

تقنية التحليل الإشعاعي الكيميائي للمواد باستخدام التبادل الأيوني

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ملخص البحث :

تم تصميم هذه التجربة لإثبات تقنية الفصل الإشعاعي الكيميائي للمواد. تم تحديد عوامل الفصل أحادية المرحلة لنظائر البورون بين (resin) كوسط التبادل الأيوني والمحلل الخارجي ، باستخدام عملية اختراق التبادل الأيوني. يتم تمرير خليط من محاليل اليورانيوم وكبريتات النحاس عبر عمود التبادل الأيوني. تم استخدام 10 % وزن/حجم من محلول كبريتات اليورانيوم ، و 10 % وزن/حجم من هيدروكسيد الأمونيوم ، بالإضافة إلى 10 % حجم/حجم من حمض الهيدروكلوريك و 1 % وزن/حجم من محلول نترات الفضة كمواد جزئية. تم فصل هذه العناصر عن عدد كبير من العناصر المتنوعة إما عن طريق الامتصاص الانتقائي أو الشطف الانتقائي بالماء أو المزلقات المحددة. تم قياس العينات باستخدام التحليل الطيفي بأشعة جاما. تشير النتائج إلى أن 16.1 % من أنيون مركب اليورانيوم بقي في الوسط.

The Technique of Radio-Chemical Separation of Materials Using Ion Exchange Resin

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Abstract

This experiment is designed to demonstrate the technique of radio-chemical separation of materials. The single-stage separation factors for boron isotopes between an ion- exchange resin and external solution were determined, using an ion- exchange breakthrough process. A mixture of uranium and copper sulphate

solutions are passed through an ion exchange column. 10 % w/v of uranyl sulphate solution, 10 % w/v of ammonium hydroxide, in addition to 10 % v/v of hydrochloric acid and 1% w/v of silver nitrate solution were used as an elutes. The separation of these elements from a large number of diverse elements was effected either by selective sorption or selective elution with water or specific elutes. The samples were measured using gamma-ray spectroscopy. The results indicate that 16.1% of uranium complex anion was stayed in the resin.

Key words - radio-chemical separation, boron, isotopes, ion-exchange, uranium, gamma-ray, spectroscopy.

1. Introduction

In multifarious hydrometallurgy processes, aqueous solutions contain big amounts of deliquesced metal ion such as cobalt, gold, uranium and copper.

This experiment relates to methods and apparatus for the treatment of metal-containing solution. More specifically, the present experiment relates to a continuous ion exchange method which is especially useful for the selected condensation and remotion of minerals from aqueous solution, either for resource inference or contamination control [1].

Improved chemical precipitation and ion exchange methods are being developed in national laboratory [2]. Ion exchange is a standard method used to separate materials by chemical means; its column contains resins, which are widely used in different separation, purification and decontamination operations. These resins contain labile ions, which are capable of undergoing

exchange with ion in the surrounding medium that is passed through the resin in solution.

In some cases ion-exchange resins were introduced in such operations as a more pliant alternative to use of natural or industrial zeolites [3].

Ion exchange is a reversible chemical reaction where dissolved ions are removed from solution and replaced with other ions of the same or similar electrical charge. Not a chemical reactant in and of itself, ion exchange resin is instead a physical medium that facilitates ion exchange reactions. The resin itself is composed of organic polymers that form a network of hydrocarbons. Throughout the polymer matrix are ion exchange sites, of either cations (positively-charged) ions or anions (negatively-charged) ions are affixed to the polymer network. These functional groups readily attract ions of an opposing charge [4,5].

An ion-exchange resin or ion-exchange polymer is a resin or polymer that acts as a medium for ion exchange. It is an insoluble matrix (or support structure) normally in the form of small (0.25–1.43 mm radius) microbeads, usually white or yellowish, fabricated from an organic polymer substrate. The beads are typically porous, providing a large surface area on and inside them the trapping of ions occurs along with the accompanying release of other ions, and thus the process is called ion exchange. There are multiple types of ion-exchange resin. Most commercial resins are made of polystyrene sulfonate. Ion exchange resins are widely used in different separation, purification, and decontamination processes. The most common examples are water softening and water purification. In many cases ion-exchange resins were introduced in such processes as a

more flexible alternative to the use of natural or artificial zeolites. Also, ion-exchange resins are highly effective in the biodiesel filtration process.

