30th Conference of the International Nuclear Target Development Society INTDS

Application of Multiple Source Preparation Procedures on Environmental Samples for Determination of Uranium In Alpha Spectrometry

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Section
01

Introduction

Introduction

Alpha-particle spectrometry in nuclear decay data measurements, geological studies, or the measurement of low levels of activity in the environment.

Water, spinach and soil used. Complete decomposition of the spinach and soil sample matrix is necessary

Aim is to investigate two different source preparation approaches for determination of uranium radioisotopes in water, spinach and soil samples

Introduction

Wet digestion used for solid samples. The preconcentration method for uranium was used by co-precipitation with $Fe(OH)_3$ at pH 9 using ammonia solution in water samples.

Electrodeposition was used for the soil and spinach samples while micro-coprecipitation with LaF_3 method was used for the tap water and waste water from the mine

These methods allow the radionuclides of interest and the tracers to be made into a thin, flat and uniform source which allows adequate transmission of the alpha-particles to the surface of the detector

Section
02

Methodology

Methodology

Spinach

STEP 1

A triplicate of 2 g of ashed spinach sample was weighed into a glass beaker and 10 mL of concentrated HNO_3 was added to make a slurry.

STEP 2

0.5 g of ^{232}U tracer was added and heated for three hours at 150°C. 1 mL H_2O_2 was added into the sample and evaporated to dryness

STEP 3

6 mL conc. HNO_3 and 1 mL H_2O_2 were added into the sample and evaporate to dryness at 250 °C. The sample was dissolved in 3 M HNO_3 and loaded on the column containing UTEVA resin..

STEP 4

The container is rinsed with 5 mL 3 M HNO_3 and loaded on a column. The column was washed with 20 mL 3 M HNO_3, 5 mL 9 M HCl, and 25 mL 5 M HCl after which uranium was eluted with 15 mL 1 M HCl into a clean tube

STEP 5

The eluted solution was evaporated to dryness. 1 mL of conc. HNO_3 and 1 mL of H_2O_2 were added and evaporated to dryness. The residue was dissolved in 1 mL of 6 M HCl and then transferred into the electrodeposition cell with 10 mL of electrolytic solution made up of 5.7% Ammonium oxalate in 0.3M HCl.

STEP 6

The electrodeposition was started with an electrical current (A) of 0.5 Ampere for 2 hours. 1 mL of ammonia was added a minute before putting off the power supply. The disc with electrodeposited radionuclides was dried and counted.

Methodology

Soil

STEP 1

A triplicate of 2 g of oven-dried soil sample was weighed into a glass beaker and 10 mL of concentrated HNO_3 was added to make a slurry.

STEP 2

0.5 g of ^{232}U tracer was added and heated for three hours at 150°C. 10 ml each of HNO_3, HF, $HClO_4$ were added to each of the samples and evaporated to dryness on a hot plate.

STEP 3

2 mL of conc. HNO_3 was added into the sample and evaporate to dryness. The sample was dissolved in 3 M HNO_3 and loaded on the column containing UTEVA resin.

STEP 4

The container is rinsed with 5 mL 3 M HNO_3 and loaded on a column. The column is washed with 20 mL 3 M HNO_3, 5 mL 9 M HCl, and 25 mL 5 M HCl after which uranium was eluted with 15 mL 1 M HCl into a clean tube.

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The eluted solution was evaporated to dryness. 1 mL of conc. HNO_3 and 1 mL of H_2O_2 were added and evaporated to dryness. The residue was dissolved in 1 mL of 6 M HCl and then transferred into the electrodeposition cell with 10 mL of electrolytic solution made up of 5.7% Ammonium oxalate in 0.3M HCl.

