International
Standard

ISO 23548

# Measurement of radioactivity — Alpha emitting radionuclides — Generic test method using alpha spectrometry

*Mesurage de la radioactivité — Radionucléides émetteurs alpha — Méthode d'essai générique par spectrométrie alpha*

First edition
2024-09

Reference number
ISO 23548:2024(en)

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Published in Switzerland

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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 document 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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This document was prepared by Technical Committee ISO/TC 85, *Nuclear energy, nuclear technologies, and radiological protection*, Subcommittee SC 2, *Radiological protection*.

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.

# Introduction

Everyone is exposed to natural radiation. The natural sources of radiation are cosmic rays and naturally occurring radioactive substances which exist in the earth and flora and fauna, including the human body. Human activities involving the use of radiation and radioactive substances add to the radiation exposure from this natural exposure. Some of those activities, such as the mining and use of ores containing naturally-occurring radioactive materials (NORM) and the production of energy by burning coal that contains such substances, simply enhance the exposure from natural radiation sources. Nuclear power plants and other nuclear installations use radioactive materials and produce radioactive effluents and wastes during operation and decommissioning. The use of radioactive materials in industry, agriculture, medicine and research is expanding around the globe.

All these human activities give rise to radiation exposures that are only a small fraction of the global average level of natural exposure. The medical use of radiation is the largest and a growing man-made source of radiation exposure in developed countries. It includes diagnostic radiology, radiotherapy, nuclear medicine and interventional radiology.

Radiation exposure also occurs as a result of occupational activities. It is incurred by workers in industry, medicine and research using radiation or radioactive substances, as well as by passengers and crew during air travel. The average level of occupational exposures is generally similar to the global average level of natural radiation exposure (see Reference [1]).

As uses of radiation increase, so do the potential health risk and the public's concerns. Thus, all these exposures are regularly assessed in order to

a) improve the understanding of global levels and temporal trends of public and worker exposure,

b) evaluate the components of exposure so as to provide a measure of their relative importance, and

c) identify emerging issues that may warrant more attention and study.

While doses to workers are mostly directly measured, doses to the public are usually assessed by indirect methods using the results of radioactivity measurements of waste, effluent and/or environmental samples.

To ensure that the data obtained from radioactivity monitoring programs support their intended use, it is essential that the stakeholders, for example, nuclear site operators, regulatory and local authorities agree on appropriate methods and procedures for obtaining representative samples and for handling, storing, preparing and measuring the test samples. An assessment of the overall measurement uncertainty also needs to be carried out systematically. As reliable, comparable and 'fit for purpose' data are an essential requirement for any public health decision based on radioactivity measurements, international standards of tested and validated radionuclide test methods are an important tool for the production of such measurement results. The application of standards serves also to guarantee comparability of the test results over time and between different testing laboratories. Laboratories apply them to demonstrate their technical competences and to complete proficiency tests successfully during interlaboratory comparisons, two prerequisites for obtaining accreditation.

Today, over a hundred International Standards are available to testing laboratories for measuring radionuclides in different matrices.

Generic standards help testing laboratories to manage the measurement process by setting out the general requirements and methods to calibrate equipment and validate techniques. These standards underpin specific standards which describe the test methods to be performed by staff, for example, for different types of sample. The specific standards cover test methods for

- 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 and $^{210}$Pb), can be found in the environmental components for natural reasons or can be released from technological processes involving naturally occurring radioactive materials (e.g. the mining and processing of mineral sands or phosphate fertilizer production and use), and

- human-made radionuclides, such as transuranium elements (americium, plutonium, neptunium, and curium), $^{3}H$, $^{14}C$, $^{90}Sr$ and gamma-ray emitting radionuclides found in waste, liquid and gaseous effluents, in environmental matrices (water, air, soil and biota), in food and in animal feed as a result of authorized releases into the environment, fallout from the explosion in the atmosphere of nuclear devices and fallout from accidents, such as those that occurred in Chernobyl and Fukushima. Natural components and foodstuff may thus contain radionuclides at activity concentrations which could present a risk to human health. In order to assess the radiological levels of environment and food, including the quality of drinking-water (mineral waters and spring waters) with respect to its radionuclide content and to provide guidance on reducing health risks by taking measures to decrease radionuclide activity concentrations, natural resources and food are monitored for their radioactivity content as recommended by the World Health Organization (WHO) and the International Atomic Energy Agency (IAEA).

An international standard on a generic test method using alpha spectrometry for the determination of the activity concentration of alpha emitting radionuclides in natural and food samples or other kind of samples from nuclear facilities is justified for test laboratories carrying out these measurements, required sometimes by national authorities, as laboratories may have to obtain a specific accreditation for radionuclide measurements in natural, food or nuclear facilities samples.

Artificial alpha emitting radionuclide activity concentrations can vary according to authorized local effluent discharges from nuclear plant and environmental characteristics.

This document is one of a set of generic International Standards on measurement of radioactivity such as ISO 19361[2] and ISO 20042[3].

# Measurement of radioactivity — Alpha emitting radionuclides — Generic test method using alpha spectrometry

## 1 Scope

This document describes a generic test method for measuring alpha emitting radionuclides, for all types of samples (soil, sediment, construction material, foodstuff, water, airborne, environmental bio-indicator, human biological samples as urine, faeces etc.) by alpha spectrometry. This method can be used for any type of environmental study or monitoring of alpha emitting radionuclides activities.

If relevant, this test method requires appropriate sample pre-treatment followed by specific chemical separation of the test portion in order to obtain a thin source proper to alpha spectrometry measurement.

This test method can be used to determine the activity, specific activity or activity concentration of a sample containing alpha emitting radionuclides such as $^{210}$Po, $^{226}$Ra, $^{228}$Th, $^{229}$Th, $^{230}$Th, $^{232}$Th, $^{232}$U,$^{234}$U, $^{235}$U, $^{238}$U, $^{238}$Pu, $^{239+240}$Pu, $^{241}$Am or $^{243+244}$Cm.

This test method can be used to measure very low levels of activity, one or two orders of magnitude less than the usual natural levels of alpha emitting radionuclides. Annexes B of UNSCEAR 2000 and UNSCEAR 2008 (References [4] and [5]) give, respectively, typical natural activity concentrations for air, foods, drinking waters and, soils and building materials. The detection limit of the test method depends on the amount of the sample material analysed (mass or volume) after concentration, chemical yield, thickness of measurement source and counting time.

The quantity of the sample to be collected and analysed depends on the expected activity of the sample and the detection limit to achieve.

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

ISO 661, *Animal and vegetable fats and oils — Preparation of test sample*

ISO 707, *Milk and milk products — Guidance on sampling*

ISO 874, *Fresh fruits and vegetables — Sampling*

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

ISO 5500, *Oilseed residues — Sampling*

ISO 5538, *Milk and milk products — Sampling — Inspection by attributes*

ISO 5555, *Animal and vegetable fats and oils — Sampling*

ISO 5667 (all parts), *Water quality — Sampling*

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

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

ISO 17604, *Microbiology of the food chain — Carcass sampling for microbiological analysis*

ISO 18400 (all parts), *Soil quality — Sampling*

ISO 18589-2, *Measurement of radioactivity in the environment — Soil — Part 2: Guidance for the selection of the sampling strategy, sampling and pre-treatment of samples*

ISO 24333, *Cereals and cereal products — Sampling*

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

## 3 Terms and definitions

For the purposes of this document, the terms and definitions given in ISO 80000-10 and ISO 11929 series and the following apply.

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

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

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

**3.1**
**thin test source**
measurement source with a very thin layer deposited on a substrate containing the radionuclides of interest in order to obtain an optimal spectral resolution previously defined by the laboratory according to the expected level of activity

**3.2**
**substrate**
medium containing the source to be measured on its surface e.g. stainless steel disk, test dish or membrane filter

**3.3**
**reference source**
radioactive secondary standard source for use in the calibration of the measuring instruments, i.e. material (solid, liquid) containing one or more radionuclides of known activity (Bq or Bq·g$^{-1}$ or Bq·ml$^{-1}$), prepared such that the activity is traceable to national or international primary standards of radioactivity

**3.4**
**alpha emitter radionuclide**
radioactive nuclide with a specified atomic number and mass which emits alpha particles

**3.5**
**energy resolution**
**FWHM**
full width at half of the maximum of the alpha peak distribution

Note 1 to entry: The width is given in kiloelectronvolts (keV).

**3.6**
**adsorption**
process in which a substance forms a very thin layer onto the surface of a solid by attraction of the molecules

**3.7**
**resolution**
ratio between *energy resolution* (3.5) and energy corresponding to the maximum of the peak distribution

Note 1 to entry: The resolution is the spectral resolution and it is given in per cent.

### 3.8
### tracer

radionuclide, which has the same chemical properties as the alpha radionuclides of interest and whose activity in the test source is used to determine the chemical yield or the total efficiency

### 3.9
### carrier

a stable chemical element which is added to the test sample to ensure that the radionuclide of interest will behave normally in the radiochemical separation procedures

## 4 Symbols and units

**Table 1 — Symbols and definitions**

| Symbol | Definition | Unit |
|---|---|---|
| $A_0$ | activity of certified calibration source, at the date of the measurement | Bq |
| $A_T$ | activity of the tracer added, at the date of measurement | Bq |
| $a$ | activity of the measured radionuclides on thin test source, at the date of measurement | Bq |
| $\tilde{a}$ | true activity (see ISO 11929-1) | Bq |
| $a^*$ | decision threshold of the activity | Bq |
| $a^\#$ | detection limit of the activity | Bq |
| $a_j$ | derived activity measured from the thin test source in the unit of $w_1$ | (see $w_1$ ) |
| $a_j^*$ | decision threshold of the derived activity in the unit of $w_1$ | (see $w_1$ ) |
| $a_j^\#$ | detection limit of the derived activity in the unit of $w_1$ | (see $w_1$ ) |
| $a_j^\triangleleft$ , $a_j^\triangleright$ | lower and upper limits of the probabilistically symmetric coverage interval of derived activity in the unit of $w_1$ | (see $w_1$ ) |
| $a_j^<$ , $a_j^>$ | lower and upper limits of the shortest coverage interval of derived activity in the unit of $w_1$ | (see $w_1$ ) |
| $\varepsilon$ | detection efficiency | |
| $G$ | geometrical factor of alpha detection as a function of the effective solid angle of measurement, $\Omega$ with $\Omega = 4\pi \cdot G$ | |
| $I_\alpha$ | summation of alpha emission intensity | |
| $k$ | coverage factor with $k = 1, 2, 3...$ | |
| $k_{1-\alpha}$ , $k_{1-\beta}$ and $k_{1-\gamma/2}$ | quantiles of the standardized normal distribution for the probabilities: $1-\alpha$, $1-\beta$ and $1-\gamma/2$ | |
| $n$ | number of counts in the region of interest of the spectrum | |
| $n_g$ | number of gross counts in the region of interest of the spectrum | |
| $n_0$ | number of counts of the global background in the region of interest of the spectrum | |
| $N$ | number of channels of alpha peak integration | |
| $R$ | total efficiency with $R = R_c \cdot \varepsilon$ | |
| $R_c$ | chemical yield | |
| $r$ | count rate in the region of interest of the spectrum | $s^{-1}$ |
| $r_0$ | background count rate of the test sample region | $s^{-1}$ |
| $r_{0T}$ | background count rate of the blank sample region | $s^{-1}$ |
| $r_{0\varepsilon}$ | background count rate of the detection efficiency region | $s^{-1}$ |

