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Nuclear fuel technology — Alpha spectrometry —

Part 1: Determination of neptunium in uranium and its compounds

Technologie du combustible nucléaire — Spectrométrie alpha —

Partie 1: Détermination du neptunium dans l'uranium et ses composés

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Foreword

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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.

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ISO 21847-1 was prepared by Technical Committee ISO/TC 85, *Nuclear energy*, Subcommittee SC 5, *Nuclear fuel technology*.

ISO 21847 consists of the following parts, under the general title *Nuclear fuel technology — Alpha spectrometry*:

— *Part 1: Determination of neptunium in uranium and its compounds*

— *Part 2: Determination of plutonium in uranium and its compounds*

— *Part 3: Determination of uranium 232 in uranium and its compounds*

Nuclear fuel technology — Alpha spectrometry —

Part 1: Determination of neptunium in uranium and its compounds

1 Scope

This part of ISO 21847 describes a method for determining trace amounts of ^{237}Np in uranium hexafluoride, uranium oxides or uranyl nitrate.

2 Normative references

The following referenced documents are indispensable for the application of this document. For dated references, only the edition cited applies. For undated references, the latest edition of the referenced document (including any amendments) applies.

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

3 Principle

UF_6 samples are hydrolyzed, and solid samples are dissolved.

Pu(VI) and Pu(IV) are reduced to Pu(III) and Np to Np(IV) by hydroxylamine hydrochloride. The Np(IV) is then extracted by thenoyltrifluoroacetone and measured by alpha spectrometry.

^{239}Np can be used as a tracer to determine the separation efficiency.

4 Reagents

Use reagents of recognized analytical grade.

4.1 **Water**, at least quality 1 as specified in ISO 3696.

4.2 **Nitric acid**, concentrated, $c(HNO_3) = 15{,}8$ mol/l.

4.3 **Nitric acid**, solution, $c(HNO_3) = 8$ mol/l.

4.4 **Nitric acid**, dilute, $c(HNO_3) = 0{,}8$ mol/l.

4.5 **Hydroxylamine hydrochloride**, solution in nitric acid, $c(HNO_3) = 1$ mol/l, $\rho(ClH_4NO) = 348$ g/l.

Prepare the solution immediately before use.

4.6 Ammonium iron(II) sulfate hexahydrate, solution in nitric acid $c(HNO_3) = 1$ mol/l, $\rho[(NH_4)_2Fe(SO_4)_2 \cdot 6\ H_2O] = 100$ g/l.

Prepare the solution immediately before use.

4.7 Thenoyltrifluoroacetone, (TTA; $C_8H_5F_3O_2S$) solution in xylene, $c(C_8H_5F_3O_2S) = 0{,}5$ mol/l.

Prepare the solution weekly.

4.8 ^{237}Np reference solution, containing 1,02 g/l obtainable from CEA/Cetama[1)].

Alternatively, dissolve certified reference material IRMM 5020, $^{237}NpO_2$, in nitric acid solution (4.3).

4.9 ^{243}Am reference solution, concentrated americium solution 10^8 Bq/l, whose specific activity in ^{243}Am is precisely given by comparison with a solution of ^{243}Am certified in activity (a standard ^{243}Am spike near 40 Bq/g can be provided by the NIST[2)]).

4.10 Anion exchange resin, 150 µm to 300 µm.

NOTE Dowex®[3)], 1 × 4 (50 to 100) or equivalent resin can be used.

4.11 ^{239}Np reference solution, prepared as follows.

a) Prepare a column containing 1 ml of resin (4.10) and pack in nitric acid solution (4.3).

b) Sample approximately 10^5 Bq to 10^6 Bq of ^{243}Am (4.9).

c) Introduce the solution into the resin column and rinse with 2 ml of nitric acid solution (4.3). Recover the first rinsing solution. (This is an ^{243}Am solution, which may be reused after ageing for five days.)

d) Rinse with 50 ml of nitric acid solution (4.3) and discard the rinsing solution.

e) Elute ^{239}Np with 10 ml of dilute nitric acid (4.4).

The volume activity of ^{239}Np of a reference solution may be about 2×10^4 Bq/ml.

4.12 Gas mixture, with a volume fraction of 90 % argon and a volume fraction of 10 % methane.

5 Apparatus

5.1 Standard laboratory equipment.

5.2 Frisch grid ionization chamber, with argon-methane gas mixture, or **semiconductor detector**.

5.3 Multichannel amplitude analyser.

5.4 Stainless steel disks, approx. 60 mm diameter, or **watch glasses**.

5.5 Epiradiator.

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1) Commission d'établissement des méthodes d'analyse of the Commissariat à l'énergie atomique (CEA), France [French Atomic Energy Commission]

2) National Institute of Standards and Technology, USA.