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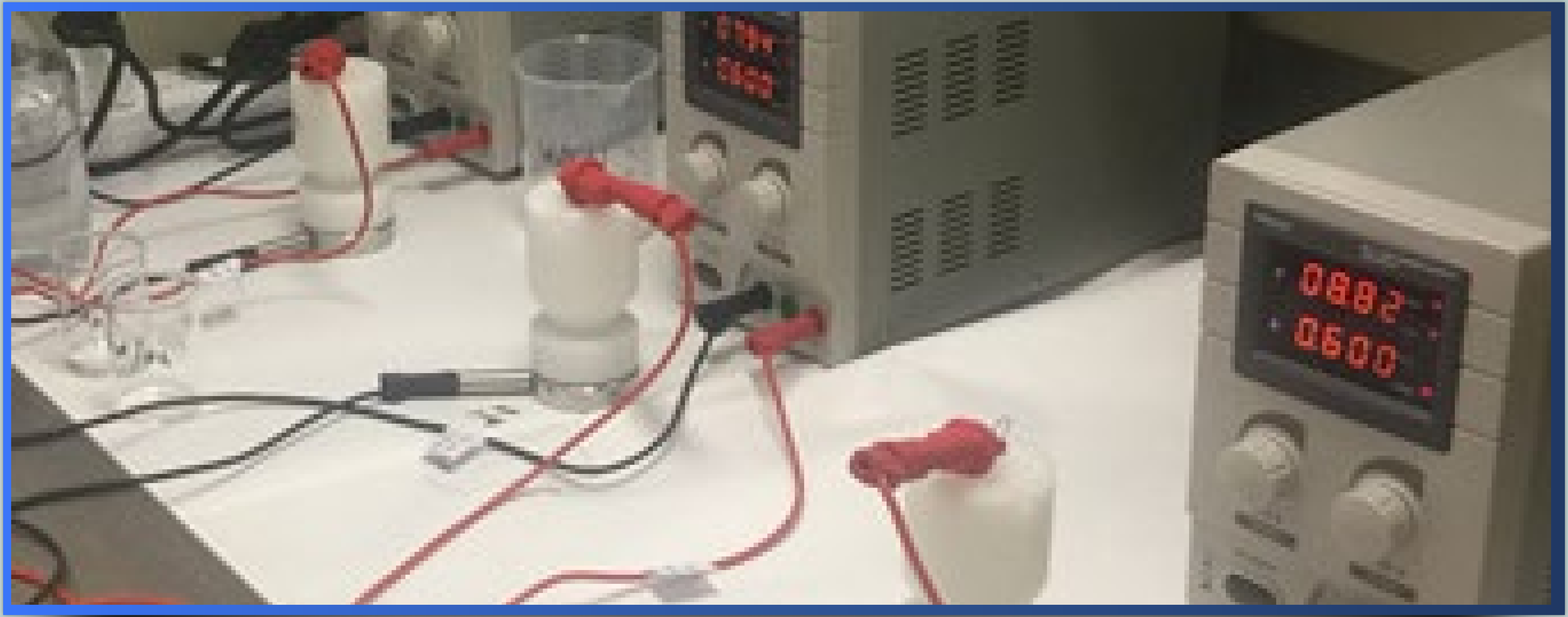

Figure 1: Samples undergoing electrodeposition

Methodology

Water

STEP 1

Three samples 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.

STEP 2

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 co-precipitation of Fe^{3+} with the radionuclides of interest.

STEP 3

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.

STEP 4

The supernatant was then decanted, centrifuged at 3500 rpm for 5 minutes.

STEP 5

The $Fe(OH)_3$ precipitate of each was then dissolved in 5 mL of 3 M HNO_3.

STEP 6

Each sample was loaded on the column. The centrifuge tubes were rinsed 2 twice with 5 mL 3 M HNO_3 and loaded 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.

Methodology

Water

STEP 7

Preconditioning of UTEVA column was done by rinsing the column with 10 mL H_2O, 10 mL 1M HNO_3 and 10 mL 3 M HNO3

STEP 8

Each sample was loaded on the column.

