
**Nuclear fuel technology — Preparation of
plutonium sources and determination of
 $^{238}\text{Pu}/^{239}\text{Pu}$ isotope ratio by alpha
spectrometry**

*Technologie du combustible nucléaire — Préparation des sources de
plutonium et détermination du rapport isotopique $^{238}\text{Pu}/^{239}\text{Pu}$ par
spectrométrie alpha*

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ISO copyright office
Case postale 56 • CH-1211 Geneva 20
Tel. + 41 22 749 01 11
Fax + 41 22 749 09 47
E-mail copyright@iso.org
Web www.iso.org

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Foreword

ISO (the International Organization for Standardization) is a worldwide federation of national standards bodies (ISO member bodies). The work of preparing International Standards is normally carried out through ISO technical committees. Each member body interested in a subject for which a technical committee has been established has the right to be represented on that committee. International organizations, governmental and non-governmental, in liaison with ISO, also take part in the work. ISO collaborates closely with the International Electrotechnical Commission (IEC) on all matters of electrotechnical standardization.

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The main task of technical committees is to prepare International Standards. Draft International Standards adopted by the technical committees are circulated to the member bodies for voting. Publication as an International Standard requires approval by at least 75 % of the member bodies casting a vote.

Attention is drawn to the possibility that some of the elements of this document may be the subject of patent rights. ISO shall not be held responsible for identifying any or all such patent rights.

ISO 11483 was prepared by Technical Committee ISO/TC 85, *Refrigeration and air-conditioning*, Subcommittee SC 5, *Nuclear fuel technology*.

This second edition cancels and replaces the first edition (ISO 11483:1994), of which it constitutes a technical revision.

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Nuclear fuel technology — Preparation of plutonium sources and determination of $^{238}\text{Pu}/^{239}\text{Pu}$ isotope ratio by alpha spectrometry

1 Scope

This International Standard describes three simple and fast procedures to prepare plutonium sources and a procedure to measure the activity ratio of ^{238}Pu to ($^{239}\text{Pu} + ^{240}\text{Pu}$) by α spectrometry. The α spectrometry method is used for the determination of isotopic abundance of ^{238}Pu in combination with mass spectrometry and eliminates the possible interferences of ^{238}U in the latter method. It applies to the analysis of purified solutions of plutonium in 2 mol/l to 4 mol/l nitric acid containing 50 μg to 200 μg of plutonium per millilitre, as may result from the chemical treatment and purification preceding plutonium isotopic analysis by mass spectrometry.

This International Standard applies to plutonium solutions free from ^{241}Am and those containing less than 10 % of other non-volatile impurities relative to the plutonium content. Anion exchange chromatography in accordance with ISO 8299:1993^[12], Subclauses 7.2 and 7.3, or solvent extraction chromatography in ISO 15366^[13] are suitable purification methods.

The methods given in this International Standard are intended for use in conjunction or in parallel with mass spectrometry for the isotopic analysis of plutonium in spent-fuel solutions or nuclear-grade plutonium products.

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2 Principle

There are two major methods to prepare plutonium sources: drop deposition and electrodeposition. Two types of drop-deposition methods are given: a drop of sample solution is mixed with wetting agent and deposited on a electrochemically polished stainless-steel disc or porcelain disc and dried. The dried residue is ignited in a flame or muffle furnace depending upon the selected types of disc to produce a thin and carrier-free source.

In the case of the electrodeposition method, the plutonium is deposited as hydrous oxide from a buffered, slightly acidic aqueous solution onto a stainless-steel disc.

The source is measured by α spectrometry with a semiconductor detector.

The spectrum is processed to obtain the activity ratio of ^{238}Pu to ($^{239}\text{Pu} + ^{240}\text{Pu}$). The tailing of the ^{238}Pu peak is corrected by the “Geometric-Progression Decrease” method or exponential extrapolation method.

The isotope ratio $^{238}\text{Pu}/^{239}\text{Pu}$ is calculated using the isotope ratio $^{240}\text{Pu}/^{239}\text{Pu}$ determined by mass spectrometry.

3 Source preparation

The source should be very thin. Its thickness should not significantly limit the α peak resolution.

Different procedures can be used to prepare α sources, such as

- sublimation in vacuum,
- electrodeposition,
- solution evaporation with a wetting agent and subsequent ignition, or
- coprecipitation of Pu(III)F_3 with lanthanide fluoride, such as NdF_3 or CeF_3

Sublimation is the preferred procedure when the aim is to study the fine structure of the plutonium α spectrum and an energy resolution of 9 keV is needed [1]. It is possible, but more difficult, to reach such a resolution with sources prepared by electrodeposition [2]. Coprecipitation is well suited to determine trace contents of plutonium in environmental or biological samples by α spectrometry [3]. However, sources prepared by coprecipitation exhibit α peaks with greater tailing.

