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Nuclear energy — Evaluation of homogeneity of Gd distribution within gadolinium fuel blends and determination of Gd_2O_3 content in gadolinium fuel pellets by measurements of uranium and gadolinium elements

Énergie nucléaire — Évaluation de l'homogénéité de la distribution du Gd dans les mélanges de combustibles au gadolinium et détermination de la teneur en Gd_2O_3 dans les pastilles combustibles au gadolinium par mesurage des éléments uranium et gadolinium

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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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Contents

Foreword

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Nuclear energy — Evaluation of homogeneity of Gd distribution within gadolinium fuel blends and determination of Gd_2O_3 content in gadolinium fuel pellets by measurements of uranium and gadolinium elements

1 Scope

This International Standard is applicable to the evaluation of the homogeneity of Gd distribution within gadolinium fuel blends, and the determination of the Gd_2O_3 content in sintered fuel pellets of Gd_2O_3+UO_2 from 1 % to 10 %, by measurements of gadolinium (Gd) and uranium (U) elements using ICP-AES.

After performing measurements of Gd and U elements using ICP-AES, if statistical methodology is additionally applied, homogeneity of Gd distribution within a Gd fuel pellet lot can also be evaluated. However, this International Standard covers the statistical methodology only on a limited basis.

NOTE 1 ISO 16796 also provides a method for Gd_2O_3 content determination by atomic emission spectrometry using an inductively coupled plasma source (ICP-AES). The methodology of ISO 16796 is different from the one of this International Standard.

NOTE 2 In this International Standard, gadolinium fuel blend represents a mixture of uranium dioxide (UO_2) powder and gadolinium oxide (Gd_2O_3) powder. The physically blended and homogenized powder may additionally contain in it rather large quantities of uranium oxide (U_3O_8) powder particles and/or the M_3O_8 powder particles obtained by oxidation of Gd pellets. In this International Standard, the symbol “M” in the chemical formula “M_3O_8” and in the terminology “O/M ratio” represents metallic elements U and Gd.

2 Normative references

The following documents, in whole or in part, are normatively referenced in this document and are indispensible for its application. For dated references, only the edition cited applies. For undated references, the latest edition of the referenced document (including amendments) applies.

ISO 3696:1987, *Water for analytical laboratory use — Specification and test methods*

3 Principle

If the Gd and U element contents and the oxygen to metal atomic ratio (commonly referred to as O/M ratio) in a gadolinium fuel pellet are measured or determined, the Gd_2O_3 content of that pellet can be determined by calculation based on the stoichiometry of the pellet. The stoichiometric compositions for Gd and U will depend upon pellet manufacturing specification. If the specification requires that the Gd_2O_3 content in the pellet be 6 % as mass fraction, after manufacturing, the ratio of total Gd mass to total U mass in that pellet will be close to 0,063.

The Gd and U element content values measured from a powder blend can make it possible to evaluate whether Gd distribution in the powder is sufficiently homogeneous. Moreover, the two values make it possible to estimate accurately the actual Gd_2O_3 content of the pellet after sintering. The estimated Gd_2O_3 content can be used to anticipate whether the Gd pellets to be produced will meet Gd_2O_3 content specifications or not.

Impurity interferences have not been observed for the usual samples of the nuclear grade material. Very small quantities of impurity elements which might be contained in the samples do not affect the principle of this International Standard.

NOTE Even in the case of M_3O_8 powder or mixture of UO_2 powder and Gd_2O_3 powder, if the Gd and U element contents and the O/M ratio are measured or determined, the Gd_2O_3 content (or Gd content) of that powder can be determined by calculation based on the stoichiometry of the powder. Using the Gd_2O_3 content (or Gd content) thus obtained and the O/M ratio, the uranium concentration factor of the powder can also be calculated so as to obtain accurate uranium accounting data.

4 Apparatus

4.1 High dispersion direct reading ICP-AES

The measured values should be indicated down to at least two decimal places when element concentrations in the range from 1 mg/l to 100 mg/l are measured. For determination of Gd and U element concentrations, 354,580 nm and 398,579 nm peaks in atomic emission spectrum are used, respectively. However, instead of the two peaks, other peaks are also available.

