Estimation of Accuracy of the Method of Gamma- Spectrometry 
for Non-Destructive Control of Gadolinium Content and 
Uranium Enrichment in Non-Irradiated VVER-type Fuel Pellets

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Abstract. The gamma-spectra of the single (UO$_2$-Gd$_2$O$_3$)-pellets were measured on the planar 
detector HPLeGe. The measurements of Gd content in (UO$_2$-Gd$_2$O$_3$)-pellets were performed by X- 
ray fluorescence method. The total count rate in the doublet 42,3 and 43,0 keV K$_{\alpha 1,2}$-X-rays from 
Gd was used as a measure of Gd content. In measurements was used $^{241}$Am as an external radiation 
source for exciting the doublet 42,3 and 43,0 keV K$_{\alpha 1,2}$-X-rays from Gd. In order to obtain the 
calibration dependence of count rate in doublet 42,3 and 43,0 keV from Gd content in (UO$_2$- 
Gd$_2$O$_3$)-pellets there were used standard (UO$_2$-Gd$_2$O$_3$)-pellets with the known Gd content, density 
and enrichment. The estimates of influence of deviations of the density of (UO$_2$-Gd$_2$O$_3$)-pellets 
and Gd content on total count rate in doublet 42,3 and 43,0 keV were performed. The «pure» gamma- 
spectra (without employing of external radiation source) were used to obtain the data on uranium 
enrichment of fuel pellets using multigroup gamma-spectra analysis code (MGAU) and additional 
information about Gd content. The dependence of $\frac{S_{185,7\,keV}}{S_{98,4\,keV}}$ from Gd content in (UO$_2$-Gd$_2$O$_3$)- 
pellets is present (the ratio of count rates for two peaks: $E_{\gamma}$=185,7 keV (the measure of $^{235}$U) and 
$E$=98,43 keV (the measure of $^{235,238}$U)).

1. Introduction
The nuclear safety of the VVER-type reactors depends from the method of compensating of initial 
reactivity excess, as well as by the reliability of the corresponding safety systems. That’s why the 
investigation of different types of technological characteristics of (UO$_2$-Gd$_2$O$_3$)-, UO$_2$-pellets is very 
important [1-6]. The applied nondestructive analysis method permits to perform continuous material 
control measurements of single pellets in laboratory conditions [7].
2. Determination of Gd content in (UO$_2$-Gd$_2$O$_3$)-pellets

2.1. The Gamma and X-rays Nuclear Data

The Gamma and X-rays Nuclear Data are present in table 1.

| Energy (keV) | Photon emission probability (%) |
|--------------|---------------------------------|
| Gd K$_{\alpha 2}$ | 42.302 | 50 |
| Gd K$_{\alpha 1}$ | 42.989 | 100 |
| U K$_{\alpha 1}$ | 98.430 | 100 |
| $\gamma$-ray $^{235}$U | 185.7 | 57.5 |
| $\gamma$-ray $^{241}$Am | 59.54 | 35.8 |

2.2. Equipment

Spectrums were taken in planar HPLEGe detector with resolution 460 eV at $E_{\gamma}$=122 keV (full width at half maximum, FWHM). The multichannel digital analyzer DSA-1000 Canberra and Genie-2000 (including MGAU-code) soft were used.

In this chapter are present experimental data obtained on the facility containing external radiation source $^{241}$Am ($A$$\approx$300 kBq). Since both the radiation from the $^{241}$Am excitation source and the Gd K$_{\alpha 1,2}$-X-rays are appreciably attenuated in typical VVER-fuel cladding material, the X-ray analysis was performed on the unclad fuel single pellets. The geometry of measurements is present on figure 1 ("side-source geometry").

![Figure 1](image)

Figure 1. The geometry of measurements of gamma-spectra of (UO$_2$–Gd$_2$O$_3$) – pellet:
1-(UO$_2$-Gd$_2$O$_3$)-pellet; 2-Pb- collimator; 3-holder for collimator; 4-gamma-radiation source $^{241}$Am; 5-Pb- plate; 6-stop for fixing equipment with external radiation source on the HPLEGe detector; 7-HPLEGe detector housing.

2.3. Experimental Results. Plotting the Calibration Dependence of the Total Count Rate in Doublet 43 keV from Gd Content

In order to obtain the calibration dependence of total count rate in doublet 43 keV from Gd content in (UO$_2$-Gd$_2$O$_3$)-pellets there were used standard (UO$_2$-Gd$_2$O$_3$)-pellets with known Gd content, density and enrichment.

