

**Procedure for determining uranium,
plutonium and americium by
extraction-chromatographic procedures**

H-U/Pu/Am-AWASS-01

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Procedure for determining uranium, plutonium and americium by means of extraction-chromatographic procedures

1 Scope

This procedure serves to simultaneously determine the uranium isotopes U-234, U-235 and U-238, the plutonium isotopes Pu-238, Pu-239 and Pu-240, as well as the americium isotope Am-241 in samples of wastewater from nuclear facilities. It furthermore offers an option of determining the curium isotopes Cm-242 and Cm-244 without further effort. For determining Pu-241, reference is made to procedure H-Pu-241-AWASS-01 of these measuring instructions.

2 Sampling

As far as sampling is concerned, reference is made to procedure H- γ -SPEKT-AWASS-01 of these measuring instructions. The sample of the wastewater to be analysed is acidified with ca. 10 ml of concentrated nitric acid ($14 \text{ mol}\cdot\text{l}^{-1}$) per litre to a pH of about 1. The stability of the acidic reaction needs to be monitored, in particular if the sample is stored for an extended period of time.

This procedure ensures that a detection limit of $0,05 \text{ Bq}\cdot\text{l}^{-1}$ for alpha-emitters is reached in a sample volume of 0,1 litres to 0,25 litres and thus complies with the nuclear safety standard 1504 of the Nuclear Safety Standards Commission (1). However, the determination of Pu-241 with a detection limit of $0,5 \text{ Bq}\cdot\text{l}^{-1}$ (2) requires a minimum volume of 0,5 litres.

3 Analysis

3.1 Principle of the procedure

The procedure is based upon the extraction-chromatographic separation of uranium, plutonium and americium ions by means of the commonly available chromatography columns UTEVA·Spec [diamyl, amyolphosphonate on an inert carrier] and TRU·Spec [octylphenyl-N,N-di-isobutyl carbamoylphosphine oxide in tri-n-butyl phosphate on a carrier]. In order to determine the yields of the radiochemical separations U-, Pu- and Am-tracers are added. The extraction-chromatographic separation will usually reach yields of 70 % to 90 %.

Following the enrichment of the radionuclides through evaporating the water sample, the prepared sample solution is loaded onto a UTEVA column with the aim to first compound the U-, Th- and Np-ions and so separate them from the trivalent Pu-, Am- and Cm-ions. The remaining sample solution is then run over a TRU-column, which will retain the Am-, Pu- and Cm-ions. After washing out interfering cations, the radionuclides to be determined are eluted with suitable solvents from the respective columns. Figure 1 provides an overview of the individual steps of the procedure.

The individual eluates (U-, Pu- and Am-fraction, respectively) are evaporated and their residues wet-ashed in order to eliminate organic constituents. The residues are then dissolved and the radionuclides electrochemically deposited on stainless steel discs. These counting sources are then measured by alpha spectrometry.

The Pu-counting source may be used to determine the Pu-241 content after the alpha spectrometric measurement has been completed.

