

ANALYSIS OF THE APPLICATION OF CDZnTE DETECTORS FOR CONTROL OF INITIAL ENRICHMENT OF FRESH NUCLEAR FUEL FOR OVERLOADING

The task of determining the initial enrichment of nuclear fuel (NF) at nuclear power plants in real time is relevant for both economic indicators of operation of the power unit and for all components of nuclear safety (nuclear safety, security and safeguards). The technology used by nuclear fuel cycle companies is not suitable for use at nuclear power plants. It is proposed to use a measuring system based on CdZnTe semiconductor detectors designed to control the burn-up depth of nuclear fuel for estimating the initial enrichment of fresh nuclear fuel in fuel assemblies (FA). The initial enrichment is estimated with help of the gamma radiation of uranium isotopes of fresh nuclear fuel. A distinctive feature of the system that has been developed is the use of passive tomography technology to take into account the self-absorption of radiation in the FA. The promise of the proposed method for estimating initial enrichment is confirmed by experimental measurements and simulation results. It is shown that the system developed makes it possible to determine the enrichment of nuclear fuel, taking into account its profiling by the volume of FA. Also, the results of measurements confirmed the possibility of using the proposed technical solutions for CdZnTe detectors to create portable monitoring systems for special nuclear material in the field. The solution of this task is of great importance for national security.

Keywords: CdZnTe detectors, gamma-ray imaging, gamma-ray spectroscopy, special nuclear materials monitoring.

Full control over the state of nuclear fuel, at all stages of its life cycle, at nuclear power plants necessarily requires an instrumental assessment of the initial enrichment of fresh fuel before it is loaded into the reactor core. Nuclear fuel (NF) is a mixture of ^{235}U isotopes with a range of content of 1.6-4.4%, therefore the initial isotope composition of nuclear materials is often the object of measurements. However, there are no such systems for determining the initial enrichment of nuclear fuel at nuclear power plants. Therefore, at present, it is necessary to rely solely on the data

declared by the manufacturer. Knowledge of the actual deviation of the initial enrichment is of great importance in order to optimize the fuel permutations, especially when operating at the final stages of the campaign.

The determination of the initial enrichment of NF (i.e., the content of the fissile isotope ^{235}U) in real time can be carried out through passive radiation measurements of the sample's self-radiation. Like all passive measurements, this method of analysis is non-destructive [1]. The determination of uranium enrichment level in samples is a key measurement in technological processes and in the production control on the uranium enrichment and nuclear fuel companies, it acts an important part in the international nuclear safeguards inspections to grantee that uranium fuel is used for peaceful purposes. The principles of enrichment measurement can be used to determine the content of any isotope if their radiation characteristics are known and some special measurement conditions [2,...,5] are met.

The main radiation used in passive non-destructive analysis of uranium samples is gamma radiation. When measuring the enrichment of ^{235}U , the gamma radiation line with a gamma-ray energy of 186 keV is most often used. It is the most distinguished single gamma-emission line for any uranium samples enriched by ^{235}U above the natural level.

The method for determining the enrichment of uranium by gamma radiation was for the first time used to control the gas cylinders with UF_6 [1, 4, 6]. The method of the measurement included recording the radiation of the uranium sample through the channel of the collimator by the detector of the gamma radiation. The enrichment was determined by the intensity of gamma radiation of the ^{235}U isotope with 186 keV energy. If the sample is thick enough, the detector detects gamma rays with an energy of 186 keV only from an insignificant part of the total sample, because of the strong absorption of low-energy quanta in the fuel assembly materials. In this sense, in the terminology of [1], fuel element is a "thick sample". The "visible volume" of the sample is characterized by the configuration of the collimator, the detector geometry and by the length of the free path of radiation with an energy of 186 keV in the sample material [1, 3, 4].