2. Theory

Uranium can be extracted from two main leach types, called acid and carbonate leaches. The uranium can be presented as $[\text{UO}_2(\text{CO}_3)_3]^{-4}$ in carbonate leaches, whereas in sulphuric acid leach solutions it can be presented as UO_2^{+2} cations or $[\text{UO}_2(\text{SO}_4)_2]^{-2}$ and $[\text{UO}_2(\text{CO}_4)_3]^{-4}$.

2.1 Ion exchange IX

IX is an exchange of ions between two electrolytes. In general, ion exchange is the term that used to import the processes of decontamination, purification and separation of ion in solution with solid aqueous [4].

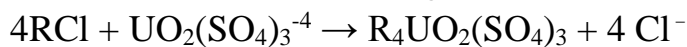
IX is a reversible interchange of charged particles-or ions-with those of like charge. The ions present on an insoluble IX exchange resin matrix effectively swap places with ions of a similar charge that are present in a surrounding solution.

The ion exchange resin functions this way because of its functional groups, which are basically fixed ions that are permanently bound within the polymer matrix of the resin. Then charged ions will simply bond with ions of an opposing charge, which are delivered through the application of a counterion solution. These reverses will bond with the functional groups until it reach equilibrium [4]. During an ion exchange cycle, the solution to be treated would be added to the ion exchange resin bed and allowed to flow through the beads. The solution moves through the ion exchange resin, then functional groups of the resin attract any reverses present in the

solution. If the functional groups have a greater affinity for the new counterions than those already present, then the ions in solution will dislodge the existing ions and take their place, bonding with the functional groups through shared electrostatic attraction. In general, the greater the size and/or valency of an ion, the greater affinity it will have with ions of an opposite charge [4,5].

Typical ion exchangers are ion exchange resins, in this experiment the resin used initially was amberlite (IRA-420(Cl⁻)), which is an anionic resin containing chloride ions Figure 1 provides ion exchange resin beads [4,5].

To perform the ion exchange separation process an amberlite resin was used (see Figure 2). Ion exchangers are either anion exchangers or cation exchangers. Following in order the equation describes the ion exchange reaction and the typical examples of ions that bind to ion exchanges:



The eluting in this experiment was ammonium nitrate.

1. Single charged monatomic ions, such as K⁺, Na⁺ and Cl⁻.
2. Double charged monatomic ions like Mg⁺², Ca⁺².
3. H⁺ (proton) and OH⁻ (hydroxide).
4. Polyatomic inorganic ions such as SO₄²⁻ [5,6].



Fig. 1 Ion exchange resin beads [7].

3. Experimental procedure

The first step as an experiment method was the calibration of system by using ^{152}Eu source. Uranyl sulphate (the yellow solution) was taken to the low background counting; it located in a transport container to record its background spectrum. A spectrum recorded for 300 seconds as a count time.

The apparatus was set up, and then 20 ml of uranyl sulphate were mixed with 80 ml of copper sulphate to result a green solution; in which uranium will be presented as a complex anion UO_2SO_4 . As well as the column was washed through with 60 ml of distilled water. This water was collected in three plastic bottles in 20 ml labeled as A1, A2 and A, this solution passed through the column at 5- 10 ml per minute, and then collected in 20 ml aliquot which labeled as B1 to B5.

120 ml of distilled water passed through the column, and each 20 ml aliquot collected C1 to C6. The aliquot C6 was tested by adding some dilute ammonia to see any copper was left in the resin. It still

blue colour when mixed with ammonia, consequently another 60 ml of water was added until the copper is completely flushed out. 200 ml of ammonium nitrate solution was passed through the column and the recollected in 20 ml aliquots as D1 to D9. finally, all the aliquots were tested by gamma ray spectrometer in a low background.