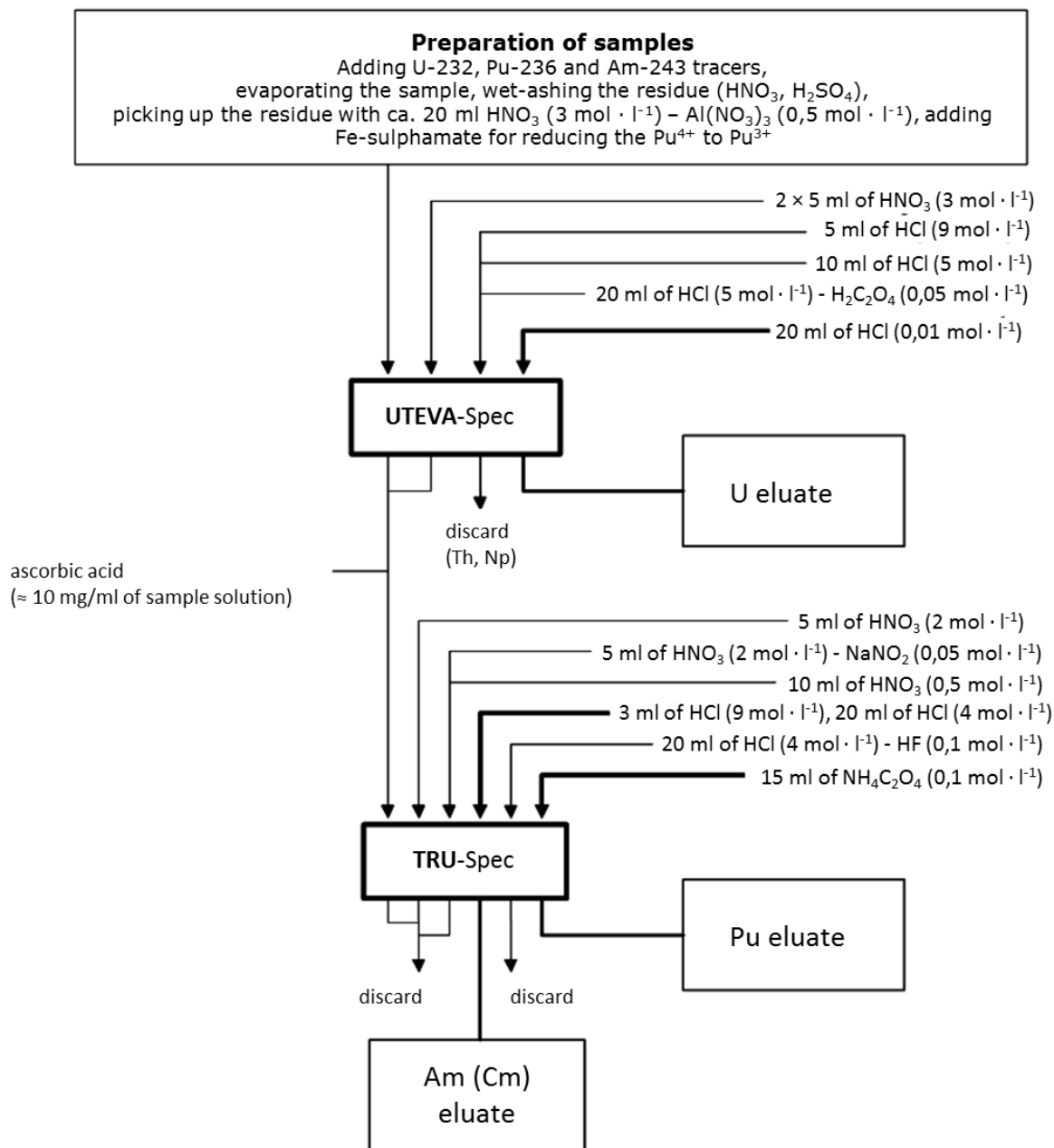


Fig. 1: Flow-chart for the separation of uranium, plutonium and americium isotopes

3.2 Sample preparation

U-, Pu- and Am-tracers are added to the water sample, with the levels of tracer activities depending on the expected activity concentrations. Suitable tracers include U-232, Pu-236, Pu-242 and Am-243, for example.

Note

Decay products of the above-mentioned tracer nuclides (U-232, Th-228 and Np-239, respectively) do not interfere with the measurement. However, if the tracer Pu-236 is used, the activity of U-232 that is added at the same time needs to be taken into consideration for determining the yield of the uranium analysis.

The water sample is evaporated to dryness and the residue fumed off several times with 10 ml of concentrated nitric acid ($14 \text{ mol}\cdot\text{l}^{-1}$) and 5 ml of concentrated sulphuric acid ($18 \text{ mol}\cdot\text{l}^{-1}$) in order to remove all organic constituents. Thereafter, the sample is picked up with about 10 ml of nitric acid ($6 \text{ mol}\cdot\text{l}^{-1}$) and dissolved under stirring in 10 ml aluminium nitrate solution ($1 \text{ mol}\cdot\text{l}^{-1}$). Insoluble constituents may be filtered off.

3.3 Radiochemical separation

3.3.1 2 ml of iron sulphamate solution ($0,6 \text{ mol}\cdot\text{l}^{-1}$) is added to the sample solution in order to reduce Pu-ions of higher oxidation states to Pu^{3+} ; to this end, the resulting sample solution should be at least 0,05 molar in terms of Fe^{2+} -ions (1 ml Fe-sulphamate solution ($0,6 \text{ mol}\cdot\text{l}^{-1}$) per 10 ml of sample solution).

3.3.2 The UTEVA-column (2 ml of column bed volume, 0,7 g of resin) is preconditioned with 5 ml of a solution composed of diluted nitric acid ($3 \text{ mol}\cdot\text{l}^{-1}$), aluminium nitrate ($0,25 \text{ mol}\cdot\text{l}^{-1}$), Fe-sulphamate ($0,01 \text{ mol}\cdot\text{l}^{-1}$) before the sample solution is applied to the column. The vessel containing the sample is washed twice with 5 ml of nitric acid ($3 \text{ mol}\cdot\text{l}^{-1}$), and the wash waters are applied to the column as well. The run-off of the sample solution and the rinsing waters contain Pu- and Am-ions (as well as Cm^{3+}) and is stored for further separation processes.