**Table 1** *(continued)*

| Symbol | Definition | Unit |
|---|---|---|
| $r_g$ | gross count rate of the test sample region | $s^{-1}$ |
| $r_{g\varepsilon}$ | gross count rate of the detection efficiency region | $s^{-1}$ |
| $r_{gT}$ | gross count rate of the tracer region | $s^{-1}$ |
| $S_{ref}$ | reference area of alpha peak integration | |
| $t$ | counting time | s |
| $t_0$ | background counting time of the test sample | s |
| $t_{0\varepsilon}$ | background counting time of detection efficiency | s |
| $t_{0T}$ | background counting time of tracer | s |
| $t_g$ | counting time of the test sample | s |
| $t_{g\varepsilon}$ | counting time of detection efficiency | s |
| $t_{gT}$ | counting time of tracer | s |
| $u(y)$ | standard uncertainty associated with parameter $y$ result $(k=1)$ in unit of the measurand $y$ | |
| $U(y)$ | expanded uncertainty calculated by $U(y)=k\cdot u(y)$ with $k>1$, in unit of the measurand $y$ | |
| $u_{rel}(y)$ | relative standard uncertainty associated with parameter $y$ result calculated by $u_{rel}(y)=u(y)\cdot y^{-1}$ | |
| $U_{rel}(y)$ | relative expanded uncertainty calculated by $U_{rel}(y)=k\cdot u_{rel}(y)$ with $k>1$ | |
| $w$ | correction factor for activity of the thin test source | (see $w_1$) |
| $w_1$ | correction factor for derived activity of the test sample in the inverse of the sample unit, i.e.:<br>— per kilogram for a solid sample for a specific activity concentration;<br>— per litre for a liquid sample for an activity concentration;<br>— per square metre for a contaminated surface sampled with a wipe-test;<br>— per cubic metre for an activity concentration of particles in air or gases sampled;<br>— per one unit for specific thin test sample directly measured. | <br>$kg^{-1}$<br>$l^{-1}$<br>$m^{-2}$<br>$m^{-3}$ |

## 5 Principle

Generally, the test sample solution, containing alpha emitter radionuclides of interest, is mixed with an aliquot of a solution of similar physico-chemical properties and containing a radioactive tracer. This solution has at least one isotope of the radionuclides of interest contained in the sample, except if the chemical properties of the tracing radionuclide are close enough to the alpha radionuclides of interest, for example as in the case of curium and americium (see Table 2). This is followed by a specific preparation according to the type of sample matrix by dissolution entailing either dissolution with an acid or fusion by melting an inorganic base (e.g. alkaline fusion).

After a valence cycle to adjust the oxidation states, chemical separation of the radionuclide is achieved by a concentration step (e.g. a precipitation) followed by one or more specific separation steps (e.g. ion exchange chromatography, extraction chromatography, liquid-liquid extraction or selective precipitation).

The sample can also be measured directly by alpha spectrometry, i.e. without sample preparation or pre-treatment, no addition of carrier or radionuclide tracer, no chemical separation, or some combination of these considerations. If relevant, in this case, the user shall ensure that the quality of the measurement source allows to quantify its activity or to qualitatively determine the energies of the alpha-emitter present with a sufficient resolution previously defined by the user without degrading the performance of the detector by radioactive contamination due to a poor quality of the deposit or by volatile decay parent products, e.g. $^{210}$Po, radon isotopes and their daughter nuclides. Other alpha emitting radionuclides can be present in the sample and can interfere with the counting of the radionuclide of interest to measure if no chemical separation is carried out to remove these interfering radionuclides from the test sample.

The thin test source is usually prepared by electro-deposition, by coprecipitation or by spontaneous deposition and assayed by alpha spectrometry using a grid chamber or a semiconductor-type device. The measurement relies on the interaction of the alpha particles with the detecting medium. This interaction creates an electric charge, which is amplified and output as a voltage pulse proportional to the deposited energy of the incoming alpha particle.

The electric pulse from the detector is analysed by the electronic systems and stored by a multichannel analyser (MCA). Data analysis software shows the alpha emitter radionuclides spectrum which is the distribution of number of pulses (counts) as a function of energy.

The analysis of the count rates in the region of interest (ROI) of the alpha spectra for the radionuclide considered allows the determination of the test sample activity concentration after correcting by blank count rate, chemical yield and detection efficiency.

The chemical yield and the detection efficiency are not necessarily determined separately. In this case, they are often obtained as of the total efficiency combined from the net count rate of the radionuclide added as a chemical yield tracer. In order to quantify any potential interference coming from the reagents, a blank sample is prepared in the same way as the test sample. This blank sample is prepared using an appropriate blank material.

For quality control, in order to quantify potential impurities in the tracer solution, another blank sample shall be prepared with addition of the tracer.

Even with a good energy resolution of approximately (20 to 30) keV (FWHM) several isotopes of one radioactive element cannot easily be resolved by alpha spectrometry due to the very close similarity of their alpha emission energies, e.g. $^{233}$U and $^{234}$U, $^{235}$U and $^{236}$U, $^{239}$Pu and $^{240}$Pu or even $^{243}$Cm and $^{244}$Cm.

The dissolution, radiochemical and evaluation parts of an analytical process are summarized in Figure A.1.

## 6 Chemical reagents and equipment

### 6.1 General

Use only reagents of recognized analytical grade.

### 6.2 Chemical reagents

#### 6.2.1 Water quality

Laboratory water is used as a blank, as free as possible from chemical and radioactive impurities, complying with ISO 3696, grade 3 or an equivalent purity (e.g. distilled or demineralized water).

#### 6.2.2 Tracer solution

The radionuclide tracer solution for determining the total yield can also be used to calculate the chemical yield. The solution is prepared by the dilution of a suitable, traceable standard. The tracer solution shall be homogeneous and chemically stable by adding suitable acids.

The concentration of the tracer solution should be chosen such that a small yet accurately determined amount may be added with activity in the range of the test sample.

It is important to check the activity and the purity of the tracer solutions periodically after preparation. Those checks which ensure method validity and performance, can be done by liquid scintillation counting, grid ionization chamber counting or alpha spectrometry. Performing a blank analysis with tracer is a potential way to identify any presence of radioactive impurity in the tracer.

For example, $^{228}$Th can be present in the $^{232}$U tracer solution and has a very close energy to that of its parent $^{232}$U. Therefore, a complete separation of Th and U prior to or during the chemical purification process is required (References [6] and [7]) to minimize the interference of $^{228}$Th so that the counting yield of $^{232}$U is not overestimated (see Clause 4). Table 2 gives a list of alpha emitter tracers with possible radioactive impurities and their decay products.

Before using a purified tracer, the user shall ensure that the purified tracer has been properly referenced to the activity in becquerel, if the activity of the tracer no longer corresponds to that of the certificate which accompanies the standardized material.

**Table 2 — Alpha emitter tracers and possible radioactive impurities**

| Radionuclide of interest | Used tracer | Alpha impurity | Origin |
|---|---|---|---|
| $^{234}$U, $^{235}$U and $^{238}$U, $^{233}$U, $^{236}$U | $^{232}$U | $^{228}$Th | $^{232}$U |
| $^{238}$Pu and $^{239+240}$Pu | $^{242}$Pu, $^{236}$Pu | $^{241}$Am | $^{241}$Pu |
| $^{241}$Am, $^{242}$Cm and $^{243+244}$Cm | $^{243}$Am[a] | | |
| $^{237}$Np | $^{235}$Np, $^{236}$Np<br>$^{238}$Np, $^{239}$Np | | |
| $^{227}$Th, $^{228}$Th, $^{230}$Th, $^{232}$Th | $^{229}$Th | Decay products of $^{229}$Th: $^{225}$Ac, $^{221}$Fr, $^{217}$At and $^{213}$Po from which $^{225}$Ac can interfere with the determination of $^{227}$Th | $^{229}$Th |
| $^{226}$Ra | $^{133}$Ba[b]<br>or<br>$^{224}$Ra (from $^{228}$Th) | | |
| $^{210}$Po | $^{208}$Po, $^{209}$Po | | |

[a] The chemical property of curium is so similar to that of americium that there is no need of a curium tracer, i.e. the americium tracer is used as a chemical yield monitor for those both radionuclides[8].

[b] The measurement of $^{133}$Ba activity is achieved by gamma spectrometry.

## 6.3 Equipment

Usual laboratory device and in particular the following.

### 6.3.1 Detector.

Alpha spectrometry can be performed with a grid chamber device (high detection efficiency, low resolution), an ion implanted semiconductor device (low detection efficiency, high resolution) or a liquid scintillation counter (near 100 % detection efficiency, low resolution). Those measurement devices operate at constant temperature following the manufacturer's instructions.

