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Nuclear fuel technology — Determination of the O/M ratio in MOX pellets — Gravimetric method

Technologie du combustible nucléaire — Détermination du rapport O/M dans les pastilles MOX — Méthode gravimétrique

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Foreword

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ISO 21484 was prepared by Technical Committee ISO/TC 85, *Nuclear energy*, Subcommittee SC 5, *Nuclear fuel technology*.

Nuclear fuel technology — Determination of the O/M ratio in MOX pellets — Gravimetric method

1 Scope

This International Standard describes a method for determining the oxygen-to-metal (O/M) ratio in mixed uranium-plutonium oxide $(U,Pu)O_{2 \pm X}$ pellets.

2 Principle

The $(U,Pu)O_{2 \pm X}$ sample is submitted to controlled oxidation-reduction under thermodynamic conditions designed to change the O/M ratio to a value of 2,000. The initial stoichiometric deviation, X, is determined from the sample mass difference before and after heat treatment.

3 Reactions

The principal reactions are as follows:

a) $(U,Pu)O_{2 \pm X} \pm {}^{x}/_{2}\, O_2 \rightarrow (U,Pu)O_{2,000}$

b) $(U,Pu)O_{2 \pm X} + xH_2 \rightarrow (U,Pu)O_{2,000} + xH_2O$

4 Reagents

During the analysis, unless otherwise stated, use only reagents of recognized analytical grade and distilled or demineralized water or water of equivalent purity.

4.1 Nitric acid solution, with a volume fraction of 50 %.

4.2 Purge gas.

4.2.1 Air, with a volume fraction of 99,99 % purity grade.

4.2.2 Argon.

4.2.3 Hydrogen or hydrogen/argon mixtures, with a volume fraction of 99,99 % purity grade, to which water vapour may be added to obtain an oxygen potential (ΔG_0) approaching $-420\ kJ \cdot mol^{-1}$ ($-100\ kcal \cdot mol^{-1}$).

5 Apparatus

5.1 Muffle furnace, having provision for measuring the temperature and sweeping the hearth with various gases.

5.2 **Crucibles**, made of platinum or quartz.

5.3 **Analytical balance**, with an accuracy of at least ± 0,1 mg.

6 Sampling

A representative sample (at least one pellet) shall be taken from the pellet batch for analysis.

If necessary, care shall be taken to avoid sample oxidation during the sampling procedure. Typical precautions include performing the sampling operation under argon atmosphere, placing the sample in an argon-filled bottle, etc. The laboratory performing the analyses shall establish the procedure.

7 Procedure

7.1 Preliminary test

The balance shall be checked at regular intervals using a standard weight.

7.2 Preparation of the crucibles

The crucibles shall be clean and weighed before use. An example of a cleaning procedure that could be used is as follows:

7.2.1 Clean the crucibles by heating them in nitric acid (4.1) to a temperature near the boiling point.

7.2.2 Rinse thoroughly with deionized water.

7.2.3 Dry the crucibles in the furnace for 30 min at 150 °C under argon atmosphere.

7.2.4 Allow the crucibles to cool to 35 °C in the furnace under argon atmosphere.

7.2.5 Remove each crucible from the furnace and weigh it to within ± 0,1 mg.

7.2.6 Record the crucible mass, m_0, in grams.

7.3 Weighing of the sample

7.3.1 Place the pellets, sampled as indicated in Clause 6, in an empty crucible.

7.3.2 Weigh the crucible containing the pellets to within ± 0,1 mg.

7.3.3 Record the mass, m_1, in grams, of the crucible containing the pellets.

7.4 Heat treatment cycles

7.4.1 General

The sample shall be heat treated in such a way that the O/M ratio is changed to exactly 2,000. The duration of the oxidation-reduction cycles, and the gas flow rates in the furnace, shall be optimized according to the nature of the furnace used, the number of sample pellets measured, the pellet composition, etc.

