)	30	30	100		3.0M HCl
6	K(I)	100	99	99	$1\text{x}10^{-3}$ M Alanine	NSPC
	Sr(II)	50	49	98		0.1MCH_3COOH
	U(VI)	50	49.5	99		3.0MCH_3COOH
	Th(VI)	30	29	97		3.0M HCl
7	Li(I)	100	100	100	$1\text{x}10^{-3}$ M Alanine	NSPC
	U (VI)	50	49	98		3.0MCH_3COOH
	Th(VI)	30	30	100		0.1MCH_3COOH
	Pb(II)	50	49	98		5.0 M H_2SO_4
8	Li(I)	100	99	99	$1\text{x}10^{-3}$ M Alanine	NSPC
	U (VI)	50	50	100		3.0MCH_3COOH
	Th(VI)	30	30	100		0.1MCH_3COOH
	Pb(II)	50	49	98		5.0 M H_2SO_4

Determination of uranium (VI) in geological sample

Monazite sand sample was brought into solution as per procedure described elsewhere.[17,18] An aliquot of sample solution was subjected for the separation of uranium(VI) as per the above procedure and then it was determined spectrophotometrically with arsenazo(III) at 660 nm.[15,16] The amount of uranium(VI) by triplicate analysis found monazite sand was 0.295% as against the standard value of 0.30%.

CONCLUSION

The important feature of this method is that using column chromatographic method and poly [dibenzo-18-crown-6] the separation of uranium(VI) from associated element in alanine medium has been achieved. The capacity of poly [dibenzo-18-crown-6] for uranium (VI) was found to be 1.092 ± 0.01 mmol/g of crown polymer. Uranium (VI) was separated from number of cations in binary as well as multicomponent mixtures. The method was extended to the determination of uranium (VI) in real sample. The aim of present investigation is to demonstrate a simple, selective and cheap method and has good reproducibility (approximately ± 2%).

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