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Second edition
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Water quality — Gross alpha and gross beta activity — Test method using thin source deposit

Qualité de l'eau — Activités alpha globale et bêta globale — Méthode d'essai par dépôt d'une source fine

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ISO copyright office
CP 401 • Ch. de Blandonnet 8
CH-1214 Vernier, Geneva
Phone: +41 22 749 01 11
Fax: +41 22 749 09 47
Email: copyright@iso.org
Website: www.iso.org

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Foreword

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The procedures used to develop this document and those intended for its further maintenance are described in the ISO/IEC Directives, Part 1. In particular the different approval criteria needed for the different types of ISO documents should be noted. This document was drafted in accordance with the editorial rules of the ISO/IEC Directives, Part 2 (see www.iso.org/directives).

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For an explanation on the voluntary nature of standards, the meaning of ISO specific terms and expressions related to conformity assessment, as well as information about ISO's adherence to the World Trade Organization (WTO) principles in the Technical Barriers to Trade (TBT) see the following URL: www.iso.org/iso/foreword.html.

This document was prepared by Technical Committee ISO/TC 147, *Water quality*, Subcommittee SC 3, *Radioactivity measurements*.

Any feedback or questions on this document should be directed to the user's national standards body. A complete listing of these bodies can be found at www.iso.org/members.html.

This second edition cancels and replaces the first edition (ISO 10704:2009), which has been technically revised. The main changes compared to the previous edition are as follows:

— Introduction: an introduction has been added;

— Clause 1: the scope has been modified to specify applicability to emergency situations and applicability of waste water as a test sample; information about the exclusion of low energy beta emitters has also been added;

— Clause 4: the filtration has been specified to be carried out at 0,45 µ;

— 5.1.2.2: ^{137}Cs has been introduced as a standard that can be used;

— 5.2.4: the recommended thickness has been increased to up to 400 µg/cm^2;

— 7.6.3.1: in order to evaluate self-absorption phenomena, spiking method has been recommended to mimic the nature of the salt;

— Clause 8:

  — a new Formula (9) has been introduced to obtain the beta activity concentration when systematic correction is not required;

  — the subsequent Formulae have been renumbered;

— Clause 9: several limitations and interferences have been given;

— 9.1: the natural radionuclides contributions have been given.

Introduction

Radioactivity from several naturally-occurring and anthropogenic sources is present throughout the environment. Thus, water bodies (e.g. surface waters, ground waters, sea waters) can contain radionuclides of natural, human-made, or both origins:

— natural radionuclides, including ^{40}K, ^{3}H, ^{14}C, and those originating from the thorium and uranium decay series, in particular ^{226}Ra, ^{228}Ra, ^{234}U, ^{238}U, ^{210}Po and ^{210}Pb can be found in water for natural reasons (e.g. desorption from the soil and washoff by rain water) or can be released from technological processes involving naturally occurring radioactive materials (e.g. the mining and processing of mineral sands or phosphate fertilizers production and use);

— human-made radionuclides such as transuranium elements (americium, plutonium, neptunium, curium), ^{3}H, ^{14}C, ^{90}Sr, and gamma emitting radionuclides can also be found in natural waters. Small quantities of these radionuclides are discharged from nuclear fuel cycle facilities into the environment as a result of authorized routine releases. Some of these radionuclides used for medical and industrial applications are also released into the environment after use. Anthropogenic radionuclides are also found in waters as a result of past fallout contaminations resulting from the explosion in the atmosphere of nuclear devices and accidents such as those that occurred in Chernobyl and Fukushima.

Radionuclide activity concentration in water bodies can vary according to local geological characteristics and climatic conditions and can be locally and temporally enhanced by releases from nuclear installation during planned, existing, and emergency exposure situations[1]. Drinking-water can thus contain radionuclides at activity concentrations which could present a risk to human health.

The radionuclides present in liquid effluents are usually controlled before being discharged into the environment[2] and water bodies. Drinking waters are monitored for their radioactivity as recommended by the World Health Organization (WHO)[3] so that proper actions can be taken to ensure that there is no adverse health effect to the public. Following these international recommendations, national regulations usually specify radionuclide authorized concentration limits for liquid effluent discharged to the environment and radionuclide guidance levels for waterbodies and drinking waters for planned, existing, and emergency exposure situations. Compliance with these limits can be assessed using measurement results with their associated uncertainties as specified by ISO/IEC Guide 98-3 and ISO 5667-20[4].

Depending on the exposure situation, there are different limits and guidance levels that would result in an action to reduce health risk. As an example, during a planned or existing situation, the WHO guidelines for guidance level in drinking water is 0,5 Bq/l for gross alpha activity and 1 Bq/l for gross beta activity.

NOTE The guidance level is the activity concentration with an intake of 2 l/d of drinking water for one year that results in an effective dose of 0,1 mSv/a for members of the public. This is an effective dose that represents a very low level of risk and which is not expected to give rise to any detectable adverse health effects[3].

Thus, the test method can be adapted so that the characteristic limits, decision threshold, detection limit and uncertainties ensure that the radionuclide activity concentrations test results can be verified to be below the guidance levels required by a national authority for either planned/existing situations or for an emergency situation[5][6][7].

Usually, the test methods can be adjusted to measure the activity concentration of the radionuclide(s) in either wastewaters before storage or in liquid effluents before being discharged to the environment. The test results will enable the plant/installation operator to verify that, before their discharge, wastewaters/liquid effluent radioactive activity concentrations do not exceed authorized limits.

The test method(s) described in this document can be used during planned, existing and emergency exposure situations as well as for wastewaters and liquid effluents with specific modifications that could increase the overall uncertainty, detection limit, and threshold.

The test method(s) can be used for water samples after proper sampling, sample handling, and test sample preparation (see the relevant part of the ISO 5667 series).

An International Standard on a test method of gross alpha and gross beta activity concentrations in water samples is justified for test laboratories carrying out these measurements, required sometimes by national authorities, as laboratories might have to obtain a specific accreditation for radionuclide measurement in drinking water samples.