#### 6.3.1.1 Grid ionization chamber.

A grid ionization chamber is a pulse ionization chamber. It detects variations in charges due to the ionization of a gas by alpha particles. Under the action of an electric field the charges created migrate to their respective electrodes. The amplitude of the corresponding signal depends on the energy of the incident particle but also on the distance of its interaction between the cathode and the anode. This difficulty was solved with a grid placed between the anode and the cathode where the alpha particles interact only in the space between the cathode and the grid. This grid subjected as an intermediate potential captures only a negligible number of electrons during their migration to the anode. Signals thus created become usable for alpha spectrometry. The energy resolution is not less than 30 keV. The detection efficiency depends on the quality of the measurement source to be measured (thickness, chemical and elemental purities) and on the region of interest (ROI) of the spectra defined by the user. As the measurement source is inside of the detector the detection efficiency can reach 50 % i.e. a detection geometry of 2π steradian or more considering possible backscatters. In addition, the grid chamber works with an ionization gas. The most used gases are nitrogen, hydrogen, hydrocarbons or rare gases such as Ar-CH$_4$ or Ar-CO$_2$. The gas consumption during the measurement is in the order of ten litres per hour with an operating pressure varying between $1 \cdot 10^5$ Pa and $1,5 \cdot 10^5$ Pa.

#### 6.3.1.2 Semiconductor.

For alpha spectrometry a semiconductor device can be used. Measurements are based on the interaction of alpha particles with ion-implanted silicon. This interaction instantly changes the conductivity of the silicon, proportional to the energy of the incident alpha particle. To achieve well-resolved spectra, the detection system needs to be maintained at a pressure <1 kPa. The resolution also depends on the quality of the measurement source and on ROI. It can be improved by increasing the distance between the measurement source and the detector to the disadvantage of the detection efficiency. In theory the energy resolution could be of 10 keV which is the intrinsic resolution of the detector. In practice it is between 20 keV and 30 keV. Under these conditions alpha spectrometry can hardly separate $^{233}$U and $^{234}$U, $^{235}$U and $^{236}$U, $^{239}$Pu and $^{240}$Pu or $^{243}$Cm and $^{244}$Cm because of the similarity of their respective alpha emission energies (see 8.6). Generally, $^{233}$U and $^{236}$U are not present inside environmental samples. If these radionuclides are not expected from a nearby source, they are often under the detection limits of the alpha spectrometry given in 9.1.1.

#### 6.3.1.3 Liquid scintillator.

The technique for measuring activity by liquid scintillation consists in transforming ionizing radiation into quantifiable light radiation which is usually detected by two facing photomultiplier tubes in coincidence. The usual commercial counters with scintillating cocktails and alpha/beta discrimination does not allow to achieve sufficient resolutions for alpha spectrometry. Otherwise, light attenuation or quenching has the consequence of shifting the energy of a given alpha emitter to low energies and generating counting losses and degrading the resolution. This effect depends on the chemical purity and on the chemical composition of the sample to be measured. However, a specific technique to alpha spectrometry was developed in the 1970s and later developed in its current form by McDowell (Reference [9]). This technique called PERALS for photon-electron rejecting liquid alpha scintillation spectrometry combines liquid scintillation instrumentation with aqueous-non-miscible extraction scintillators specific to the alpha-emitter elements and a pulse shape discrimination electronics.

This technique is faster because the radioactive element of interest is simply extracted from the test sample by the liquid-liquid scintillating extractor exchanges. Despite a detection efficiency close to 100 %, this technique does not allow to obtain energy resolutions below 200 keV to 300 keV (References [10] and [11]). This energy resolution is sufficient to determine the total activity of most naturally radioactive chemical elements but it limits the separation power of alpha emitters whose emission energies are close (e.g. for isotopes 241 and 243 of americium or for isotopes 208, 209 and 210 of polonium). Therefore, it is often necessary to know the total yield a priori.

### 6.3.2 Pipette.

The pipette shall be suitable for the accurate transfer of radioactive tracer solutions (e.g. 100 µl) with a total precision within ±1 %.

### 6.3.3 Scale.

A calibrated scale shall be used for measurements of mass (e.g. a balance capable to achieve ±0,1 mg precision).

### 6.3.4 Coprecipitation device for deposition.

The coprecipitation unit consists of the usual laboratory equipment and in particular the following:

- **membrane filter**, (e.g. made from cellulose ester, polyethersulfone, polycarbonate or PTFE) with pore diameter of 0,1 µm for uranium isotopes or 0,22 µm for other actinides isotopes such as americium, curium and plutonium isotopes;
- **filtration system**;

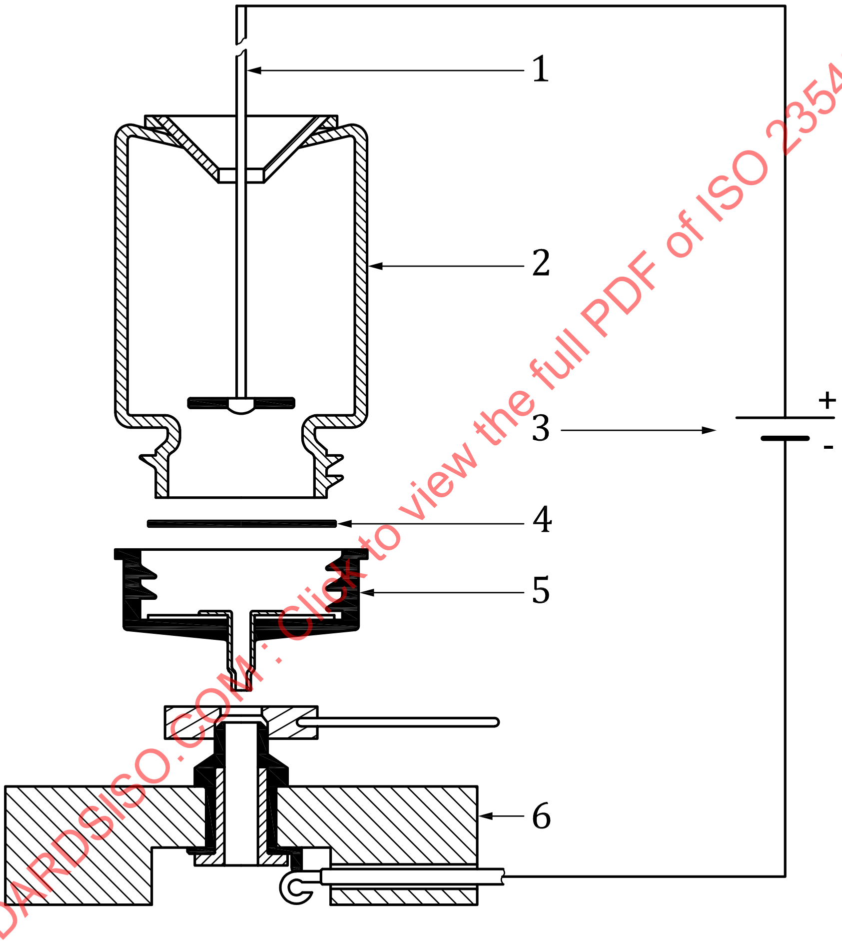
- **test dish or stainless-steel disk**, with a diameter adapted to the spectrometry device.

### 6.3.5 Electrodeposition device

The electrodeposition device is generally composed of the following components:

- glass, Teflon$^{TM}$ or polyethylene electrodeposition cell containing the analyte;
- platinum wire (anode);
- stainless steel disk (cathode) with a diameter adapted to the electrodeposition cell;
- DC power supply.

Figure 1 shows an example of the main components of an electrodeposition device.



**Key**

1 platinum wire (anode)
2 electrodeposition cell
3 DC generator
4 disk (cathode)
5 cap assembly
6 base

**Figure 1 — Diagram of an electrodeposition device**

## 7 Sampling and samples

If appropriate, samples shall be collected in accordance with the relevant standard (See Table 3).

The following information shall be recorded:

- a unique identifier for the sample;
- location;
- reference to the sampling procedure followed;
- sampling date and time;
- sample description (physical, chemical, biological form, mass and volume, as appropriate);
- name of the sampling operator.

Special handling of samples can be required to avoid degradation if there is risk of a long delay (several days) between sampling and measurement. If samples are perishable, it is recommended that they are transported and stored in the absence of light and at a temperature between 1 °C and 5 °C before processing and measurement.

Conditions of sampling depend on sample types. It is important that the laboratory receive a representative sample, unmodified during transport or storage and in an undamaged container. If required, the sample shall be sampled and/or stored according to corresponding ISO standards given in Table 3.

**Table 3 — Standards for sampling and/or storage of different types of samples**

| Type of sample | Standards |
|---|---|
| **Water** | ISO 5667 series on potable water, sea water, rainwater, ground water, waste waters, sludges, effluents, suspended solids and sediments |
| **Foodstuff** | ISO 707 and ISO 5538 on milk and milk products<br>ISO 874 on fresh fruits and vegetables<br>ISO 17604 on meat of slaughtered animals<br>ISO 24333 on cereals and cereal products |
| **Oils and fats** | ISO 661 and ISO 5555 on animal and vegetable fats and oils<br>ISO 5500 on oilseeds residue |
| **Soils and mineral materials** | ISO 18400 series on soil in the landscape, soil stockpiles, potentially contaminated sites, agricultural soils, landfills, and forest soils<br>ISO 18589-2 on soil, rock, bedrock, building materials |

## 8 Procedure

### 8.1 General

The quality of the alpha spectrum depends on the quality of the deposit made for alpha spectrometry and/or on the chemical purity of the solution containing the alpha-emitter radionuclides. As the linear energy transfer of alpha-particles is very high (very short path through matter) the presence of chemical impurities in the deposit increases the FWHM resulting in worse energy resolution. To improve the energy resolution, the laboratory shall use sample dissolution processes in order to obtain a liquid aqueous sample usable in the different steps of the specific radiochemical separation procedures. Then the test sample realized becomes usable for deposition techniques to obtain a thin test source with the most uniform surface distribution possible or for direct counting by liquid scintillation. The dissolution, radiochemical and evaluation parts of an analytical process are summarized in Figure A.1.

NOTE Specific methods for dissolution, specific chemical separation, preparation of measurement source, and evaluation of the result of measurement of alpha emitters in particular sample types can be used according to the requirements of ISO/IEC 17025. Proposed methods published either in international standards, in relevant scientific texts or journals, or as specified by the equipment manufacturer are listed in the bibliography in References [12] to [13] and [33] to [41].

## 8.2 Calcination

The sample to be measured after chemical preparation of calcined ashes is homogenized, placed in a container and introduced into a calibrated oven.

The container (made for example of platinum) shall be non-vulnerable under the test conditions, have a suitable size to provide the largest possible surface area for oxidation allowing a homogeneous distribution of the test sample to be obtained and shall be designed to prevent cross contamination.