The calibration dependence of total count rate in doublet 43 keV from Gd content in (UO$_2$-Gd$_2$O$_3$)-pellets is present in figure 2.
On the base of obtained experimental data there was obtained expression for the dependence of the total count rate in doublet 43 keV from Gd content:

$$S_{43\text{keV}} = (7.3\pm0.3) \times 10^{-3} + \left(4.2\pm0.1\right) \times 10^{-2} \cdot \text{w.f.Gd}_2\text{O}_3 \quad (1)$$

2.4. The Estimates of Influence of Deviations of the Density of (UO$_2$-Gd$_2$O$_3$)-Pellets and Gd Content on Total Count Rate in Doublet 43 keV

The calculation of the ratio $I_1/I_2$ of total count rate in doublet 43 keV was performed for three values of the density ($\gamma$) of (UO$_2$-Gd$_2$O$_3$)-pellets: 10.5, 10.1 and 9.6 g/cm$^3$. The weight fraction of Gd$_2$O$_3$ in (UO$_2$-Gd$_2$O$_3$)-pellet is 8 %. The data are shown in table 2.

| $\gamma$, g/cm$^3$ | $I_1/I_2$ |
|-------------------|-----------|
| 10.5              | 1.0006    |
| 10.1              | 1.0000    |
| 9.6               | 0.9976    |

The changes in $I_1/I_2$ are little ($\pm0.05\%$). Its explain by the features of the geometry of the measurements: $K_{\alpha_{1,2}}$-X-rays from Gd 43 keV escape from very thin surface layer (d<0.1 mm) from (UO$_2$-Gd$_2$O$_3$)-pellet to the direction of the sensitive volume of detector.

The next calculation of the ratio $I_1/I_2$ of total count rate in doublet 43 keV was performed for three values of the weight fractions of Gd$_2$O$_3$ in (UO$_2$-Gd$_2$O$_3$)-pellet: 7%, 8%, and 9%. The data are shown in table 3.
Table 3 The changes of the ratio $I_1/I_2$ of total count rate in the doublet 43 keV from the weight fraction of Gd$_2$O$_3$ in (UO$_2$-Gd$_2$O$_3$)-pellet.

| w.f. of Gd$_2$O$_3$, % | $I_1/I_2$ |
|----------------------|-----------|
| 7.0                  | 0.992     |
| 8.0                  | 1.000     |
| 9.0                  | 1.008     |

When we change the concentration of Gd$_2$O$_3$ in (UO$_2$-Gd$_2$O$_3$)-pellets step by step on 1%, the total count rate in the doublet 43 keV also change on $\approx$1%. So, the obtained data in total count rate in doublet 43 keV are sensitive to the change of weight fraction of Gd$_2$O$_3$.

2.5. The Procedure of Obtaining of the Estimates of Influence of Deviations of the Density of (UO$_2$-Gd$_2$O$_3$)-Pellets and Gd Content on Total Count Rate in Doublet 43 keV

The ratio of the total count rate $I_1/I_2$ in the doublet 43 keV for two types ($i=1, 2$) of (UO$_2$-Gd$_2$O$_3$)-pellets with different technological data was calculated using the next expression:

$$
\frac{I_1}{I_2} = \frac{\gamma_{(UO_2-Gd_2O_3)_i} \left[ \mu_i(59,54) + \mu_i(43) \right] (1 - e^{\rho_i(59,54)} + e^{\rho_i(43)}) x_i}{\gamma_{(UO_2-Gd_2O_3)_i} \left[ \mu_i(59,54) + \mu_i(43) \right] (1 - e^{\rho_i(59,54)} + e^{\rho_i(43)}) x_i} 
$$

(2)

were $X_i$ – thickness, $\gamma_i$ - density, $\mu_0$ - linear attenuation coefficient of fuel pellets with certain technological data ($i=1, 2$). The following expression was used to calculate linear attenuation coefficients in materials with complex chemical composition:

$$
\mu_m = \sum_i (\mu_{m_i}(E) \cdot \rho_i)
$$

(3)

where $\mu_{m_i}$ – mass attenuation coefficient [10].