This document is one of a set of International Standards on test methods dealing with the measurement of the activity concentration of radionuclides in water samples.

Water quality — Gross alpha and gross beta activity — Test method using thin source deposit

WARNING — Persons using this document should be familiar with normal laboratory practice. This document does not purport to address all of the safety problems, if any, associated with its use. It is the responsibility of the user to establish appropriate safety and health practices.

IMPORTANT — It is absolutely essential that tests conducted according to this document be carried out by suitably trained staff.

1 Scope

This document specifies a method for the determination of gross alpha and gross beta activity concentration for alpha- and beta-emitting radionuclides. Gross alpha and gross beta activity measurement is not intended to give an absolute determination of the activity concentration of all alpha and beta emitting radionuclides in a test sample, but is a screening analysis to ensure particular reference levels of specific alpha and beta emitters have not been exceeded. This type of determination is also known as gross alpha and gross beta index. Gross alpha and gross beta analysis is not expected to be as accurate nor as precise as specific radionuclide analysis after radiochemical separations.

Maximum beta energies of approximately 0,1 MeV or higher are well measured. It is possible that low energy beta emitters can not detected (e.g. ^{3}H, ^{55}Fe, ^{241}Pu) or can only be partially detected (e.g. ^{14}C, ^{35}S, ^{63}Ni, ^{210}Pb, ^{228}Ra).

The method covers non-volatile radionuclides, since some gaseous or volatile radionuclides (e.g. radon and radioiodine) can be lost during the source preparation.

The method is applicable to test samples of drinking water, rainwater, surface and ground water as well as cooling water, industrial water, domestic and industrial wastewater after proper sampling, sample handling, and test sample preparation (filtration when necessary and taking into account the amount of dissolved material in the water).

The method described in this document is applicable in the event of an emergency situation, because the results can be obtained in less than 1 h. Detection limits reached for gross alpha and gross beta are less than 10 Bq/l and 20 Bq/l respectively. The evaporation of 10 ml sample is carried out in 20 min followed by 10 min counting with window-proportional counters.

It is the laboratory's responsibility to ensure the suitability of this test method for the water samples tested.

2 Normative references

The following documents are referred to in the text in such a way that some or all of their content constitutes requirements of this document. For dated references, only the edition cited applies. For undated references, the latest edition of the referenced document (including any amendments) applies.

ISO 3696, *Water for analytical laboratory use — Specification and test methods*

ISO 5667-1, *Water quality — Sampling — Part 1: Guidance on the design of sampling programmes and sampling techniques*

ISO 5667-3, *Water quality — Sampling — Part 3: Preservation and handling of water samples*

ISO 11929, *Determination of the characteristic limits (decision threshold, detection limit and limits of the confidence interval) for measurements of ionizing radiation — Fundamentals and application*

ISO 80000-10, *Quantities and units — Part 10: Atomic and nuclear physics*

ISO/IEC 17025, *General requirements for the competence of testing and calibration laboratories*

ISO/IEC Guide 98-3, *Uncertainty of measurement — Part 3: Guide to the expression of uncertainty in measurement (GUM:1995)*

3 Terms, definitions and symbols

For the purposes of this document, the terms, definitions and symbols given in ISO 80000-10 and the following apply.

ISO and IEC maintain terminological databases for use in standardization at the following addresses:

— ISO Online browsing platform: available at https://www.iso.org/obp

— IEC Electropedia: available at http://www.electropedia.org/

A	activity of the calibration source	Bq
A_a	activity spiked in sample a, prepared for self-absorption estimation purposes	Bq
c_A	activity concentration	Bq l^{-1}
c_A^*	decision threshold	Bq l^{-1}
$c_A^\#$	detection limit	Bq l^{-1}
$c_A^\triangleleft, c_A^\triangleright$	lower and upper limits of the confidence interval	Bq l^{-1}
$f_{a\alpha}, f_{a\beta}$	self-absorption factor of sample a for α and β, respectively	—
m_d	mass of the deposit	mg
m_p	mass of the planchet	mg
m_{pd}	mass of the planchet and the deposit	mg
m_{pf}	mass of the planchet and the filter	mg
m_{pfd}	mass of the planchet, the filter and the deposit	mg
$r_{0\alpha}$, $r_{0\beta}$	background count rate from the α and β windows, respectively	s^{-1}
$r_{a\alpha}$, $r_{a\beta}$	self-absorption sample a count rate from the α and β windows, respectively	s^{-1}
$r_{g\alpha}$, $r_{g\beta}$	sample gross count rate from the α and β windows, respectively	s^{-1}
$r_{s\alpha}$, $r_{s\beta}$	calibration count rate from the α and β windows, respectively	s^{-1}
t_0	background counting time	s
t_g	sample counting time	s
t_s	calibration counting time	s
U	expanded uncertainty calculated by $U = k \cdot u(c_A)$ with $k = 1, 2, \ldots$	Bq l^{-1}

$u(c_A)$	standard uncertainty associated with the measurement result	Bq l^{-1}
V	volume of test sample	l
ε_α, ε_β	counting efficiency for α and β, respectively	—
$\varepsilon_{a\alpha}$, $\varepsilon_{a\beta}$	counting efficiency of sample a for α and β, respectively	—
χ	alpha-beta crosstalk	—

4 Principle

The gross alpha and gross beta activity of the deposit is measured by counting in an alpha- and beta-particle detector or counting system previously calibrated against alpha- and beta-emitting standards. In order to obtain a thin and homogeneous deposit directly on a planchet, the sample can be progressively evaporated to dryness at a temperature below about 85 °C. Alternatively, for the gross alpha determination, radionuclides can be concentrated via a co-precipitation, the filtered co-precipitate deposited on the planchet being measured[8].

When suspended matter is present, filtration through 0,45 µm filter media is required and the gross alpha and gross beta activity can also be determined for the material retained on the filter.