3.3.3 The UTEVA-column is then washed successively with the following solutions in succession:

- 5 ml of hydrochloric acid ($9 \text{ mol}\cdot\text{l}^{-1}$),
- 10 ml of hydrochloric acid ($5 \text{ mol}\cdot\text{l}^{-1}$), and
- 20 ml of a solution of hydrochloric acid ($5 \text{ mol}\cdot\text{l}^{-1}$) and oxalic acid ($0,05 \text{ mol}\cdot\text{l}^{-1}$).

This serves to separate Th-, Np- and remaining Fe-ions. The resulting wash waters are discarded.

3.3.4 The uranyl-ions are eluted with 20 ml of hydrochloric acid ($0,01 \text{ mol}\cdot\text{l}^{-1}$) from the UTEVA-column, and the eluate is processed further for electrolysis.

Note

The UTEVA-column may be reused several times, but needs to be washed once with, e. g., 5 ml of nitric acid ($0,1 \text{ mol}\cdot\text{l}^{-1}$) before it is stored. The column is capped and left standing with a little supernatant solvent.

3.3.5 The TRU-column (2 ml of column bed volume, 0,7 g of resin) is preconditioned with 5 ml of nitric acid ($2 \text{ mol}\cdot\text{l}^{-1}$).

3.3.6 The remaining sample solution from step 3.3.2 is mixed with 300 mg of ascorbic acid. The solution obtained should be at least 0,05 molar in ascorbic acid in order to maintain the Fe-ions at an oxidation state of +2. This solution is applied to the preconditioned TRU-column. The sample vessel is washed with 5 ml of nitric

acid ($2 \text{ mol}\cdot\text{l}^{-1}$) and the wash water applied to the column as well. The remaining sample solution and flush water may then be discarded.

3.3.7 The TRU-column is washed successively with the following solutions:

- 5 ml of a nitric acid ($2 \text{ mol}\cdot\text{l}^{-1}$) and sodium nitrite ($0,05 \text{ mol}\cdot\text{l}^{-1}$) to oxidise Pu^{3+} Pu^{4+} ,
- 10 ml of nitric acid ($0,5 \text{ mol}\cdot\text{l}^{-1}$) to separate Fe-ions that have remained.

These wash solutions are then discarded.

3.3.8 Subsequently, Am^{3+} (together with Cm^{3+}) is eluted from the column with 3 ml of hydrochloric acid ($9 \text{ mol}\cdot\text{l}^{-1}$) and another 20 ml of hydrochloric acid ($4 \text{ mol}\cdot\text{l}^{-1}$). The eluate is processed further for electrolysis.

3.3.9 The TRU-column is washed with 20 ml of a mixture of hydrochloric acid ($4 \text{ mol}\cdot\text{l}^{-1}$) and hydrofluoric acid ($0,1 \text{ mol}\cdot\text{l}^{-1}$) to separate Th-ions that have remained; the wash water is discarded.

3.3.10 Another clean beaker is placed below the drain of the column and Pu^{4+} is eluted with 15 ml of ammonium hydrogen oxalate ($\text{NH}_4\text{HC}_2\text{O}_4$) solution ($0,1 \text{ mol}\cdot\text{l}^{-1}$); the eluate is then processed further for the electrochemical deposition of plutonium.

3.4 Counting source preparation

The individual eluates (U-, Pu- and Am-fractions) are spiked with 1 ml of sodium hydrogen sulphate solution ($0,1 \text{ mol}\cdot\text{l}^{-1}$), evaporated to dryness on a sand bath and their residues fumed off with 10 ml of concentrated nitric acid ($14 \text{ mol}\cdot\text{l}^{-1}$) and 5 ml of concentrated sulphuric acid ($18 \text{ mol}\cdot\text{l}^{-1}$). This ensures the decomposition of organic substances that might possibly have been washed off the column.

The dry residues are dissolved under heating with 10 ml of sulphuric acid ($1,5 \text{ mol}\cdot\text{l}^{-1}$), the solutions cooled to room temperature, and then transferred to the respective electrolytic cells. The sample vessels are washed twice with 2 ml of sulphuric acid ($0,5 \text{ mol}\cdot\text{l}^{-1}$) and the wash waters added to the corresponding cells as well. Following the addition of one drop of methyl-red solution, a few drops of concentrated ammonia solution ($13,5 \text{ mol}\cdot\text{l}^{-1}$) are added until the colour turns to yellow. The pH is adjusted to 2,4 to 2,5 (colour change to red) by adding sulphuric acid ($1,5 \text{ mol}\cdot\text{l}^{-1}$) drop by drop. The radionuclides are then deposited electrochemically on the cathode at 300 mA for 4 hours. About 1 minute before the electrolytic process is completed, 1 ml of concentrated ammoniac solution is added. The cell is dismantled; the steel disc is rinsed with distilled water and thereafter dried by rinsing it with acetone. The counting sources are labelled and used for measuring their activity.