3) Dowex® is an example of a suitable product available commercially. This information is given for the convenience of users of this part of ISO 21847 and does not constitute an endorsement by ISO of this product.

6 Preparation of test sample

6.1 Hydrolyze the UF_6 sample and prepare uranyl solution.

6.2 Dissolve the oxide sample in nitric acid (4.2) to obtain uranium nitrate solution.

7 Procedure

7.1 Test sample

7.1.1 Place a test portion containing about 2 g of uranium in a beaker.

7.1.2 Evaporate the solution until dry.

7.1.3 Repeat three times: add 10 ml of concentrated nitric acid (4.2) to the residue, then evaporate until dry.

7.1.4 Dissolve the residue in 20 ml of dilute nitric acid (4.4).

7.1.5 If necessary, add about 2 500 Bq to 10 000 Bq of ^{239}Np using solution (4.11). Dilute to 25 ml with dilute nitric acid (4.4), then count the resulting solution by gamma spectrometry at 277 keV for 30 min. Record the measured activity of ^{239}Np as A_1.

7.1.6 Add 1 ml of ammonium iron sulfate (4.6) and 3 ml of hydroxylamine hydrochloride (4.5). Boil for 15 min, then allow to cool for 30 min.

7.2 Extraction

7.2.1 Pour the solution into a separatory funnel, rinse the beaker three times with 1 ml of dilute nitric acid (4.4).

7.2.2 Add the rinsing solutions to the test solution.

7.2.3 Add 10 ml of TTA (4.7) and agitate mechanically for 15 min, then allow to settle.

7.2.4 Allow the phases to separate.

7.2.5 Add 10 ml of dilute nitric acid (4.4) and 1 ml of hydroxylamine hydrochloride (4.5) to the organic phase agitate mechanically for 2 min, and then allow settling.

In the case of enriched uranium, up to four rinses can be required to eliminate interference by ^{234}U.

7.2.6 Allow the phases to separate.

7.2.7 If the ^{239}Np tracer is used: Combine the aqueous phases. Dilute to 25 ml and count by gamma spectrometry for 30 min. Record the measured residual activity of ^{239}Np as A_2.

7.3 Preparation of source sample

7.3.1 Place 0,2 ml of the organic phase (or the aqueous phase after stripping) on a stainless steel disk or a watch glass (5.4).

7.3.2 Evaporate under an epiradiator (5.5) until dry (about 15 min).

7.4 Measurement

7.4.1 Place the source in the Frisch grid ionization chamber or semiconductor detector (5.2).

7.4.2 Create a vacuum in the chamber, and then fill it with the gas mixture (4.12).

7.4.3 Set the multichannel analyser to the 4 MeV to 6 MeV energy band and record an alpha spectrum.

NOTE The counting time depends on the source activity and the detection system.

7.4.4 Periodically perform background counts for the same time on a stainless steel disk without any deposit.

8 Expression of results

Subtract the background count from the source sample count for the ^{237}Np peak at 4,79 MeV.

Calculate the massic activity, a_{237Np}, of ^{237}Np, expressed as becquerels per gram uranium, by Equation (1):

$$a_{237Np} = \frac{A_s \times 10^3 \times V_{ph}}{t \times \eta_e \times \eta_d \times V_{tp} \times V_s \times \rho_U} \quad (1)$$

where

A_s is the number of pulses during the counting time;

t is the counting time, expressed in seconds;

η_e is the extraction efficiency of TTA, calculated by measuring the standard prepared from a reference solution and measured under the same conditions (7.1) (~ 0,92);

η_d is the detection efficiency of the source (~ 0,1 to 0,5);

V_{tp} is the volume, expressed in millilitres, of the test portion of UF_6 hydrolyzate or uranyl nitrate;

ρ_U is the mass concentration, expressed in grams per litre, of uranium in the UF_6 hydrolyzate or uranyl nitrate;

V_s is the volume, expressed in millilitres, of the organic or aqueous phase taken for the source sample on stainless steel disk (equal to 0,2 ml);

V_{ph} is the volume, expressed in millilitres, of organic phase used for the extraction or the volume of the aqueous phase after stripping (7.2).

If ^{239}Np is used, η_e can be calculated directly from Equation (2):

$$\eta_e = \frac{A_1 - A_2}{A_1} \quad (2)$$

9 Precision and bias

Reproducibility standard deviation is 2,5 Bq/g at a level of 35 Bq/g of U.

Reproducibility standard deviation is 5,5 Bq/g at a level of 153 Bq/g of U.

A yield equal to or higher than 90 % indicates that the method is working well (that there is no bias).