IMPORTANT — Gross alpha and gross beta determinations are not absolute determinations of the sample alpha and beta radioactive contents, but relative determinations referenced to specific alpha and beta emitters that constitute the standard calibration sources.

5 Chemical reagents and equipment

5.1 Reagents

5.1.1 General

All reagents shall be of recognized analytical grade and shall not contain any detectable alpha and beta activity, except for radioactive standards solutions.

5.1.2 Standard solutions

5.1.2.1 Alpha standard.

The choice of alpha standard depends on the knowledge of the type of radioactive contaminant likely to be present in the waters being tested. In general, this leads to a choice between naturally occurring and man-made alpha emitters.

Commonly used standards of artificial alpha-emitting radionuclides employed for this purpose are ^{241}Am solutions and ^{239}Pu solutions. When ^{239}Pu is used, the presence of ^{241}Pu as an impurity shall be taken into account as it leads to growth of ^{241}Am in prepared standard solutions of sources. When ^{241}Am is used, take into account the interferences of its x and γ emission.

NOTE A uranium compound of certified natural or known isotopic composition has one arguable advantage, in that its specific activity can be calculated from established physical constants and isotopic abundance date which are independent of the calibration procedures of a particular organization. However, a uranium compound of known isotopic composition is difficult to obtain. Furthermore, since the energies of the alpha emissions from uranium isotopes are less than those from the artificial transuranic nuclides, the use of a uranium standard tends to give a high result for transuranic elements.

5.1.2.2 Beta standard.

The choice of beta standard depends on knowledge of the type of radioactive contaminant likely to be present in the waters being tested.

As a natural material, ^{40}K as potassium chloride, dried to constant mass at 105 °C, can be used. Standard solutions of artificial beta-emitting radionuclides $^{90}Sr/Y$ in equilibrium or ^{137}Cs are commonly used.

5.1.3 Wetting or surfactant agents

5.1.3.1 Vinyl acetate.

5.1.4 Volatile organic solvents

5.1.4.1 Ethyl alcohol.

5.1.5 Water

5.1.5.1 Water, complying with the requirements of ISO 3696, grade 3.

5.1.6 Specific reagents for alpha-emitting radionuclides co-precipitation

5.1.6.1 Ammonium hydroxide solution, $c(NH_4OH)$ = 6 mol/l.

5.1.6.2 Nitric acid, concentrated, $c(HNO_3)$ = 15,8 mol/l.

5.1.6.3 Sulfuric acid solution, $c(H_2SO_4)$ = 1 mol/l.

5.1.6.4 Iron carrier, solution of 5 mg of iron per millilitre.

5.1.6.5 Barium carrier, solution of 5 mg of barium per millilitre.

5.2 Equipment

5.2.1 Laboratory equipment for direct evaporation

Usual laboratory apparatus to store and prepare the sample as specified in ISO 5667-3.

A hot plate, an automatic evaporator or any other appropriate apparatus.

5.2.2 General equipment

5.2.2.1 Filters, of pore size 0,45 µm.

5.2.2.2 Planchet (counting trays).

The planchet shall be lipped and of stainless steel. The diameter of the planchet is determined taking account of the detector diameter and source holder dimensions of the counter used. In the specific case of co-precipitation, an annular support is used to fix the filter on to a filter holder or on to the planchet.

As the source, test portion and standard, is spread directly on to the planchet for evaporation, it is easier to produce an even deposit on a roughened metal surface; sand blasting or chemical etching can be applied for this purpose, alternatively, a rippled planchet can be used.

5.2.3 Special equipment for alpha-emitting radionuclide co-precipitation

5.2.3.1 Hot plate with stirring equipment.

5.2.3.2 Infrared lamp.

5.2.3.3 Vacuum filtration system.

5.2.3.4 Filters, of pore size 0,45 µm.

5.2.4 Measurement equipment

5.2.4.1 Alpha-beta counter.

Gross alpha and gross beta activity can be measured using either a silicon surface barrier (SSB) detector or a proportional counter (windowless). Ion-implanted Si detectors and window-proportional counters (between 80 µg cm^{-2} to 400 µg cm^{-2}) may also be used. Gross alpha and gross beta activity can also be counted using a silver-activated zinc sulfide scintillation screen and plastic scintillation detector, respectively.

6 Sampling

Sample, handle and store water samples in accordance with ISO 5667-1 and ISO 5667-3. Additional information on sampling of different types of waters can be found in the relative parts of the ISO 5667 series[9][10][11][12][13][14][15][16].

The laboratory sample is not usually acidified as the test portion is directly evaporated on the planchet. Acidification minimizes the loss of radioactive material from solution by adsorption on the wall of the vial, but is done after filtration, as otherwise it desorbs radioactive material already adsorbed on the particulate material and, also, increases the salt content of the test sample, and thus the thickness of the deposit. If necessary, concentrated nitric acid can be used (it is recommended to avoid hydrochloric acid).

7 Procedure

7.1 Preliminary

Calculate the volume of laboratory sample for gross alpha measurement, i.e. the volume of the test portion, in order to produce a deposit with a surface density lower than 5 mg cm^{-2} on the planchet. For deposits of surface density below 5 mg cm^{-2}, self-absorption phenomena can be neglected for gross beta measurement except when using low energy beta emitter such as ^{137}Cs for calibration[17].

When using the same deposit for the simultaneous gross alpha and gross beta measurement, the planchet surface density limit for alpha activity determinations applies.

7.2 Source preparation

IMPORTANT — Due to the ingrowth of radon decay products over time, the results are dependent on the time elapsed between sample preparation and measurement. For comparison purposes, it is recommended that the measurement be performed at the same time after the preparation of the sample.

7.2.1 Preparation of planchet

Degrease planchets (5.2.2.2) using a solvent or a surfactant to ensure that the test portion is well distributed over the entire surface and consequently that there is a deposit of uniform surface density

bonded to the planchets. Some suppliers degrease planchets at the end of a cycle of fabrication and deliver, on demand, a certificate of attestation.