STEP 9

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

STEP 10

Each of the samples was left for a minimum of 30 minutes in an ice bath.

STEP 11

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.

STEP 12

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.

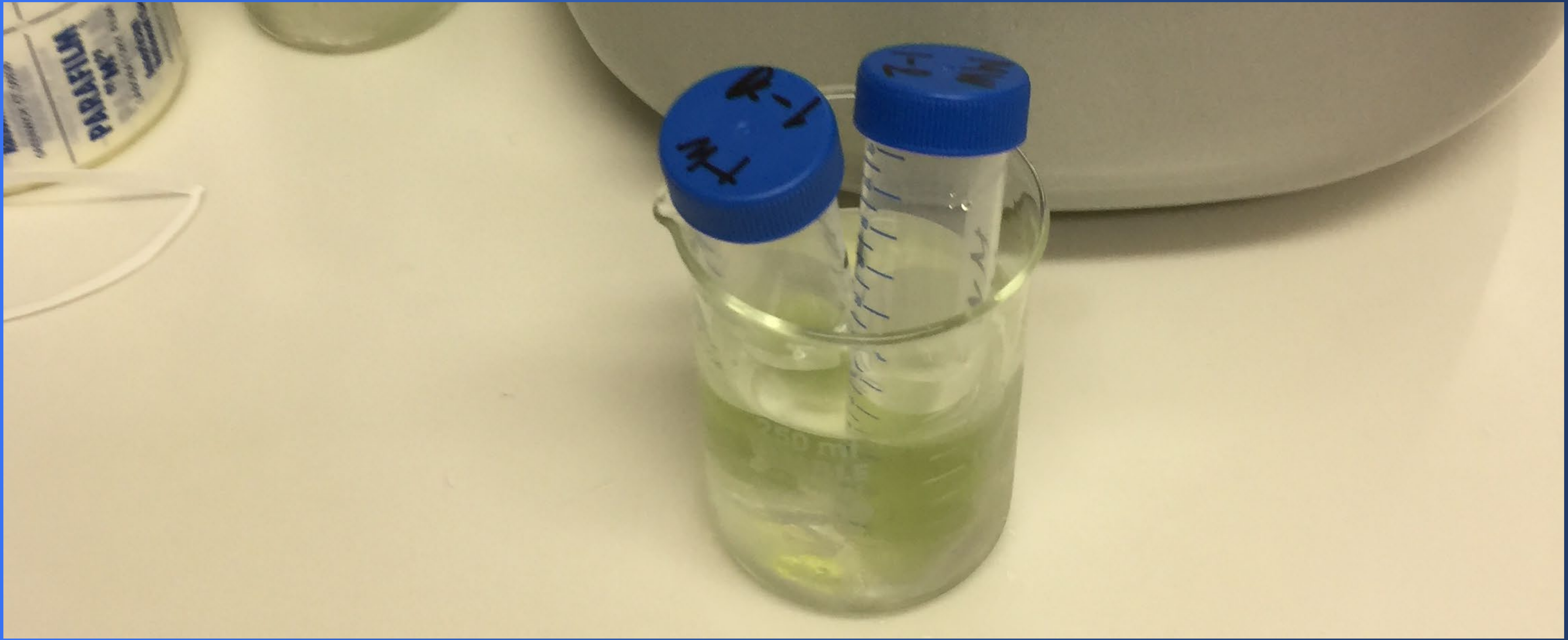

Figure 2: Samples undergoing Microprecipitation

Methodology

Counting

Figure 3: Alpha spectrometry counting system

Passivated Implanted Planar Silicon

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}Pu, ^{241}Am.

