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Re-sintering test for UO_2, $(U,Gd)O_2$ and $(U,Pu)O_2$ pellets

Test de refrittage pour pastilles UO_2, $(U,Gd)O_2$ et $(U,Pu)O_2$

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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*.

Re-sintering test for UO_2, $(U,Gd)O_2$ and $(U,Pu)O_2$ pellets

1 Scope

This International Standard describes a procedure for measuring the densification of UO_2, $(U,Gd)O_2$, and $(U,Pu)O_2$ pellets, achieved by heat treatment under defined conditions.

The densification of fuel in power operation is an important design feature. Essentially, it is dependent on structural parameters such as pore size, spatial pore distribution, grain size, and in the case of $(U,Gd)O_2$ and $(U,Pu)O_2$, oxide phase structure. A thermal re-sintering test can be used to characterize the dimensional behaviour of the pellets under high temperature. The results of this test are used by the fuel designer to predict dimensional behaviour in the reactor, because thermal densification in the reactor is also dependent on these structural parameters, albeit in a differing manner in terms of quantity.

On the assumption of the prediction, it is necessary to correlate the results of this test by some correlation rules, because the results of this test vastly depend on the re-sintering conditions (such as heat treatment temperature, treatment time, gas content, and partial oxygen pressure).

2 Brief description of procedure

The density of nuclear fuel pellets is measured before the re-sintering test. Afterwards, the pellets are subjected to heat treatment in a furnace under specified conditions with regard to temperature, time, and sintering atmosphere. After cooling, the density is remeasured.

The oxygen/metal molar ratio should remain constant during the re-sintering test. The difference between the two density measurements shall be used to assess the thermal stability of the pellet lot.

3 Incidents

Minor chipping can occur during pellet handling. Densities of visibly chipped pellets shall not be measured using the geometric method, as the results will be inaccurate.

Densities of such chipped pellets can be measured using the buoyancy method.

4 Apparatus

Specifically for $(U,Pu)O_2$, all operations shall be performed in glove boxes.

4.1 Equipment for measuring density

The same method shall be used before and after the re-sintering of pellets.

In 4.1.1 and 4.1.2, two different measurement methods are mentioned as examples. Other methods can be used if they meet the customer uncertainty requirement.

4.1.1 Geometric measurement of density

A dial test indicator or micrometer with precision of 1 µm shall be used in order to measure the diameter and the height.

Analytical balance with readability of ±1 mg for weighing the pellets.

4.1.2 Measurement of density using buoyancy method

See ISO 3369.

4.2 Heat treatment furnace

A programmable furnace able to reach temperatures of about 1 800 °C under gas scavenging and to hold a homogeneous temperature of the sintering area shall be used. It is advisable to use two different thermocouples for furnace control and monitoring.

5 Reagents

5.1 Gas.

The furnace gas shall ensure a constant oxygen/heavy metal ratio. Usually, the re-sinter test employs a similar furnace atmosphere to that used when the pellets were sintered during manufacture. The test furnace shall be qualified to prove that the oxygen/heavy metal ratio of the fuel tested does not change by more than ±0,005. The following are examples of furnace atmospheres that can be used.

NOTE Heavy metal = U, U+Pu, or U+Gd, depending on the concerned product.

— For UO_2 fuel:

  — pure hydrogen (purification better than a volume fraction of 99,99 %);

  — mixture of noble gas (purification better than a volume fraction of 99,99 %) with at least 3 % hydrogen.

— For $(U,Pu)O_2$ and $(U,Gd)O_2$ fuels:

  — pure argon (purification better than a volume fraction of 99,99 %);

  — pure argon (purification better than a volume fraction of 99,99 %) with 7 % hydrogen, humidified as required to maintain fuel stoichiometry;

  — argon with 4 % hydrogen and CO_2 as required to maintain fuel stoichiometry;

  — dry cracked ammonia gas (75 % H_2-25 % N_2 by volume) with gas mixture purity of at least 99,9 % by volume and a dew point less than −40 °C.

5.2 Degassed water, distilled and degassed (e.g. by boiling) water as buoyancy test fluid, or distilled water to which a wetting agent (soap) has been added to eliminate any microbubbles.

6 Sampling

In case of using the geometric method for the density measurement, the selected pellets shall be free of any visible defects liable to bias the density measurements (e.g. chipping, cracking, thumbnail cracks, etc.). In case of using the buoyancy method for the density measurement, the pellets with these visible defects can be kept.

7 Procedure

7.1 Density measurements before heat treatment

The density of the fuel pellets has to be measured prior to the heat treatment. Of all the procedures available, geometric density measurements and the buoyancy method have been proven most satisfactory for routine quality control inspection. The geometric method is preferable where the pellets have a large fraction of open pores (i.e. pores having a connection to the pellet surface). Pellets that have sustained

chipping are better measured using the buoyancy method or the penetration immersion method as defined in ISO 9278.

7.2 Heat treatment

Following the density measurement, the pellets, after position identification to ensure traceability, are subjected to heat treatment in the furnace using the following parameters.

- Heating rate: <15 °C/min (at higher rates the pellets could burst). A heating rate of 5 °C/min is recommended in order to be sure not to burst the pellets.
- Heat treatment temperature: shall be specified jointly by the customer/fuel designer and the fuel vendor. As example, temperatures of the order of 1 700 °C are generally applied.
- Treatment time: shall be specified jointly by the customer/fuel designer and the fuel vendor. As example, residence time of the order of 24 h is generally applied.
- Suitable treatment atmosphere (see Examples 1 to 3).

