Application of gamma-ray spectrometry for non-destructive determination of $^{235}$U enrichment and mass of uranium in non-irradiated VVER-type fuel pellets

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Application of gamma-ray spectrometry for non-destructive determination of $^{235}$U enrichment and mass of uranium in non-irradiated VVER-type fuel pellets

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Abstract. In order to determine weight fractions of $^{235}$U, $^{238}$U and $^{234}$U in VVER-type fuel pellets there were taken gamma spectra of these UO$_2$- and (UO$_2$-Gd$_2$O$_3$)- pellets using planar detector (HPLeGe) and multi-group gamma-spectra analysis code (MGAU) was used. The mass of $^{238}$U VVER-type fuel pellets was determined as a result of comparison of the count rates in gamma-peaks with $E_G=1001$ keV (the measure of $^{238}$U): in fuel pellet and in thin sample from metal uranium with known characteristics ($^{238}$U mass and weight fractions of $^{235}$U, $^{238}$U and $^{234}$U). The mass of U in fuel pellets was determined on the base of the data of $^{238}$U mass (obtained by gamma-spectrometry method), weight fractions of $^{235}$U, $^{238}$U and $^{234}$U. The possibility of using scintillation spectrometer for measurement of count rates in gamma-peaks with $E_G=1001$ keV in fuel pellet and in thin U-samples was performed. The NaI(Tl) crystal $\varnothing 63*63$ mm contain the wall (measurement hole $\varnothing 10*40$ mm). During the measurements of gamma-spectra, the fuel pellets and thin U-samples were placed on the bottom of the wall of NaI(Tl) crystal. The identification of UO$_2$- and (UO$_2$-Gd$_2$O$_3$)- pellets was performed on the base of corresponding differences in gamma-spectrums, obtained on planar detector (presence of 43 keV K$_\alpha$Gd -peak in (UO$_2$-Gd$_2$O$_3$)- pellet gamma-spectra). The accuracy of the determination of U mass in VVER-type fuel pellets must be sufficient in order to determine the loss of U mass in (UO$_2$-Gd$_2$O$_3$)- pellet (8% wt Gd$_2$O$_3$) with respect to U mass of UO$_2$ pellet (Gd replace U in (UO$_2$-Gd$_2$O$_3$)- pellet).

1. Introduction
Nondestructive gamma-spectrometry methods are widely use for the comprehensive study of nuclear materials. These methods are useful for nuclear material control and accounting, and also for future improvements of technical and economical characteristics of nuclear power reactors [1]. Nuclear safety of the VVER-type reactors depends from the method of the compensation of initial excess reactivity. That’s why the investigation of different types of technological characteristics of (UO$_2$-Gd$_2$O$_3$)-, UO$_2$- pellets is very important [2-5].
2. Determination of weight fractions of $^{235}$U, $^{238}$U and $^{234}$U in fuel pellets

2.1. Equipment
Gamma-spectrums were taken in planar HPLeGe detector with resolution 460 eV at $E_g=122$ keV (full width at half maximum, FWHM). The multi-channel digital analyzer DSA-1000 Canberra and Genie-2000 (including MGAU-code) soft were used.

The outstanding features of use the MGAU-code are present in [6-11].

2.2. The Results of the Experimental Investigation
Gamma-spectrum of UO$_2$- pellet (x=2.4 wt %) is present on Figure 1. It was taken on HPLGe planar detector.

![Gamma-spectrum of UO$_2$- pellet (x=2.4 wt %)](image1.png)

Gamma-spectrum of (UO$_2$-Gd$_2$O$_3$)- pellet (x=6.5 wt %, 8 wt % Gd$_2$O$_3$) is present on Figure 2. The presence of K$_x$Gd double peak with $E=42.3$ keV and 42.9 keV allow to identify the presence of Gd in (UO$_2$-Gd$_2$O$_3$)- pellets.

![Gamma-spectrum of (UO$_2$-Gd$_2$O$_3$)- pellet (x=6.5 wt %, 8 wt % Gd$_2$O$_3$)](image2.png)
The results of weight fraction of $^{235}\text{U}$, $^{238}\text{U}$ and $^{234}\text{U}$ in U- samples, UO$_2$-, (UO$_2$-Gd$_2$O$_3$)- pellets were obtained by use of MGAU-code. The results of weight fractions of $^{235}\text{U}$, $^{238}\text{U}$ and $^{234}\text{U}$ in U- sample (standard sample) are present in Table 1.