We propose a measuring system based on CdZnTe detectors [7, 8, 9], created for the burn-up analysis of spent nuclear fuel, can be used to estimate the initial enrichment of an overloaded fresh fuel. In this case, the main problem may be the low detection efficiency of the detectors used. To test the possibility of using CdZnTe detectors, fuel pellets with an initial enrichment of 4.4% were measured. One of the obtained spectra is shown in Fig. 1 and Fig. 2, using different scales of the axes for displaying the count results (i.e. measured intensity) in the channels of the multichannel analyzer. It is done to provide greater visibility of the peak of the total absorption with gamma radiation with energy of ^{235}U 205.3 keV. In the spectrum, well-identifiable lines of 186 keV and 205 keV of self-radiation of ^{235}U are observed. The lines of sample self-radiation of ^{238}U with energies of 742 keV, 766 keV, 786 keV and 1001 keV are visible as well. The 1001 keV line is usually used for analytical purposes.

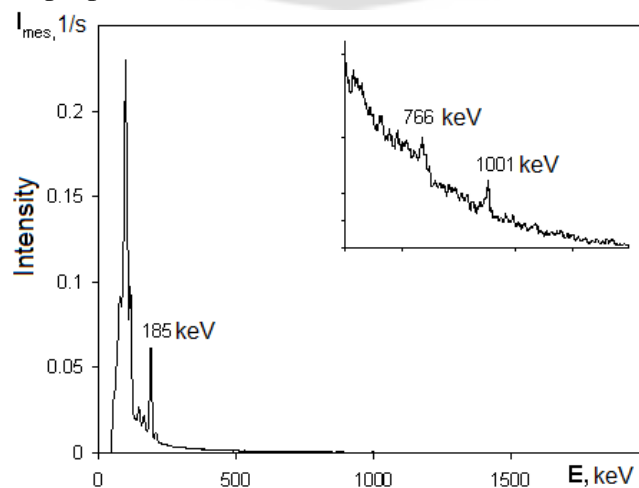


Fig. 1. The spectrum of gamma self-radiation of fuel pellets with initial enrichment of 4.4%

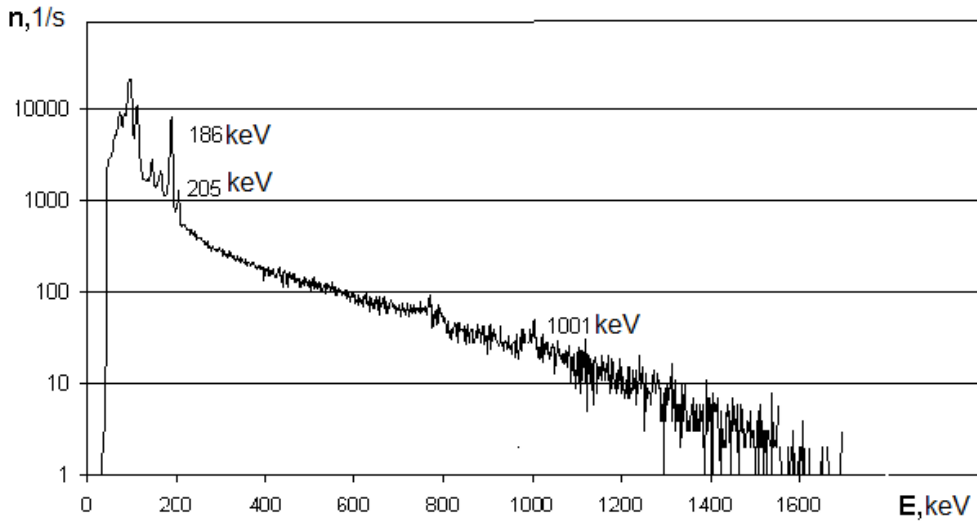


Fig. 2. The spectrum of gamma self-radiation of fuel pellets with an initial enrichment of 4.4% on a semilogarithmic scale

For comparison, Fig. 3 shows the spectrum of the self-radiation of a metal sample of depleted uranium. The mass of uranium in both series of measurements was approximately the same. It can be seen that the analytical lines of the ^{235}U isotope are absent, and the radiation intensity is much lower. In both cases, the value of the parameter of the thickness of the sample is the "infinite thickness".