Methodology

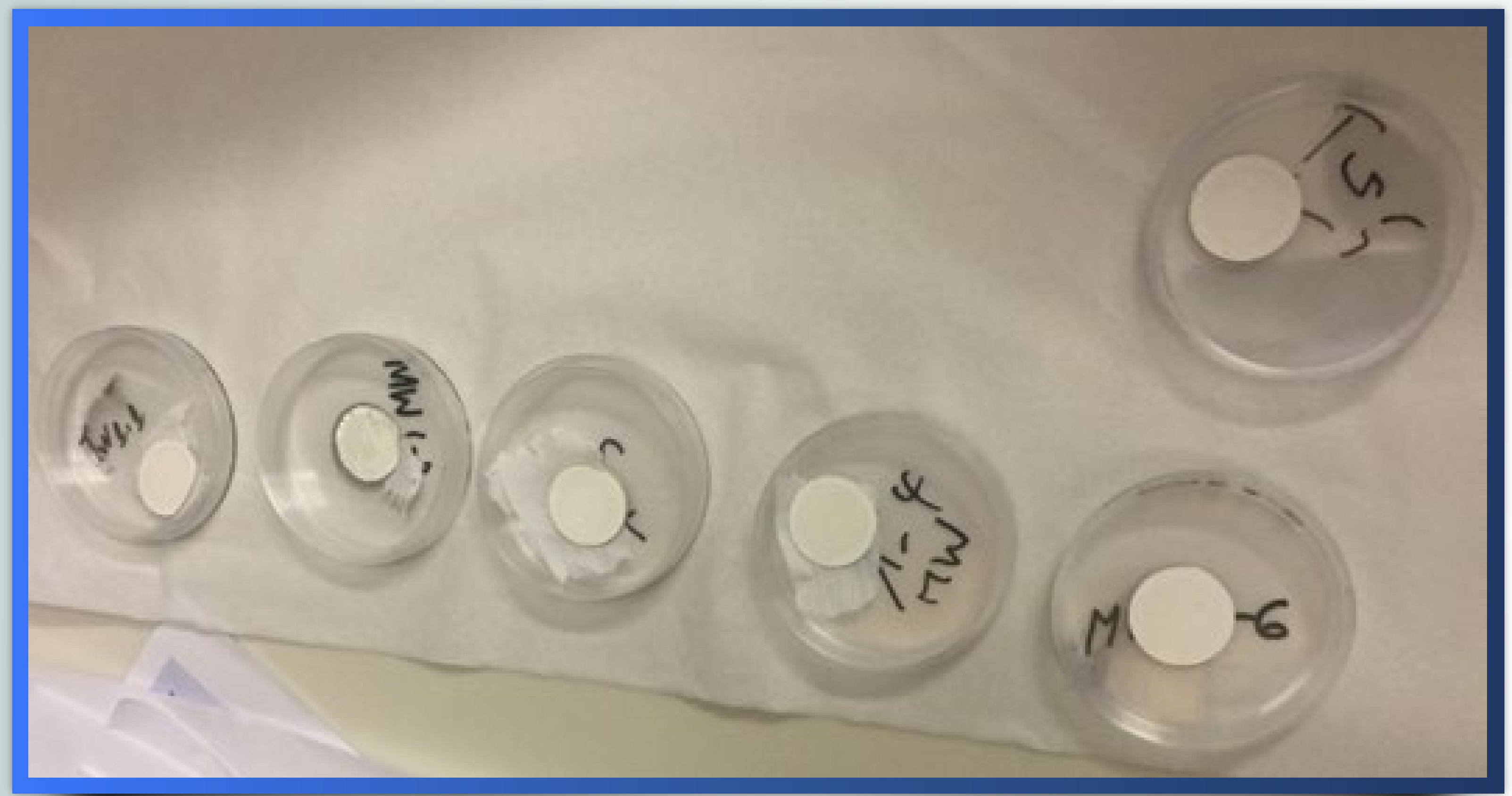

Figure 4a: Samples prepared using Microprecipitation ready for counting.