4.2 Analytical balance

The sensitivity of the balance is ± 0,1 mg.

4.3 Small stainless steel spoon

The small spoon is able to accommodate powder in the range from 1 mg to 100 mg.

4.4 Hot plate.

4.5 Glass beakers or polytetrafluoroethylene (PTFE) beakers.

4.6 Micropipettes.

4.7 Volumetric flasks.

4.8 Percussion mortar.

5 Reagents

5.1 Nitric acid, aqueous solution at a volume fraction of 50 %.

5.2 Water, distilled water or water complying with grade 3 of ISO 3696:1987.

6 Reference solutions

6.1 Gadolinium element reference solutions

Three Gd reference solutions whose Gd concentrations are 5 mg/l, 10 mg/l and 15 mg/l, respectively, are used for calibration. These Gd reference solutions[1] are the ones that have been manufactured by diluting a reference material with the water specified in 5.2.

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[1] A certified gadolinium reference material from NIST (National Institute of Standards and Technology, USA) is an example of a suitable product available commercially. This information is given for the convenience of users of this document and does not constitute an endorsement of this product by ISO.

6.2 Uranium element reference solutions

Three U reference solutions whose U concentrations are 50 mg/l, 70 mg/l and 100 mg/l, respectively, are used for calibration. These U reference solutions[2] are the ones that have been manufactured by diluting a reference material with the water specified in 5.2.

7 Sample preparation

7.1 Sample preparation for evaluation of Gd homogeneity in gadolinium fuel blend

Using a small spoon, take randomly at least five different powder samples from the blended powder, then dissolve each of the samples in the nitric acid solution of 5.1. Number each beaker individually. The U concentrations of the sample solutions prepared should be somewhere in the range from 50 mg/l to 100 mg/l.

To evaluate the homogeneity of the Gd distribution within a Gd fuel pellet lot, take at least five different pellet samples from the lot and then crush each of the pellets into small pellet fragments as described in 7.2.

NOTE 1 If about 2 mg of the powder sample is dissolved in 20 ml of the nitric acid solution, the required uranium concentration range will be obtained. Alternatively, if about 100 mg of sample is dissolved and then diluted by proper pipetting and addition of distilled water, the concentration range can also be obtained.

7.2 Sample preparation for determining the Gd_2O_3 content of Gd fuel pellets

After crushing a gadolinium fuel pellet using a percussion mortar, randomly take a small sample from the pellet fragments, and dissolve it in the nitric acid solution. The U concentration should be somewhere in the range from 50 mg/l to 100 mg/l.

8 Calibration and analysis of the samples

8.1 Calibration of Gd peak line intensity

The three Gd reference solutions of 6.1 are used to calibrate the intensity of the Gd peak line. The Gd peak line intensity is repeatedly measured to establish an intensity-versus-concentration curve (regression curve) needed for determination of Gd concentrations. See A.1.

8.2 Calibration of U peak line intensity

The three U reference solutions of 6.2 are used to calibrate the intensity of the U peak line. The U peak line intensity is repeatedly measured to establish an intensity-versus-concentration curve (regression curve) needed for determination of U concentrations. See A.2.

8.3 Evaluation of Gd homogeneity in gadolinium fuel blend

8.3.1 Estimated Gd_2O_3 content

Measure the concentrations of Gd and U elements contained in the sample solutions (at least five different solutions) that were prepared according to 7.1. Each sample solution is repeatedly measured six times, and the six measurement values of Gd and U each are averaged. Each sample solution will be given a Gd

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[2] A certified uranium reference material from NIST (National Institute of Standards and Technology, USA) is an example of a suitable product available commercially. This information is given for the convenience of users of this document and does not constitute an endorsement of this product by ISO.