The oven, the temperature of which shall be adjustable and programmable, shall allow the gradual destruction of organic matter by heating until ashes are obtained. The maximum temperature must be reached gradually through increasing temperature steps. This protocol makes it possible to limit the risks of projection and self-ignition of the sample.

Depending on the type of chemical preparation applied, either partial or complete calcination of the samples can be carried out. In the case of complete calcination of the sample, the mineral residue forming the ash obtained shall be free of carbonaceous particles which is characterised by the appearance of coloured ash (e.g., red-brown ash indicating the presence of iron) or white ash, without any carbonaceous particles in the residue. If carbonaceous residues (e.g., black particles) are present, a second calcination should be performed. Upon removal from the oven the mineral residue obtained after calcination under the specified conditions can be totally used for the chemical preparation. Otherwise, sample ashes can be stored temporarily according to laboratory specifications.

## 8.3 Sample dissolution processes

The radionuclides contained in the solid or liquid organic samples shall be dissolved in aqueous solution before the chemical separation steps. Different methods for dissolving solids exist and the choice of method will depend on the nature of the solid samples and the radionuclides to be analysed.

The mass of the test portion depends on the activity level of the sample, the sample type and the expected decision threshold or detection limit. In practice, it varies from 0,1 g to 500 g or more of the sample for each test.

Sample dissolution can be total or partial (leaching). Total dissolution of the solid sample is not necessary if it can be demonstrated that the radionuclides of interest are completely solubilized in aqueous solution.

Usually, the solids are completely or almost completely dissolved by a solution or a mixture of nitric acid, hydrochloric acid, hydrofluoric acid, sulfuric or perchloric acid. It should be ensured that the volume of the acid solution is sufficient for dissolving the quantity of test sample.

The complete dissolution process may include stages of dissolution or leaching by acids, separation of solid and liquid phases in the case of leaching, dry evaporation of the liquid, re-dissolution of the dry residue with a suitable acid composition and optionally followed by a step of adjusting the acid concentration in the final step.

Solids which are difficult to dissolve with acids can be melted by the alkaline fusion technique using suitable fluxes to obtain substances which can be solubilized easily by acids. Examples of melting conditions are presented in Annex B.

A higher temperature increases the reaction rate but with a risk of vaporizing certain radionuclides.

Sometimes a combination of acid dissolution and fusion is performed. It consists of a first dissolution of as many samples as possible by mineral acids including hydrofluoric acid. The insoluble residue is then dissolved by alkaline fusion. This treatment sequence is often practical to perform in the laboratory.

The particular technique of dissolution using microwaves in an open or closed device (under pressure) makes it possible, under these conditions, to greatly improve the kinetics of dissolution.

## 8.4 Specific radiochemical separation procedures

The principle of the chemical separation methods used in radiological analysis consists in shifting the equilibrium of the chemical species to another phase of the system. In general, the separation phases are solid/liquid (precipitation, coprecipitation, separation resins) or liquid/liquid (liquid-liquid extraction).

The separation is based on the differences in the physical and chemical properties of the species and on the shifting the equilibrium of the species from one phase to another. Shifting the equilibrium can involve modifying parameters such as the pH, acid concentration, oxidation state of the elements, complexation, solubility product, solubility in the organic phase, etc.

The solution intended for chemical separations shall not contain organic matter which may react with radionuclides and which may interfere with the chemical separation steps. If organic matter is present, the appropriate chemical treatments by dissolution processes shall be carried out before the chemical separation steps. The absence of organic matter can be verified by observing a clear white or light grey colour of the solid residue.

$Fe(OH)_3$, $CaHPO_4$, $LnF_3$ (Ln = La, Nd, Ce) are often used to co-precipitate actinides and $BaSO_4$ is used to co-precipitate radium. Separation by liquid-liquid extraction uses extractants, the most common of which are tributyl phosphate (TBP), bis(2-ethylhexyl) phosphoric acid (HDEHP), tri-n-octylamine (TnOA) or TIOA, Aliquat-336, thenoyltrifluoroacetone (TTA). Those extractants are diluted in an organic solvent to extract the actinides.

The chemical separation or purification steps of actinides mainly use ion exchange resins and chromatographic resins.

Ion exchangers are classified as cation exchangers or anion exchangers, according to their affinity for negative or positive counterions respectively. They are further subdivided into strongly or weakly ionized groups. Most cation exchangers contain sulfonic acid groups, whereas typical anion exchangers have quaternary amine groups with replaceable hydroxyl ions (see Table 4 where R is alkyl group).

**Table 4 — Typical functional groups of ion-exchange resins**

| Cation-exchange resin | Anion-exchange resin |
|---|---|
| R- $SO_3H$ | R- $NH_2$ |
| R- COOH | R- NHR |
| R- OH | R- $NR_2$ |
| R- SH | R- $NR_3^+$ |

The most used chromatographic resins are presented in Table 5.

**Table 5 — Extraction chromatography resins and resins designed for the separation of specific radionuclides**

| Chromatographic resin | Extraction reagent | Use |
|---|---|---|
| TEVA | Aliphatic quaternary amine | For tetravalent actinides $Th^{4+}$, $U^{4+}$, $Np^{4+}$, and $Pu^{4+}$ |
| TRU | Octylphenyl-N,N-diisobutyl carbamoylphosphine oxide (CMPO) | For tri and tetravalent actinides $Th^{4+}$, $U^{4+}$, $Np^{4+}$, $Pu^{4+}$, $Pu^{3+}$, $Am^{3+}$, $Ac^{3+}$, and hexavalent uranium $UO_2^{2+}$. |
| UTEVA | Diamyl, amylphosphonate (DAAP) | For tetravalent actinides $Th^{4+}$, $U^{4+}$, $Np^{4+}$, $Pu^{4+}$, and hexavalent uranium $UO_2^{2+}$ |
| DGA | N,N,N',N'-tetra-n-octyldiglycolamide | For tri and tetravalent actinides $Th^{4+}$, $U^{4+}$, $Pu^{4+}$, $Pu^{3+}$, $Am^{3+}$, and hexavalent uranium $UO_2^{2+}$ |
| $MnO_2$ | Manganese Dioxide ($MnO_2$) | For $Ra^{2+}$ |
| LN | Bis(2-ethylhexyl)phosphoric acid (HDEHP) | For $Ra^{2+}$ and rare earth elements (REE) $Ln^{3+}$ |

In chemical separation steps, the oxidation state of some elements can be modified. Oxidizing ($NaNO_2$ or $H_2O_2$) and reducing ($Fe(NH_2SO_3)_2$, $NaHSO_3$, $Na_2S_2O_3$, $TiCl_3$, hydroxylamine hydrochloride, ascorbic acid) agents can be used to adjust the oxidation state of actinides.

The sequence of the different chemical separation steps will depend on the chemical composition of the separation solution and the alpha emitter radionuclides to be analysed. The laboratory should choose the most appropriate separation method in different separation protocols proposed by the literature and/or by specific ISO standards according to its objectives and the nature of the samples.

## 8.5 Methods of deposition

### 8.5.1 Direct deposition

Direct deposition consists of depositing a known quantity of the solution to be analysed on the substrate with a pipette, a pycnometer or a dropper bottle. For measurements in defined geometry, the deposit shall be centred and shall not contain fractions of the solution outside the deposit area.

The geometry obtained by direct deposition is generally not reproducible in terms of surface and thickness because the uniformity of the deposits depends on the liquid-solid surface tension at the interface of the substrate and the liquid solution. The spreading of the deposit on the substrate is improved by adding a wetting agent. It can be centred by applying the wetting agent with a calibrated pad on top of the substrate surface.

Evaporation shall be carried out without exceeding the boiling point of the solution to avoid splashing outside the deposit area. When the residue is dry, its calcination by pyrolysis process leads to better adhesion of the deposit on the substrate. It also eliminates volatile and carbonaceous impurities. The calcination temperature shall be compatible with the material of the substrate and shall not generate diffusion of the deposit into the substrate. For example, for stainless steel, the maximum of pyrolysis temperature is about 500 °C.

Direct deposition is a fast but non-selective method. When the deposit is made with a non-purified solution, the non-volatile impurities present in the solution are also present in the deposit. This will degrade the resolution of the alpha spectrum.

### 8.5.2 Spontaneous deposition of polonium

Radiochemical separation of polonium can be quite simple. It consists of its spontaneous deposition on metal disks such as silver or nickel in acidic chloride, nitric or acetic acid medium. The medium shall be between 0,1 M and 1 M for HCl and $HNO_3$ and between 1 M and 2 M for $CH_3COOH$. Spontaneous deposition is furthered if the acidic solution is stirred and heated with a temperature less than 90 °C. Spontaneous deposition on silver can be inhibited in the presence of $Hg^{2+}$ and $Fe^{3+}$ ions. Ferric ions inhibit deposits on silver when their concentrations are greater than $5 \cdot 10^{-4}$ mol·l$^{-1}$. Deposition on a silver disk eliminates elements such as Bi, Cu, Pb, Zn and natural radionuclides, especially $^{210}$Bi and $^{210}$Pb. During the dissolution part, the sample should be heated at high temperatures only by wet methods (i.e. by acid dissolution or by alkaline fusion) because the polonium readily volatilizes.

### 8.5.3 Microprecipitation

Precipitation depends on the solubility product of the compound which is proportional to the solubility of the compound in solution. If a suitable isotopic carrier for a radionuclide is not available precipitation shall be performed with a non-isotopic carrier to achieve a coprecipitation. An example of microprecipitation procedure is presented in Annex C.

The coprecipitation process associates a micro-component (radionuclide) with a macro-component (carrier) to generate an insoluble homogeneous salt. The precipitation assumes that the solubility of the formed salt is sufficiently low to ensure quantitative coprecipitation of the radionuclide by adsorption on the surface of the precipitate grains. The quality of the deposit depends on parameters given in Table 6.

After precipitation, the precipitate is collected on a membrane filter, dried and fixed on a substrate which shall be adapted for alpha counting. However, even if the layer of the deposit is thicker compared to sources gained by auto-deposition or by electrodeposition, the resolution obtained by this rapid technique of deposition is acceptable for alpha spectrometry (References [11] and [18]).

**Table 6 — Influence of precipitation conditions**

| Influence parameter | Effect on adsorption [a] |
|---|---|
| High carrier concentration (over saturation) | + |
| Speed of precipitation | + |
| Agitation | – |
| Heating | – |
| Extended wait | – |
| Washing | – |

[a] + or – facilitate or not the coprecipitation.

