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Abstract

In this study, the effectiveness of micro coprecipitation technique with LaF_3 was tested using tap water samples and waste water from the mine prepared for the determination of ^{234}U , ^{235}U and ^{238}U , and their recoveries in alpha spectrometry. The actinides were separated by UTEVA resin in columns. The actinides of interest were then deposited on a suitable substrate prior to measurement by alpha spectrometry. In order to minimize energy degradation and self-absorption effect, thin and uniform sources had to be prepared. Therefore micro co-precipitation technique was used on each of the samples for the source preparation. The activity concentrations for the waste water from the mine ranged from (911.44 – 1093.47) mBq/L for ^{238}U , (60.98 – 282.16) mBq/L for ^{235}U and (910.73 – 1154.44) mBq/L for ^{234}U . For the tap water, it ranged from (1.28 – 1.60) mBq/L for ^{238}U , (0.36 – 0.75) mBq/L for ^{235}U and (3.67 – 4.11) mBq/L for ^{234}U . Good recoveries were obtained using the micro co-precipitation technique in the range of (68.9% – 83.6%) for the waste water from the mine, while recoveries for the tap water samples were in the range of (54.5% – 64.6%). This demonstrates the capability of micro co-precipitation with LaF_3 as a good substitute to electrodeposition in alpha spectrometry.



Figure 1: Tap water and Mine water samples ready for counting

Introduction

The major stages in alpha spectrometry are pre-concentration, radiochemical separation, source preparation, and counting which are time consuming. It is therefore very important to develop effective techniques to reduce the time at each stage. Some current techniques are effective but can take up to several days. There is a need for a fast, simple method, in which the radionuclides of interest and the tracers are made into a thin, flat and uniform source which allows adequate transmission of the alpha-particles to the detector surface, hence the development of micro co-precipitation technique with LaF_3 to produce a homogeneously thin source for counting. In this study, the method was used for water samples only but can be extended on solid matrix such as sediments, plant, soil, food, etc. to obtain very good recovery, in some measurements as high as 90%.

Methodology

Three sample each of volume 4 L of tap water and three samples each of volume 100 mL of waste water from the mine were measured into different beakers. Each of the samples was acidified with 12 mL of conc. HNO_3 after which 0.5 g of ^{232}U tracer was added into each sample. 8 mL of Fe^{3+} carrier (5 mg of Fe/mL) was added to every sample to initiate the coprecipitation of Fe^{3+} with the radionuclides of interest. The pH of the samples was changed to 9.6 by gradually adding conc. NH_3 . Each solution was stirred for 3 hours and allowed to settle down by leaving them overnight. The supernatant was then decanted, centrifuged at 3500 rpm for 5 minutes and rinsed until neutral pH was obtained. The $\text{Fe}(\text{OH})_3$ precipitate of each was then dissolved in 5 mL of 3 M HNO_3 . Preconditioning of UTEVA column was done by rinsing the column with 10 mL H_2O , 10 mL 1 M HNO_3 and 10 mL 3 M HNO_3 .

Methodology

Each sample was loaded on the column. The centrifuge tubes were rinsed 2 twice with 5 mL 3 M HNO_3 and load on every column after which the column was rinsed with 20 mL 3 M HNO_3 , 5 mL 9 M HCl , 25 mL 5 M HCl . Uranium was finally eluted with 15 mL of 1 M HCl into a clean tube. 0.1 mL of lanthanum tracer La^{3+} (1 mg/mL), 1 mL TiCl_3 and 1 mL conc. HF were added in the uranium fraction. Each of the samples was left for a minimum of 30 minutes in an ice bath. A filter paper of size 0.22 μm was placed on a funnel for vacuum filtration after which it was rinsed with deionised water and 10 mL of LaF_3 suspension of 0.2 mg/mL. The sample was passed through the filter paper and filtrated. The tube was rinsed twice with 5 mL 0.58 M HF , deionised water and filtrated. Each filter paper was then removed from the funnel, dried under an IR lamp and stuck onto aluminium disk for counting. The measurements were done using 450 mm^2 active surface Passivated Implanted Planar Silicon (PIPS) semiconductor detectors installed in the 12-chamber Alpha Analyst System (Canberra). The measurements were carried out at a source to detector distance of about 5 cm. The accumulation and analysis of Alpha spectra was done using Genie 2000 software with measurement time of about 144,000s. The calibration of the detectors was made with a standard radionuclide source containing ^{238}U , ^{234}U , and ^{239}P , ^{241}Am .