Keep planchets that are not to be used immediately in a dessicator to prevent any modification by ambient atmosphere in the laboratory.

Weigh the planchets before use, and record the mass, m_p. If a co-precipitation method is used, weigh the filter (5.2.2.1) with the planchet before use, and record the mass, m_{pf}.

Avoid reuse of planchets to minimize cross-contamination. If the planchets are reused, their freedom from contamination shall be demonstrated.

7.2.2 Evaporation

Transfer the test portion on to the planchet using automatic or non-automatic equipment with a known uncertainty (pipette, water distribution system) and carefully evaporate to dryness.

The residue deposited should form a thin layer of uniform surface density to limit self-absorption phenomena and to ensure similarity with the calibration source geometry.

After cooling the planchets to ambient temperature, weigh them and record the mass, m_{pd}. The mass deposited, m_d, is given by Formula (1):

$$m_d = m_{pd} - m_p \quad (1)$$

If hygroscopic salt deposit is expected the planchet can also be weight at the end of the measurement.

To minimize any loss by spitting, maintain the temperature below about 85 °C over the entire planchet surface to avoid any overheated areas.

Before evaporating the test portion to dryness on the planchet, pre-evaporation can be performed with appropriate equipment (5.2.1).

A homogeneous deposit is best achieved on etched or sandblasted planchets. If the deposit is not homogeneously spread, add a wetting agent or surfactant (5.1.3).

7.2.3 Co-precipitation

The recommended working volume is 500 ml.

If a test portion of lower volume is to be analysed, make up to 500 ml with water.

If a test portion of higher volume is to be analysed, concentrate it by evaporation (5.2.1) to 500 ml.

Adjust the pH of the working volume to 7,0 ± 0,5.

Add 20 ml of sulfuric acid (5.1.6.3) and boil for 5 min on a hot plate while stirring.

At a temperature of approximately 50 °C, add 1 ml of the barium carrier solution (5.1.6.5) and stir for 30 min.

The barium sulfate precipitates.

Then add 1 ml of the iron carrier solution (5.1.6.4).

Adjust the pH with ammonium hydroxide, (5.1.6.1) drop by drop until iron(III) hydroxide precipitates.

Continue stirring for 30 min.

Filter (5.2.3.4) the co-precipitates.

Place the filter on to the identified planchet and fix it by an annular support to avoid deformation while drying.

Dry at moderate temperature.

After cooling the planchet and the filter to ambient temperature, weigh them and record their mass, m_{pfd}. Determine the mass deposited, m_d, using Formula (2):

$$m_d = m_{pfd} - m_{pf} \tag{2}$$

NOTE Radium, polonium and actinides co-precipitate quantitatively with barium sulfate or iron(III) hydroxide[7].

7.3 Counting stage

Following evaporation (7.2.2) or co-precipitation (7.2.3), if the counting is not performed immediately the planchet with the deposit can be stored in a desiccator.

The measurement of the residue activity on the planchet is performed by counting for an appropriate duration to reach the required characteristic limits and depending on the test portion and background count rates.

The counting strategy depends on the objectives of the measurements and the regulator requirements. To monitor natural radionuclides ingrowth or decay (see 9.11), counting procedures should be repeated periodically over a period of one month. If specific counting condition is applied, it is recommended to mention it in the report.

When the counting strategy is defined, the laboratory shall apply it systematically for comparison purposes.

7.4 Background and blank determination

Measure the background activity using a clean planchet (5.2.2.2) under conditions representative of the measurement method. Repeated counts confirm the stability of the background level.

If reagents are used, measure the blank activity using a clean planchet and reagents under conditions representative of the measurement method. Repeated counts confirm the stability of the blank level.

7.5 Preparation of counting standard for calibration

Prepare a geometry- and matrix-matched calibration source [planchet (5.2.2.2) or filter (5.2.3.4) with precipitates and annular attachment to the planchet] to closely mimic the procedure applied to test portions in order to obtain the same retro-diffusion effect.

Add an accurately known amount (about 5 Bq to 10 Bq) of a standard solution (5.1.2.1) to the starting volume of water and use the same source preparation procedure or directly add the standard solution to the planchet.

These standard sources are measured in the detector with a counting duration that leads to a counting uncertainty of 1 % (more than 10 000 counts should be recorded).

The counting efficiency is calculated by dividing the net count rate (test portion minus background) by the activity of the calibration source, as shown by Formula (3):

$$\varepsilon_\alpha = \frac{r_{s\alpha} - r_{0\alpha}}{A} \tag{3}$$

A commercially available certified calibration source may be used.

7.6 Preparation of calibration source for self-absorption determination

7.6.1 General

As alpha-particle counting efficiency is directly dependent on the source thickness, estimate the self-absorption factor, defined by Formula (4):

$$f_{a\alpha} = \frac{\varepsilon_{a\alpha}}{\varepsilon_{\alpha}} \tag{4}$$

Two possibilities are given:

— spiking a single sample (7.6.2);

— spiking a set of samples of increasing mass to plot a self-absorption curve.

For beta particle counting efficiency, self-absorption phenomena are negligible $\varepsilon_{\beta} = \varepsilon_{a\beta}$ with ($f_{a\beta} = 1$).

7.6.2 Spiking one of two test portions

Two test portions from the same laboratory sample are prepared under the same conditions with one test portion spiked with a known activity of the standard solution.

The specific efficiency is calculated by dividing the net count rate (spiked test portion minus unspiked test portion) by the activity of sample a Formula (5):

$$\varepsilon_{a\alpha} = \frac{r_{a\alpha} - r_{g\alpha}}{A_a} \tag{5}$$

7.6.3 Self-absorption curve

7.6.3.1 Alpha self-absorption correction

A set of calibration sources with the same standard activity and increasing mass is prepared with the same sample preparation procedure to allow the determination of a series of absorption factors, $f_{a\alpha}$. A function relating the absorption factor to the mass can be derived from the measurement data. Several mathematical models are proposed to describe self-absorption phenomena such as linear, hyperbolic, exponential, polynomial, power laws[18] to [24]. Commonly used curve-fitting techniques can be employed to derive the function and associated uncertainty.