The determination of the initial enrichment using the ^{235}U self-radiation intensity measurements with 186 keV energy is carried out according to the following formula [1, 11]:

$$R = a \cdot I_{mes} \cdot F \cdot \exp(\mu_c \rho_c t_c) = F \cdot \exp(\mu_c \rho_c t_c) (a \cdot C_1 - a \cdot f \cdot C_2), \quad (1)$$

where I_{mes} is the net counting rate at the peak of full absorption;

C_1 and C_2 are the total counting rate in the peak of the total absorption and the counting rate of the background and the continuous component of the Compton distribution, respectively;

$F \cdot \exp(\mu_c \rho_c t_c)$ are the experimentally determined coefficients for taking into account self-absorption and scattering of radiation in the fuel matrix material and fuel cladding walls.

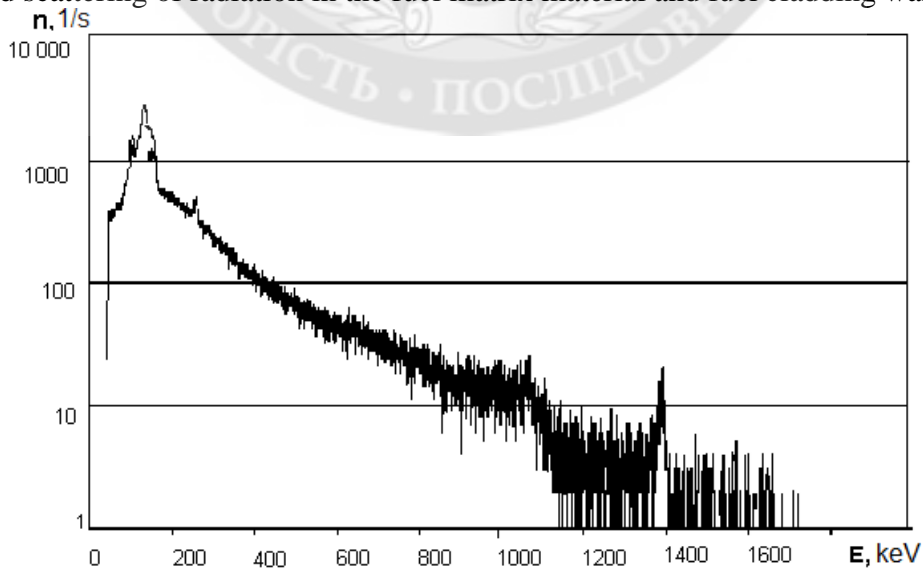


Fig. 3. Spectrum of the self-radiation of a metal sample of depleted uranium

Such a method is fairly simple when carrying out measurements of typical homogeneous samples, for example, in the measurement of barrels containing uranium dioxide. However, FA is an object with a complex geometry and in this case it cannot be considered as a "thick" bung. As a rule, it can be assumed that the main contribution to the counting rate of the full-absorption peak with 186 keV energy is given by one, or at most two rows of external fuel rods. To increase the recording efficiency and reduce the measurement time, the detector should be positioned as close as possible to the fuel assemblies, but in this case, there are observation points where so-called "firing" from fuel rods located in the inner rows are recorded, while the gamma radiation intensity is very uneven depending on the viewing angle [10]. An example of such a phenomenon, obtained by the modeling described below, is shown in Fig. 4. This leads to a strong dependence of the obtained results on the location of the detector relative to the measured FA. This effect is even more pronounced when measuring fuel assemblies containing fuel elements with different enrichment (for example, fuel assemblies marked "ED" contain an external row of fuel rods with an enrichment of 3.6%, and internal fuel rods have an enrichment of 4.4%).