This method saves time for source preparation and is easier to set up for batch processing with high sample analysis throughput. It may be affected by the presence of impurities in the sample matrix. For example, when the sample contains calcium concentrations in milligrams the alpha source is thicker which degrades the resolution of the alpha spectra.

### 8.5.4 Electrodeposition

After the purification of the interfering elements and the selection of the radionuclides of interest by the chemical separation techniques (see 8.3), the deposit is carried out by electrodeposition on a disk. The disk must be thoroughly cleaned, for example with ethanol and/or acetone, to remove deposits of organic impurities. An example of an electrodeposition procedure is presented in Annex D.

A DC generator is used to apply different voltages between two electrodes, resulting in the reduction of the metal cations dissolved in the electrolyte. The reduction that takes place at the cathode leads to the formation of a deposit of actinides in hydroxide form.

With a constant redox current density of about 150 mA·cm$^{-2}$ this technique can produce sources which achieve very good intrinsic resolutions less than 15 keV improving the deconvolution of the spectra.

The pH of the solution must not hydrolyse the actinide ions except when these ions are complexed. The pH of the electrolyte must be greater than 1,5 and to avoid shifts in pH during electrolysis, a buffered system is required.

At the end of the electrodeposition process, a hydroxide deposit can be dissolved by the acidic electrolyte when the current is stopped. Therefore, the little amounts of a base (e.g. ammonia, sodium hydroxide or potassium hydroxide) are added while maintaining the current for a further half to one minute.

Americium and curium may be slightly dissolved when rinsing the source with water. In this case, the deposit should be rinsed with ethanol.

In addition, calcining the source improves its intrinsic resolution.

This method can be affected by the presence of inorganic and organic impurities in the sample matrix which could result in a lower counting efficiency and a worse resolution for alpha source.

## 8.6 Measurement

### 8.6.1 Measurement geometry

#### 8.6.1.1 Grid ionization chamber

Since the grid chamber has a detection geometry of 2π and the test sample with a given surface density is placed inside the volume of the detection gas, the form of the deposits and the position of the source compared to the centre of the detector support do not affect the spectrum shape, the resolution and, the detection efficiency.

#### 8.6.1.2 Semiconductor detector

For semiconductor detectors the test sample with a given surface density is placed outside the detection volume. Consequently, the form of the test samples (i.e. diameter and uniformity of the surface distribution of the source) as well as the source-detector distance define the solid detection angle. This solid angle is inversely proportional to the source-detector distance; when this distance increases, the resolution improves to the detriment of the detection efficiency and vice versa.

### 8.6.2 Spectrum exploitation

#### 8.6.2.1 General

After the spectrum acquisition the gross counts $n_g$ of the radionuclides are calculated by integrating the number of counts $n$ in the regions of interest (ROI) of the corresponding peaks and if used, in the ROI assigned to the tracer peak.

#### 8.6.2.2 Selection of alpha peak areas

##### 8.6.2.2.1 General

The spectrum stored in the multichannel analyser shows one or several alpha peaks. Referring to the peak maximum the alpha peaks approximately show a Gaussian shape on the right side (higher channels) and an exponential tailing shape on the left side (lower channels). This low-energy distortion is due to energy loss. It depends on self-absorption, emission angle and energy lost during the path of the alpha particle from the source to the detector. The channel number of the maximum alpha peak depends on the energy of the alpha particle detected. The net alpha peak area is proportional to the number of alpha particles of that energy which have interacted with the detector during the counting period. The net alpha peak area can be determined in the analysis software package by fitting or by summation of gross area after manual selection of the ROI.

##### 8.6.2.2.2 Fitting method

Spectra processing software uses two methods of spectra deconvolution:

- automatic adjustment by the least squares method, or
- semi-automatic methods where the user manually adjusts a peak and its tailing which is taken as a reference.

In the second case, the modelled shape of the peak is superimposed on each peak of the spectrum. Then, the software adjusts the amplitude and position parameters of each peak using the χ2 method for gaussian fluctuations (least squares method). This method is adapted to deconvolute the spectra with mixtures of alpha radionuclides whose energies are too close to be separated by simplest methods. At the end of the calculations, the overall envelope of the initial spectrum is compared to the sum of all the modelled peaks. When the user considers the result unacceptable, the shape of the reference peak tailing must be readjusted for a new peak adjustment calculations.

#### 8.6.2.2.3 Gross area method

The gross area method assigns a radionuclide to an alpha peak and determines the number of gross counts in the peak. Then, the activity is calculated taking into account the total number of gross counts of the peak and an alpha intensity corresponding to the sum of the main energy and associated fine structures.

The manual integration of a spectrum consists of the determination of the number of alpha peaks separated from each other in the area of the energy range of the spectrum. The area associated with a selected alpha peak is the integral of the number of counts below the alpha peak in the defined ROI. In this case, the software does not correct the global background. If relevant, this subtraction has to be done manually.

Due to the asymmetrical shape of the alpha peaks, more channels should be selected to the left of the maximum of the alpha peak than to the right. Generally, the ratio on the right is 1/3 and 2/3 on the left. Those ratios can be different depending on the quality of the spectrum due to the geometry of the measurement, the shape and the quality of the sample, the measurement device electronics and mainly the number of counts in each channel.

If a tracer is used for the measurement, the alpha peak area can be estimated in the same way for all peaks, including the tracer peak, using a fixed number of channels previously defined by the user. Then, the estimation error due to the selection of the tracer peak is reproduced on all other peaks and it is taken into account for the activity calculations. When using this method alpha emissions of different radionuclides should not be included in the same selection.

For other methods, each alpha peak range can be selected by minimizing integration uncertainty. A procedure of this method (Reference [11]) is presented in Annex E.

### 8.6.2.3 Count rate

The count rate $r$ is given by Formula (1):

$$r = \frac{n}{t} \tag{1}$$

where $t$ is the counting time.

The corresponding uncertainty $u(r)$ is given by Formula (2):

$$u(r) = \sqrt{\frac{r}{t}} \tag{2}$$

The net count rate is calculated by the ratio between the subtraction of the number of gross counts, $n_g$, with the number of counts of the global background of the detection system, $n_0$, and the counting time.

The activity of the different radionuclides on the measurement source is calculated by dividing the net count rate by the detection efficiency and, if relevant by the alpha emission intensity.

### 8.6.3 Energy calibration

Energy calibration identifies the energy peaks of the alpha emitter radionuclides detected in the spectrum. This calibration establishes the relationship between the channel number of a peak maximum and the known energy of the corresponding alpha emitter radionuclides. The energy calibration shall be established using a source containing a mixture of alpha emitter radionuclides that cover the energy range of interest (e.g. $^{233}$U, $^{239+240}$Pu, $^{241}$Am, $^{244}$Cm, etc.). Energy calibration shall be performed periodically according to quality control procedures to take account any drifts a priori. After the counting period of a test sample in a given solid angle with a too large quantity of matter, the intensity of the alpha peaks weakens due to the self-absorption, thus, the peaks move to lower channels as a function of the energy, broaden (Reference [18]) and the exponential tailing increases a posteriori.

### 8.6.4 Efficiency calibration

#### 8.6.4.1 General

The estimation of the detection efficiency is not necessary if a radionuclide tracer is used but, it is needed to determine the absolute activity, the chemical yield, and the decision threshold values. In this aim, there are two ways to assess the detection efficiency by a defined solid angle (DSA) or by direct comparison with a certified reference source.

#### 8.6.4.2 Defined solid angle

The defined solid angle consists of a collimator placed in the alpha detection chamber between the source and the semiconductor detector. This device allows to precisely define the solid angle of detection. The measurement in defined geometry is based on the following four parameters:

- the diameter of the collimation diaphragm;
- the distance between the source and the detector;
- the diameter of the deposited source; and
- the eccentricity of the source relative to the centres of source and detector.

From the known values of the collimator radius, the distance of source to detector, the source radius and its eccentricity, a geometrical factor G can be defined as a function of the effective solid angle of measurement $\Omega$ such as $G=\Omega/4\pi$. Then, the activity of the source is calculated from the product of the net count rate per 1/G.

This absolute method relative to time units of length allows estimation of alpha activities of primary sources with relative standard uncertainties in the order of 0,1 %, including the uncertainty from weighing the dispensed amounts in the sources. More explanations are summarized in References [19], [20] and [21].

#### 8.6.4.3 Comparison with a certified reference source

The comparison of the count rate given by the detector with the activity of a certified reference source allows the quantification of alpha activity from each test sample prepared as much as possible in the same manner as the reference source, i.e. identical layer thickness and diameter deposited on the same type of substrate. For example, a thin source of $^{239+240}$Pu can be used for this purpose; alpha emissions are in the energy range between 5,10 MeV and 5,20 MeV and there is no significant decay over the life of the source. Moreover, the detection efficiency by alpha spectrometry is practically independent of the alpha energy particles and their emission intensity. Under this condition, detection efficiency is considered as constant in the energy range of interest.

The measurement time shall be sufficient to record a large number of counts to minimize or to neglect the standard uncertainty due to the number of pulses stored in each alpha peak used for calibration.

NOTE An alpha peak area of 10 000 counts is normally sufficient.

The efficiency calibration shall be performed and checked periodically according to quality control procedures to take into account any anomaly. A different source should be used for checking the efficiency calibration. A record of previous values of detection efficiency (e.g. a Shewhart control charts given in Reference [22]), should be used to identify deviations which have to be investigated and corrected if necessary. Any deviations shall be investigated and corrective action taken if necessary.

The detection efficiency $\varepsilon$ can be calculated using Formula (3):

$$\varepsilon = \frac{r_{g\varepsilon} - r_{0\varepsilon}}{A_0 \cdot I_\alpha} \tag{3}$$

The corresponding relative uncertainty is given by Formula (4):

$$u_{\text{rel}}(\varepsilon) = \sqrt{\frac{u^2(r_{g\varepsilon}) + u^2(r_{0\varepsilon})}{(r_{g\varepsilon} - r_{0\varepsilon})^2} + u_{\text{rel}}^2(A_0) + u_{\text{rel}}^2(I_\alpha)} \tag{4}$$

where $I_\alpha$ corresponds to the summation of alpha emission intensity in the selected region of interest.