The use of gross alpha self-absorption curves prepared with salts that mimic as closely as possible the salt content and salt nature of the test sample, improves the accuracy of results [25][26]. That consideration apply typically for unknown sample, the evaluation of the self-absorption by the spiking method is recommended.

WARNING — This procedure requires the preparation of a dry deposit spiked with alpha-emitting radionuclides. Avoid accidental ingestion of radioactive particulates by use of a sample closed cabinet glove box for the preparation of the spiked sources. Alternatively, a laboratory fume cupboard may be used, provided that the extract draught is not excessive and liable to create disturbance or carry fine powder particles into the air.

7.6.3.2 Beta self-absorption correction

Beta self-absorption phenomena can be neglected for gross beta measurement when deposits of surface density is below 5 mg cm^{-2} except when using low energy beta emitter such as ^{137}Cs for calibration.

8 Expression of results

8.1 General

An example of parameter values obtained for a specific test that can be used to check the computation results obtained with the different formulae of this document is given in Table A.1 of Annex A.

8.2 Alpha activity concentration

The alpha activity concentration, c_A, can be obtained using Formula (6):

$$c_A = \frac{r_{g\alpha} - r_{0\alpha}}{V\,\varepsilon_\alpha\,f_{a\alpha}} = (r_{g\alpha} - r_{0\alpha})w \tag{6}$$

with

$$w = \frac{1}{V\,\varepsilon_\alpha\,f_{a\alpha}}$$

It is advisable to choose the alpha and beta windows to reduce the alpha-beta crosstalk to a minimum.

8.3 Beta activity concentration

The beta activity concentration, c_A, can be obtained using Formula (7):

$$c_A = \frac{r_{g\beta} - r_{0\beta} - \chi(r_{g\alpha} - r_{0\alpha})}{V\,\varepsilon_\beta\,f_{a\beta}} = \left[r_{g\beta} - r_{0\beta} - \chi(r_{g\alpha} - r_{0\alpha})\right]w \tag{7}$$

with

$$w = \frac{1}{V\,\varepsilon_\beta}$$

and

$$f_{a\beta} = 1$$

Take any alpha-beta crosstalk into account, in order to derive the correct beta count rate. If needed, the alpha-beta crosstalk correction factor, χ, can be calculated using Formula (8):

$$\chi = \frac{r_{s\alpha\to\beta}}{r_{s\alpha}} \tag{8}$$

where $r_{s\alpha\to\beta}$ is the count rate in the beta window when the alpha calibration source is measured.

Because the alpha-beta crosstalk is specific to the alpha standard used for the calibration, the systematic correction is not required, the beta activity concentration, c_A, can be obtained using Formula (9):

$$c_A = \frac{r_{g\beta} - r_{0\beta}}{V\,\varepsilon_\beta} = \left[r_{g\beta} - r_{0\beta}\right]w \tag{9}$$

If the laboratory neglect alpha-beta crosstalk, it has nevertheless to be aware of the orders of magnitude.

8.4 Standard uncertainty of the alpha activity concentration

As specified in ISO/IEC Guide 98-3 (see also ISO 11929), the standard uncertainty of c_A is calculated using Formula (10):

$$u(c_A)=\sqrt{w^2\left[u^2(r_{g\alpha})+u^2(r_{0\alpha})\right]+c_A{}^2\,u_{rel}^2(w)}=\sqrt{w^2\left(\frac{r_{g\alpha}}{t_g}+\frac{r_{0\alpha}}{t_0}\right)+c_A{}^2\,u_{rel}^2(w)} \tag{10}$$

As the uncertainty of the counting time and the co-precipitation stage are negligible in comparison with other sources of uncertainty[7], it can be neglected and the relative standard uncertainty of w can be calculated using Formula (11):

$$u_{rel}^2(w)=u_{rel}^2(\varepsilon_\alpha)+u_{rel}^2(V)+u_{rel}^2(f_{a\alpha}) \tag{11}$$

and the relative standard uncertainty of ε_α is calculated using Formula (12):

$$u_{rel}^2(\varepsilon_\alpha)=u_{rel}^2(r_{s\alpha}-r_{0\alpha})+u_{rel}^2(A_\alpha)=\frac{(r_{s\alpha}/t_s)+(r_{0\alpha}/t_0)}{(r_{s\alpha}-r_{0\alpha})^2}+u_{rel}^2(A_\alpha) \tag{12}$$

All the uncertainties related to the calibration source are included in $u_{rel}^2(A_\alpha)$, i.e. those of the standard solution and the preparation of the calibration source.

For the calculation of the detection limits, the term $\tilde{u}(\tilde{c}_A)$ is required (as specified in ISO 11929); the standard uncertainty of c_A as a function of its true value, calculated using Formula (13):

$$u(\tilde{c}_A)=\sqrt{w^2\left[\frac{\tilde{c}_A/w+r_{0\alpha}}{t_g}+\frac{r_{0\alpha}}{t_0}\right]+\tilde{c}_A^{\,2}\,u_{rel}^2(w)} \tag{13}$$