Load the furnace with the crucible containing the sample pellets. Apply the desired heat treatment cycle.

7.4.2 Example 1

7.4.2.1 Under argon sweeping, raise the temperature to 900 °C ± 30 °C and hold for 5 min.

7.4.2.2 Under air sweeping, maintain the furnace temperature for 1 min to 7 min at 900 °C ± 30 °C.

7.4.2.3 Under argon sweeping, maintain the furnace temperature for 5 min at 900 °C ± 30 °C.

7.4.2.4 Under argon +5 % H_2 sweeping, maintain the furnace temperature for 8 h to 13 h at 900 °C ± 30 °C.

7.4.2.5 Under argon +5 % H_2 sweeping, shut off the furnace heating and allow the temperature to drop to 150 °C.

7.4.3 Example 2

7.4.3.1 Heat the samples for 16 h at 800 °C, in an atmosphere comprising a volume fraction of 4 % H_2 in argon saturated with water at room temperature.

7.4.3.2 Cool under dry argon containing a volume fraction of 4 % H_2.

7.5 Final weighing

7.5.1 Remove the crucible from the furnace.

7.5.2 Weigh the crucible to within 0,1 mg.

7.5.3 Record the mass, m_2, in grams, of the crucible after oxidation-reduction heat treatment.

8 Calculation

8.1 Mean atomic mass of oxide after heat treatment

Calculate the mean atomic mass of oxide after heat treatment, when O/M = 2,000 exactly, using Equation (1):

$$A_t = \frac{[A_t(\mathrm{Pu})]\,\mathrm{Pu}\,\% + [A_t(\mathrm{U})]\,\mathrm{U}\,\% + [A_t(\mathrm{Am})]\,\mathrm{Am}\,\%}{\mathrm{Pu}\,\% + \mathrm{U}\,\% + \mathrm{Am}\,\%} + 2A_t(\mathrm{O}) \qquad (1)$$

where

A_t is the mean atomic mass of the oxide of heavy metals;

A_t(Pu) is the mean atomic mass of plutonium in the oxide;

A_t(U) is the mean atomic mass of uranium in the oxide;

A_t(Am) is the mean atomic mass of americium in the oxide;

Pu % is the mole fraction, in percentage, of plutonium in the oxide;

U % is the mole fraction, in percentage, of uranium in the oxide;

Am % is the mole fraction, in percentage, of americium in the oxide;

A_t(O) is the atomic mass of oxygen (15,999 4).

8.2 Calculation of O/M ratio

Calculate the O/M ratio using Equation (2):

$$O/M = 2{,}000 - \frac{A_t \times (m_2 - m_1)}{A_t(O) \times (m_2 - m_0)} \quad (2)$$

where

m_0 is the mass, in grams, of the empty crucible;

m_1 is the mass, in grams, of the crucible with test sample before oxidation-reduction;

m_2 is the mass, in grams, of the crucible with test sample after oxidation-reduction heat treatment;

$A_t(O)$ is the atomic mass of oxygen (15,999 4);

A_t is the mean atomic mass of heavy metals in the oxide.

9 Precision

9.1 Accuracy

Thirty determinations on stoichiometric mixed oxide pellets containing approximately 7 % plutonium give a mean O/M ratio of 2,000.

9.2 Precision

The standard deviation calculated from 30 determinations by two operators on a control sample of stoichiometric mixed oxide pellets containing approximately 7 % plutonium is better than 0,001.

9.3 Sensitivity

Typically, for a 15 g sample, a change in weight of 0,5 mg results in a change in O/M ratio of 0,001.

10 Test report

The test report shall contain the following information:

— all information necessary for the identification of the sample tested;

— reference to this International Standard;

— test method used;

— results of the test, including the results of the individual determinations and their mean, calculated as specified in Clause 8;

— any deviations from the procedure specified in this International Standard;

— any unusual features (anomalies) observed during the test;

— date of the test.