Depositing and drying a small drop of solution of the purified sample onto an inert support produce satisfactory sources for the purpose of plutonium accountancy measurements. Two examples of the procedure of drop-deposition are described below. These alternatives were selected because they are convenient and in common use when the sample is measured in parallel by α and mass spectrometry. It is also easy, with these procedures, to prepare five to twenty sources simultaneously. Drop deposition yields sources of appropriate properties when using semiconductor detectors with an intrinsic energy resolution of 20 keV or less for 5,50 MeV alpha-particle energy of ^{238}Pu .

The source support shall have a clean and perfectly smooth surface, which shall remain physically and chemically inert during the preparation of the sources. In particular, the material of the support shall not melt and shall be resistant against nitric acid, plutonium nitrate, the wetting agent and their decomposition products, as well as against atmospheric corrosion.

3.1 Source preparation on electrochemically polished stainless-steel disc [4]

In the following procedure, the sources shall be prepared in a fume cupboard or a glove box designed for work with α -emitting radioisotopes where the use of a gas burner is authorized.

3.1.1 Apparatus

3.1.1.1 Electrochemically polished discs, of heat-resistant stainless steel, about 25 mm in diameter and 0,5 mm thick; stainless steels of the following composition would be suitable: Cr 17 % to 20 %; Ni 8 % to 15 %; Mo 2,5 % to 4 %; C 0,12 % max.; Si 1,0 % max.; Mn 2,0 % max.; S 0,030 % to 0,045 %; P 0,045 %.

3.1.1.2 Burner

3.1.1.3 Piston pipette, of capacity 10 μl , with disposable tip.

3.1.1.4 Sample holder, made of a 50 mm diameter and 5 mm thick stainless-steel plate with a 25 cm long insulated handle. A 0,2 mm to 0,3 mm deep recess shall be provided in the centre of the plate with a diameter 1 mm greater than the diameter of the disc.

3.1.2 Reagents

3.1.2.1 Ethanol, analytical grade.

3.1.2.2 Colourless varnish

3.1.2.3 Tetraethylene glycol, analytical grade.

3.1.3 Procedure

3.1.3.1 Clean the disc with the ethanol (3.1.2.1).

3.1.3.2 Draw a circular rim, of about 12 mm diameter, on the polished surface of the disc with a varnish (3.1.2.2) and let it dry.

NOTE An artist's brush can be useful for this operation.

3.1.3.3 Place the disc in the recess of the sample holder.

3.1.3.4 Pipette 10 µl of the tetraethylene glycol (3.1.2.3) inside the circular rim.

3.1.3.5 Change the pipette tip and pipette 10 µl of the sample solution inside the circular rim.

3.1.3.6 Gently heat the holder and the disc with a gas burner, swirling slowly over the flame to mix the sample well with the tetraethylene glycol. The solution should be evaporated slowly, without boiling.

3.1.3.7 When the disc is dry, heat more strongly to burn off the rim of varnish.

3.1.3.8 Finally, introduce the disc into the reducing zone of the flame and bring the source to a dull red heat for approximately 30 s.

3.1.3.9 Let the holder and the disc cool to ambient temperature. The source is ready for measurement.

3.2 Source preparation on porcelain disc ^[5]

3.2.1 Apparatus

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3.2.1.1 Glazed porcelain disc, of about 29 mm diameter and 5,5 mm thickness. A circular recess of 8,5 mm diameter and 1 mm depth shall be provided in the centre of the disc to receive the sample solution, which defines the source area. The glazing shall not melt below 1 000 °C.

3.2.1.2 Hotplate, with a maximum temperature of 250 °C.

NOTE A surface thermometer may be used to monitor the temperature of the plate.

3.2.1.3 Muffle furnace, controllable to 1 000 °C and equipped with a removable quartz boat or tube, about 100 mm to 200 mm long and 30 mm in diameter. The boat shall be wide enough to accept the porcelain discs.

3.2.1.4 Piston pipette, of capacity 10 µl, with disposable tip.

3.2.1.5 Pencil, to mark the porcelain disc. The marking shall still be readable after ignition at 850 °C.

3.2.2 Reagents

3.2.2.1 Ethanol, analytical grade.

3.2.2.2 Tetraethylene glycol, analytical grade.

3.2.3 Procedure

3.2.3.1 Clean the surface of the porcelain disc using a tissue paper moistened with ethanol (3.2.2.1). Write the identification of the sample with a pencil (3.2.1.5) on the reverse side of the disc.