Taking into account the weight fractions for each element in the pellet composition, the linear attenuation coefficients for energies 59,54 keV and 43 keV were calculated as follows:

$$
\mu_i(59,54 \text{ keV}) = \gamma_{(UO_2-Gd_2O_3)} \cdot (\mu^{(59,54)}_{m_i} \cdot \text{w.f.}(U) + \mu^{(59,54)}_{m_{Gd}} \cdot \text{w.f.}(Gd) + \mu^{(59,54)}_{m_O} \cdot \text{w.f.}(O))
$$

(4)

$$
\mu_i(43 \text{ keV}) = \gamma_{(UO_2-Gd_2O_3)} \cdot (\mu^{(43)}_{m_i} \cdot \text{w.f.}(U) + \mu^{(43)}_{m_{Gd}} \cdot \text{w.f.}(Gd) + \mu^{(43)}_{m_O} \cdot \text{w.f.}(O))
$$

(5)

where $\gamma_{(UO_2-Gd_2O_3)}$ is the density of the (UO$_2$-Gd$_2$O$_3$)-pellet; $\mu_{m_{U_i}}(E_i)$, $\mu_{m_{Gd}}(E_i)$, $\mu_{m_{U}}(E_i)$ are the mass attenuation coefficients of the U, Gd, and oxygen for energies of 43 and 59,54 keV; w.f.(U), w.f. (Gd), w.f. (O)-weight fractions of the U, Gd, and oxygen in (UO$_2$-Gd$_2$O$_3$)-pellets.

The density of (UO$_2$-Gd$_2$O$_3$)-pellet was calculated using the expression:

$$
\gamma_{UO_2-Gd_2O_3} = \gamma_{UO_2} \cdot (1 - \varepsilon_v) + \gamma_{Gd_2O_3} \cdot \varepsilon_v
$$

(6)

where $\varepsilon_v$ is the volume fraction of Gd$_2$O$_3$ in the (UO$_2$-Gd$_2$O$_3$)-pellet.
The weight fractions of the U, Gd, and oxygen in the (UO$_2$-Gd$_2$O$_3$)-pellet were calculated using the following expressions:

$$w.f.(U) = \frac{m_U}{m_U + m_{Gd} + m_O}; \quad w.f.(Gd) = \frac{m_{Gd}}{m_U + m_{Gd} + m_O}; \quad w.f.(O) = \frac{m_O}{m_U + m_{Gd} + m_O}$$ (8–10)

Based on the results of the above calculations, the dependence of the density of (UO$_2$-Gd$_2$O$_3$)-pellets from the weight fraction of Gd$_2$O$_3$ is present on Figure 3. The dependences of the weight fractions of uranium, gadolinium and oxygen from the weight fraction of Gd$_2$O$_3$ in the (UO$_2$-Gd$_2$O$_3$)-pellet are present on the figures 4, 5 and 6.

**Figure 3.** Dependence of the density of the (UO$_2$-Gd$_2$O$_3$)-pellet from the weight fraction of Gd$_2$O$_3$.

**Figure 4.** Dependence of the weight fraction of uranium from the weight fraction of Gd$_2$O$_3$ in (UO$_2$-Gd$_2$O$_3$)-pellet.
As mentioned earlier, the total count rate in the doublet 43 keV Gd X-ray determine by two portions:

- X-ray count rate Gd, due to the "external" gamma source (241Am);
- X-ray counting rate Gd, due to the "internal" gamma source (decay products of uranium in the pellet).

When we use the "side source geometry" the pellet moves away from the horizontal surface of the sensitive volume of the detector. The contribution to the total count rate in the doublet 43 keV from the «internal» sources should decrease and will negligible when the activity of the external source and geometry of measurements chose properly.

In general, the value of the effect of the influence of the "internal" source on the total count rate in the doublet 43 keV depends from the activity of the "external" and "internal" source, uranium enrichment, density pellet, measurement geometry.

3. Determination of the Dependence of the Ratio of Count Rate in Peaks with $E_\gamma=185,7$ keV and $E_{\text{x-ray}}=98,43$ keV From the Gd Content in (UO$_2$-Gd$_2$O$_3$)-Pellets.

In this chapter are present the data obtained on the facility when external radiation source is absent. So, only the internal gamma radiation sources are present in the (UO$_2$-Gd$_2$O$_3$)-pellets (decay products of gamma radiation emitters of U).

The analysis is showing, that the mass attenuation coefficient for Gd is bigger of mass attenuation coefficient for U in the energy range above the K-absorption edge of Gd (50,23 keV) and below the K-absorption edge of U (115,6 keV). Therefore, the more Gd concentration in (UO$_2$-Gd$_2$O$_3$)-pellets, the smaller the intensity of the U- radiation in this energy range because of both decrease of U concentration and increase of the summary mass attenuation coefficient.