8.5 Standard uncertainty of the beta activity concentration

As specified in the ISO/IEC Guide 98-3 (see also ISO 11929), the standard uncertainty of c_A is calculated using Formula (14) to (16):

$$u(c_A)=\sqrt{w^2\left[u^2(r_{g\beta})+u^2(r_{0\beta})+T(\chi)\right]+c_A{}^2\,u_{rel}^2(w)}=\sqrt{w^2\left[\frac{r_{g\beta}}{t_g}+\frac{r_{0\beta}}{t_0}+T(\chi)\right]+c_A{}^2\,u_{rel}^2(w)} \tag{14}$$

with

$$T(\chi)=\left(r_{g\alpha}-r_{0\alpha}\right)^2u^2(\chi)+\chi^2\left(\frac{r_{g\alpha}}{t_g}+\frac{r_{0\alpha}}{t_0}\right) \tag{15}$$

and

$$u(\chi)=\sqrt{\frac{\chi(\chi+1)}{r_{s\alpha}\,t_{s\alpha}}} \tag{16}$$

As the uncertainty of the counting time is small in comparison to other sources of uncertainty, it can be neglected and the relative standard uncertainty of w is calculated using Formula (17):

$$u_{\mathrm{rel}}^2(w) = u_{\mathrm{rel}}^2(\varepsilon_\beta) + u_{\mathrm{rel}}^2(V) \tag{17}$$

and the relative standard uncertainty of ε_β is calculated using Formula (18):

$$u_{\mathrm{rel}}^2(\varepsilon_\beta) = u_{\mathrm{rel}}^2(r_{s\beta} - r_{0\beta}) + u_{\mathrm{rel}}^2(A_\beta) = \frac{(r_{s\beta}/t_s) + (r_{0\beta}/t_0)}{(r_{s\beta} - r_{0\beta})^2} + u_{\mathrm{rel}}^2(A_\beta) \tag{18}$$

All the uncertainties related to the calibration source are included in $u_{\mathrm{rel}}^2(A_\beta)$, i.e. those of the standard solution and the preparation of the calibration source.

For the calculation of the detection limits, the term $\tilde{u}(\tilde{c}_A)$ is required (as specified in ISO 11929); the standard uncertainty of c_A as a function of its true value, calculated by Formula (19):

$$\tilde{u}(\tilde{c}_A) = \sqrt{w^2\left[\frac{\tilde{c}_A/w + \chi(r_{g\alpha} - r_{0\alpha}) + r_{0\beta}}{t_g} + \frac{r_{0\beta}}{t_0}\right] + \tilde{c}_A^2\, u_{\mathrm{rel}}^2(w)} \tag{19}$$

If the alpha-beta crosstalk is neglected, as specified in ISO/IEC Guide 98-3 (see also ISO 11929), the standard uncertainty of c_A is calculated using Formula (20):

$$u(c_A) = \sqrt{w^2\left[u^2(r_{g\beta}) + u^2(r_{0\beta})\right] + c_A^2\, u_{\mathrm{rel}}^2(w)} = \sqrt{w^2\left[\frac{r_{g\beta}}{t_g} + \frac{r_{0\beta}}{t_0}\right] + c_A^2\, u_{\mathrm{rel}}^2(w)} \tag{20}$$

As the uncertainty of the counting time and the co-precipitation stage are negligible in comparison with other sources of uncertainty[7] it can be neglected and the relative standard uncertainty of w can be calculated using Formula (21):

$$u_{\mathrm{rel}}^2(w) = u_{\mathrm{rel}}^2(w\varepsilon_\beta) + u_{\mathrm{rel}}^2(V) \tag{21}$$

and the relative standard uncertainty of ε_α is calculated using Formula (22):

$$u_{\mathrm{rel}}^2(\varepsilon_\beta) = u_{\mathrm{rel}}^2(r_{s\beta} - r_{0\beta}) + u_{\mathrm{rel}}^2(A_\beta) = \frac{(r_{s\beta}/t_s) + (r_{0\beta}/t_0)}{(r_{s\beta} - r_{0\beta})^2} + u_{\mathrm{rel}}^2(A_\beta) \tag{22}$$

All the uncertainties related to the calibration source are included in $u_{\mathrm{rel}}^2(A_\beta)$, i.e. those of the standard solution and the preparation of the calibration source.

For the calculation of the detection limits, the term $u(\tilde{c}_A)$ is required (as specified in ISO 11929); the standard uncertainty of c_A as a function of its true value, calculated using Formula (23):

$$\tilde{u}(\tilde{c}_A) = \sqrt{w^2\left[\frac{\tilde{c}_A/w + r_{0\beta}}{t_g} + \frac{r_{0\beta}}{t_0}\right] + \tilde{c}_A^2\, u_{\mathrm{rel}}^2(w)} \tag{23}$$

8.6 Decision threshold

8.6.1 Decision threshold of the alpha activity concentration

The decision threshold, c_A^*, is obtained from Formula (13) for $\tilde{c}_A = 0$ (see ISO 11929):

$$c_A^* = k_{1-\alpha}\, \tilde{u}(0) = k_{1-\alpha}\, w \sqrt{\frac{r_{0\alpha}}{t_g} + \frac{r_{0\alpha}}{t_0}} \tag{24}$$

$\alpha = 0{,}05$ with $k_{1-\alpha} = 1{,}65$ are often chosen by default.

8.6.2 Decision threshold of the beta activity concentration

The decision threshold, c_A^*, is obtained from Formula (19) for $\tilde{c}_A = 0$ (see ISO 11929). This yields:

$$c_A^* = k_{1-\alpha}\, \tilde{u}(0) = k_{1-\alpha}\, w \sqrt{\frac{\chi(r_{g\alpha} - r_{0\alpha}) + r_{0\beta}}{t_g} + \frac{r_{0\beta}}{t_0} + \mathrm{T}(\chi)} \tag{25}$$

$\alpha = 0{,}05$ with $k_{1-\alpha} = 1{,}65$ are often chosen by default.

If the alpha-beta crosstalk is neglected, the decision threshold, c_A^*, is obtained from Formula (23) for $\tilde{c}_A = 0$ (see ISO 11929):

$$c_A^* = k_{1-\alpha}\, \tilde{u}(0) = k_{1-\alpha}\, w \sqrt{\frac{r_{0\beta}}{t_g} + \frac{r_{0\beta}}{t_0}} \tag{26}$$

$\alpha = 0{,}05$ with $k_{1-\alpha} = 1{,}65$ are often chosen by default.