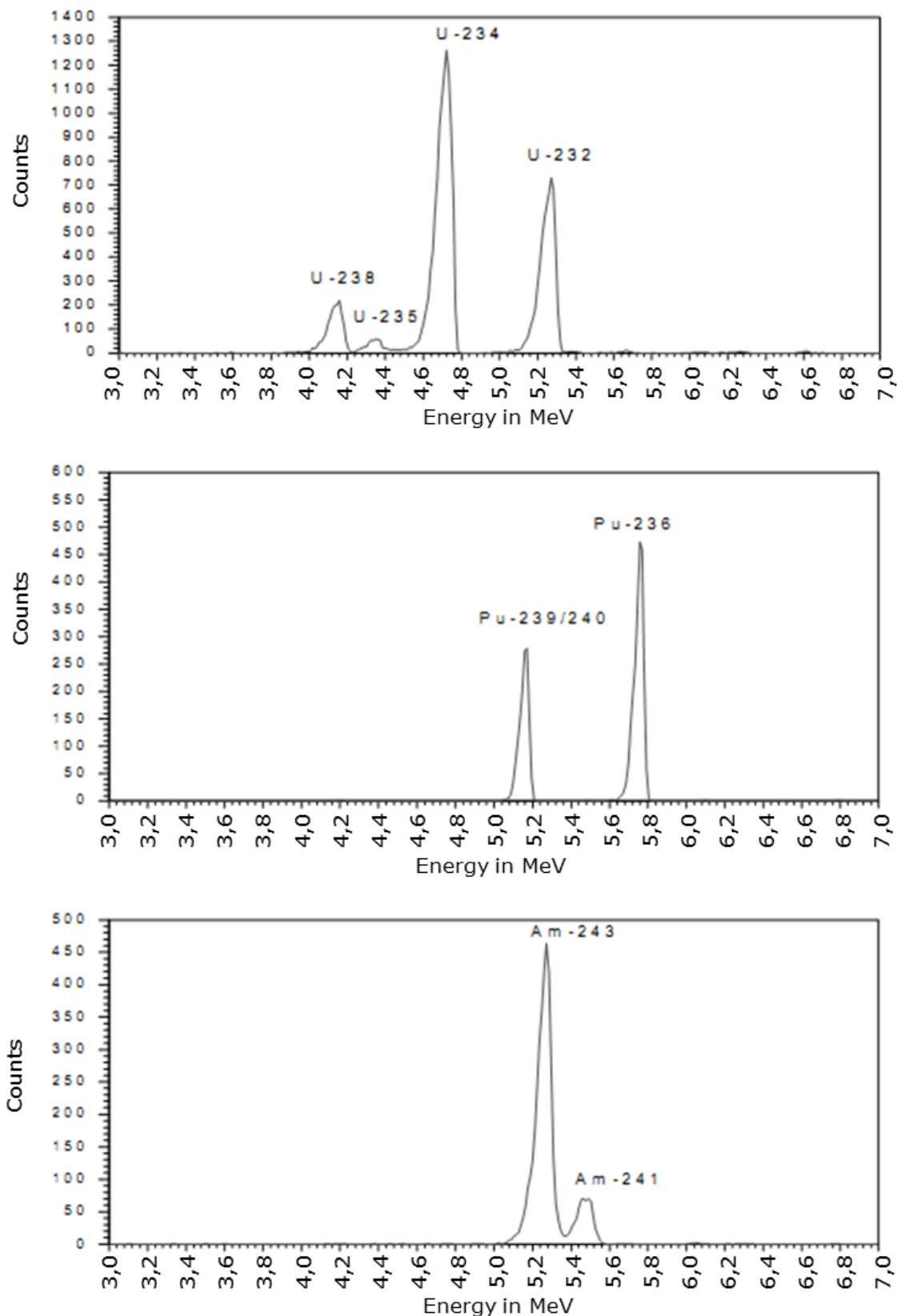


Fig. 2: Alpha spectra of the counting sources obtained from U-, Pu- and Am-eluates of a sample

4 Measuring the activity

Si-semiconductor detectors are employed to measure alpha activity. For basic information on alpha spectrometry, please refer to chapter IV.2 or this procedures manual.

4.1 Calibration

The measuring configuration is calibrated with the aid of calibration sources (e. g., U-233 and Am-241) with known activities and layers of negligible thickness. It is important, though, that the diameters of the active layers of the calibration and the counting sources are identical. Being the quotient of activity (at an emission probability of 100 %) and the measured count rate, the calibration factor, φ_A , is constant throughout the relevant energy spectrum from 4 MeV through 7 MeV.

