Procedure for determining uranium isotopes in drinking water and ground water by alpha spectrometry

 $H-\alpha$ -SPEKT-TWASS-01

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ISSN 1865-8725

Version September 1992

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1 Scope

The procedure described here serves to simultaneously determine the uranium isotopes U-234, U-235 and U-238 in drinking and ground water. The principles of the procedure may also be projected to other waters, including, e.g., raw water in water purification plants, spring and seepage waters, mineral water etc. This procedure largely conforms to procedure H- α -SPEKT-AWASS-01 for wastewater.

The chemical yields from waters that are very high in electrolytes, like mineral and seepage waters from landfill sites (with a specific electrical conductivity > 2000 μ S·cm⁻¹) must be expected to be lower compared to the other waters mentioned above.

Note

One detail with regard to the activity ratio between the uranium isotopes U-238 and U-234 in drinking and ground waters needs to be pointed out. In general, the activity ratio between the parent nuclide U-238 and its daughter nuclide U-234 in ground water ranges from 1:1,2 to 1:1,5 and thus deviates to a larger or smaller extent from a perfect equilibrium of 1:1. In individual cases, the activity ratio in ground water may even be at 1:5. In contrast, the activity ratio between U-238 and U-235 remains unchanged at a ratio of 21,7:1.

2 Sampling

An adequately large volume of representative sample water (normally 1 l to 10 l) is collected. Details on the selection of samples and sampling can be found in procedure H- γ -SPEKT-TWASS-01. The water samples are stabilised with about 1 ml of nitric acid (14 mol·l⁻¹) per litre of water and may be stored in closed polyethylene vessels until they can be processed.

3 Analysis

3.1 Principle of the procedure

The principle of the procedure is illustrated in Figure 1. Uranium is separated from a hydrochloric acid solution that will remove accompanying elements such as plutonium, thorium and iron as a hexachlorido complex by anion exchange. The activity is determined by alpha spectrometry of thin-film counting sources that are manufactured by electrochemical deposition of uranium in hydroxide form on stainless steel discs. For determining the chemical yield, a U-232 tracer with known activity is added.

Note

The extraction process using trioctylmethylammonium nitrate/xylene described in procedure H- α -SPEKT-AWASS-01 can usually be skipped here.

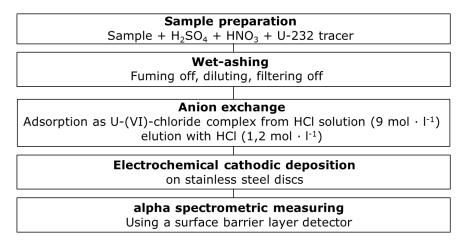


Fig. 1: Principle of determining uranium isotopes in drinking water

3.2 Sample preparation

The water sample is spiked with about 0,5 Bq of U-232 tracer, 5 ml of sulphuric acid (18 mol·l⁻¹), and 10 ml of nitric acid (14 mol·l⁻¹) are added, evaporated, and fumed off to dryness. Once cooled, 2 ml of sulphuric acid (18 mol·l⁻¹) and 5 ml of nitric acid (14 mol·l⁻¹) are added to the residue and once more fumed off to dryness. This step needs to be repeated until the residue will no longer contain organic constituents.

The sample is then processed further according to procedure H- α -SPEKT-AWASS-01, step 3.3.5 ff.

4 Measuring the activity

For measuring the activity, reference is made to procedure H- α -SPEKT-AWASS-01, section 4.

A typical alpha spectrum of a ground water sample is illustrated in Figure 2 and clearly demonstrates that the activity ratio between U-238 and U-234 does not lie at 1:1, but rather at about 1:2.