**Table 1. Weight fractions of $^{235}\text{U}$, $^{238}\text{U}$ and $^{234}\text{U}$ in U- sample**

|          | $^{235}\text{U}$,wt% | $^{238}\text{U}$,wt% | $^{234}\text{U}$,wt% |
|----------|-----------------------|-----------------------|-----------------------|
|          | 0,702 ± 0,005         | 99,293 ± 0,005        | 0,005 ± 0,001         |

The results of weight fraction of $^{235}\text{U}$ in UO$_2$- pellets are present in Figure 3.

![Figure 3. The results of weight fraction of $^{235}\text{U}$ in different UO$_2$- pellets](image)

3. **Determination of $^{238}\text{U}$ mass in fuel pellets**

$^{238}\text{U}$ – is a weak gamma-emitter ($E_{\gamma}=49.5$ keV (0.005%), 113.5 keV (0.01%)), therefore these gamma lines are not used in order to detect of $^{238}\text{U}$. The best measure of $^{238}\text{U}$ can be the gamma line with $E_{\gamma}=1001$ keV (0.84 %) belonging to $^{234}\text{mPa}$ ($T_{1/2}=1.17$ min) – the second decay product of $^{238}\text{U}$. The activity of $^{234}\text{mPa}$ in equilibrium with $^{238}\text{U}$ was measured in present work. Equilibrium ($^{238}\text{U} - ^{234}\text{mPa}$) is achieved in 4 months because $^{234}\text{Th}$ has short half-life ($T_{1/2}=24.1$ days). So, the gamma-peak 1001 keV is the measure of $^{238}\text{U}$.

Comparison of gamma spectra, taken on NaI(Tl)-detector and HPGe coaxial detector are present in Figure 4.
3.1. Equipment
The scintillation type of detector with inorganic NaI(Tl) crystal was used in measurements in order to determine the relative count rates in gamma peak with $E_g = 1001$ keV in fuel pellets. Two types of NaI(Tl) crystals were used in scintillation spectrometer (Figure 5, Figure 6). Technical characteristics of used scintillation detectors are present in Table 2.

Figure 4. Gamma-spectra of VVER-type fuel pellet ($x=2,4$ wt %), taken on two types of gamma-spectrometers: 1-HPGe coaxial detector; 2- NaI(Tl)- detector

Figure 5. The view of photomultiplier and NaI(Tl) crystal: 1- crystal with wall; 2- crystal without wall

Figure 6. The view of the scintillation facility with shielding
Table 2. Technical characteristics of used scintillation detectors

|                                | NaI(Tl) crystal with wall | NaI(Tl) crystal without wall |
|--------------------------------|----------------------------|------------------------------|
| The range of detected energies of gamma-radiation | (0.05 – 3) MeV             | (0.05 – 3) MeV               |
| The relative energy resolution for 662 keV ($^{137}$Cs), % | ≈8.3                       | ≈7                           |
| Geometrical dimensions of crystal, mm | Ø63*63; wall Ø10*36 (cylindrical hole) | Ø63*63                      |
| Count pulse rate in the peak with $E_r=1001$ keV, $\frac{\text{count}}{s \cdot g^{238U}}$ | ≈ 48.3                     | ≈ 8.7                       |

The aim of the experiments with different types of scintillation detectors is the investigation of the next important problems. 1) Sum peak effect. When measuring sample closed to the detector, the count rate $S^{1001keV}$ may be underestimated. This underestimation is caused by true coincidence between different gamma-peaks. These coincidences result in a decrease of $S^{1001keV}$. 2) Influence of beta-particles on the peak count rate $S^{1001keV}$ [12-14].

The geometry of gamma-spectra fuel pellet measurements is present at Figure 7.

![Figure 7](image-url)

1- fuel pellet; 2 - NaI(Tl) crystal without wall.

The distance from the VVER-type fuel pellet to the top of NaI(Tl) crystal $h=50$ mm

The VVER-type fuel pellet is at the bottom of the wall.

**Figure 7.** Two types of the geometry of gamma-spectra fuel pellet measurements
Axial distributions of relative count rates in gamma-peaks from the distance «point» source - wall bottom for the wall of NaI(Tl) crystal are present on Figure 8. Measurements were performed by use $^{60}$Co «point» source.

**Figure 8.** Axial distributions of relative count rates in gamma-peaks from the distance «point» source - wall bottom for the wall of NaI(Tl) crystal: 1- $E_p=1333$ keV; 2- $E_p=1173$ keV

Given that the nominal geometric dimensions of non-irradiated VVER-type pellets are $7.6*12$ mm (within technological uncertainties), it can be concluded that the optimal $4\pi$-geometry of measurements is realized in experiments with the selected NaI(Tl) crystal with wall.

