DETERMINATION OF URANIUM CONTENT IN AEROSOL FILTERS BY ALPHA-SPECTROMETRY

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ABSTRACT

The procedure for determination of uranium content in aerosol filters by alpha spectrometry is an appropriate method of monitoring the releases in the environment. The procedure is based on the use of UTEVA resin to separate the uranium. For sample preparation, the filter is dissolved with concentrated nitric acid and hydrogen peroxide, at boiling temperature. Separation yield of actinides is monitored with addition of a known amount of ^{233}U tracer, to the initial sample. Source preparation for alpha spectrometry is performed by micro-precipitation with cerium fluoride. The sample is filtered by using a 0,1 micron 25 mm filter, followed by the mount on stainless steel plate, for subsequent counting by alpha-spectrometry. The separation yields for the analytes were compared, as concerns the recoveries and the counting efficiencies, with used reference materials.

Key words: uranium, aerosol filters, alpha-spectrometry, radiochemistry.

Introduction

Nuclear activities suppose the release of trace amounts of radioactive elements into the environment, so it is necessary to ensure that all this activities have no major effect on the environment and on public health in the close proximity of the nuclear site. By monitoring the radioactive releases in the atmosphere, we ensure that the legal requirements regarding maximum levels of radionuclides, in air, are complied.
Uranium isotopes are alpha emitters and have a particular biological effect. Due to the potential public health effects of releases of uranium to the environment, isotopic determination and measurement of this radionuclide in environmental samples is very important. To achieve this goal, monitoring programs for this radionuclide are necessary and are applied in many countries. The procedure for determination of uranium content in aerosol filters by alpha-spectrometry is an appropriate method of monitoring the releases in the environment.
Measurements of actinides concentration is usually based on detection of their alpha particles emission, which is a very sensitive process requiring preconcentration and separation of the analytes from the matrix and other radioactive isotopes. Determination of uranium isotopic composition in the environmental matrices by alpha-spectrometry requires high degree of chemical purification.
This method is based on sequential separations of uranium by using chromatographic UTEVA resin (Uranium and Tetravalent Actinide Specific Resin) prior to determination of uranium content in aerosol filters by alpha-spectrometry. UTEVA is a resin able to separate and concentrate uranium and tetravalent

actinides from aqueous solutions. Separation yield of actinides is monitored with addition of a well-known amount of ^{233}U tracer, to the initial sample. Usually, ^{232}U is used for tracer in alpha-spectrometry and permits uranium isotopes identification. The aerosol filters, which we use for samples, are results of a technological practice who involve only natural uranium. Source preparation for alpha spectrometry was performed by micro-precipitation with cerium fluoride.

Materials and method

Samples. The experiments were conducted using cellulose aerosols filters with potential content of natural uranium, from technological nuclear activities that suppose the release of trace amounts of radioactive elements into the environment

Tracers. Radioactive tracer solutions containing ^{233}U and ^{232}U were prepared from activity standard solutions. Mass concentrations of these solutions were gravimetrically determined, using the information supplied by initial solutions calibrating certificate. The concentrations of tracer solutions used were verified with alpha – LSC (Liquid Scintillation Counting). We used a solution of ^{233}U in 1M HNO_3 1M with an activity concentration of 371 ± 4 Bq/g, with NIST traceability to prepare a working solution of 3.77 Bq/g activity. For ^{232}U, a working solution with an activity of 0.711 Bq/g was prepared from a standard solution with concentration of 38.22 ± 0.09 Bq/g (**Table 1**).

Table 1 *The concentration of radioactive tracer solutions*

Tracer	C_g(Bq/g)
^{233}U	3.781 ± 0.036
^{232}U	0.711 ± 0.002

C_g – gravimetrically determined concentration

High-efficiency alpha-particle spectrometry. Measurement of uranium content was carried out by using an NIM SOLOIST module, which contains a PIPS detector (passivated implanted planar silicon). The spectrometer was energy and efficiency calibrated by using electroplated standard sources of ^{230}Th, ^{241}Am, ^{244}Cm and ^{233}U.