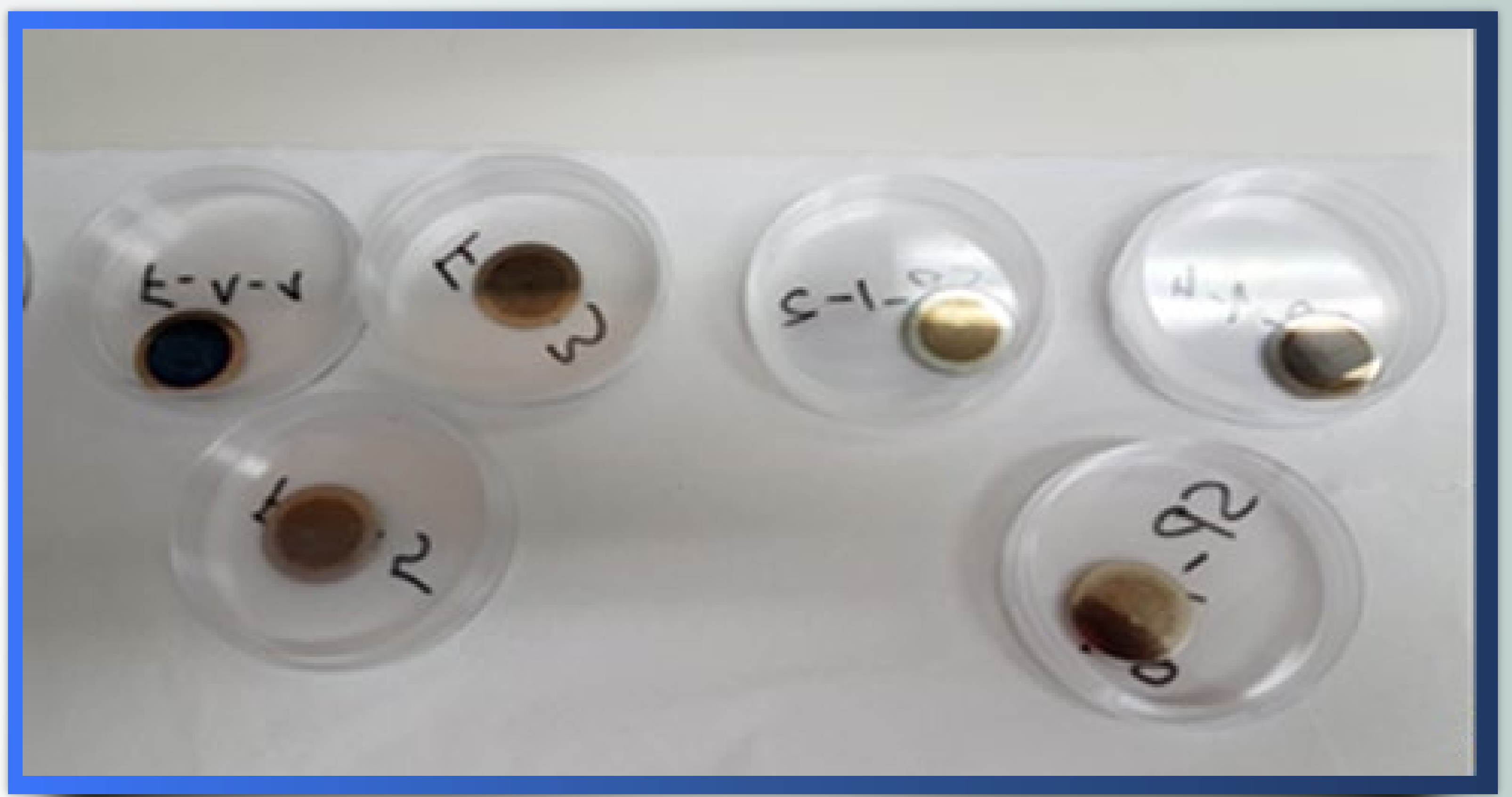

Figure 4b: Samples prepared using Electrodeposition ready for counting.

