SEPARATION OF RHENIUM(VII) FROM TUNGSTEN(VI)

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Abstract

Examined were the conditions for an effective separation of tungsten(VI) and rhenium(VII) on alumina if the solution of 0.20 mol dm^{-3} NaCl, pH=2-6, is used as the aqueous phase. Under the given experimental conditions alumina was found to be much better adsorbent for tungsten than for rhenium. The breakthrough and saturation capacities of alumina at pH = 2 are 24 and 78 mg W/g Al_2O_3, respectively. With the increase of pH these values decrease. So, at pH = 6 they are only 4 and 13 mg W/g Al_2O_3, respectively. The elution volume for rhenium for the given column dimensions and the quantity of the adsorbent, is about 16 ml. These results were confirmed by the experiments of the radiochemical separations. Tungsten-187 remains firmly bound to alumina. The radionuclidic purity of the eluted ^{186,188}Re at pH = 2 is very high.

Introduction

Therapy in nuclear medicine gained a renewed interest and investigations are devoted to the introduction of new radionuclides. Particular emphasis is given to ^{188}Re which is the decay product of its parent ^{188}W($T_{1/2}$=69 d) formed in nuclear reactor during the irradiation of tungsten targets. However, the isolation of rhenium from tungsten is not easy. Many efforts are devoted to the development of an efficient routine separation procedure based on alumina as the adsorbent [1,2].

Experimental

Sodium tungstate ($Na_2WO_4 \cdot 2H_2O$, p.a., Fluka) and potassium perrhenate ($KReO_4$, p.a., Aldrich) were commercially purchased. Alumina for column chromatography (Alumina N- Super I, ICN Biomedicals), grain size 40-140 µm, is used without any pretreatment.

The concentrations of tungsten and rhenium in the solutions were determined by the direct current argon arc plasma atomic emission spectroscopy (DCP-AES) with aerosol supply [3].

The column experiments were performed for the system: alumina-0.20 mol dm^{-3} NaCl. The breakthrough and saturation capacities of alumina for W(VI) were determined in the glass column (8 mm diameter 40 mm length) containing 1 g of Al_2O_3. The concentration of tungsten was 5.4x10^{-3} mol dm^{-3} in 0.20 mol dm^{-3} NaCl, pH = 2-6 (±0.2). The flow rate was 2.5 ml min^{-1}cm^{-2}. According to the previous results [2] the capacity of alumina for rhenium is low. Therefore its breakthrough and saturation capacities were not determined.

The elution volumes of rhenium were determined in the glass column (10 mm diameter; 100 mm length) containing 3.5 g Al_2O_3. After sorption of 1 ml of rhenium solution (9×10^{-3} mol dm^{-3} in 0.2 mol dm^{-3} NaCl), rhenium was eluted by 0.20 mol dm^{-3} NaCl, pH = 2-6, (±0.2). The flow rate of the eluence solution was 1.5 ml min^{-1}cm^{-2}. The elution volumes for tungsten were not determined.

For the irradiations the samples containing 50 mg of $KReO_4$ and H_2WO_4, respectively, were used. The irradiations were performed in the thermal core No. 114 of zero power heavy water reactor RB (Vinča Institute of Nuclear Sciences) at fission power equal 40 W (neutron flux 3.6×10^{8} n cm^{-2} s^{-1}) for 60 minutes [4]. The gamma activities of the irradiated samples were measured by using Canberra XtRa (Extended Range) coaxial Ge-detector in the Canberra 777 ultra low-background shield and the ISOCS (In Situ Object Counting System) Calibration Software [5]. Neutron activation of tungsten ^{186}W gives ^{187}W ($T_{1/2}$ = 23.8 h) while the products of the reaction ^{187}Re(n,γ) are ^{186}Re ($T_{1/2}$ = 90.6 h) and ^{188}Re ($T_{1/2}$ = 16.9 h).

After irradiation, Re samples were dissolved in water and diluted to 5 ml by 0.20 mol dm^{-3} NaCl (pH = 2). Tungsten samples were dissolved in 1 mol dm^{-3} NaOH and the excess alkali was neutralized by HF. Finally they were diluted to 10 ml by 0.20 mol dm^{-3} NaCl (pH = 2). Both solutions were mixed and passed through alumina column (10 mm diameter, 100 mm lenght) containing 6 g Al_2O_3. The elutions were performed by 0.20 mol dm^{-3} NaCl (pH = 2), flow rate 1 ml min^{-1}.

Results and Discussion

The breakthrough and saturation capacities of alumina for tungstate anion and the elution volumes of inactive rhenium and ^{186,188}Re are shown in Table I. Data in Table I present the adsorption characteristics of alumina when the aqueous phase is 0.20 mol dm^{-3} NaCl. It can be seen that they depend on pH. Both the breakthrough and saturation capacities decrease with the increase of pH.

The results for the elution volumes of inactive rhenium and ^{186,188}Re refer to the applied experimental conditions. It can be concluded that they do not depend on pH. For inactive rhenium the value is about 15 ml 0.20 mol dm^{-3} NaCl throughout the examined pH range. These results were tested also with the solutions containing ^{187}W, ^{186}Re and ^{188}Re. The radiochemical separation of rhenium and tungsten radioisotopes was achieved on alumina column containing 6 g Al_2O_3. Under the given experimental conditions W is efficiently adsorbed and its elution volumes are high. In 100 ml of the eluence passed through the column only the peaks beloging to rhenium radioisotopes were observed. For the elution of rhenium activity (^{186}Re+^{188}Re) 23 ml of the eluence was needed. Since no activity of ^{187}W was detected, the radionuclidic purity of the eluted rhenium is very high.

Table I. Breakthrough and saturation capacities of alumina for tungsten and elution volumes of inactive rhenium and $^{186,188}Re$

Column: 8 mm diameter, 40 mm lenght; bed: 1 g Al_2O_3 N Super I (ICN Biomedicals)

Rhenium elution volume-Column: 10 mm diameter, 100 mm length; bed: 3.5 g Al_2O_3 N Super I (ICN Biomedicals)

$^{186,188}Re$ elution volume-Column: 10 mm diameter, 100 mm length; bed: 6 g Al_2O_3 N Super I (ICN Biomedicals)

PH of 0.20 mol dm^{-3} NaCl	Breakthrough capacity (mg W/g Al_2O_3)	Saturation capacity (mg W/g Al_2O_3)	Elution volume of ReO_4^- (ml)	Elution volume of $^{186,188}ReO_4^-$ (ml)
6	4	13	15	-
4	17	26	15	-
2	24	78	16	23

Conclusion

Sorption studies for WO_4^{2-} and ReO_4^- were carried out by using alumina and aqueous solution of 0.20 mol dm^{-3} NaCl. Our experiments confirm that alumina is, under examined experimental conditions, much better adsorbent for tungstate than for perrhenate anions. The breakthrough and saturation capacities of alumina for tungsten are relatively high. For the given column dimensions and the quantity of alumina used in the experiments, it was found that rhenium can be separated with a relatively low volume of NaCl solution. These results were tested by using the solutions containing ^{187}W, ^{186}Re and ^{188}Re. The radiochemical separations confirm that tungsten is strongly adsorbed on alumina. The radionuclidic purity of separated $^{186,188}Re$ is very high.

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