average and a U average. After all of the Gd concentration average and U concentration average sets have been obtained, the estimated Gd_2O_3 content for each sample is calculated by the following formula:

$$w_{\mathrm{Gd2O3,est}} = \frac{(\frac{1}{2})(100)[\frac{(15{,}999\ 4\times3)}{157{,}25}+2](\bar{\rho}_{\mathrm{Gd}})}{(\bar{\rho}_{\mathrm{Gd}}+\bar{\rho}_{\mathrm{U}})+(2)(15{,}999\ 4)\left(\frac{\bar{\rho}_{\mathrm{Gd}}}{157{,}25}+\frac{\bar{\rho}_{\mathrm{U}}}{238{,}03}\right)}$$

$$= \frac{(0{,}5)(100)(2{,}305\ 2\,\bar{\rho}_{\mathrm{Gd}})}{(\bar{\rho}_{\mathrm{Gd}}+\bar{\rho}_{\mathrm{U}})+(2)\left(0{,}101\ 7\,\bar{\rho}_{\mathrm{Gd}}+0{,}067\ 2\,\bar{\rho}_{\mathrm{U}}\right)}$$

where

$w_{\mathrm{Gd2O3,est}}$ is the estimated Gd_2O_3 content, mass fraction (%) as given by $Gd_2O_3/(Gd,U)O_2$;

157,25 is the atomic mass of gadolinium (Gd);

238,03 is the atomic mass of uranium (U);

15,9994 is the atomic mass of oxygen (O);

$\bar{\rho}_{\mathrm{Gd}}$ is the Gd concentration average in mg/l, indicated down to two decimal places;

$\bar{\rho}_{\mathrm{U}}$ is the U concentration average in mg/l, indicated down to two decimal places.

If five different sample solutions have been prepared and measured, five estimated values of Gd_2O_3 content will be obtained by this procedure.

NOTE The estimated Gd_2O_3 content represents the most possible Gd_2O_3 content of the expected Gd pellet to be manufactured from the powder blend. As a general rule, after the powder blend has been sintered, the O/M ratio of manufactured Gd pellet will be 2 or very close to 2. Therefore, the actual Gd_2O_3 content of the pellet will be very close to this estimated Gd_2O_3 content.

8.3.2 Evaluation of homogeneity of Gd distribution

Evaluate the homogeneity of Gd distribution within the blended powder by applying a statistical methodology to the estimated contents of Gd_2O_3 obtained in Clause 8.3.1. The average and the standard deviation calculated from at least five estimated values of Gd_2O_3 content will be used for the evaluation.

EXAMPLE If the homogeneity specification of the blended powder requires that one standard deviation of estimated Gd_2O_3 content values of five blended powder samples randomly taken shall be within 1 % relative from the mean of the five values, and if the mean and the standard deviation of the five estimated Gd_2O_3 contents are 6,03 % and 0,04 %, respectively, it can be said that the blended powder meets the homogeneity specification, because all of the related inequalities, i.e. 6,03 – (6,03 × 0,01) < 6,03 – 0,04 and 6,03 + 0,04 < 6,03 + (6,03 × 0,01), are correct.

NOTE The statistical methodology and limitations will depend on the quality assurance policy and goals of the organization concerned. To find detailed information on the applicable statistical methods other than the above evaluation example, see ISO 3951-4.

8.4 Determination of Gd_2O_3 content in gadolinium fuel pellet

8.4.1 Preliminary Gd_2O_3 content

Repeatedly measure six times Gd and U concentrations in the sample solution prepared according to Clause 7.2, and calculate the concentration average of Gd and U each. The preliminary Gd_2O_3 content in the pellet can be determined by the following formula:

$$w_{\mathrm{Gd_2O_3,pre}} = \frac{(100)(157{,}25\times2+15{,}999\ 4\times3)(Z)}{2(1-Z)(238{,}03+15{,}999\ 4\times2)+(157{,}25\times2+15{,}999\ 4\times3)(Z)}$$

$$Z=\frac{\left(\frac{\bar{\rho}_{\mathrm{Gd}}}{157{,}25}\right)}{\left(\frac{\bar{\rho}_{\mathrm{Gd}}}{157{,}25}+\frac{\bar{\rho}_{\mathrm{U}}}{238{,}03}\right)}$$

where

$w_{\mathrm{Gd2O3,pre}}$ is the preliminary Gd_2O_3 content, mass fraction in %;

$\bar{\rho}_{\mathrm{Gd}}$ is the Gd concentration average in mg/l, indicated down to two decimal places;

$\bar{\rho}_{\mathrm{U}}$ is the U concentration average in mg/l, indicated down to two decimal places.