### 3.2. The Procedure of the Experimental Investigations

The expression for determining the mass of $^{238}$U in a fuel pellet:

$$M_{fuel\ pellet}^{(238}U) = M_{st.\ sample}^{(238}U) \cdot \frac{S_{fuel\ pellet}^{1001keV}}{S_{st.\ sample}^{1001keV}} \cdot \frac{k_{fuel\ pellet}}{k_{st.\ sample}}$$

were $M_{st.\ sample}^{(238}U)$ - the $^{238}$U mass of thin sample from uranium metal with a known weight fractions of $^{235}$U, $^{233}$U, $^{234}$U (standard sample); $\frac{S_{fuel\ pellet}^{1001keV}}{S_{st.\ sample}^{1001keV}}$ - the ratio of count rates in peaks with $E_p=1001$ keV for the fuel pellet and for standard sample; $\frac{k_{fuel\ pellet}}{k_{st.\ sample}}$ - the ratio of self-absorption factors of gamma-radiation in fuel pellet and standard sample [1].
3.3. Experimental results

Gamma-spectrometric measurements were performed for the standard thin sample from U-metal. The count pulse rate in the peak with \( E_1 = 1001 \text{ keV} \) was obtained: \( S^{1001\text{keV}} = (10,8 \pm 0,3) \frac{\text{count}}{s \cdot g \cdot ^{238}\text{U}} \)

The next experimental parameters were determined (Figure 9-11)

The dependence of the count pulse rate in the peak of 1001 keV (normalized by the ligature weight of the sample):
- for thin samples from U, UO₂, (UO₂-Gd₂O₃);
- for «pellets» made up of a different number of thin samples;
- for full-size pellets from UO₂, (UO₂-Gd₂O₃).

Estimation of the specific counting rate at peak 1001 keV, normalized by weight \(^{238}\text{U}\) for thin samples from U, UO₂, (UO₂-Gd₂O₃) - based on the calculated mass fractions of \(^{238}\text{U}\)

![Graph](image)

**Figure 9.** Dependence of count rate in the peak with \( E_1 = 1001 \text{ keV} \) (normalized by the ligature weight of the sample) for the different thin samples from UO₂ (\( x = 2,4 \% \)) from number of sample
Figure 10. Dependence of the count rate in the peak with $E_0=1001$ keV (normalized by the ligature weight of the sample) for the «pellets» made up of different number of thin samples from UO$_2$ ($x=2.4$ wt %) from mass of «pellet»

Figure 11. Dependence of the count rate in the peak with $E_0=1001$ keV (normalized by the ligature weight of the sample) for the full-size pellets from UO$_2$ ($x=2.4$ wt %) from number of pellet

4. Conclusion
1. The weight fractions of $^{235}\text{U}$, $^{238}\text{U}$ and $^{234}\text{U}$ were determined in VVER-type fuel pellets using planar detector (HPLeGe) and multi-group gamma-spectra analysis code (MGAU) was used.
2. The identification of UO₂- and (UO₂-Gd₂O₃)- pellets was performed on the base of corresponding differences in gamma-spectrums, obtained on planar detector (presence of 43 keV Kα,Gd -peak in (UO₂-Gd₂O₃)- pellet gamma-spectra).
3. The possibility of using of scintillation spectrometer for measurement of count rates in gamma-peaks with Eₙ=1001 keV in fuel pellet and in thin U-samples was performed. The NaI(Tl) crystal Ø63*63 mm containing the wall Ø10*40 mm was used.
4. The mass of ²³⁸U VVER-type fuel pellets was determined as a result of comparison of the count rates in gamma-peaks with Eₙ=1001 keV (the measure of ²³⁸U): in fuel pellet and in thin sample from metal uranium with known characteristics (²³⁸U mass and weight fractions of ²³⁵U, ²³⁸U and ²³⁴U). The mass of U in fuel pellets was determined on the base of the data of ²³⁸U mass (obtained by gamma-spectrometry method), weight fractions of ²³⁵U, ²³⁸U and ²³⁴U.
5. Gamma-spectrometric measurements were performed for the standard thin sample from U-metal. The count pulse rate in the peak with Eₙ=1001 keV was obtained: 

\[ S^{1001 \text{keV}} = (10,8 \pm 0,3) \frac{\text{count}}{s \cdot g^{238U}} \]

6. Preliminary results were obtained: the dependence of the count pulse rate in the peak of 1001 keV (normalized by the ligature weight of the sample): for thin samples from U, UO₂, (UO₂-Gd₂O₃); for «pellets» made up of a different number of thin samples; for full-size pellets from UO₂, (UO₂-Gd₂O₃).

7. The accuracy of the determination of U mass in VVER-type fuel pellets must be sufficient in order to determine the loss of U mass in (UO₂-Gd₂O₃)- pellet (8% wt Gd₂O₃) with respect to U mass of UO₂ pellet (Gd replace U in (UO₂-Gd₂O₃)- pellet)

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