Note

The emission probability, $p_{\alpha,r}$, is of only of relevance for the calculation of the activity or activity concentration, respectively, in the case of the uranium isotope U-235. Approx. 25 % of the numerous alpha energy lines of U-235 lie outside the analysed energy range of the main lines ($p_{\alpha,U-235} = 0,75$).

The energy calibration is verified at regular intervals, using, e. g., standard sources that contain several alpha emitters.

4.2 Measurement

The measurement of the electrolysis counting sources is based on the same geometric configuration that was used for calibration. Chapter IV.6 of this procedures manual contains a list of the relevant energy lines of alpha emitters that are to be determined.

The alpha spectra of the three counting sources obtained from the analysis of a wastewater sample are illustrated in Figure 2; U-232, Pu-236 and Am-243 were used as tracers here. Measurement periods were 80000 and 100000 seconds, respectively.

5 Calculation of the results

Calculating the activity concentrations of the U-, Pu- and Am-isotopes, c_r , in the wastewater sample follows equation (1):

$$c_r = \varphi_A \cdot \frac{R_{gr} - R_{0r}}{V \cdot \eta_B \cdot p_{\alpha}} \quad (1)$$

with the chemical yields, η , being determined according to equation (2):

$$\eta_B = \varphi_A \cdot \frac{R_{gr} - R_{0r}}{A_{Tr}} \quad (2)$$

where:

c_r activity concentration of the isotope r , in $\text{Bq} \cdot \text{l}^{-1}$;

φ_A calibration factor for a certain measuring geometry, in $\text{Bq} \cdot \text{s}$;

p_{α} emission probability for the alpha radiation of the analysed energy of the radionuclide;

- R_{gr} gross count rate at the main energy of the alpha radiation of the radionuclide r , in s^{-1} ;
- R_{0r} background count rate within the energy range used for measuring the gross count rate, in s^{-1} ;
- V sample volume used, in l;
- η_B chemical yield for the respective element B;
- A_{Tr} activity added as tracer, in Bq.

The relative standard deviations of the activity concentrations of the individual isotopes are calculated according to equation (3):

$$\frac{s(c_r)}{c_r} = \sqrt{\left(\frac{s(R_{nr})}{R_{nr}}\right)^2 + \left(\frac{s(\varphi_A)}{\varphi_A}\right)^2 + \left(\frac{s(\eta_B)}{\eta_B}\right)^2} \quad (3)$$

with

- $\frac{s(c_r)}{c_r}$ relative standard deviation of the activity concentration of the radionuclide r ;
- $\frac{s(R_{nr})}{R_{nr}}$ relative standard deviation of the net count rate, R_{nr} ;
- $\frac{s(\varphi_A)}{\varphi_A}$ relative standard deviation of the calibration factor, φ_A ;
- $\frac{s(\eta_B)}{\eta_B}$ relative standard deviation of the chemical yield of the respective element analysis, η_B .

The relative standard deviation of the net count rate is calculated according to equation (4):

$$\frac{s(R_{nr})}{R_{nr}} = \frac{1}{R_{nr}} \cdot \sqrt{\frac{R_{gr}}{t_m} + \frac{R_{0r}}{t_0}} \quad (4)$$

in which the following additional symbols are introduced:

- R_{nr} net count rate of the radionuclide r , in s^{-1} ;
- t_m duration of the counting source measurement, in s;
- t_0 duration of the background measurement, in s.

Experiences suggest that the relative standard deviation of the calibration factor ranges from 5 % to 10 %, and that of the chemical yield around 10 %.

5.1 Worked example

The following values obtained from analysing a wastewater sample (the corresponding alpha-spectra are illustrated in Figure 2) are used to calculate the activity concentrations of U-, Pu- and Am-isotopes and the respective standard deviations of the activity concentrations:

Tracer activities: 0,28 Bq U-232; 0,10 Bq Pu-236; 0,19 Bq Am-243

$$V = 0,25 \text{ l};$$

$$\varphi_A = 3,67 \text{ Bq}\cdot\text{s};$$

$$\frac{s(\varphi_A)}{\varphi_A} = 0,1;$$

$$\frac{s(\varphi_B)}{\varphi_B} = 0,1.$$

Determining the uranium isotopes

$$t_0 = 500000 \text{ s};$$

$$t_m = 80000 \text{ s};$$

$$R_{0 \text{ U-232}} = 1,8 \cdot 10^{-5} \text{ s}^{-1};$$

$$R_{0 \text{ U-234}} = 8,0 \cdot 10^{-6} \text{ s}^{-1};$$

$$R_{0 \text{ U-235}} = 2,0 \cdot 10^{-6} \text{ s}^{-1};$$

$$R_{0 \text{ U-238}} = 8,0 \cdot 10^{-6} \text{ s}^{-1};$$

$$R_{g \text{ U-232}} = 7,185 \cdot 10^{-2} \text{ s}^{-1};$$

$$R_{g \text{ U-234}} = 1,291 \cdot 10^{-1} \text{ s}^{-1};$$

$$R_{g \text{ U-235}} = 6,20 \cdot 10^{-3} \text{ s}^{-1};$$

$$R_{g \text{ U-238}} = 2,165 \cdot 10^{-2} \text{ s}^{-1}.$$

The chemical yield of determining the uranium, η_{uranium} , is derived from the quotient between the activity measured and that added as a tracer in the shape of U-232 according to equation (2).

$$\eta_{\text{uranium}} = \frac{3,67 \cdot (7,185 \cdot 10^{-2} - 1,8 \cdot 10^{-5})}{0,28} = 0,94$$

As far as the uranium isotopes are concerned, the following activity concentrations and standard deviations are calculated according to equations (1), (3) and (4):

$$c_{\text{U-234}} = (2,02 \pm 0,283) \text{ Bq}\cdot\text{l}^{-1}$$

$$c_{\text{U-235}} = (0,129 \pm 0,019) \text{ Bq}\cdot\text{l}^{-1}$$

$$c_{\text{U-238}} = (0,338 \pm 0,047) \text{ Bq}\cdot\text{l}^{-1}.$$

Determining the plutonium isotopes

$$t_0 = 500000 \text{ s};$$

$$t_m = 100000 \text{ s};$$

$$R_{0 \text{ Pu-236}} = 2,2 \cdot 10^{-5} \text{ s}^{-1};$$

$$R_{0 \text{ Pu-238}} = 2,8 \cdot 10^{-5} \text{ s}^{-1};$$

$$R_{0 \text{ Pu-(239+240)}} = 6,0 \cdot 10^{-6} \text{ s}^{-1};$$

$$R_{g \text{ Pu-236}} = 2,395 \cdot 10^{-2} \text{ s}^{-1};$$

$$R_{g \text{ Pu-238}} = 3,20 \cdot 10^{-5} \text{ s}^{-1};$$

$$R_{g \text{ Pu-(239+240)}} = 1,401 \cdot 10^{-2} \text{ s}^{-1}.$$

The chemical yield of determining the plutonium, η_{Pu} , is calculated according to equation (2) and yields a value of 0,88. Pu-238 was not found in the sample selected as an example here (cf. section 6).

Applying the standard deviation of the net count rate obtained according to equation (4), the activity concentration of Pu-(239+240) and the standard deviation of the activity concentration are obtained from:

$$\frac{s(R_n)}{R_n} = \frac{1}{1,4004 \cdot 10^{-2}} \cdot \sqrt{\left(\frac{1,401 \cdot 10^{-2}}{10^5} + \frac{6 \cdot 10^{-6}}{5 \cdot 10^5}\right)} = 0,027$$

$$c_{\text{Pu-(239+240)}} = 3,67 \cdot \frac{1,401 \cdot 10^{-2} - 0,0006 \cdot 10^{-2}}{0,25 \cdot 0,88} = 0,234 \text{ Bq}\cdot\text{l}^{-1}$$

$$\frac{s(c_{\text{Pu-(239+240)}})}{c_{\text{Pu-(239+240)}}} = \sqrt{0,027^2 + 0,1^2 + 0,1^2} = 0,14$$

The result of the analysis thus reads:

$$c_{\text{Pu-(239+240)}} = (0,234 \pm 0,033) \text{ Bq}\cdot\text{l}^{-1}.$$

Determining the Am-241

$$\begin{array}{ll} t_0 & = 500000 \text{ s}; & t_m & = 80000 \text{ s}; \\ R_{0 \text{ Am-243}} & = 2,2 \cdot 10^{-5} \text{ s}^{-1}; & R_{0 \text{ Am-241}} & = 6,0 \cdot 10^{-6} \text{ s}^{-1}; \\ R_{g \text{ Am-243}} & = 4,261 \cdot 10^{-2} \text{ s}^{-1}; & R_{g \text{ Am-241}} & = 7,712 \cdot 10^{-3} \text{ s}^{-1}. \end{array}$$

Based upon a chemical yield of determining the Am, η_{Am} , of 0,82 and a standard deviation of the net count rate of 3,6 % $\left[\frac{s(R_n)}{R_n} = 0,036\right]$, the result of the analysis shows as follows:

$$c_{\text{Am-241}} = (0,138 \pm 0,021) \text{ Bq}\cdot\text{l}^{-1}$$

5.2 Consideration of uncertainties

Basic information on estimating uncertainties and analysing alpha spectra, please refer to chapter IV of this procedures manual.