Section
03

Results & Discussion

Results & Discussion

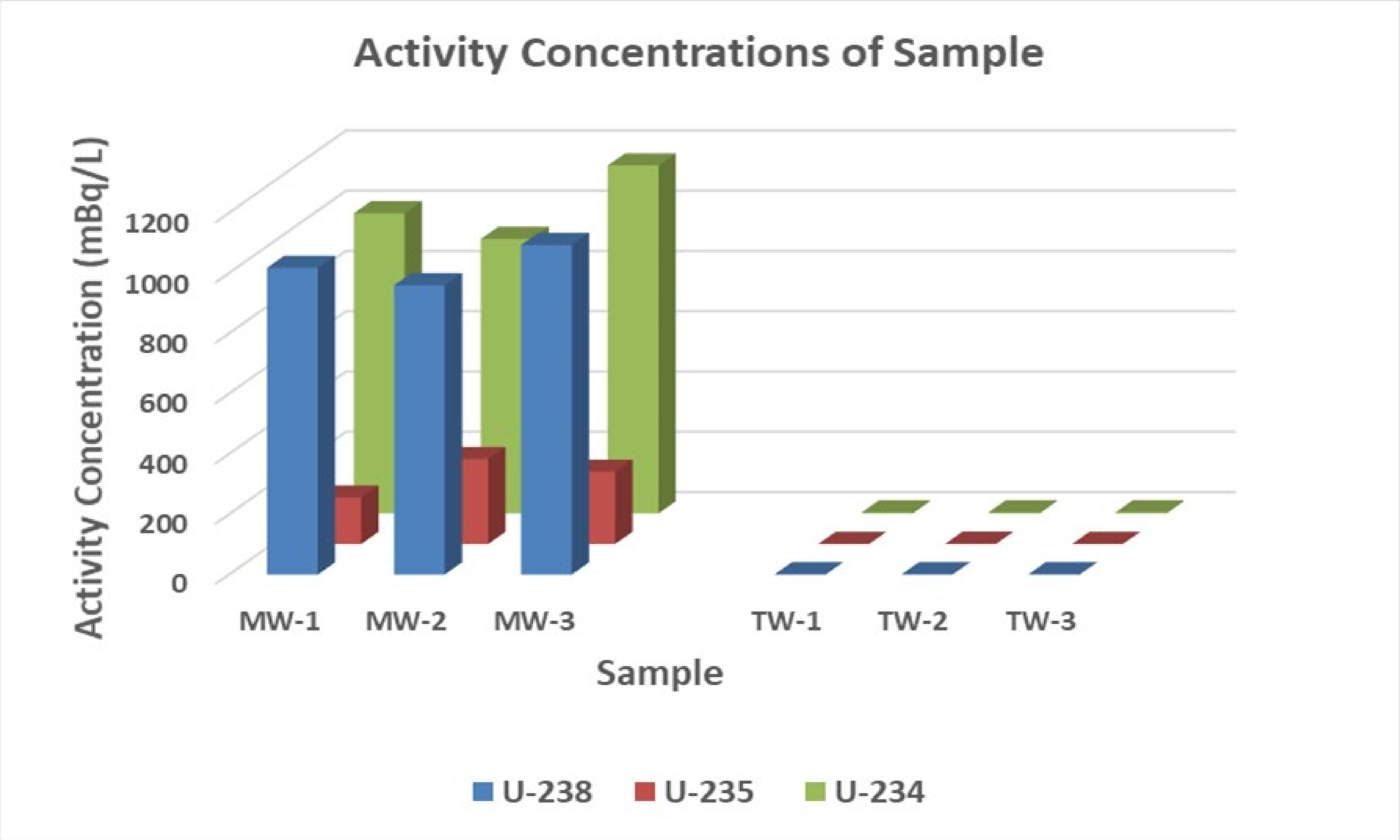

Figure 5: Graphical representation of Activity concentrations of the water samples