Sample preparation. The ^{233}U tracer solution was added to about 0.5 g of each sample before filters digestion in order to measure chemical recovery, to correct the results and to improve precision. Wet digestions were performed in glass beakers on hot plates. After the aerosol filters were dissolved in concentrated nitric acid, at boiling temperature, hydrogen peroxide was added progressively for removal of organic material. The solution was evaporated to dryness, and then the system converted to nitric form by adding concentrated nitric acid (**Figure 1**).

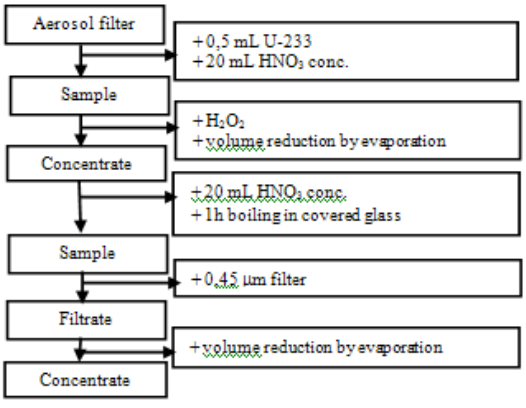

Figure 1 *Sample preparation procedure*

Covered glasses containing sample were maintained on the hot plate for about one hour. The obtained solution was filtered using a 0.45 microns filter and concentrated by evaporation to dryness. The procedure is schematically shown in the figure below.

Uranium separation. Uranium was separated and purified by extraction chromatography with UTEVA resin, product of (Eichrom Industries Inc.). UTEVA is a resin able to separate and concentrate uranium and tetravalent actinides from aqueous solutions. The sample solution was adjusted to 3M HNO_3 and loaded onto a 2 mL prepacked UTEVA column, pre-conditioned with 3M HNO_3. In this step polonium was removed. The column was then rinsed with 3M HNO_3 and washed with 9M and 5M HCl to remove thorium. Finally, the uranium radioisotopes were eluted in a clean beaker, with 15 mL of 1M HCl (**Figure 2**).

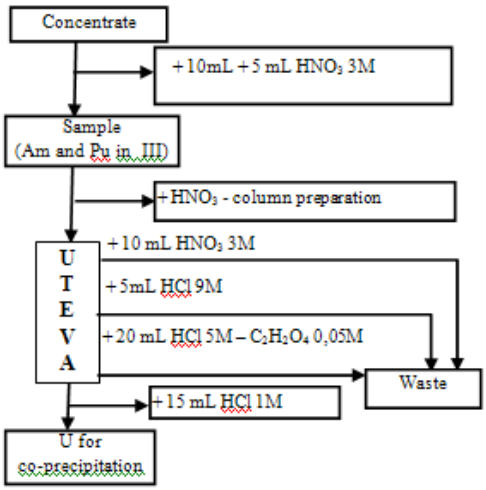

Figure 2 *Uranium separation procedure.*

Co-precipitation and source preparation. After separation, the actinides were co-precipitated with cerium fluoride method. The method suppose the addition of about 0.1 mg $Ce(NO_3)_3$ 0.004M to the uranium stripping solution, followed by the addition of 0.5 mL $TiCl_3$ 10% w/w and 1 mL concentrated HF. After 30 minutes, the cerium fluoride suspension was filtered trough polypropylene Resolve™ Filter (Eichrom Industries Inc.) with 25 mm in diameter and a pore size of 0.1 µm, which was dried then on an IR lamp. The dried filter was mounted onto a stainless steel disc (**Figure 3**). The activities of the uranium radioisotopes were measured by alpha-particle spectrometry.

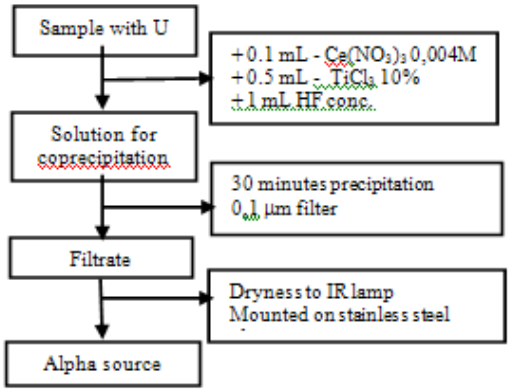

Figure 3 *Uranium co-precipitation and source preparation procedure*

Calculations of the uranium concentration. Activity concentration of ^{238}U in the sample (Bq/mc) was calculated using the formula presented below.