The calculation of standard deviations of the activity concentrations takes uncertainties in the measuring process, uncertainties in calibration, and uncertainties in determining the chemical yields into account as major contributors to the total uncertainty. Additional sources of uncertainties may be encountered in individual analyses: Imperfect separation of the individual radionuclide fractions from the sample matrix and/or interfering radionuclides may sometimes contribute substantially to the uncertainty of the results. Indicators in such a case are low yields and/or spectra that can hardly be analysed as a result of thick layers or overlapping lines.

In practice, it is not usually possible to quantify interferences of this kind, and the analysis will have to be repeated using a smaller volume of the original material.

6 Characteristic limits of the procedure

Calculating the detection limits of U-, Pu- and Am-activities is based on time preselection, low number of background counts, and different measurement periods for the counting sources prepared from the sample material and the background effects according to equation (5):

$$G_r = \frac{\varphi_A}{p_\alpha} \cdot \left[(k_{1-\alpha} + k_{1-\beta}) \sqrt{R_{0r} \cdot \left(\frac{1}{t_0} + \frac{1}{t_m} \right)} + \frac{1}{4} \cdot (k_{1-\alpha} + k_{1-\beta})^2 \left(\frac{1}{t_0} + \frac{1}{t_m} \right) \right] \quad (5)$$

with $p_\alpha = 1$ for all radionuclides analysed except U-235 ($p_\alpha = 0,75$).

Aside from the symbols already used before:

G_r detection limit of the activity of the respective radionuclide, in Bq;

$k_{1-\alpha}$, $k_{1-\beta}$ quantiles of the normal distribution.

The detection limits of the activity concentrations, g_r ($\text{Bq} \cdot \text{l}^{-1}$), are calculated according to equation (6):

$$g_r = \frac{G_r}{V \cdot \eta_B} \quad (6)$$

Using the values specified in section 5.1 and $k_{1-\alpha} + k_{1-\beta} = 4,645$, the detection limits yield:

$$G_{\text{U-234}} = 3,67 \left[4,645 \cdot \sqrt{\frac{8 \cdot 10^{-6}}{5 \cdot 10^5} + \frac{8 \cdot 10^{-6}}{8 \cdot 10^4}} + \frac{1}{4} \cdot 4,645^2 \cdot \left(\frac{1}{5 \cdot 10^5} + \frac{1}{8 \cdot 10^4} \right) \right] = 4,69 \cdot 10^{-4} \text{Bq}$$

$$g_{\text{U-234}} = \frac{4,69 \cdot 10^{-4}}{0,25 \cdot 0,94} = 1,99 \cdot 10^{-3} \text{Bq} \cdot \text{l}^{-1}$$

$G_{\text{U-234}}$	= 0,47 mBq;	$g_{\text{U-234}}$	= 2,0 mBq·l ⁻¹ ;
$G_{\text{U-235}}$	= 0,51 mBq;	$g_{\text{U-235}}$	= 2,2 mBq·l ⁻¹ ;
$G_{\text{U-238}}$	= 0,47 mBq;	$g_{\text{U-238}}$	= 2,0 mBq·l ⁻¹ ;
$G_{\text{Pu-238}}$	= 0,55 mBq;	$g_{\text{Pu-238}}$	= 2,5 mBq·l ⁻¹ ;
$G_{\text{Pu-(239+240)}}$	= 0,38 mBq;	$g_{\text{Pu-(239+240)}}$	= 1,7 mBq·l ⁻¹ ;
$G_{\text{Am-241}}$	= 0,45 mBq;	$g_{\text{Am-241}}$	= 2,2 mBq·l ⁻¹ .

7 Catalogue of chemicals and equipment

7.1 Chemicals

The extraction-chromatographic separation process requires ready-to-use columns or loose resin that are available under the following trade names (Eichrom Industries Inc.):

- UTEVA Resin
- TRU Resin

All chemicals used should furthermore be of the purity grade "pro analysi":

