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Determination of solubility in nitric acid of plutonium in unirradiated mixed oxide fuel pellets (U, Pu) O2

Détermination de la solubilité dans l'acide nitrique du plutonium des pastilles (U, Pu) O2 de combustibles d'oxydes mixtes non irradiés

Reference number
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Foreword

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The committee responsible for this document is ISO/TC 85, *Nuclear energy, nuclear technologies and radiological protection*, Subcommittee SC 5, *Nuclear fuel cycle*.

This first edition of ISO 21483 cancels and replaces ISO 12184:1994, which has been technically revised.

Determination of solubility in nitric acid of plutonium in unirradiated mixed oxide fuel pellets (U, Pu) O2

1 Scope

This International Standard specifies an analytical method for determining the solubility in nitric acid of plutonium in pellets of unirradiated mixed oxide fuel (light-water reactor fuels). The results provide information about the expected dissolution behaviour of irradiated pellets under industrial reprocessing conditions. In this aspect, the specific conditions (e.g. time of the test) may vary depending upon the need to match to a specific reprocessor's requirements. The test is aimed at determining solubility under equilibrium conditions rather than the kinetics of dissolution and hence allows for a second dissolution period.

2 Principle

A specified number of mixed oxide pellets of known plutonium content and mass are dissolved in a boiling nitric acid solution. The initial concentration of the nitric acid, the final content of U, Pu and the boiling time are carefully controlled. The undissolved residue is then dissolved quantitatively by boiling in a mixture of nitric acid and hydrofluoric acid. The plutonium content of this residue is determined by an appropriate analytical method. The solubility is expressed by the ratio of the amount of plutonium dissolved in nitric acid to the amount of plutonium in the sample.

3 Interferences

The dissolution apparatus (Clause 5) and the reagents shall not be contaminated with fluoride, as fluoride can cause an increase in the solubility of the pellets in nitric acid.

4 Reagents

Use only reagents of analytical grade and distilled or demineralised water or water of equivalent purity. Prepare the reagents in compliance with the local laboratory safety instructions.

4.1 Concentrated nitric acid

ρ = 1,40 g/ml or more

4.2 Nitric acid high

Solution [$c(HNO_3)$ = 10 mol/l]

4.3 Nitric acid low

Solution [$c(HNO_3)$ = 0,5 mol/l].

4.4 Concentrated hydrofluoric acid

ρ = 1,13 g/ml

4.5 Mixture of acid

Nitric acid, solution [$c(HNO_3)$ = 14,4 mol/l] and hydrofluoric acid, solution [$c(HF)$ = 0,05 mol/l].

4.6 Sodium hydroxide

Solution [c(NaOH) = 2 mol/l] (optional).

5 Apparatus

Usual laboratory apparatus and, in particular, the following.

5.1 Analytical balance

Balance with a 0,1 mg accuracy or better.

5.2 Dissolution apparatus with total reflux condenser

Consisting of dissolver flask (glass borosilicate or high density polyethylene HDPE are suitable), heater, total reflux condenser and gas inlet tube. The volume capacity of the flask shall reflect the sample size selected in Clause 6.

5.3 Dissolution apparatus without total reflux condenser

Consisting of dissolver flask (glass borosilicate or high density polyethylene HDPE are suitable), heater, condenser and gas washer bottle. The volume capacity of the flask shall reflect the sample size selected in Clause 6.

5.4 Second dissolution apparatus (polytetrafluoroethylene, PTFE, high density polyethylene, HDPE)

For the dissolution with nitric acid/hydrofluoric acid mixture. This second dissolution is carried out with total reflux condenser (Clause 5).

5.5 Filter apparatus

Apparatus consisting of a vacuum funnel and a nitric-acid-resistant filter of a pore size less or equal to 1 micron (e.g. nitrocellulose or polycarbonate membrane filter 1 µm).