EXAMPLE 1 For UO_2 pellets, pure hydrogen (5.1) or mixtures of noble gas with at least 3 % hydrogen (5.1) can be used.

EXAMPLE 2 For $(U, Gd)O_2$ and $(U, Pu)O_2$ pellets, a shift in stoichiometry of pellets occurs during treatment when using dry hydrogen or dry hydrogen/noble gas mixtures. This can be avoided by using a gas atmosphere with a suitable oxygen partial pressure which keeps the stoichiometry constant during re-sintering. Adjusting the oxygen partial pressure can be done by admixing CO_2 in appropriate proportions to the re-sinter atmosphere or by using wet hydrogen or wet hydrogen/noble gas mixtures.

EXAMPLE 3 A pure argon atmosphere can be used.

NOTE For $(U, Gd)O_2$ pellets, if necessary, an adjustment of the initial oxygen/heavy metal (U+Gd) ratio can be performed by heat treatment of the pellets at 150 °C in air during a holding time of 5 h. The heat-up rate shall be low enough to avoid cracking especially at high Gd contents. The annealing step ensures the adjustment of the equilibrium stoichiometry for $(U,Gd)O_2$ fuel pellets with a Gd content up to 10 % by mass.

- Cool down rate: <15 °C/min.
- Records: For each re-sintering run, the temperature profile is recorded. If the temperature profile satisfies the heat treatment condition specified above, the results of the re-sintering test are accepted.

7.3 Density measurements after heat treatment

Following the cooling, the pellets are removed from the furnace and their densities shall be remeasured with the same method used before the heat treatment.

8 Evaluation

The re-sinter densification is determined by calculating the difference between the densities before and after the heating treatment. When measuring density, the mass of the pellets shall be measured prior to and upon completion of the heat treatment.

To check that the O/M ratio has not changed in excess of the tolerance of ±0,005, it is necessary to verify that the pellet mass difference, before and after the heat test, is below the following limit:

$$\text{Mass difference} \leq \frac{\text{Mass pellet before test}}{\text{Oxide molar mass}} \times \text{O}_2\,\text{molar mass} \times \text{O/M Changing tolerance} \quad (1)$$

The demonstration of Formula (1) is in Annex A.

In case of mixed oxide (MOX) pellets, the calculation is performed using Formula (2):

$$\text{Mass difference} \leq \frac{\text{Mass pellet before test}}{(238+2\times16)}\times(2\times16)\times0{,}005 \quad (2)$$

$$\text{Mass difference} \leq 0{,}000\,59 \times \text{Mass pellet before test}$$

It is a good practice to monitor the mean change of all measured pellet mass as a result of re-sintering and check that this does not differ statistically significantly from zero. A significant change would indicate a fault with the furnace atmosphere (e.g. due to leakage). The absolute value of the difference from zero should be lower than the maximum mass difference accepted for one pellet [see Formula (1)].

The density can be calculated through the ratio of measured density to the theoretical density.

The density measurement shall be made according to the specified methods as mutually agreed upon between the buyer and the seller (see ISO 9278).

9 Precision of the procedure

The precision of the overall procedure is determined by the precision of the method used to measure density. The repeatability standard deviation in DIN 55 350 for the buoyancy and the penetration immersion method (in turn defined in ISO 9278) is 0,1 %.

Nevertheless, due to the difficulty in weighing a pellet in liquid, in the course of the buoyancy density measurement method, the uncertainty can reach 0,2 %.

10 Test report

The test report with reference to this International Standard shall contain the following information:

— all data necessary for the identification of the sample;

— the result of test;

— the location and date of test;

— the recorded profile of temperature.

Annex A
(informative)

Relationship between pellet mass evolution and O/M ratio evolution

Assumption 1: It is considered that UO_2 pellets and MOX pellets are equivalent with regard to the O/M ratio evolution, because MOX pellets contains around 90 % of UO_2 and because the molar weight of U is near that of Pu.

Assumption 2: The maximum acceptable evolution of the O/M ratio during the re-sintering test is 0,01.

Chemical formula:

$$UO_2 + 0{,}01 \times \frac{1}{2} O_2 \rightarrow UO_{2{,}01} \tag{A.1}$$

Molar balance:

$$n_{UO_2} = n_{UO_{2,01}} \tag{A.2}$$

$$\frac{m_{UO_2}}{M_{UO_2}} = \frac{m_{UO_{2,01}}}{M_{UO_{2,01}}} \tag{A.3}$$

$$m_{UO_{2,01}} = \frac{m_{UO_2}}{M_{UO_2}} \times M_{UO_{2,01}} \tag{A.4}$$

Mass difference:

$$\Delta m = m_{UO_{2,01}} - m_{UO_2} \tag{A.5}$$

$$\Delta m = \frac{m_{UO_2}}{M_{UO_2}} \times M_{UO_{2,01}} - m_{UO_2} \tag{A.6}$$

$$\Delta m = m_{UO_2} \times \left(\frac{M_{UO_{2,01}}}{M_{UO_2}} - 1 \right) \tag{A.7}$$

$$\Delta m = m_{UO_2} \times \left(\frac{M_{UO_2} + 0{,}01 \times M_O - M_{UO_2}}{M_{UO_2}} \right) \tag{A.8}$$

$$\Delta m = m_{UO_2} \times \frac{0{,}01 \times M_O}{M_{UO_2}} \tag{A.9}$$

$$\Delta m = m_{pellet} \times \frac{0{,}01 \times 16}{270} \tag{A.10}$$