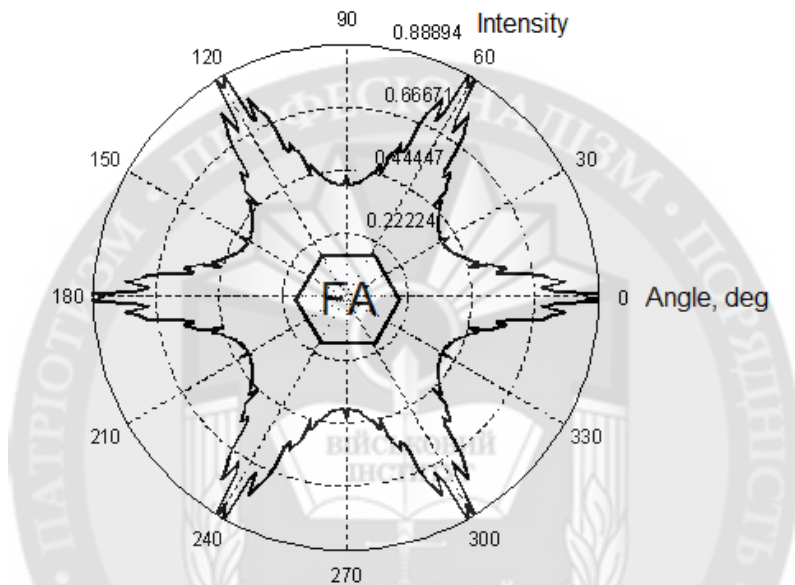


Fig. 4. The intensity of the gamma radiation of ^{235}U 186 keV, depending on the angle of the detector (the distance "FA axis – detector" is 14 cm).

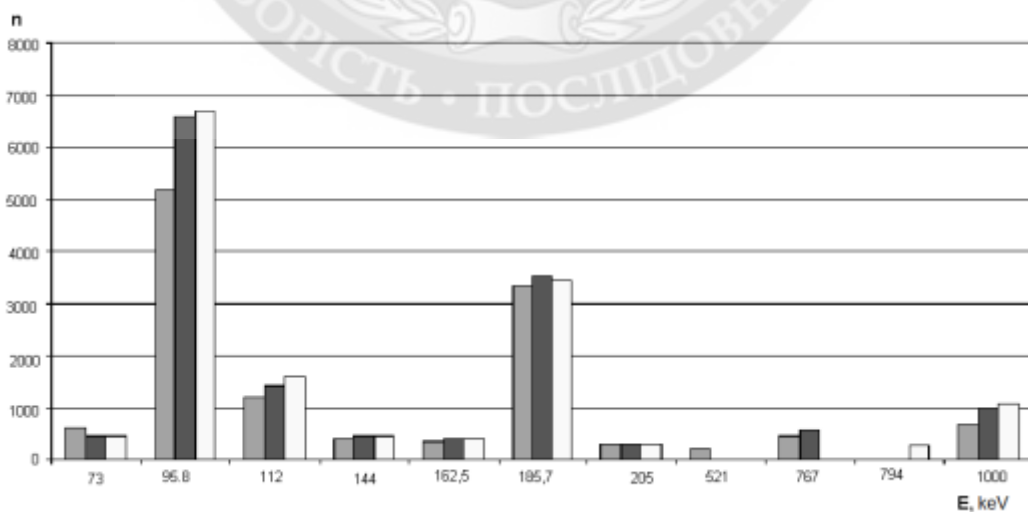


Fig. 5. Intensities of the gamma self-radiation during measurements from one, two and three layers of fuel pellets with an initial enrichment of 4.4%

To reduce the measurement error, we took into account the effect of self-absorption and scattering of radiation in the material of the fuel matrix and fuel cladding walls, enrichment can be determined on the basis of data on the ratio of the counting rate at the peak of full-energy absorption of 186 keV of the ^{235}U self-radiation and the counting rate at the peak of full absorption of 1001 keV of self-radiation of ^{238}U . Figure 5 shows the results of measurements of gamma-radiation intensity of one (gray bars), two (black bars), and three (white bars) layers of VVER-1000 fuel pellets. It is clearly seen that an increase in the thickness of the measured layer leads to a significant increase in the emission intensity of the 1001-keV line of self-radiation of ^{238}U and secondary X-ray radiation due to the interaction of the primary radiation with the fuel matrix.