### 8.6.5 Global background

The global background is a combination of the detector background and the radiochemical blank. Under routine (or usual) measurement conditions the detector background results from two inseparable parameters:

- electronic and/or digital noises and/or perturbations of signal processing materials and
- the potential presence of alpha emitter radionuclides contamination of the measurement chamber.

The radiochemical blank depends on the radioactive purity of reagents and materials used, such as filters, and on the cross contamination of the laboratory equipment by the radionuclides of interest.

The optimum counting time for the measurement of the detector background and radiochemical blank depends on decision threshold or detection limit required by the customer. It shall not be less than the measurement time of a very low activity source.

The detector background count rate of each detector is determined with an empty source substrate which is also used for the measurement of real sample sources. It shall be regularly checked for contaminations which could be brought on the detector surface by impurities on the measurement sources or by decay products of the radionuclides of interest.

If used, the radiochemical blank shall be carried out without adding tracer and with identical clean water, reagents and materials as filters (i.e. containing no detectable nuclide of interest). The radiochemical blank value shall be compared to the detector background values obtained from the same detector. This value can be comparable to the background value measured with the empty source substrate in the energy regions of radionuclide of interest and of the tracer if there are no reagent or laboratory equipment contaminations. If the radiochemical blank and the detector background values are similar then, the global background can take one or the other value.

In all cases, if a significant number of background counts is observed, it shall be taken into account for the calculation of the net count rate in the regions of interest defined by the user.

### 8.6.6 Chemical yield and total efficiency

The chemical yield can be considered as an important quality control parameter for the complete radiochemical method covering dissolution, separation/purification and preparation of the measurement source. In general, the chemical yield obtained is more than 60 % (References [23] and [24]).

For very low chemical yields, the laboratory can decide to repeat the chemical separation and deposition.

If the total efficiency is lower than a previously defined level and if it is relevant the laboratory should take appropriate actions as repeating the separation possibly using alternative procedures which have to be validated in accordance with the requirements of ISO/IEC 17025.

The chemical yield $R_c$ of the process can be calculated using Formula (5):

$$R_c = \frac{R}{\varepsilon} \tag{5}$$

The total efficiency $R$ is the product of the chemical yield $R_c$ and the detection efficiency $\varepsilon$. It is determined from the activity $A_T$ of the tracer added, and the net count rate using Formula (6):

$$R = \frac{\left(r_{gT} - r_{0T}\right)}{A_T} \tag{6}$$

The corresponding relative uncertainty is given by Formula (7):

$$u_{rel}(R) = \sqrt{\frac{u^2\left(r_{gT}\right) + u^2\left(r_{0T}\right)}{\left(r_{gT} - r_{0T}\right)^2} + u_{rel}^2\left(A_T\right)} \tag{7}$$

where

$$u^2\left(r_{gT}\right) + u^2\left(r_{0T}\right) = \frac{r_{gT}}{t_{gT}} + \frac{r_{0T}}{t_{0T}} \tag{8}$$

and

$$u_{rel}^2\left(A_T\right) = \frac{u^2\left(A_T\right)}{{A_T}^2} \tag{9}$$

### 8.6.7 Quality control sources

Measurement sources for quality control shall regularly be measured to verify that the measurement equipment is performing within agreed limits.

A measurement source of one or more known radionuclides with alpha emissions in a very close energy range (e.g. $^{239+240}$Pu, $^{242}$Pu, $^{241}$Am, $^{243}$Am) and with a sufficiently long half-life (i.e. no notable decay over the lifetime of the measurement source) should regularly be measured to check the correctness of the peak position and/or to carry out an energy (re)calibration.

## 8.7 Recommended nuclear decay data

Nuclear decay data (half-lives, alpha energy and emission probabilities) are available from the scientific literature, databases and other publications. There are often differences between the values depending on the original data used and the evaluation method.

To ensure consistent results from alpha spectrometry measurements, nuclear decay data used for instrument calibration or for estimating the activity content of samples, shall therefore be taken from the Decay Data Evaluation Project (http://www.lnhb.fr/nuclear-data/). If no evaluation of the nuclear decay data of the radionuclide of interest is available in this database, the following databases should be consulted:

- Joint Evaluated Fission and Fusion (JEFF) Library (https://www.oecd-nea.org/dbdata/);
- National Nuclear Data Center database (https://www.nndc.bnl.gov/);
- Evaluated Nuclear Data File (ENDF) (https://www-nds.iaea.org/).

# 9 Expression of results

## 9.1 Calculation of activity

### 9.1.1 General

The used symbols are defined in Table 1.

The activity of the radionuclide of interest is calculated by integrating the number of counts in the corresponding energy region of the alpha emission spectrum of the sample. The results of these integrations, divided by the counting time, are the gross count rates, $r_g$ which are corrected by the background contribution and, if needed, for the contribution of the tailing of higher-energy peaks, which depends on the detector characteristics and on the quality of the measured source.

Background count rates are calculated from the alpha emission spectrum of the detector background or a blank sample by integrating the number of counts in the regions of interest (ROI) in which the peaks appear in the sample spectrum.

Combined absolute or relative uncertainties shall be calculated in accordance with ISO/IEC Guide 98-3.

The characteristic limits (decision threshold, detection limit and limits of the coverage interval) are calculated in accordance with ISO 11929-1.

The detection limit of the test method depends on the amount of the analysed aliquot of the sample material (mass or volume) after radiochemical process (see Annex A) and counting time. Typically, with an initial sample material of $5 \cdot 10^{-3}$ kg of soil or of 5 litres of water, a chemical yield of 80 %, an energy resolution of 20 keV to 30 keV, and a counting time of 36 000 s to 200 000 s the detection limit of the test method is $5 \cdot 10^{-2}$ Bq·kg$^{-1}$ to $5 \cdot 10^{-4}$ Bq·l$^{-1}$.

### 9.1.2 Alpha activity

The activity of the radionuclide of interest is given by Formula (10):

$$a = \left(r_g - r_0\right) \cdot w \qquad (10)$$

where

$w = R^{-1}$ if the activity is determined by spiking with a certified tracer source and chemical separation

$w = (I_\alpha \cdot \varepsilon)^{-1}$ if the activity is directly determined from the detection efficiency

If the radiochemical blank and the detector background values are similar, then $r_0$ can take one or the other value.

### 9.1.3 Uncertainty

The standard uncertainty of measured activity, $a$, is given by Formula (11) where the following parameters are considered:

- the gross count rates from measured radionuclide of interest $r_g$;
- the global background count rates of measured radionuclide $r_0$;
- the conversion factor to express derived activity per unit of test sample $w$ and
- the counting time uncertainty is neglected.

$$u(a)=\sqrt{w^2\cdot\left(u^2\left(r_g\right)+u^2\left(r_0\right)\right)+a^2\cdot u_{\mathrm{rel}}^2(w)} \tag{11}$$

where

$$u^2\left(r_g\right)+u^2\left(r_0\right)=\frac{r_g}{t_g}+\frac{r_0}{t_0} \tag{12}$$

The relative standard uncertainty, $u_{\mathrm{rel}}^2(w)$, of the conversion factor, $w$, is given by Formula 13 if the activity is determined by spiking with a certified tracer source or by Formula 14 if the activity is directly determined from the efficiency calibration.

$$u_{\mathrm{rel}}^2(w)=u_{\mathrm{rel}}^2(R) \tag{13}$$

$$u_{\mathrm{rel}}^2(w)=u_{\mathrm{rel}}^2\left(I_\alpha\right)+u_{\mathrm{rel}}^2(\varepsilon) \tag{14}$$

$u(a)$ includes all uncertainties related to the tracer activity: the standard solution activity, preparation of the tracer solution, addition of the tracer solution to the sample, correction for interferences, etc. For the calculation of the characteristic limits according to ISO 11929-1, $\tilde{u}(\tilde{a})$, i.e. the standard uncertainty of $a$ as a function of its assumed true value is given by the Formula (15):

$$\tilde{u}(\tilde{a})=\sqrt{w^2\cdot\left[\frac{\left(\frac{\tilde{a}}{w}+r_0\right)}{t_g}+\frac{r_0}{t_0}\right]+\tilde{a}^2\cdot u_{\mathrm{rel}}^2(w)} \tag{15}$$

### 9.1.4 Decision threshold

The decision threshold, $a^*$, in becquerel, obtained from Formula (15) for $\tilde{a}=0$ in accordance with ISO 11929-1, is given by Formula (16):

$$a^*=k_{1-\alpha}\cdot\tilde{u}(0)=k_{1-\alpha}\cdot w\cdot\sqrt{r_0\cdot\left(\frac{1}{t_g}+\frac{1}{t_0}\right)} \tag{16}$$

$\alpha=0{,}05$ and then, $k_{1-\alpha}=1{,}65$ is often chosen by default.

### 9.1.5 Detection limit

The detection limit can be calculated by solving Formula (17) for $a^\#$, or more simply by iteration with a starting approximation $a^\#=2\cdot a^*$ in the term of the right-hand side of the Formula (17).

$$a^\#=a^*+k_{1-\beta}\cdot\tilde{u}\left(a^\#\right)=a^*+k_{1-\beta}\cdot\sqrt{w^2\cdot\left(\frac{\left(\frac{a^\#}{w}+r_0\right)}{t_g}+\frac{r_0}{t_0}\right)+a^{\#2}\cdot u_{\mathrm{rel}}^2(w)} \tag{17}$$

where $\beta=0{,}05$ and then, $k_{1-\beta}=1{,}65$ is often chosen by default.

If $k_{1-\alpha}=k_{1-\beta}=k$ the detection limit is given by Formula (18):

$$a^\#=\frac{2\cdot a^*+k^2\cdot\frac{w}{t_g}}{1-k^2\cdot u_{\mathrm{rel}}^2(w)} \tag{18}$$

## 9.2 Calculation of sample activity

### 9.2.1 General

To convert alpha activity on the thin source, in becquerel, into derived activity, a conversion factor, $w_1$ is introduced. The unit of the conversion factor is the inverse unit of the sample, i.e. per kilogram for a solid sample with a specific activity, per litre for a liquid sample with a volume activity, per square metre for a contaminated surface sampled with a wipe-test, per cubic metre for a volume activity of particles in air or gases sampled with a filter or other per one unit for specific thin test samples directly measured by alpha spectrometry. In that last case, the uncertainty of the unit is null.