NOTE The preliminary Gd_2O_3 content represents the interim value before obtaining the final Gd_2O_3 content value of the Gd pellet. From the aspect of the stoichiometry of the Gd pellet, this preliminary Gd_2O_3 content value is not complete theoretically.

8.4.2 O/M ratio of the Gd pellet

The O/M ratio, *B*, of the pellet is determined by the following formula:

$$B = 2-\frac{(m_2-m_1)}{(m_2)(0{,}0592)+(m_1)(w_{\mathrm{Gd_2O_3,pre}})(0{,}0264)(0{,}01)}$$

where

B is the O/M ratio, and will be indicated down to at least two decimal places;

m_1 is the sample mass before equilibration in grams, indicated down to four decimal places;

m_2 is the sample mass after equilibration in grams, indicated down to four decimal places;

$w_{\mathrm{Gd2O3,pre}}$ is the preliminary Gd_2O_3 content (mass fraction of Gd_2O_3 in %) obtained in 8.4.1.

NOTE 1 The above formula is used to determine the O/M ratio by an atmospheric equilibration method. For application of this method, a Gd pellet goes through an oxidation-reduction process at 900 °C ± 20 °C for 4 h in a mixed gas of argon (or nitrogen) at a volume fraction of 96 % and hydrogen at a volume fraction of 4 %; the mixed gas also contains a small volume fraction of water vapor. The Gd pellet masses before and after equilibration by oxidation-reduction and Gd_2O_3 content value are used to determine the O/M ratio. See ASTM C1430-07.[4]

NOTE 2 The derivation of the formulas for O/M and O/U ratios of Gd pellet can be seen in Annex B.

8.4.3 Final Gd_2O_3 content

The Gd_2O_3 content in the sampled pellet is finally determined by the following formula:

$$w_{\mathrm{Gd_2O_3,fin}}=\frac{\left(\frac{1}{2}\right)(100)\left[\frac{(15{,}999\,4\times 3)}{157{,}25}+2\right](\bar{\rho}_{\mathrm{Gd}})}{(\bar{\rho}_{\mathrm{Gd}}+\bar{\rho}_{\mathrm{U}})+(\mathrm{B})(15{,}999\,4)\left(\frac{\bar{\rho}_{\mathrm{Gd}}}{157{,}25}+\frac{\bar{\rho}_{\mathrm{U}}}{238{,}03}\right)}$$

$$=\frac{(0{,}5)(100)(2{,}305\,2\,\bar{\rho}_{\mathrm{Gd}})}{(\bar{\rho}_{\mathrm{Gd}}+\bar{\rho}_{\mathrm{U}})+(B)(0{,}101\,7\,\bar{\rho}_{\mathrm{Gd}}+0{,}067\,2\,\bar{\rho}_{\mathrm{U}})}$$

where

$w_{\mathrm{Gd2O3,fin}}$	is the final Gd_2O_3 content in %, i.e. mass fraction of Gd_2O_3 (%) as given by $Gd_2O_3/(Gd,U)O_2$;
$\bar{\rho}_{\mathrm{Gd}}$	is the Gd concentration average that was obtained in 8.4.1;
$\bar{\rho}_{\mathrm{U}}$	is the U concentration average that was obtained in 8.4.1;
B	is the O/M ratio that was obtained in 8.4.2.

Although nominal O/M ratio 2 is substituted into the above formula in order to determine the final Gd_2O_3 content, instead of the O/M ratio B that was directly measured in 8.4.2, the results will almost be the same. Therefore, the use of nominal O/M ratio may be permitted. In this case, the formula will become the same as the one that was introduced to obtain the estimated Gd_2O_3 content in 8.3.1.

9 Precision

The possible uncertainty of Gd_2O_3 content determination using the method of this International Standard, excluding systematic uncertainty such as calibration uncertainty, is about 0,2 % relative (0,012 % absolute) at nominal Gd_2O_3 content of 6 %.

To find more information regarding precision of Gd_2O_3 content determination using the method of this International Standard, see Annex C.

10 Test report

The test report shall include at least the following information:

a) identification of sample, i.e. lot number of powder or pellet, etc.;

b) the reference of the method used;

c) the measurement results and their units;

d) any unusual features noted during the measurements;

e) any operations not included in this International Standard.