8.7 Detection limit

8.7.1 Detection limit of the alpha activity concentration

The detection limit, $c_A^\#$, is calculated using Formula (27) (see ISO 11929):

$$c_A^\# = c_A^* + k_{1-\beta}\, \tilde{u}(c_A^\#) = c_A^* + k_{1-\beta} \sqrt{w^2 \left[\frac{c_A^\# / w + r_{0\alpha}}{t_g} + \frac{r_{0\alpha}}{t_0} \right] + c_A^{\#2}\, u_{\mathrm{rel}}^2(w)} \tag{27}$$

$\beta = 0{,}05$ with $k_{1-\beta} = 1{,}65$ are often chosen by default.

The detection limit can be calculated by solving Formula (27) for $c_A^\#$ or, more simply, by iteration with a starting approximation $c_A^\# = 2\, c_A^*$.

When taking $\alpha = \beta$ then $k_{1-\alpha} = k_{1-\beta} = k$ and the solution of Formula (27) is given by Formula (28):

$$c_A^\# = \frac{2\, c_A^* + (k^2 w / t_g)}{1 - k^2\, u_{\mathrm{rel}}^2(w)} \tag{28}$$

8.7.2 Detection limit of the beta activity concentration

The detection limit, $c_A^{\#}$, is calculated using Formula (29) (see ISO 11929):

$$c_A^{\#} = c_A^{*} + k_{1-\beta}\,\tilde{u}(c_A^{\#}) = c_A^{*} + k_{1-\beta}\sqrt{w^2\left[\frac{c_A^{\#}/w + \chi\left(r_{g\alpha} - r_{0\alpha}\right) + r_{0\beta}}{t_g} + \frac{r_{0\beta}}{t_0} + T(\chi)\right] + c_A^{\#2}\,u_{rel}^{2}(w)} \quad (29)$$

β = 0,05 with $k_{1-\beta}$ = 1,65 are often chosen by default.

If the alpha-beta crosstalk is neglected, the detection limit, $c_A^{\#}$, is calculated using Formula (30) (see ISO 11929):

$$c_A^{\#} = c_A^{*} + k_{1-\beta}\,\tilde{u}(c_A^{\#}) = c_A^{*} + k_{1-\beta}\sqrt{w^2\left[\frac{c_A^{\#}/w + r_{0\beta}}{t_g} + \frac{r_{0\beta}}{t_0}\right] + c_A^{\#2}\,u_{rel}^{2}(w)} \quad (30)$$

The detection limit can be calculated by solving Formula (29) or (30) for $c_A^{\#}$ or, more simply, by iteration with a starting approximation $c_A^{\#} = 2c_A^{*}$.

When taking $\alpha = \beta$ then $k_{1-\alpha} = k_{1-\beta} = k$ and the solution of Formula (29) and (30) is given by Formula (31):

$$c_A^{\#} = \frac{2c_A^{*} + (k^2\,w/t_g)}{1 - k^2\,u_{rel}^{2}(w)} \quad (31)$$

8.8 Confidence limits

The lower, $c_A^{\triangleleft}$, and upper, $c_A^{\triangleright}$, confidence limits are calculated using Formulae (32) and (33) (as specified in ISO 11929):

$$c_A^{\triangleleft} = c_A - k_p\,u(c_A)\;p = \omega\left(1 - \frac{\gamma}{2}\right) \quad (32)$$

$$c_A^{\triangleright} = c_A + k_q\,u(c_A)\;q = 1 - \frac{\omega\,\gamma}{2} \quad (33)$$

where $\omega = \Phi[y/u(y)]$ in which Φ is the distribution function of the standardized normal distribution.

Set ω = 1 if c_A W 4 $u(c_A)$. In this case:

$$c_A^{\triangleleft\triangleright} = c_A \pm k_{1-(\gamma/2)}\,u(c_A) \quad (34)$$

γ = 0,05 with $k_{1-(\gamma/2)}$ = 1,96 are often chosen by default.

9 Control of interferences

9.1 General

The problems discussed below should be recognized as inherent interferences and limitations of the method.

9.2 Relative humidity

If the evaporated sample contains significant levels of hygroscopic material, then this may absorb moisture from the atmosphere, which increases the counting source mass and this, in turn, leads to inconsistent detection efficiencies for the source. Although the use of a desiccator may reduce this effect, the sample may still absorb significant amounts of moisture when it is removed from the desiccator and placed in the sample measurement system. The sample mass before and after the counting can be recorded.

To deal with this issue, several strategies are available. One is to spike the sample with internal standard and to count spiked and non-spiked planchets successively with the same detector; this method is reliant on the relative humidity remaining stable. Alternatively, both planchets may be counted using two detectors in parallel.

Additionally, the sample deposit may be stabilized by treating it with a polymer solution such as collodion or poly(vinyl acetate).

If the techniques described in ISO 9696 and ISO 9697 are used, then the hygroscopic deposit is transformed into a hydrophobic deposit by the sulfation stage. ISO 11704 which is based on liquid scintillation counting, may also be used.

Whatever technique is used, the efficiency and crosstalk determined should be based on the sample configuration.

The counterpoint, dry samples that have obtained a static charge, may ionize the carrier gas in the gas proportional counting system yielding increased counts above the level of radioactivity that is actually present. Accumulation of static charge may occur when conditions of low humidity (<60 % relative humidity) exist in the laboratory. If such conditions exist, the static charge may be successfully removed by connecting a small wire to a grounded metal structure and touching the planchet with the wire before counting.

A usual method[27] suggested to convert salts to oxides is by heating the sample intensely until it glows with a characteristic dull-red colour. This process is not recommended due to the potential radionuclides losses such as ^{137}Cs.

9.3 Geometry of the deposit

Non-uniformity of the sample residue in counting planchet interferes with the accuracy and precision of the method. The requirements also apply for calibration using self-absorption mass curves. For instance, sources made with calcium carbonate or sodium sulfate can induce homogeneous or non-homogenous deposit, respectively[28][29].