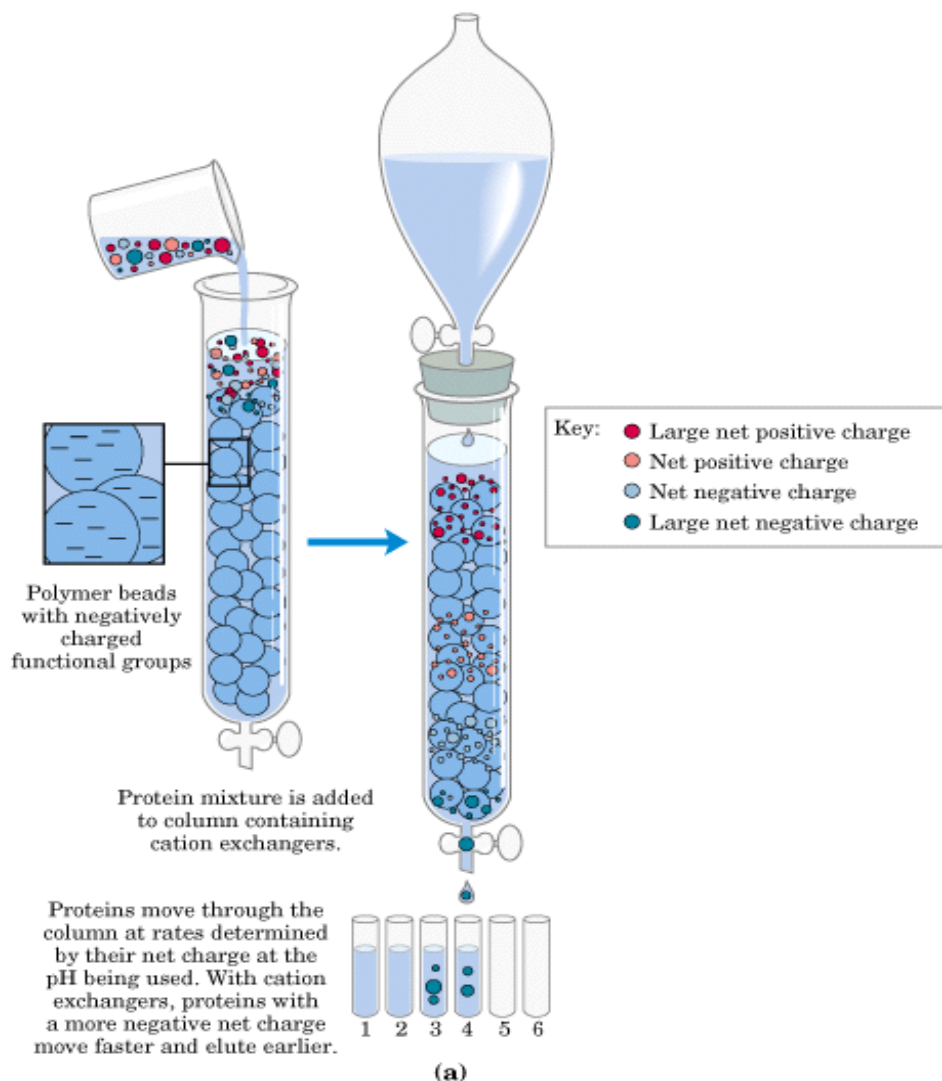


Fig. 2. Ion exchange separation process [8].

4. Result & Discussion

In the system calibration, the ^{152}Eu source was used for the energy of each peak. Table 1 contains on the main four peaks were chosen, when the aliquots compared with the sample of original uranyl sulphate solution.

Table 1. Gamma- rays Energy Peaks in the Original Uranyl Sulphate Solution

γ -rays peak (keV)	Counts (in 300s)
^{234}Th (62.97 keV)	$1353 \pm 4.01\%$
^{234}Th (92.38 keV)	$5885 \pm 1.98\%$
^{235}U (134.7 keV)	$260 \pm 10.91\%$
^{235}U (185.57 keV)	$1175 \pm 3.85\%$