3.2.3.2 Pipette 10 µl, of tetraethylene glycol (3.2.2.2) into the recess in the centre of the porcelain disc and spread it, with the pipette tip, over the entire surface of the recess.

3.2.3.3 Change the pipette tip and pipette 10 µl of the sample solution into the recess in the centre of the disc, previously moistened with tetraethylene glycol; mix both solutions well with the tip of the pipette.

3.2.3.4 Place the porcelain disc on the hotplate (3.2.1.2), set the temperature to 100 °C and warm it up for 1 h; then increase the temperature to 150 °C for another hour; after 2 h, the disc should be dry.

3.2.3.5 Introduce the porcelain disc into the quartz tube or boat of the muffle furnace (3.2.1.3), set the temperature control to 850 °C and ignite at this temperature for 20 min; the furnace reaches 850 °C in about 20 min.

3.2.3.6 Switch off the furnace and let it cool for about 20 min before removing the source.

The source is ready for measurement.

3.3 Source preparation by electrodeposition [2]

3.3.1 Apparatus

3.3.1.1 Stainless-steel discs, as described in 3.1.1.1. The diameter has to fit the electroplating cell.

3.3.1.2 Device for electroplating, with adjustable voltage of up to 12 V and adjustable current of up to 200 mA/cm² of area of deposition. The anode must be rotating to serve as a stirrer.

3.3.1.3 Disposable electroplating cell, of 10 ml capacity and diameter compatible with the diameter of the stainless-steel discs (3.1.1.1).

3.3.1.4 Pt-anode

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3.3.2 Reagents

3.3.2.1 Nitric acid, analytical grade, concentrated, 3 mol/l, 0,1 mol/l, 0,001 mol/l.

3.3.2.2 H₂O₂, analytical grade, 30 %.

3.3.2.3 Ammonium nitrate, analytical grade, saturated solution in distilled water.

3.3.2.4 Ammonia solution, analytical grade, 5 % in distilled water.

3.3.2.5 Acetone, analytical grade.

3.3.3 Procedure

3.3.3.1 A redox treatment is necessary to ensure isotopic homogenization if the Pu was mixed from different origins. This can be done by first treating the mixture twice with concentrated nitric acid (HNO₃) to break the polymer and then adding H₂O₂ in 3 mol/l HNO₃ medium. Then evaporate to dryness and redissolve in 0,1 mol/l HNO₃ to adjust the concentration to about 1 µg Pu/10 µl.

3.3.3.2 Add 5 ml of 0,001 mol/l HNO₃ to the electrodeposition cell. Transfer 10 µl of the Pu solution to the cell.

3.3.3.3 Connect the mirror-polished stainless-steel disc and the platinum stirrer to act as cathode and anode, respectively. It is important to stir the solution during electrodeposition, since there is gas evolution during the process.

3.3.3.4 Now start the electrodeposition by maintaining a voltage of 10 V to 12 V and adjusting the current density to about 200 mA/cm² by the addition of saturated solution of ammonium nitrate.

3.3.3.5 Run the deposition for about 1 h.

3.3.3.6 Before stopping the electrodeposition, add a few drops of diluted ammonia solution.

3.3.3.7 Remove the electrical connections and wash the electrodeposited source with water and acetone and keep for drying.

3.3.3.8 Wash the Pt electrode thoroughly with concentrated nitric acid (HNO₃). Always use a fresh cell for a new source.

4 Alpha spectrometry

4.1 Apparatus

4.1.1 Vacuum chamber, equipped with a vacuum gauge and a pump providing pressures of 5 Pa or less.

4.1.2 Si detector, surface barrier passivated or ion-implanted, energy resolution (FWHM) 20 keV or better, active area about 100 mm².

Passivated ion-implanted detectors have a resolution of 9 keV to 10 keV, but are not required unless the source is prepared by vacuum sublimation or electrodeposition and the aim is to reach a much better accuracy or to study the fine structure of the plutonium α spectrum.

The detector shall be mounted in the vacuum chamber at a distance of about 15 mm from the α source, i.e. about 1,5 times the diameter of the source. <https://standards.iteh.ai/catalog/standards/sist/23f8bebc-e748-4b37-8aca-5cb1ac14375d/iso-11483-2005>

4.1.3 Electronic modules

Bias voltage supply, adjustable up to 200 V, preamplifier and spectrometry amplifier, 1 024 or more memory multichannel analyser. The amplifier gain is adjusted in such a way that the peaks of ²³⁸Pu and ²³⁹Pu + ²⁴⁰Pu are displayed on the upper energy side of the screen; the energy scale should be about 1,5 keV/channel for the 1 024 multichannel analyser.