For gamma- and X- spectra, obtained for each single (UO$_2$-Gd$_2$O$_3$)-pellet the ratio of count rates in peaks $E_\gamma=185,7$ keV (the measure of $^{235}$U) and $E_{\text{x-ray}}=98,43$ keV (the measure of $^{235,238}$U) was determined. The dependence of $\frac{S_{185,7\text{keV}}}{S_{98,43\text{keV}}}$ from Gd content in (UO$_2$-Gd$_2$O$_3$)-pellets is present on the figure 7.
4. Determination of uranium enrichment in (UO$_2$-Gd$_2$O$_3$)-pellets

The certified method of measurement of the uranium enrichment in (UO$_2$-Gd$_2$O$_3$)-pellets using multi-group analysis of the spectra (MGAU code) was used.

For non-irradiated (UO$_2$-Gd$_2$O$_3$)-pellets with passport nominal data of the uranium enrichment $x=6.50\%$ there was obtained in confirming measurements of uranium enrichment by the use of MGAU code $x=(6.53\pm0.03)\%$.

The outstanding features of uranium enrichment measurements by use the MGAU code was presented in [9, 10].

5. Conclusion

The unified gamma-spectrometric method of performing confirmatory measurements of Gd$_2$O$_3$ content and uranium enrichment in non-irradiated single (UO$_2$-Gd$_2$O$_3$)-pellets was developed. In gamma-spectrometric measurements was used planar detector. The Gd$_2$O$_3$ content in (UO$_2$-Gd$_2$O$_3$)-pellets was determined on the basis of the total count rate in doublet 43 keV K$_{\alpha1,2}$-X-rays from Gd. There was used «side-source geometry» as the most practicable source-specimen-detector arrangement. $^{241}$Am is convenient and efficient radiation source for exciting the 43 keV K$_{\alpha}$-X-rays from Gd. Since both the radiation from the $^{241}$Am excitation source and the Gd K$_{\alpha1,2}$-X-rays are appreciably attenuated in typical VVER-fuel cladding material, the X-ray analysis was performed on the unclad fuel pellets. The mean square error in determining of the Gd content in (UO$_2$-Gd$_2$O$_3$)-pellets is $\pm2\%$ (using standard (UO$_2$-Gd$_2$O$_3$)-pellets with the known Gd content, density and enrichment). The accuracy of obtaining results depends from the different parameters: Gd content, density and uranium enrichment of (UO$_2$-Gd$_2$O$_3$)-pellets, activity of the external radiation source, time of the measurement of X-ray spectra, efficiency of HPLeGe detector, measurement geometry, effect/background ratio. An experimental calibration dependence of total count rate in doublet 43 keV from Gd content in (UO$_2$-Gd$_2$O$_3$)-pellets is present by the expression: $S^{43keV}=(7.3\pm0.3)\cdot10^{-3}+(4.2\pm0.1)\cdot10^{-2}\cdot w.f. Gd_2O_3$. Control productivity 1-2 pellet/hour (the activity $\approx$300 kBq for used $^{241}$Am).

The estimates of influence of deviations of the density and Gd content of (UO$_2$-Gd$_2$O$_3$)-pellets on total count rate in doublet 43 keV were performed.

The information obtained on the base of «pure» gamma-spectra of (UO$_2$-Gd$_2$O$_3$)-pellets (without employ of external radiation source) was used to obtain uranium enrichment and to obtain the dependence

![Figure 7. The calibration dependence of $S^{185.7 keV} / S^{98.93 keV}$ from Gd content in (UO$_2$-Gd$_2$O$_3$)-pellets.](image-url)
of \( \frac{S_{185.7\,\text{keV}}}{S_{98.43\,\text{keV}}} \) from Gd content in (UO\(_2\)-Gd\(_2\)O\(_3\))-pellets. The experimental value of \( \frac{S_{185.7\,\text{keV}}}{S_{98.43\,\text{keV}}} \) depends not only from Gd content, but uranium enrichment and density of (UO\(_2\)-Gd\(_2\)O\(_3\))-pellets.

Pellets from UO\(_2\) and (UO\(_2\)-Gd\(_2\)O\(_3\)) can be identified on the basis of differences in the corresponding gamma-ray spectra. In the spectra of (UO\(_2\)-Gd\(_2\)O\(_3\)) - pellets there is an X-ray peak of 43 keV (Gd K\(\alpha_1,2\)). The certified method of measurement of the uranium enrichment in (UO\(_2\)-Gd\(_2\)O\(_3\))-pellets using multi-group analysis of the spectra MGAU was used.

For nominal passport data of the uranium enrichment x=6.50 % in non-irradiated (UO\(_2\)-Gd\(_2\)O\(_3\))- pellets there was obtained in confirming measurements of uranium enrichment by the use of MGAU code \( x=(6.53\pm0.03)\% \).

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