Results and Discussion

Alpha detector efficiencies of the detectors in chambers containing MW1, MW2 and MW3 are 0.1268, 0.1302 and 0.1239 respectively while alpha detector efficiencies of the detectors in chambers containing TW1, TW2 and TW3 are 0.128, 0.1331 and 0.1322. After counting for a relatively long period of time to obtain enough counts for both the background and the samples, the activity concentrations of all the samples were obtained. The activity concentrations of the two types of water samples are shown in the graph below:

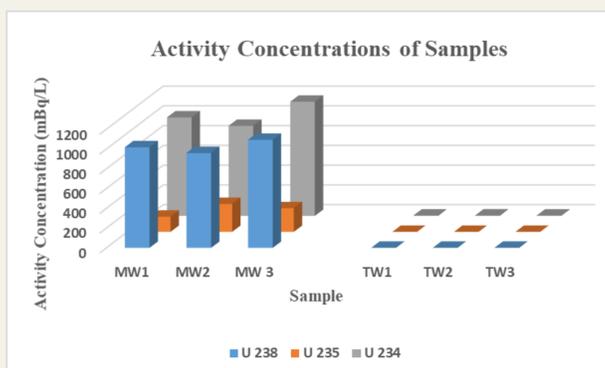


Figure 2: Graphical representation of Activity concentrations of samples

The graph above shows the activity concentrations of the mine water samples being greater than the activity concentrations of the tap water samples as predicted. The average activity concentrations of ^{238}U , ^{235}U and ^{234}U in the Mine Water are (1060.0 \pm 46.7) mBq/L, (225.3 \pm 18.6) mBq/L and (1042.0 \pm 51.2) mBq/L respectively while the average activity concentrations of ^{238}U , ^{235}U and ^{234}U in the Tap Water are (1.5 \pm 0.2) mBq/L, (0.5 \pm 0.1) mBq/L and (3.8 \pm 0.3) mBq/L respectively. In the first set of samples (MW), ^{238}U and ^{234}U have similar activity concentrations with ^{235}U having the lowest as expected. In the second set of samples (TW) the activity concentration of ^{238}U is almost half of that of ^{234}U with ^{235}U having the lowest concentration.

Results and Discussion

Table 1: Activity Concentrations of ^{238}U , ^{235}U and ^{234}U in Mine Water

| | MW1 | MW2 | MW3 |
|----------------------------|------|-----|------|
| U-238 (mBq/L) | 1018 | 960 | 1093 |
| Uncertainty | 34 | 50 | 56 |
| expanded uncertainty (k=2) | 68 | 101 | 112 |
| U-235 (mBq/L) | 154 | 282 | 240 |
| Uncertainty | 14 | 22 | 20 |
| expanded uncertainty (k=2) | 28 | 43 | 40 |
| U-234 (mBq/L) | 996 | 911 | 1154 |
| uncertainty | 47 | 48 | 58 |
| expanded uncertainty (k=2) | 94 | 97 | 117 |

Table 2: Activity Concentrations of ^{238}U , ^{235}U and ^{234}U in Tap Water

| | TW1 | TW2 | TW3 |
|----------------------------|-----|-----|-----|
| U-238 (mBq/L) | 1.3 | 1.6 | 1.5 |
| Uncertainty | 0.2 | 0.2 | 0.2 |
| expanded uncertainty (k=2) | 0.3 | 0.4 | 0.4 |
| U-235 (mBq/L) | 0.4 | 0.4 | 0.8 |
| Uncertainty | 0.1 | 0.1 | 0.1 |
| expanded uncertainty (k=2) | 0.2 | 0.2 | 0.3 |
| U-234 (mBq/L) | 3.7 | 4.1 | 3.7 |
| uncertainty | 0.3 | 0.4 | 0.3 |
| expanded uncertainty (k=2) | 0.6 | 0.7 | 0.7 |

The figure below shows the percentage radiochemical recovery for the method described. Good radiochemical recoveries were obtained for this particular method (recovery > 40%). The average radiochemical recovery obtained using this method for mine water (74.4%) is higher than the average radiochemical recovery obtained using this method for Tap water (59.5). This perhaps gives an indication of the method being more effective on water samples with high uranium concentration.

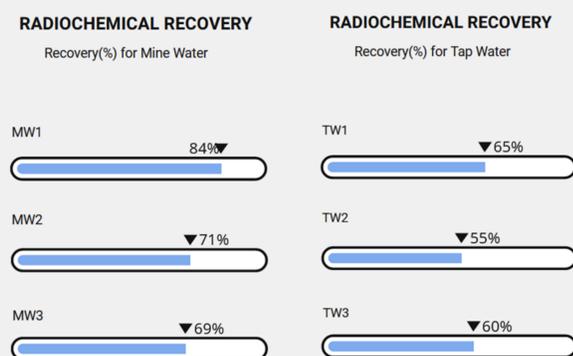


Figure 3: Radiochemical recoveries of all samples

Conclusion

This work presents the effectiveness of the micro coprecipitation technique with LaF_3 following column separation using UTEVA resin for two types of water samples; tap water and waste water from the mines. The results obtained show that the method allows for a fast and efficient determination of uranium activity concentrations in all of the analyzed samples. The radiochemical recoveries determined by the method are greater than the threshold recovery (40%) for acceptance.

References

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