4.1.4 Small computing device, separate or incorporated in the multichannel analyser.

4.2 Measurement

4.2.1 Place the source in the vacuum chamber and pump until a vacuum of about 5 Pa (5×10^{-2} mbar) is obtained. Then set the detector voltage to the value recommended by the manufacturer.

4.2.2 Record the spectrum of α particle radiation, collecting whenever practical at least 500 000 counts in the smaller of the two peak groups [corresponding to ²³⁸Pu or (²³⁹Pu + ²⁴⁰Pu) peaks]. The spectrum recorded in the semi-logarithmic scale shall be similar to the one presented in Figure 1. The upper energy sides of the two peak groups shall be smooth, while the lower energy sides shall decrease monotonically, except for the two small peaks indicated in Figure 1. At mid-height, the 5,5 MeV peak of ²³⁸Pu shall have a width of 20 keV or less. The counts per channel in the valley between the two groups shall be no greater than 1 % of the counts at the ²³⁸Pu peak top.

4.3 Spectra evaluation

A number of computer software have been developed to perform elaborate evaluations of α spectra [6, 7, 8]. Some can be used on personal computers. However, sources with very high resolution are needed to draw a

significant advantage out of such sophisticated evaluations, compared to the simple empirical procedure described in 4.3.2.

4.3.1 Principle

Very simple models are very efficient for accurately correcting the tailing of the ^{238}Pu peak, as long as the spectrum is well resolved and the tailing is small.

The following assumptions are then generally applicable:

- a) single α peaks all have the same shape;
- b) the tails are the sum of the tailings of all single peaks of higher energies;
- c) in the flat portion of the tail, peak tailing contributions are proportional to the corresponding peak areas.

The above assumptions form the basis of the procedures which are described below.

4.3.2 Geometric-progression decrease method [9]

The method involves two assumptions:

- the self absorption of the alpha particles of ^{238}Pu , ^{239}Pu and ^{240}Pu is the same;
- the counts in the tail of a peak decrease in geometric progression.

Its main features are as follows:

- a) equal intervals of integration are selected for the ^{238}Pu and ($^{239}\text{Pu} + ^{240}\text{Pu}$) peaks;
- b) each interval is selected to include all the relevant single peaks, which contribute at least 0,05 % of the total activity of the group of interest;
- c) the intervals of integration of the ^{238}Pu peaks include only contributions from ^{238}Pu (and ^{241}Am) isotopes;
- d) the interval of integration of the ($^{239}\text{Pu} + ^{240}\text{Pu}$) peaks excludes all the major ^{242}Pu and ^{241}Pu peaks;
- e) tail-contribution estimates also exclude the contributions of the major ^{242}Pu and ^{241}Pu peaks and the minor ^{238}Pu peak at 5,357 9 MeV.

4.3.2.1 Divide the plutonium α -spectrum into four regions: A, B, C and D, each region having an equal number of channels (see Figure 1).

4.3.2.2 Select the high-energy end of region A: $E_{\text{max}}^A = 5,499\ 0\ \text{MeV} + \text{FWHM}$, where FWHM(MeV) is the full width at half-maximum of the ^{238}Pu peak at the energy 5,499 0 MeV.

4.3.2.3 Fix the low-energy end of the region A: $E_{\text{min}}^A = E_{\text{max}}^A - 0,267\ 7\ \text{MeV}$.

4.3.2.4 Select the high-energy and the low energy ends of region B: $E_{\text{max}}^B = 5,168\ 2\ \text{MeV} + \text{FWHM}$ and $E_{\text{min}}^B = E_{\text{max}}^B - 0,267\ 7\ \text{MeV}$.

4.3.2.5 Select the high-energy and low-energy ends of the region C: $E_{\text{max}}^C = 4,856\ 2\ \text{MeV} - \text{FWHM}$ and $E_{\text{min}}^C = E_{\text{max}}^C - 0,267\ 7\ \text{MeV}$.

4.3.2.6 Select the high-energy and low-energy ends of the region D:

$$E_{\text{max}}^D = E_{\text{max}}^B - (E_{\text{max}}^A - E_{\text{max}}^C) \text{ and } E_{\text{min}}^D = E_{\text{max}}^D - 0,267\ 7\ \text{MeV}.$$

4.3.2.7 The interval width 0,267 7 MeV is the energy interval between the ^{240}Pu peak at 5,168 2 MeV and the ^{242}Pu peak at 4,900 5 MeV (see Table 1).

4.3.2.8 Obtain the sums of the counts in the channels of regions A, B, C and D. Call the corresponding counts a , b , c and d respectively.