If relevant, the conversion factor shall include specific corrections and associated uncertainty such as sampling efficiency of air or when derived activity per unit of volume is converted by dividing by the density $\rho$ in kilogram per litre, for example.

Combined absolute or relative uncertainties are calculated in accordance with ISO/IEC Guide 98-3.

The characteristic limits, decision threshold, detection limit and coverage interval limits are calculated in accordance with ISO 11929-1.

### 9.2.2 Calculation of derived alpha activity

The derived activity of the radionuclide of interest is given by Formula (19):

$$a_j = a \cdot w_1 \tag{19}$$

Table 7 gives examples of derived activity calculations.

**Table 7 — Examples of derived activity calculations**

| Derived activity $a_j$ | Expression of $w_1$ | Description |
|---|---|---|
| for specific activity for solid sample (Bq·kg$^{-1}$) | $\frac{1}{m}$ | $m$ is the mass of the test sample expressed in kilogram |
| for volume activity for a liquid sample (Bq·l$^{-1}$) | $\frac{1}{V}$ | $V$ is the volume of the test sample expressed in litre |
| for volume activity of particles in air or gases sampled (Bq·m$^{-3}$) | $\frac{1}{V_{air} \cdot \varepsilon_T \cdot (1-\varepsilon_S)}$ | $V_{air}$ is the volume of air sampled expressed in cubic metre<br>$\varepsilon_T$ is the trapping efficiency of the media filter<br>$\varepsilon_S$ is the loss yield due to the deposition along the sampling time |

### 9.2.3 Relative uncertainty

The combined relative uncertainty of derived activity $a_j$ is given by Formula (20):

$$u_{rel}(a_j) = \sqrt{u_{rel}^2(a) + u_{rel}^2(w_1)} \tag{20}$$

### 9.2.4 Decision threshold

The decision threshold $a_j^*$ is given by Formula (21):

$$a_j^* = a^* \cdot w_1 \tag{21}$$

### 9.2.5 Detection limit

The detection limit $a_j^{\#}$ is given by Formula (22):

$$a_j^{\#} = a_j^{*} \cdot \frac{\frac{a^{\#}}{a^{*}} \cdot \left[1 - k^2 \cdot u_{\text{rel}}^2 (w_1)\right]}{1 - k^2 \cdot \left[u_{\text{rel}}^2 (a) + u_{\text{rel}}^2 (w_1)\right]} \tag{22}$$

### 9.2.6 Coverage interval limits

#### 9.2.6.1 Probabilistically symmetric coverage interval

The lower $a_j^{\triangleleft}$ and upper $a_j^{\triangleright}$ coverage interval limits are calculated using Formulae (23) and (24) (see ISO 11929-1):

$$a_j^{\triangleleft} = a_j \left[1 - k_p \cdot u_{\text{rel}} \left(a_j\right)\right] \; ; p \; = \; \omega \; \cdot \; (1 - \gamma / 2) \tag{23}$$

$$a_j^{\triangleright} = a_j \left[1 + k_q \cdot u_{\text{rel}} \left(a_j\right)\right] \; ; q \; = \; 1 - \omega \; \cdot \; \gamma / 2 \tag{24}$$

where

$\omega = \phi \left( \frac{a_j}{u\left(a_j\right)} \right)$, $\phi$ being the distribution function of the standardized normal distribution;

$(1 - \gamma / 2)$ is the probability for the coverage interval of the measurand;

$\omega = 1$ may be set if $a_j \geq 4 \cdot u\left(a_j\right)$.

In this case, the probabilistic coverage interval is symmetric and given by Formula (25):

$$a_j^{\triangleleft}, a_j^{\triangleright} = a_j \cdot \left[1 \pm k_{1-\gamma/2} \cdot u_{\text{rel}} \left(a_j\right)\right] \tag{25}$$

$\gamma = 0{,}05$ and then $k_{1-\gamma/2} = 1{,}96$ is often chosen by default.

#### 9.2.6.2 Shortest coverage interval

The lower limit of the shortest coverage interval, $a_j^{<}$, and the upper limit of the shortest coverage interval, $a_j^{>}$, calculated from a primary measurement result, $a_j$, of the measurand and the standard uncertainty, $u\left(a_j\right)$, associated with $a_j$, are given by Formulae (26) or (27) (see ISO 11929-1):

$$a_j^{<}, a_j^{>} = a_j \cdot \left[1 \; \pm \; k_p \; \cdot u_{\text{rel}} \left(a_j\right)\right]; \; p = [1 + \omega \; \cdot \; (1 - \gamma)] / 2 \tag{26}$$

Or if $a_j^{<} < 0$, then $a_j^{<} = 0$ and

$$a_j^{>} = a_j \cdot \left[1 \; \pm \; k_q \; \cdot u_{\text{rel}} \left(a_j\right)\right]; \; q = 1 - \omega \cdot \gamma \tag{27}$$

$\omega = \phi \left[ \frac{a_j}{u\left(a_j\right)} \right]$, $\phi$ being the distribution function of the standardized normal distribution.

The relation $0 \leq a_j^{<} < a_j^{>}$ apply and the approximation of Formula (25) is valid.

## 10 Test report

The test report shall conform to ISO/IEC 17025 requirements. It shall contain the following information:

a) reference to this document, i.e. ISO 23548:2024;

b) identification of the sample;

c) units in which the results are expressed;

d) test result:

   1) when the derived alpha activity, $a_\mathrm{j}$, is compared with the decision threshold:

      — if the result is less than the decision threshold, the result of the measurement is expressed as $\leq a_\mathrm{j}^*$;

      — if the result is greater than the decision threshold, the result of the measurement is expressed as $a_\mathrm{j} \pm u(a_\mathrm{j})$ or $a_\mathrm{j} \pm U(a_\mathrm{j})$ with the associated $k$ value;

   2) when the derived alpha activity, $a_\mathrm{j}$ is compared with the detection limit;

      — if the result is less than the detection limit, the result of the measurement is expressed as $\leq a_\mathrm{j}^\#$:

      — if the result is greater than the detection limit, the result of the measurement is expressed as $a_\mathrm{j} \pm u(a_\mathrm{j})$ or $a_\mathrm{j} \pm U(a_\mathrm{j})$ with the associated $k$ value.

Complementary information can be provided such as:

— the uncertainty can also be expressed as the limits of the probabilistically symmetric coverage interval and/or the limits of the shortest coverage interval;

— probabilities $\alpha$, $\beta$ and $(1-\gamma/2)$;

— decision thresholds and the detection limits;

— if the detection limit exceeds the guideline value, it shall be documented that the method is not suitable for the measurement purpose;

— mention of any relevant information likely to affect and/or to explain the results;

NOTE Occasionally, it is requested by the customer or regulator to compare the primary measurement results, $a_\mathrm{j}$, with the respectively detection limit, $a_\mathrm{j}^\#$, in order to decide whether the physical effect is recognized or not. Such stipulations are not in accordance with ISO 11929-1. They have the consequence that it is decided too frequently that the physical effect is absent when in fact it is not absent.