Annex A
(informative)

Calibration and Gd and U measurement uncertainties

A.1 Calibration of Gd line intensity and residual mean square

Table A.1 — Gd line intensity-versus-concentration of Gd reference solution

Measurements	Measured intensity 5 mg/l solution	Measured intensity 10 mg/l solution	Measured intensity 15 mg/l solution
1st measurement	572 478,511	1 127 995,396	1 682 373,714
2nd measurement	568 482,882	1 118 771,581	1 692 754,967
3rd measurement	567 587,661	1 124 687,706	1 678 890,058
4th measurement	573 010,558	1 119 946,173	1 674 248,737
5th measurement	567 155,019	1 114 052,914	1 685 414,131
6th measurement	568 116,061	1 131 790,625	1 688 138,089
Mean Value	**569 471,782**	**1 122 874,065**	**1 683 636,616**
Standard Deviation	**2 580,558**	**6 515,181**	**6 615,467**

Using the least square method, the regression curve equation of Gd line intensity-versus-concentration can be obtained as follows:

$$\rho_{\mathrm{Gd}} = a + bI_{\mathrm{Gd}} = -\,0{,}1 + 8{,}975 \times 10^{-6} I_{\mathrm{Gd}}$$

where

I_{Gd} is the measured Gd line intensity in generic units;

ρ_{Gd} is the Gd concentration of the solution in mg/l;

a and b are the coefficients of the regression curve ($a = -\,0{,}100$ and $b = 8{,}975 \times 10^{-6}$).

The residual mean square and regression standard deviation are calculated as follows:

$$(\sigma_{\mathrm{Gd}})^2 = \{\sum(\rho_{\mathrm{Gd}\,i} - a - bI_{\mathrm{Gd}\,i})^2\}/(n-2),\ (n \text{ is } 3, \text{ and } \sum \text{ is the summation from } i = 1 \text{ to } 3.)$$

$$= 0{,}000\,728 \text{ (residual mean square)}$$

$$\therefore \sigma_{\mathrm{Gd}} = 0{,}027 \text{ (mg/l) (regression standard deviation)}$$

A.2 Calibration of U line intensity and residual mean square

Table A.2 — U line intensity-versus-concentration of U reference solution

Measurements	Measured intensity 50 mg/l solution	Measured intensity 70 mg/l solution	Measured intensity 100 mg/l solution
1st measurement	53 409,807	73 762,528	107 245,053
2nd measurement	53 509,371	73 878,279	107 085,192
3rd measurement	53 300,306	73 633,468	108 205,616
4th measurement	52 518,142	73 598,608	107 540,834
5th measurement	53 102,651	74 037,009	107 383,643
6th measurement	53 208,716	74 061,585	107 607,147
Mean Value	**53 174,832**	**73 828,580**	**107 511,248**
Standard Deviation	**352,271**	**197,747**	**390,098**

Using the least square method, the regression curve equation of U line intensity-versus-concentration can be obtained as follows:

$$\rho_U = a + bI_U = 1{,}627 + 9{,}173 \text{ x } 10^{-4} I_U$$

where

I_U is the measured U line intensity in generic units;

ρ_U is the U concentration of the solution in mg/l;

a and b are the coefficients of the regression curve ($a = 1{,}627$ and $b = 9{,}173 \text{ x } 10^{-4}$).

The residual mean square and regression standard deviation are calculated as follows:

$$(\sigma_U)^2 = \{\sum(\rho_{U\,i} - a - bI_{U\,i})^2\}/(n-2), \text{ (} n \text{ is 3, and } \sum \text{ is the summation from } i = 1 \text{ to 3.)}$$

$$= 0{,}647\,036 \text{ (residual mean square)}$$

$$\therefore \sigma_U = 0{,}804 \text{ (mg/l) (regression standard deviation)}$$

NOTE 1 If only one of the three reference solutions is used for calibration of the Gd line intensity, the regression standard deviation σ_{Gd} (0,027 mg/l) can simply be regarded as calibration uncertainty of the Gd line intensity. Similarly, the regression standard deviation σ_U (0,804 mg/l) can also be regarded as calibration uncertainty of the U line intensity.

NOTE 2 The calibration uncertainties in this International Standard are mainly due to the equipment's random errors and pipetting errors that arise while manufacturing the reference solutions from the certified reference materials.