The dependence of enrichment on the ratio of the self-radiation intensities of ^{235}U and ^{238}U for FA will obviously be nonlinear due to the different absorption of gamma quanta with energies of 186 keV and 1001 keV. For the empirical determination of this dependence, it is necessary to carry out a sufficient number of measurements for a fuel assembly with a different enrichment in a statistical sense, which in principle is not a problem under NPP conditions [10, 12]:

To confirm the possibility of estimating the initial enrichment of nuclear fuel, a two-dimensional plane model of the field of the gamma radiation of uranium isotopes contained in the fuel assemblage was constructed, according to which the location of the detector at the n -th observation point at a distance R_n from the FA axis the gamma-radiation of j -th isotope of uranium with energy E_j at the location of the detector is:

$$I_n^{jk} = \sum_m A_{mj} k_{jk} w_{mn}, \quad (2)$$

where A_{mj} is the activity of the j -th uranium isotope for m -th fuel element taking into account the actual value of enrichment;

k_{jk} is the quantum yield of the k -th gamma line for the j -th isotope;

w_{mn} is the coefficient of the contribution of the m -th fuel element to the intensity of the gamma radiation of the j -th isotope with energy E_k , taking into account the effects of attenuation when the gamma-ray beam propagates from the m -th fuel element to the n -th point of observation, $m = 1, \dots, 312$.

Taking into account the registration efficiency, the measured intensity (counting rate) $\varepsilon(E_k)$ for an isotope with energy E_k the measured gamma radiation intensity of the j -th isotope with energy E_k is expressed as

$$I_n^{jkm\epsilon s} = \sum_m A_{mj} k_{jk} w_{mn} \varepsilon(E_k). \quad (3)$$

In the future, we omit the indices jk , since we are talking about the intensity of gamma radiation at the point of registration of the j -th isotope with energy E_k , i.e. in the simulation it is assumed that the peaks of the gamma spectrum are isolated and processed.

The attenuation in the propagation of gamma radiation is mainly connected with two physical mechanisms:

– geometric attenuation associated with the spatial divergence of the beam

$$P_{geom} = \frac{S_{det}}{4\pi R_{mn}^2}, \quad (4)$$

where S_{det} is the active area of the detector;

R_{mn} is the distance from the center of the m -th fuel rod to the n -th location of the detector;

– physical attenuation of gamma radiation during passage through various media (in particular, when passing through UO_2 in other fuel elements located on the path of gamma-ray run to the detector, in their zirconium shells, and also in the water environment – both inside the FA and

outside of it, in the overload pool); the physical attenuation of radiation from the m -th fuel element is expressed as

$$P_{phys} = \exp\left(-\sum_l \mu_l l_l\right), \quad (5)$$

where μ_l is the coefficients of linear attenuation of gamma quanta with energy E_k for the corresponding media;

l_l is the length of run of gamma radiation in a medium of type l (uranium dioxide, zirconium, or water).

Thus, the contribution coefficient of the m -th fuel element for the n -th position of the detector, associated with attenuation, is

$$w_m = \frac{S_m}{4\pi R^2 \sum_l \mu_l l_l} \quad (6)$$

According to the formulated model, the fields of gamma self-radiation with an energy of 185 keV of the ^{235}U isotope and with an energy of 1001 keV for ^{238}U at different locations of the detector were calculated. All the results presented below are calculated from the relations (2), (3), (6) for the "FA axis – detector" distance of 22.5 cm. This is because of the fact that the developed equipment for monitoring the fuel burn-up depth was specially designed for placement on the working bar of the reloading machine (RM), and the specified size is determined by the diameter of the working rod of the RM. The angular diagram of the intensity distribution of gamma radiation with an energy of 185 keV ^{235}U is shown in fig. 6. It can be seen that the intensity distribution practically repeats the shape of the fuel assembly. At separate viewing angles, there are irregularities and "sharp increases", the magnitude of which is small and does not exceed 7-8% of the mean. There are smooth maxima at the angles corresponding to FA edges.