# Annex A
(informative)

# Generic alpha test method processes

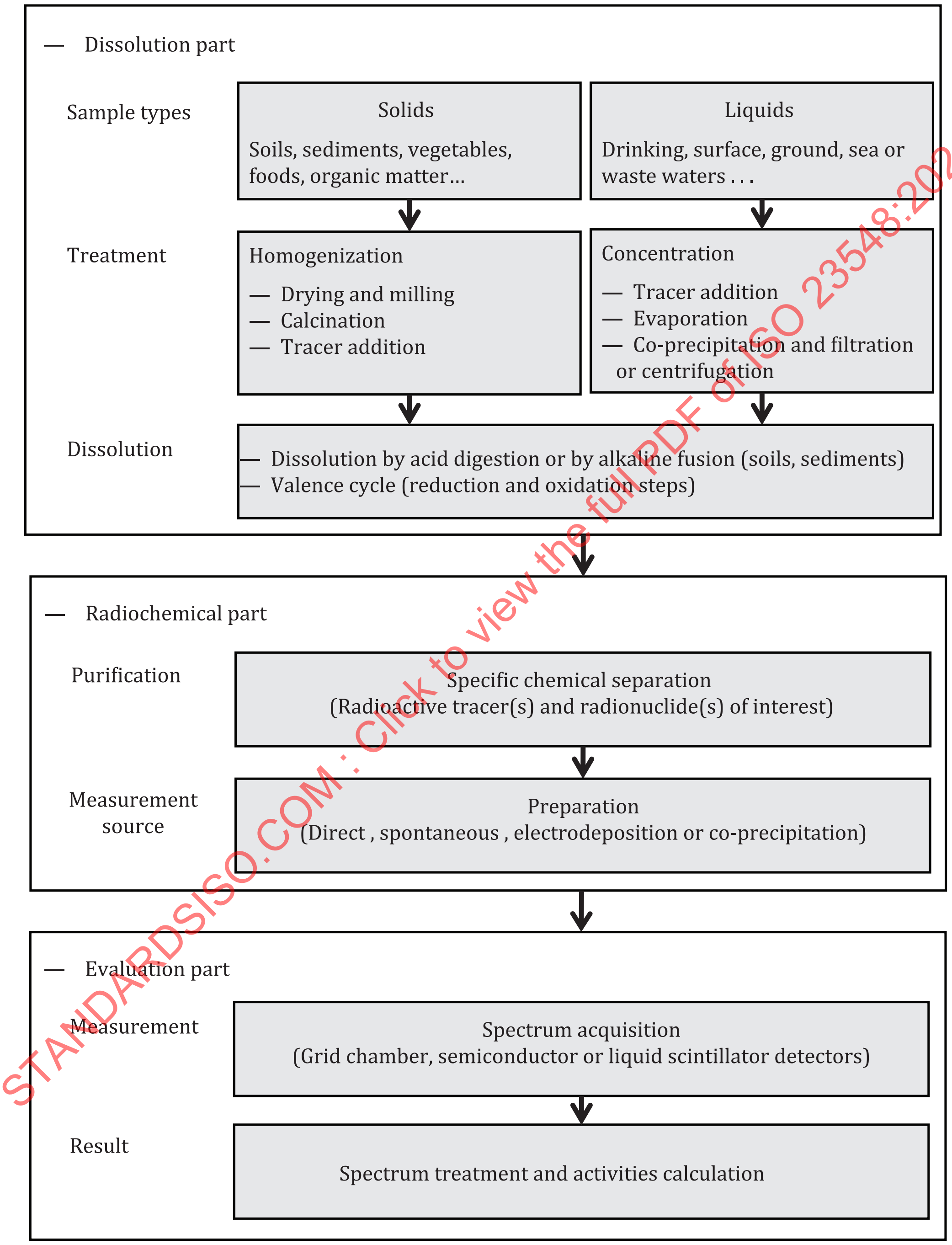


**Figure A.1 — Schematic diagram of generic alpha test method processes**

# Annex B
(informative)

# Examples of melting flux

### Table B.1 — Common flux and melting conditions for various kinds of samples (Reference [25])

| **Flux** mp[a], °C | **Fusion temperature °C** | **Type of crucible** | **Types of decomposed sample** |
|---|---|---|---|
| $Na_2S_2O_7$<br>or<br>$K_2S_2O_7$ | Up to red heat | Pt, quartz, porcelain | For insoluble oxides and oxide containing samples, particularly those of Al, Be, Ta, Ti, Zr, Pu, and the rare earths. |
| NaOH (321)<br>or<br>KOH (404) | 450 - 600 | Ni, Ag, glassy carbon | For silicates, oxides, phosphates, and fluorides. |
| $Na_2CO_3$ (853) or<br>$K_2CO_3$ (903) | 900 - 1 000 | Ni<br>Pt for short periods (use lid) | For silicates and silica containing samples (clays, minerals, rocks, glasses), refractory oxides, quartz, and insoluble phosphates and sulfates. |
| $Na_2O_2$ | 600 | Ni, Ag, Au, Zr<br>Pt (<500 °C) | For sulfides; acid-insoluble alloys of Fe, Ni, Cr, Mo, W, and Li; Pt alloys; Cr, Sn, and Zn minerals. |
| $H_3BO_3$ | 250 | Pt | For analysis of sand, aluminum silicates, titanite, natural aluminum oxide (corundum), and enamels. |
| $Na_2B_4O_7$ (878) | 1 000 - 1 200 | Pt | For $Al_2O_3$; $ZrO_2$ and zirconium ores, minerals of the rare earths, Ti, Nb, and Ta, aluminum containing materials; iron ores and slags. |
| $Li_2B_4O_7$ (920)<br>or<br>$LiBO_2$ (845) | 1 000 - 1 100 | Pt, graphite | For almost anything except metals and sulfides. The tetraborate salt is especially good for basic oxides and some resistant silicates. The metaborate is better suited for dissolving acidic oxides such as silica and $TiO_2$ and nearly all minerals. |
| $NH_4HF_2$ (125),<br>NaF (992),<br>KF (857)<br>or<br>$KHF_2$ (239) | 900 | Pt | For the removal of silicon, the destruction of silicates and rare earth minerals, and the analysis of oxides of Nb, Ta, Ti, and Zr. |

[a] Melting point.

# Annex C
(informative)

# Preparation of the source by coprecipitation

## C.1 Principle

The isotopes of interest are recovered by coprecipitation using lanthanum or cerium fluorides.

## C.2 Equipment

Usual laboratory equipment and in particular the following:

**C.2.1 Membrane filter**, membrane filter with a diameter adapted for alpha spectrometry and filtration system, pore diameter (see 6.3.4):

- 0,1 µm for uranium, thorium and radium isotopes and
- 0,22 µm for other actinides such as americium, curium, plutonium and, neptunium isotopes.

**C.2.2 Test dish or stainless-steel disk** with a diameter adapted to the spectrometry device.

**C.2.3 Filtration system.**

**C.2.4 Petri dishes.**

**C.2.5 Gamma spectrometry** for the determination of the chromatographic separation yield of radium using the $^{133}$Ba tracer.

## C.3 Reagents

Unless otherwise stated, use only reagents of recognized analytical grade and water complying with ISO 3696, grade 3 or equivalent purity (e.g. distilled or demineralized waters).

**C.3.1 Laboratory water**, used as a blank, as free as possible of chemical or radioactive impurities specially for radionuclides of interest, complying with ISO 3696, grade 3 or equivalent purity, e.g. distilled or demineralized water.

**C.3.2 Hydrochloric acid**, $c(\text{HCl}) = 0,2\ \text{mol}\cdot\text{l}^{-1}$.

**C.3.3 Concentrated ammonia**, $w(\text{NH}_4\text{OH}) = 25\ \%$ mass fraction.

**C.3.4 Lanthanum oxide**, $w(\text{La}_2\text{O}_3) = 99,999\ \%$ mass fraction.

**C.3.5 Cerium (III) nitrate hexahydrate**, $w(\text{Ce(NO}_3)_3\cdot 6\text{H}_2\text{O}) = 99,999\ \%$ mass fraction.

**C.3.6 Hydrofluoric acid**, $w(\text{HF}) = 40\ \%$ mass fraction.

**C.3.7 Ammonium fluoride solution**, $w(\text{NH}_4\text{F}) = 10\ \%$ mass fraction.

**C.3.8 Lanthanum carrier solution**, 29 mg of lanthanum oxide (C.3.4) dissolved in 100 ml of 0,2 molar HCl (C.3.2).

**C.3.9 Cerium carrier solution**, 155 mg of cerium (III) nitrate hexahydrate (C.3.5) dissolved in 100 ml of water (C.3.1).

**C.3.10 Sodium nitrite**, ($NaNO_2$), crystallized.

**C.3.11 Titanium trichloride solution**, $w(TiCl_3) = 15\ \%$ mass fraction.

**C.3.12 Ethanol** 80 %.

**C.3.13 $^{133}$Ba tracer** (~50 $Bq{\cdot}ml^{-1}$).

**C.3.14 Ammonium sulfate**, $(NH_4)_2SO_4$.

**C.3.15 Barium chloride dihydrate**, $BaCl_2{\cdot}2H_2O$.

**C.3.16 Barium carrier** (0,75 $mg{\cdot}ml^{-1}$).

Dissolve 338 mg reagent grade barium chloride, dihydrate (C.3.15) in water (C.3.1) and dilute to 250 ml with water (C.3.1).

**C.3.17 Isopropanol**.

## C.4 Procedures

### C.4.1 Uranium, Thorium

This procedure is based on the References [25], [26] and [27].

Using the fraction obtained from the chemical separation, carry out, for example, the following operations in HCl solution.

- Set up the filter and wash it with 5 ml 80 % ethanol (C.3.12);
- Add 0,2 ml of the cerium carrier solution (C.3.9);
- Add 0,5 ml of the titanium trichloride solution (C.3.11);
- Add 1 ml of HF (C.3.6) or 1 ml of $NH_4F$ (C.3.7) solutions;
- Adjust the pH to 1,9 by adding $NH_4OH$ (C.3.3);
- Stir, then leave to settle for 15 min;
- Recover the precipitate by light vacuum filtering (C.2.3) on the membrane filter (C.2.1) without forgetting to collect the deposit remaining on the side of the beaker in water (C.3.1), and wash the filtered material with 5 ml ethanol (C.3.12);
- Leave the precipitate to dry in open air, then attach the membrane filter to the test dish or stainless-steel disk (C.2.2).

The source is ready to be measured by alpha spectrometry.

### C.4.2 Americium, Curium, Plutonium, Neptunium

Using the fraction obtained from chromatographic elution, re-extraction or oxalate decomposition and carrying out the following operations:

- Adjust the pH to 1,9 by adding $NH_4OH$ (C.3.3);
- Add, for a deposit of 20 $cm^2$, the equivalent of 250 µg of lanthanum, (e.g. 1 ml of lanthanum carrier solution (C.3.8) or 0,5 ml of cerium carrier solution (C.3.9);
- Add 1 ml of HF (C.3.6) or $NH_4F$ solutions (C.3.7);
- Adjust plutonium to the oxidation state IV by adding several milligrams of $NaNO_2$ (C.3.10);
- Stir, then leave to settle for 10 min;
- Recover the precipitate by light vacuum filtering (C.2.3) on the membrane filter (C.2.1) without forgetting to collect the deposit remaining on the side of the beaker in water (C.3.1), and wash the filtered material with 5 ml ethanol (C.3.12);
- Leave the precipitate to dry in open air, then attach the membrane to the test dish or stainless-steel disk (C.2.2).

The source is ready to be measured by alpha spectrometry.

### C.4.3 Radium

This procedure is based on the References [28] and [29].

Using the fraction obtained from chromatographic separation elution (LN Resin, $MnO_2$ resin, DGA resin, ...) carrying out the following operations:

- Count vials containing Ra/Ba fractions for $^{133}Ba$ by gamma spectrometry for tracer recovery;
- Add 3,0 g of ammonium sulfate (C.3.14) to the Ra/Ba sample tubes. Mix to dissolve;
- Add 100 µl of barium carrier (C.3.16) to each sample. Swirl to mix;
- Add 5 ml of isopropanol (C.3.17) to each sample. Swirl to mix;
- Place tubes in an ‘ice-cold’ water bath for at least 30 minutes;
- Prepare alpha spectrometry sources on resolve filters as outlined in Method SPA01 (Reference [30]);

**Option 1:** Place a 0,1 µm/25 mm polypropylene filter (C.2.1) on a filter apparatus (C.2.3), with 50 ml polysulfone funnel. Insert the stem of the filter apparatus into a vacuum box outer tip. Insert the outer tip into a hole on a hole vacuum box. Repeat for each filter assembly available.

**Option 2:** Make sure that an 0,1 µm/25 mm polypropylene filter (C.2.1) in disposable funnel assembly is fit together tightly with the filter properly set in the bottom of the funnel. Insert the stem of the disposable filter into an outer tip. Insert the outer tip into a hole on a hole of vacuum box. Repeat for each filter assembly to be used.

NOTE The vacuum box assembly in either option 1 or 2 can be run with individual 50 ml plastic centrifuge tubes in a vacuum box rack below each filter or a single vacuum box inner liner. Either option is normally effective. However, using individual 50 ml centrifuge tubes provides additional insurance, should issues arise during the filtration of the $BaSO_4$ micro precipitate.

- Apply vacuum (~34 kPa). Add 3-5 ml of ethanol (C.3.12) to wet each filter. Make sure that there are no leaks along the sides of the filter assembly. Allow all liquid to pass through filter;
- Add 2-3 ml of water (C.3.1) to each filter. Allow all liquid to pass through filter;