A.3 Measurement uncertainties of Gd and U concentrations

After the correlation coefficient between *a* and *b,* and the uncertainties of regression coefficients *a* and *b* were calculated and the rules for error propagation were applied to the equations $\rho_{Gd} = -\,0{,}1 + 8{,}975 \text{ x } 10^{-6} I_{Gd}$ and $\rho_U = 1{,}627 + 9{,}173 \text{ x } 10^{-4} I_U$ to obtain the combined uncertainties of Gd and U concentration measurements. The combined uncertainties thus obtained can be seen in Tables A.3 and A.4.

Table A.3 — Measurement uncertainty of Gd concentration

Intended Gd concentration of reference solution mg/l	Calibration uncertainty mg/l	Random uncertainty mg/l	Combined uncertainty mg/l
5	0,024 3	0,009 5	0,026 1
10	0,014 6	0,023 9	0,028 0
15	0,023 7	0,024 3	0,033 9

Table A.4 — Measurement uncertainty of U concentration

Intended U concentration of reference solution mg/l	Calibration uncertainty mg/l	Random uncertainty mg/l	Combined uncertainty mg/l
50	0,696 0	0,131 9	0,708 4
70	0,473 2	0,074 1	0,478 9
100	0,765 5	0,146 1	0,779 3

NOTE When calculating the above measurement uncertainties, the uncertainties of the certified reference materials that were used to manufacture the reference solutions were disregarded, because they were small.

Annex B
(informative)

Derivation of O/M and O/U ratio formulas

B.1 Derivation of O/M ratio formula

Based on the stoichiometry of sintered Gd fuel pellet, the following two simultaneous Formulae (B.1) and (B.2) can be established. From the equations, the formula for the O/M ratio, *B*, is derived as follows:

$$m_1 = (m_\mathrm{U} + m_\mathrm{Gd}) + \{(m_\mathrm{U}/238{,}03) + (m_\mathrm{Gd}/157{,}25)\}\,(B)\,(15{,}999\ 4) \qquad \text{(B.1)}$$

$$m_2 = (m_\mathrm{U} + m_\mathrm{Gd}) + \{(m_\mathrm{U}/238{,}03) + (m_\mathrm{Gd}/157{,}25)\}\,(2)\,(15{,}999\ 4) \qquad \text{(B.2)}$$

where

m_1	is the mass of the Gd pellet sample before equilibration, in grams;
m_2	is the mass of the Gd pellet sample after equilibration, in grams;
m_U	is the quantity of uranium contained in the sample, in grams;
m_Gd	is the quantity of gadolinium contained in the sample, in grams;
238,03	is the atomic mass of uranium;
157,25	is the atomic mass of gadolinium;
15,999 4	is the atomic mass of oxygen;
B	is the O/M ratio of Gd pellet before equilibration

$$m_2 - m_1 = \{(m_\mathrm{U}/238{,}03) + (m_\mathrm{Gd}/157{,}25)\}\,(2)\,(15{,}999\ 4)$$

$$- \{(m_\mathrm{U}/238{,}03) + (m_\mathrm{Gd}/157{,}25)\}\,(B)\,(15{,}999\ 4)$$

$$\therefore B = 2 - (m_2 - m_1) \,/\, [\{(m_\mathrm{U}/238{,}03) + (m_\mathrm{Gd}/157{,}25)\}\,(15{,}999\ 4)]$$

$$= 2 - (m_2 - m_1) \,/\, \{0{,}067\ 2\,(m_\mathrm{U}) + 0{,}101\ 7\,(m_\mathrm{Gd})\}$$

$$m_\mathrm{Gd} = \{(314{,}5)/(362{,}498\ 2)\}\,(w_\mathrm{Gd2O3})\,(0{,}01)\,(m_1) = 0{,}008\ 676\,(w_\mathrm{Gd2O3})\,(m_1)$$

w_Gd2O3 = the Gd_2O_3 mass fraction (%) as given by $Gd_2O_3/(Gd,U)O_2$ before equilibration

$$m_2 = \{1 + (2 \times 15{,}999\ 4)/(238{,}03)\}\,(m_\mathrm{U}) + \{1 + (2 \times 15{,}999\ 4)/(157{,}25)\}\,(m_\mathrm{Gd})$$

$$\therefore m_\mathrm{U} = 0{,}881\ 5\ m_2 - 0{,}920\ 42\,(0{,}01)\,(w_\mathrm{Gd2O3})\,(m_1)$$

$$\therefore \mathrm{B} = 2 - (m_2 - m_1)/\{(0{,}059\ 2)(m_2) + (0{,}026\ 4)\,(w_\mathrm{Gd2O3})\,(m_1)\} \times 0{,}01$$