Table 2. Counts on each aliquot and their errors

Aliquot No:	^{234}Th (62.97keV)	^{234}Th (92.38keV)	^{235}U (143.7keV)	^{235}U (185.57keV)
A1	56 ± 14.39	213 ± 7.38	35 ± 19.05	197 ± 7.39
A2	32 ± 19.05	57 ± 15.28	35 ± 17.85	114 ± 9.75
A3	2 ± 93.99	16 ± 27.44	6 ± 46.65	30 ± 18.54
B1	14 ± 29.28	44 ± 15.85	9 ± 39.82	27 ± 19.57
B2	17 ± 28.52	181 ± 7.92	5 ± 52.69	17 ± 24.91
B3	27 ± 22.62	31 ± 34.85	3 ± 93.13	8 ± 47.21
B4	74 ± 12.85	4 ± 268.48	3 ± 88.60	2 ± 162.26
B5	39 ± 29.52	0	4 ± 75	5 ± 70.29
C1	32 ± 31.35	0	0	6 ± 48.54
C2	30 ± 25.49	0	0	0
C3	56 ± 15.54	0	4 ± 60.81	2 ± 99.41
C4	76 ± 12.88	303 ± 6.04	0	3 ± 135.26
C5	63 ± 14.30	209 ± 7.58	2 ± 204	7 ± 45.63
C6	0	15 ± 36.38	2 ± 108.37	1 ± 177.9
C7	48 ± 15.63	139 ± 9.41	3 ± 125.80	12 ± 31.57
C8	17 ± 44.83	192 ± 8.05	1 ± 25	14 ± 26.73

C9	18 ± 28.42	80 ± 11.73	4 ± 94.29	9 ± 37.52
D1	134 ± 10.24	540 ± 4.66	16 ± 33.06	45 ± 16.34
D2	180 ± 8.62	634 ± 4.34	39 ± 20.23	133 ± 9.14
D3	136 ± 9.46	448 ± 5.91	38 ± 20.68	1 ± 100.00
D4	90 ± 12.07	378 ± 5.91	44 ± 16.40	167 ± 8.15
D5	147 ± 9.77	576 ± 4.68	24 ± 33.09	170 ± 8.00
D6	88 ± 12.33	275 ± 6.82	42 ± 19.23	152 ± 8.50
D7	59 ± 14.56	148 ± 9.92	18 ± 36.95	132 ± 8.96
D8	31 ± 22.56	128 ± 9.91	11 ± 51.36	78 ± 12.19
D9	6 ± 78.09	37 ± 18.15	5 ± 73.95	21 ± 23.50

The above table shows that, the four main peaks of gamma-rays for all aliquots. However, you have to note that, the aliquot (C6) was mixed with an ammonia, because we have gotten a little bit of solution in C6 aliquot. Therefore, the dilute ammonia was mixed in the same aliquot of C6; the table also provides the variation of each aliquot to another.

In addition to an acceptable qualitative visual impression of the relative abundances of ^{235}U and ^{234}Th that was provided in (Figure 3). The graph also shows the high concentration of ^{234}Th .