$$A_{^{238}U} = \frac{P_{^{238}U} \times m_{^{233}U} \times A_{^{233}U}}{P_{^{233}U} \times V_{sample}}$$

$P_{^{238}U}$ - net peak area of ^{238}U; $A_{^{238}U}$ - activity concentration of ^{238}U; $m_{^{233}U}$ - mass of added tracer (g);

$P_{^{233}U}$ - net peak area of ^{232}U; V_{sample} - volume of the sample (mc).

Results and discussions

The analytical method described above was used since June 2012, by the Radiation Protection Laboratory, to determine uranium content in aerosol filters from effluents monitor of a technological installation from Institute for Nuclear Research. The following table contains data related to the performance of the method, the average value and its range of variation for measured samples.

Table 2 *Analysis results for 2012 and 2013*

Month	η %	MDA* (mBq)	Mean ^{238}U activity (mBq)	Range of ^{238}U activity (mBq)
June	86 -100	1	157	65 - 288
July	60 -100	1	140	30 - 255
August	71 -95	1	149	72 - 320
September	60 - 99	1	28	8 - 105
October	90 - 100	2	91	49 - 160
November	95 - 100	1	21	17 - 24
January	72 - 97	4	47	24 - 112
February	72 - 100	2	73	22 -156
March	85 - 97	2	131	29 -317

*MDA – minimum detectable activity

The overall recovery measured using ^{233}U was between 60% and 100%. The minimum detectable activity (MDA) for filter samples was in the range of 1 – 4 mBq, (**Table 2**), these results show that the method of ^{238}U is reliable, robust and very sensitive.

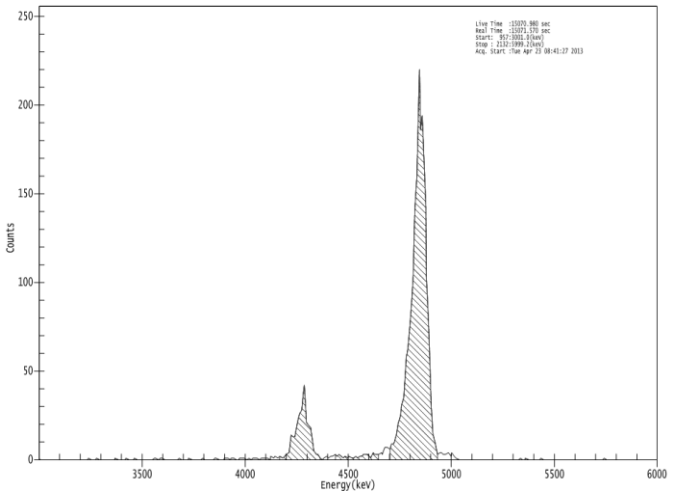

Figure 2 *Alpha spectra of a sample*

The detection efficiency for alpha spectrometry measurement of sources was determined and has a value of 21%.
The use of ^{233}U as tracer for uranium content determination is appropriate, in this case, for the reason that it is well-known that the filters, which we use for samples, are results of a technological practice that involve only natural uranium and is not needed the isotopic discrimination.

Conclusions

Determination of uranium content in aerosol filters is a part of the regular monitoring programme for some nuclear facilities. The procedure for determination of uranium content in aerosol filters by alpha-spectrometry is an appropriate method for monitoring the releases in the environment.
The procedure developed for determination of uranium in aerosol filters, implemented by the Radiation Protection Laboratory, is a selective and accurate method and is suitable for routine measurements of aerosol filters in monitoring programmes. A rapid and precise measurement method was developed for determination of natural uranium in environmental monitoring samples, which is cheaper and more efficient than the traditional techniques based on ion exchange, solvent extraction or precipitation.
Technical capabilities and competence area in the field of effluent monitoring for the Radiation Protection Laboratory as notified testing laboratory were improved through method validation and implementation of this procedure for uranium content determination in aerosol filters by alpha-spectrometry.

References

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