B.2 Derivation of O/U ratio formula

If the O/U ratio is defined as the ratio of the number of oxygen atoms to the number of uranium atoms contained in the Gd fuel pellet, the formula for the O/U ratio, *D*, is derived as follows:

$N_{\text{oxygen}} = \{m_1 - (m_U + m_{Gd})\}/(15{,}999\ 4)$

$= m_1\{0{,}062\ 5 + (0{,}003\ 301)\ (0{,}01)\ (w_{Gd2O3})\} - m_2\ (0{,}055\ 10)$

$N_{\text{uranium}} = \{(0{,}881\ 50)\ (m_2) - (0{,}920\ 42)\ (0{,}01)\ (w_{Gd2O3})\ (m_1)\}/(238{,}03)$

$\therefore D = [\{(0{,}062\ 5) + (0{,}003\ 301)\ (0{,}01)\ (w_{Gd2O3})\}\ (m_1) - (0{,}055\ 10)\ (m_2)]\ /\ \{(0{,}003\ 703)\ (m_2) - (0{,}003\ 867) \times (0{,}01)\ (w_{Gd2O3})\ (m_1)\}$

where

N_{oxygen} is the number of oxygen atoms contained in Gd fuel pellet;

N_{uranium} is the number of uranium atoms contained in Gd fuel pellet;

D is O/U ratio

Annex C
(informative)

Evaluation of Gd_2O_3 measurement precision

C.1 Experiment for Gd_2O_3 measurement precision evaluation

A gadolinium oxide pellet having 6 % (mass fraction) of nominal Gd_2O_3 content was used in an experiment to evaluate the precision of Gd_2O_3 content determined by measurements of the Gd and U elements. A small quantity of sample was taken from that crushed pellet and the sample was dissolved in a nitric acid solution at a volume fraction of 50 %. Then, the Gd concentration and the U concentration of the solution were measured six times by ICP-AES, respectively. The data obtained from the experiment can be seen in Table C.1.

Table C.1 — Measurement data obtained from experiment for precision evaluation

Measurements	**Gd concentration** ρ_{Gd}, mg/l	**U concentration** ρ_{U}, mg/l
1st measurement	5,32	85,74
2nd measurement	5,38	85,46
3rd measurement	5,33	85,49
4th measurement	5,36	85,52
5th measurement	5,36	85,55
6th measurement	5,37	85,41
Mean Value	**5,353**	**85,528**
Standard Deviation	**0,023**	**0,14**

From the two averaged values, i.e. the averaged Gd concentration value 5,353 mg/l and the averaged U concentration value 85,528 mg/l, the Gd_2O_3 content of the sample pellet was determined at 5,963 % (mass fraction). When determining the Gd_2O_3, nominal value 2 was used as O/M ratio B. From the above data, it can be said that because the average of six measurement values has been used, the standard deviation of the Gd concentration mean is $0{,}023/(6)^{1/2} = 0{,}009\,4$ (mg/l). By similar procedure, the standard deviation of the U concentration mean is $0{,}114/(6)^{1/2} = 0{,}046$ (mg/l).

C.2 Calculation of combined uncertainty using error propagation rules

It was assumed that measurements of the Gd and U concentrations and determination of the O/M ratio are independent from one another. It was also assumed that the uncertainty of the O/M ratio determination is 0,01 for convenience of calculation. Strictly speaking, from the equation used to obtain the O/M ratio in 8.4.2, the O/M ratio determination is dependent upon the measurements of Gd and U concentrations. However, because the extent of uncertainty of O/M ratio determination does not exceed 0,01 at any rate empirically, such assumptions will be conservative when evaluating the extent