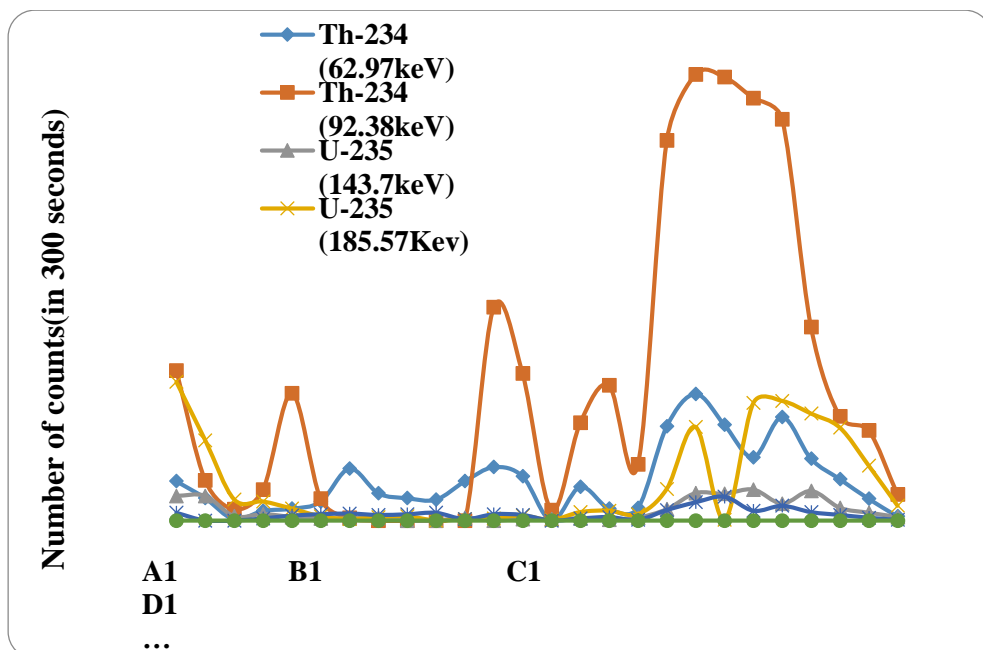


Fig. 3. Aliquot Gamma ray Activity during the Ion Exchange Separation of Uranium

4.1 The dilution factor of uranium

When 20 ml of uranyl sulphate (10% w/v) was added to 80 ml of copper sulphate (10% w/v), and the addition of the copper sulphate would have diluted the uranyl sulphate to 2% in the solution. This indicates that the outset concentration of uranyl sulphate was 2% w/v.

On the other hand, 200 ml of an ammonium nitrate were passed through the column, the average concentration of uranium in the elute would be 1% if all obtainable uranium complex anion was recorded in the 200 ml of the ammonium nitrate. This means that, the uranium would not be concentrated in the resin, it would have been diluted.

In this part of the result, the percentage uranium concentration was calculated, by comparing the amounts of the uranium, we can use

the number of counts for gamma- rays' peaks in the original uranyl sulphate solution and any aliquot, which is A1 in the calculation.

The original U1 = 127 ± 10.91 (counts in 300s).

In aliquot A1, U1 = 35 ± 17.85 % (counts in 300s),

The percentage uranium concentration = $\frac{35(\pm 17.85)}{127(\pm 10.91)} \times 100 =$

$27.55 \pm 1.63\%$

As well as, for U2:

The percentage uranium concentration = $\frac{179(\pm 7.39)}{726(\pm 3.85)} \times 100 =$

$27.134 \pm 1.91\%$

Consequently, the concentration of U1 is, almost 13.7 times higher than it in the original uranyl sulphate (2%), which passed through the column.

Whereas, 13.5 times higher for U2 peak in the uranium concentration compared to the original uranyl sulphate solution.

Now, the percentage recovery can be calculated by compare the total number of counts from all D aliquots.

Percentage recovery for

$$U1 = \frac{237 \pm 274.35}{127 \pm 10.91} \times 100 = 186.61\% \pm 25.14\%$$

Percentage recovery for

$$U_2 = \frac{899 \pm 164.18}{726 \pm 3.85} \times 100 = 123.82\% \pm 42.64\%$$

The percentage recovery of

$$\text{uranium} = \frac{186.61 + 123.82}{2} = 155.21\% \pm 33.89\%$$

5. Conclusion

In conclusion, this experiment substantiated that; the method for continuous ion exchange for metal recovery is disclosed in which a feed solution and liquid ion exchange are separated by porous barrier. The lighter isotope ^{235}U was clearly found to be enriched in the uranyl malate species in the solution phase at all temperatures after separating most floc, the water is filtered as the final step to remove remaining suspended particles and unsettled floc. Most of the uranium was retained in the resin as a complex anion of $\text{UO}_2(\text{SO}_4)_3$.

In the second part of the experiment the uranium was flushed out when an ammonium nitrate was passed through the resin the nitrate anion displaces the uranium complex anion.

On the other hand, the separation of uranium ions from copper was done successfully, the main uranium daughter product that copper solution was ^{234}Th . The separation of uranium from its daughters was less clear than it for the copper. Figure 3 provides that, whereas the uranium was standed firm the resin, the uranium-238 daughter and ^{234}Th were allowed to pass through the column to a much stronger extent. Finally, the experiment was depended on

making comparisons between solutions, and it also was undertaken to search the separation of daughters.

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