- Concentrated nitric acid, HNO_3 ($14 \text{ mol}\cdot\text{l}^{-1}$)
moreover HNO_3 solutions with the following concentrations:
 $6 \text{ mol}\cdot\text{l}^{-1}$ (420 ml of concentrated HNO_3 per litre),
 $3 \text{ mol}\cdot\text{l}^{-1}$ (210 ml of concentrated HNO_3 per litre),
 $2 \text{ mol}\cdot\text{l}^{-1}$ (140 ml of concentrated HNO_3 per litre)
- Concentrated hydrochloric acid ($10 \text{ mol}\cdot\text{l}^{-1}$)
and HCl solutions with the following concentrations:
 $9 \text{ mol}\cdot\text{l}^{-1}$ (880 ml of concentrated HCl per litre),
 $5 \text{ mol}\cdot\text{l}^{-1}$ (490 ml of concentrated HCl per litre),
 $4 \text{ mol}\cdot\text{l}^{-1}$ (390 ml of concentrated HCl per litre),
 $0,01 \text{ mol}\cdot\text{l}^{-1}$ (1 ml of concentrated HCl per litre)
- Concentrated sulphuric acid ($18 \text{ mol}\cdot\text{l}^{-1}$)
and H_2SO_4 solutions with the following concentrations:
 $1,5 \text{ mol}\cdot\text{l}^{-1}$ (83 ml of concentrated H_2SO_4 per litre),
 $0,5 \text{ mol}\cdot\text{l}^{-1}$ (28 ml of concentrated H_2SO_4 per litre)
- Hydrofluoric acid 40 %, HF ($23 \text{ mol}\cdot\text{l}^{-1}$)
- Ammonia solution 25 %, NH_3 ($13,5 \text{ mol}\cdot\text{l}^{-1}$)
- Sodium hydrogen sulphate solution, NaHSO_4 ($0,1 \text{ mol}\cdot\text{l}^{-1}$):
12 g of NaHSO_4 in 1 l of H_2O
- Methyl-red solution (0,5 %, in ethanol)
- $\text{Al}(\text{NO}_3)_3$ solution ($1 \text{ mol}\cdot\text{l}^{-1}$): 75 g of $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ dissolved in 200 ml of H_2O
- HCl ($5 \text{ mol}\cdot\text{l}^{-1}$) containing 0,05 M of oxalic acid ($0,05 \text{ mol}\cdot\text{l}^{-1}$):
3,15 g oxalic acid di-hydrate ($\text{H}_2\text{C}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$) dissolved in 500 ml of 5 M of HCl
- Ammonium hydrogen oxalate solution, $\text{NH}_4\text{HC}_2\text{O}_4$ ($0,1 \text{ mol}\cdot\text{l}^{-1}$):
2,52 g oxalic acid di-hydrate ($\text{H}_2\text{C}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$) and 2,84 g of di-ammonium oxalate mono-hydrate [$(\text{NH}_4)_2\text{C}_2\text{O}_4 \cdot \text{H}_2\text{O}$] dissolved in 180 ml of H_2O ; the solution might need to be filtered and diluted with H_2O to a volume of 200 ml

Moreover, the following, freshly prepared chemicals are required for the separation process (with the amounts of solutions being calculated to last for 2 or 3 analyses):

- Fe-sulphamate solution ($0,6 \text{ mol}\cdot\text{l}^{-1}$), **prepared fresh every week**
(The concentration specified refers to Fe; $\text{H}_2\text{NSO}_3\text{H}$ is contained in excess)
Dissolve 5,7 g of sulphamic acid in 15 ml of H_2O and add 700 mg of Fe-filings in portions under stirring at ca. 70 °C, filter solution, and replenish with H_2O to a total of 20 ml.
- HNO_3 ($2 \text{ mol}\cdot\text{l}^{-1}$) – NaNO_2 ($0,05 \text{ mol}\cdot\text{l}^{-1}$), **prepared fresh every day**
52 mg of NaNO_2 dissolved in 15 ml of 2 M HNO_3
- HCl ($4 \text{ mol}\cdot\text{l}^{-1}$) - HF ($0,1 \text{ mol}\cdot\text{l}^{-1}$), **prepared fresh every day**
250 μl of concentrated HF (40 %) mixed with 60 ml of 4 M HCl ,

- HNO_3 ($3 \text{ mol}\cdot\text{l}^{-1}$) – $\text{Al}(\text{NO}_3)_3$ ($0,25 \text{ mol}\cdot\text{l}^{-1}$) – Fe-sulphamate ($0,01 \text{ mol}\cdot\text{l}^{-1}$),
prepared fresh every week
Dilute 10 ml of HNO_3 ($6 \text{ mol}\cdot\text{l}^{-1}$) and 2,5 ml of 1 M of $\text{Al}(\text{NO}_3)_3$ ($1 \text{ mol}\cdot\text{l}^{-1}$) to 20 ml and add 300 μl of Fe-sulphamate solution ($0,6 \text{ mol}\cdot\text{l}^{-1}$) to the solution.

7.2 Equipment

- Stainless steel discs (V2A, \varnothing 25 mm, thickness 0,3 mm);
- Electrolysis cell (see chapter IV.2 alpha-spectrometry);
- Measuring station for alpha spectrometry;
- Basic equipment of a radiochemical laboratory.

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