9.4 Crosstalk

In general, the determination of alpha activity in the presence of beta activity is not subject to significant interference. Although crosstalk cannot be eliminated when counting on the beta plateau, beta to alpha crosstalk can be minimized during set-up of the instrument by adjusting the alpha and beta discriminators. While minimizing crosstalk also minimizes corrections needed for most samples, it should be kept in mind that if there is a significant difference in alpha and beta activities significant crosstalk may still be present. In cases where unusually high ratios of beta activity to alpha activity are present in samples, determining the alpha activity of samples in a separate measurement from the beta by counting on the alpha plateau provides the most effective discrimination against beta-to-alpha crosstalk and the most accurate measurement of alpha activity in samples.

The issue of alpha to beta crosstalk is different. Decay of alpha emitting radionuclides is often accompanied by the emission of conversion electrons, auger electrons and X-rays, which produce pulses indistinguishable from beta pulses, thus causing significant alpha to beta crosstalk[30]. Consequently, the crosstalk correction is only appropriate if the radionuclides assayed are exactly the same as used for calibration, otherwise an unpredictable bias can be expected for gross beta determination.

When the correction strategy is defined the laboratory shall apply it systematically for comparison purposes.

9.5 Gamma emitters

Moderate to high levels of gamma radiation can be misinterpreted as alpha or beta activity by gas-flow proportional counters. This interference can be minimized by using a guard detector and employing anti-coincident counting techniques.

9.6 Low beta energy

Proportional counters operated in the "windowed" mode configuration are limited to detecting (beta) particles with an energy of 0,1 MeV or greater. To detect "soft" betas, the counter needs to be operated in the "windowless" mode configuration.

9.7 Chlorides

If the sample is known or suspected to contain significant chloride (acid or salts), the chloride should be converted to nitrate before transferring to a stainless steel planchet. (Chlorides attack the stainless steel, depositing heavy metal ions on top of the sample, thereby increasing inelastic scattering. Subsequently, no correction can be made for this effect[27]).

9.8 Organic matter

If the sample contains organic matter, the evaporation residue can be inappropriate for counting. The sample can be treated with acids to oxidize organic matter[27]. The additional step may induce losses of ^{14}C. Another solution is to use liquid scintillation counter (see ISO 11704[31]).

9.9 Contamination

Check the contamination of reagents by evaporating the volumes of the reagents used in the procedures on to separate planchets. Ensure that the activities are negligible compared with the activity of the test sample.

If the activity is not negligible, select reagents with lower radioactivity, or include a blank determination in the main procedure.

9.10 Losses of activity

Volatile radioisotopes of C, ^{3}H, I, Po, and adsorbed Rn may be lost when samples are heated.

9.11 Contribution of the natural radionuclides

The most common natural radiological contaminants found in samples are the uranium isotopes (^{234}U, ^{235}U, and ^{238}U), the radium isotopes (^{224}Ra, ^{226}Ra, ^{228}Ra), lead isotope ^{210}Pb and polonium isotope ^{210}Po.

Isotopes of radon can be lost by volatilization during the evaporation step. Once the residue forms, ^{222}Rn and ^{220}Rn produced by ^{226}Ra and ^{224}Ra respectively are trapped in the residue[32][33].

The gross alpha and gross beta activity are differently affected.

— The contribution of ^{224}Ra decay products to the gross alpha activity decreases with the time between collection and analysis and becomes negligible (less than 2,5 %) at 21 days.

— The contribution of the ^{228}Ra decay products to the gross alpha activity increases with the time between collection and analysis and can become significant (9 %) after three months.

— The contribution of the ^{226}Ra decay products to the gross alpha activity increases with the time between preparation and analysis, becomes more than 16 % after one day and reaches 95 % after 16 days. It can show an ingrowth of four times the initial count.

— The contribution of the ^{210}Pb decay product ^{210}Po to the gross alpha activity increases with the time between collection and analysis, becomes more than 11 % after one month, and reaches 50 % after 5 months.

— The contribution of the ^{238}U decay products to the gross beta activity increases with the time between preparation and analysis becomes more than 2 % after one day and reaches 95 % after 3 months days.

— The contribution of the ^{226}Ra decay products to the gross beta activity increases with the time between preparation and analysis, becomes more than 16 % after one day and reaches 95 % after 16 days.

— The contribution of ^{224}Ra decay products to the gross beta activity decreases with the time between collection and analysis and becomes negligible (less than 2,5 %) at 21 days.

— The contribution of the ^{228}Ra decay product ^{228}Ac to the gross beta activity increases rapidly with the time between collection and analysis and reaches 93 % after one day.

— The contribution of the ^{210}Pb decay product ^{210}Bi to the gross beta activity increases with the time between collection and analysis, becomes more than 13 % after one day, and reaches 95 % after 22 days.

The problem is complicated when seasonal variations of the concentration of natural radionuclides are observed in the water sampled[34].

When the correction strategy is defined, the laboratory shall keep it for comparison purposes.

9.12 Losses of activity

Volatile radioisotopes of C, ^{3}H, I, Po, and adsorbed Rn may be lost when samples are heated.

If an oxidation step is required, it may induce losses of ^{14}C.

Some alpha-emitting polonium isotopes, which occur naturally as members of the uranium and thorium decay series, may contribute to a significant proportion of the gross alpha activity of some waters. The element and some of its compounds, particularly the halides which sublime at relatively low temperatures, may be volatilized[35]. The problem can be solved by converted halide to nitrate[27].

10 Test report

The test report shall conform to the requirements of ISO/IEC 17025 and shall contain at least the following information:

a) a reference to this document, i.e. ISO 10704:2019;

b) all information necessary for the complete identification of the sample;

c) units in which the results are expressed;

d) test result, $c_A \pm u(c_A)$ or $c_A \pm U$, with the associated k value.

Complementary information can be provided, such as the following:

e) probabilities α, β and $(1 - \gamma)$;

f) decision threshold and the detection limit;