Results & Discussion

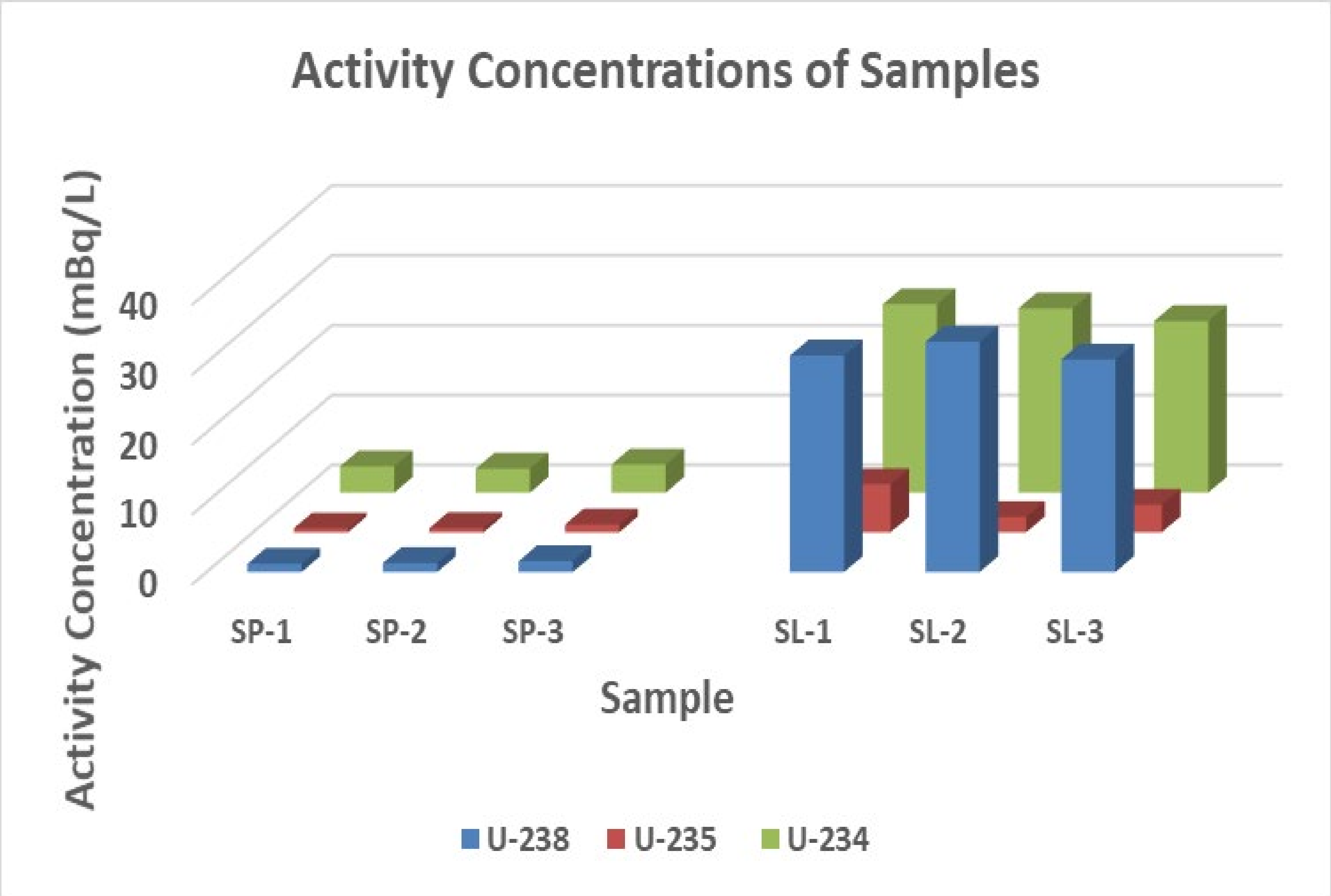

Figure 6: Graphical representation of Activity concentrations of the spinach and soil samples