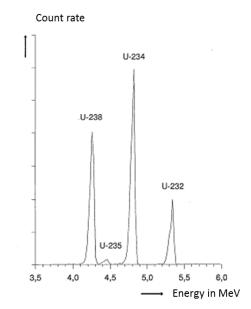


Fig. 2: Alpha spectrum of uranium isotopes in a sample of ground water

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5 Calculation of the results

The activity concentrations, c_r , of the individual uranium isotopes, r, are calculated according to equation (1):

$$c_{\rm r} = \frac{\varphi_{\rm A}}{V \cdot \eta \cdot p_{\alpha,\rm r}} \cdot \left(R_{\rm g} - R_{\rm 0}\right)$$
(1)

where

 c_r activity concentration of the uranium isotope r, in Bq·l⁻¹;

 φ_A the calibration factor, in Bq·s;

 $p_{\alpha,r}$ emission probability of alpha radiation of the nuclide r;

 η chemical yield; count < 1;

- R_g gross count rate in the region of the line at E_{α} , in s⁻¹;
- R_0 mean background count rate in the region of the base width of the line at E_{α} , in s⁻¹;
- *V* volume of the sample used, in I.

5.1 Worked example

The following data are available for determining the uranium content of a sample of drinking water:

$arphi_{A}$	= 5,46 Bq·s;	
η	= 0,871 (87,1%);	
$p_{lpha r}$	= 0,74 for U-235;	
$p_{lpha r}$	= 1,00 for U-234 ar	nd U-238;
t _m	= 55885 s;	
R_0	= 2,0·10 ⁻⁴ s ⁻¹	in the region of the U-234 line;
R_0	= 7,5·10 ⁻⁵ s ⁻¹	in the region of the U-235 line;
R_0	$= 1, 1 \cdot 10^{-4} \text{ s}^{-1}$	in the region of the U-238 line;
R_{g}	= 1,14·10 ⁻² s ⁻¹	in the region of the U-234 line;
Rg	= 2,86·10 ⁻⁴ s ⁻¹	in the region of the U-235 line;
R_{g}	= 7,87·10 ⁻³ s ⁻¹	in the region of the U-238 line;
R n	= 1,12·10 ⁻² s ⁻¹	in the region of the U-234 line;
R _n	= 2,11·10 ⁻⁴ s ⁻¹	in the region of the U-235 line;
R _n	= 7,76·10 ⁻³ s ⁻¹	in the region of the U-238 line;
V	= 5,0 l.	

where R_n is the net count rate.

According to equation (1), the following values are obtained for the activity concentration of the U-isotopes:

$$\begin{aligned} c_{U^{-234}} &= 1, 4 \cdot 10^{-2} \text{ Bq} \cdot \text{I}^{-1} \text{ (14 mBq} \cdot \text{I}^{-1}); \\ c_{U^{-235}} &< 5 \cdot 10^{-4} \text{ Bq} \cdot \text{I}^{-1} \text{ (see section 6)}; \\ c_{U^{-238}} &= 9, 7 \cdot 10^{-3} \text{ Bq} \cdot \text{I}^{-1} \text{ (9,7 mBq} \cdot \text{I}^{-1}). \end{aligned}$$

5.2 Consideration of uncertainties

For a consideration of uncertainties, reference is made to procedure H- α -SPEKT-AWASS-01, section 5.2.

6 Characteristic limits of the procedure

For calculating the detection limits, *G*, reference is made to chapter IV.5, equation (2.5) of this procedures manual. The calculation of achievable detection limits is discussed in procedure H- α -SPEKT-AWASS-01.

At measuring periods, t_m (sample) and t_0 (background effect) of 84600 s each, a value of k = 4,65 ($k_{1-\alpha} = 3,00$; $k_{1-\beta} = 1,65$), and a calibration factor of $\varphi_A = 5,46$ Bq·s, detection limits of the activity of 1,8 mBq are obtained for U-234 and U-238 each and 2,3 mBq for U-235. Using an original sample volume, V, of 5 l of drinking water and a radiochemical yield, η , of 87 %, the detection limit of the activity concentration is calculated according to equation (2):

$$g = \frac{G}{V \cdot \eta \cdot p_{a,r}}$$
(2)

and will give a value of 0,5 mBq \cdot I⁻¹ for each individual uranium isotope.

The minimum detection limit of 10 mBq \cdot l⁻¹ prescribed for measuring programmes for drinking water by the Precautionary Radiation Protection Act ("Strahlenschutz-vorsorgegesetz", StrVG) may be reached with a sample volume of 0,25 l under the same conditions.

7 Catalogue of chemicals and equipment

The chemicals and equipment required for this procedure are listed in procedure H- α -SPEKT-AWASS-01, section 7.

References

For references, see procedure H- α -SPEKT-AWASS-01.