6 Sampling

The sample size shall be large enough to be representative of the lot, both whole and broken pellets are acceptable for this test. The minimum sample size could be chosen as a function of the pellet composition and the laboratory's detection limit. In any way, the minimum size must be equal or greater than 1 pellet.

7 Procedure

7.1 Preparation of the sample

Weigh the pellets and transfer quantitatively into the dissolver flask (5.2 and 5.3). In order to reach a heavy metal concentration of around 1 mol/l, the total volume V, in litres, of nitric acid solution (4.2) to be used is given by

$$V = \frac{m}{cM}$$

where

m is the sum of the mass of uranium and plutonium in the sample in grams,

M is the average atomic weights of the metals, in grams per mole;

c is the concentration of the uranium and plutonium in the solution, in moles per litre (c = 1 mol/l). This value corresponds to the practice of the "reprocessing dissolution of spent fuel".

7.2 Dissolution procedure

Introduce the calculated volume of nitric acid solution (4.2) into the flask containing the sample pellets. Boil the contents of the flask under reflux for the specified period (examples are 10 h and 18 h). The time required for the solubility test is dependent upon the commercial contract and agreed with the commercial customer. Boiling shall begin within 30 min. Cooling to below around 50 °C should be ended within 30 min. To avoid a delay in cooling, compressed air may be passed through the solution. The arising nitrous vapour may be trapped in a gas washer bottle filled with sodium hydroxide solution (4.6)

7.3 Treatment of the residue

Quantitatively transfer the solution and the undissolved residue into the filter apparatus and filter the solution using the vacuum funnel. Thoroughly rinse the residue on the filter with nitric acid solution (4.3)

If the filtered residue is seen to contain fragments of pellets, this may be indicative of a problem with the manufacture of MOX material. Repeat the test using a tared filter to weigh the residue (7.5).

Place the filter and the residue in a dissolver flask (5.4) and add around 25 ml of the nitric acid/hydrofluoric acid (4.5). Heat the acid to it's boiling point and pursue ebullition under reflux until the residue is completely dissolved and at least during 2,5 h.

In general, after this duration, no residue still remains with HNO_3/HF dissolution. If residue still remains, then it is possible to add hydrofluoric acid (0,05 mol/l) in order to obtain total dissolution.

7.4 Plutonium determination

Make up the solution resulting from the treatment of the undissolved residue to a known volume or known mass. Take samples and analyse their plutonium content by a suitable method. The method shall have an uncertainty of 20 % or lower, even in the presence of higher level impurities contained in the dissolved material. The spectrophotometry method described in ISO 9463 is adequate. Other methods like mass spectrometry (TIMS or ICPMS) can also be used.

7.5 Repeat solubility test

If visible pellet fragments are observed in the residue of the first dissolution test (7.2), perform a new test on a fresh sample. For this test, the test duration (up to 18 h), concentration of nitric acid (up to 10 mol/l)

and U+Pu concentration (0,9 mol/l) shall be defined before carrying out the test. The conditions of each test and its results should be reported.

8 Expression of results

8.1 Method of calculation

The plutonium solubility, L, expressed as mass fraction in percent, is calculated according the following equation:

$$L = \{1- \frac{m_2}{m_1 W_{Pu}}\} \times 100$$

where

m_1 is the mass of the sample, in grams;

m_2 is the mass of plutonium in the residue, in grams;

W_{Pu} is the mass fraction of the plutonium in the sample.

8.2 Total uncertainty of the Pu measurement

The total uncertainty is dependent upon the mass fraction of the plutonium W_{Pu} and of the analytical method used to determine the plutonium content in the residue. Typically, for a material with a mass fraction greater than 5 % and a sample size of at least 6 g, a total uncertainty of 20 % can be achieved.

9 Test report

The test report shall include the following information:

a) a reference to this International Standard;

b) date and place of the test carried out;

c) identification and mass of the sample;

d) the reference of the analytical method used;

e) the method of calculation of the results including the boiling time;

f) the results and their units;

g) any unusual features observed during the test;

h) potential increasing of dissolution time;

i) any operation regarded as optional.