4.3.2.9 The $^{238}\text{Pu}/(^{239}\text{Pu} + ^{240}\text{Pu})$ alpha activity ratio, $R(\alpha)$, is calculated from

$$R(\alpha) = \frac{a}{b - \frac{ac}{b + \frac{ad}{c}}} \quad (1)$$

See reference [9] for more details.

4.3.3 Exponential decrease method

4.3.3.1 Divide the plutonium α spectrum into two regions: A and B as described in 4.3.2 (see Figure 1).

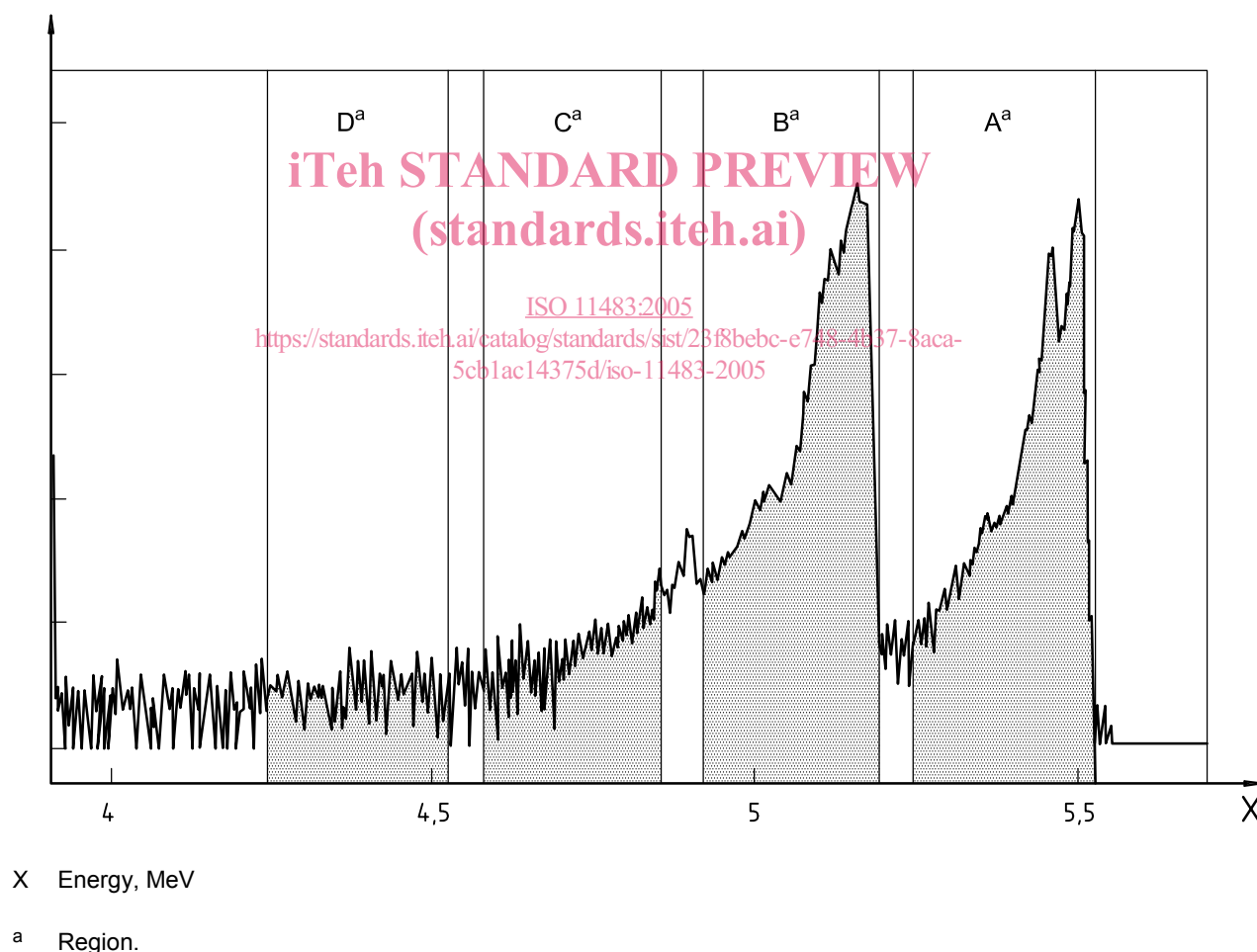


Figure 1 — Example of α spectrum

4.3.3.2 Use the 96 keV interval above region B for fitting an exponential function

$$Y_i = A_0 \cdot e^{a_1 X_i} \quad (2)$$