Results & Discussion

The average activity concentrations of ^{238}U, ^{235}U and ^{234}U in the Mine Water are (1023.7 ± 46.7) mBq/L, (225.3 ± 18.6) mBq/L and (1020.4 ± 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 ± 0.2) mBq/L, (0.5 ± 0.1) mBq/L and (3.8 ± 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 & Discussion

The average activity concentrations of ^{238}U, ^{235}U and ^{234}U in the Soil are (31.5 ± 5.2) Bq/kg, (4.3 ± 1.6) Bq/kg and (26.0 ± 4.6) Bq/kg respectively while the average activity concentrations of ^{238}U, ^{235}U and ^{234}U in the Spinach are (1.3 ± 0.2) Bq/kg, (0.8 ± 0.1) Bq/kg and (3.7 ± 0.3) Bq/kg respectively.

With regards to the activity concentrations of the soil samples, ^{238}U and ^{234}U have similar activity concentrations with ^{235}U having the lowest

However, the activity concentration of ^{234}U in the spinach samples is almost three times that of ^{238}U with ^{235}U having the lowest concentration.

Results & Discussion

	U-238 (mBq/L)	Uncertainty	Expanded Uncertainty (k=2)	U-235 (mBq/L)	Uncertainty	k=2	U-234 (mBq/L)	Uncertainty	Expanded Uncertainty k=2
TW-1	1.28	0.17	0.34	0.36	0.09	0.18	3.67	0.31	0.63
TW-2	1.55	0.20	0.39	0.75	0.13	0.27	3.71	0.33	0.66
TW-3	1.60	0.21	0.42	0.36	0.09	0.19	4.12	0.37	0.74
MW-1	1017.78	33.75	67.51	153.98	13.95	27.90	995.94	46.87	93.75
MW-2	959.85	50.33	100.67	282.16	21.68	43.35	910.73	48.37	96.74
MW-3	1093.46	55.96	111.93	239.83	20.11	40.22	1154.44	58.34	116.68
	U-238 (Bq/kg)	**Uncertainty**	**Expanded Uncertainty (k=2)**	**U-235 (Bq/kg)**	**Uncertainty**	**k=2**	**U-234 (Bq/kg)**	**Uncertainty**	**Expanded Uncertainty k=2**
SP-1	1.18	0.15	0.30	0.56	0.10	0.20	3.76	0.30	0.60
SP-2	1.28	0.15	0.29	0.60	0.10	0.20	3.43	0.26	0.53
SP-3	1.55	0.27	0.54	1.12	0.22	0.45	4.02	0.48	0.96
SL-1	30.97	5.26	10.52	6.88	2.11	4.21	26.96	4.79	9.59
SL-2	32.96	5.38	10.77	2.20	1.12	2.24	26.37	4.63	9.26
SL-3	30.43	4.93	9.86	3.93	1.44	2.89	24.54	4.26	8.52

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

Results & Discussion

The average radiochemical recovery obtained using this method for Mine Water, Tap Water, Spinach and Soil are 74.4%, 59.5%, 64.7% and 77.4% respectively.

Soil samples are often considered to be one of the most difficult matrices to digest. The Soil average radiochemical recovery of 77.4% being the highest indicates that both the digestion and the source preparation procedures were very effective and points to the fact that this method also works for samples with matrices that are tough to digest.

Section
04

Conclusion

Conclusion

This work presents two of the most effective ways of source preparation; micro co-precipitation technique with LaF_3 following column separation using UTEVA resin for two types of water samples; tap water and waste water from the mine, and electrodeposition for spinach and soil samples.

The results obtained show that the method allows for a fast and efficient determination of uranium activity concentrations in all of the samples analyzed.

The radiochemical recoveries determined by the method are greater than the threshold recovery (40%) for acceptance.

References

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That’s all. Thank you very much! ☺