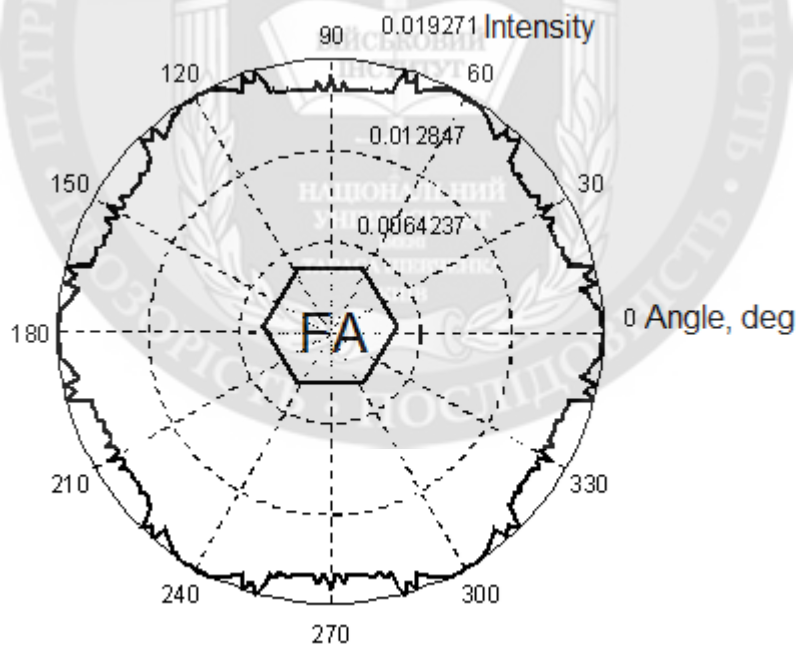


Fig. 6. Angular distribution of intensity of gamma self-radiation for ^{235}U 185 keV (fresh fuel assembly)

The coefficients of the contributions of individual fuel elements w_{mm} are investigated by the relation (6). For the energy 185 keV of ^{235}U isotope in the fig. 7, the inverse values of the contributions are given in a logarithmic scale. The type of the contribution pattern is typical for low-energy isotopes, in particular, at the observation angles corresponding to the FA edges, there is an acute "ridge", and a pronounced "multi-ridge structure" is observed on the midpoints of the faces.

Figure 8 shows the angular distribution of the intensity of gamma radiation with an energy of 1001 keV of the ^{238}U isotope. The angular distribution is fairly smooth, the nonuniformity does not exceed 5-7%. The corresponding energies of 1001 keV ^{238}U of the inverse values which are inverse to fuel element contribution coefficients are shown in fig. 9. Their appearance is also quite typical for high-energy gamma quanta: in particular, the "ridge structure" is much less pronounced than in fig. 7.

To research the practical possibility of determining the initial burn-up of fresh nuclear fuel, the ratio of measured intensities of ^{238}U with energy of 1001 keV and ^{235}U with an energy of 185 keV is modelled, taking into account the registration efficiency $\varepsilon(E_k)$ [7] depending on the angle of the detector location (fig. 10). It can be seen from the figure that the ratio of the measured intensities repeats to a considerable extent the picture in fig. 6: there are irregularities and small "sharp increases" observed in the form of peaks, and besides the peaks being concentrated at the viewing angles corresponding to the midpoints of edges. However, the absolute value of the peaks is small, which allows us to assume that the detectors are arranged exactly opposite to the edges.

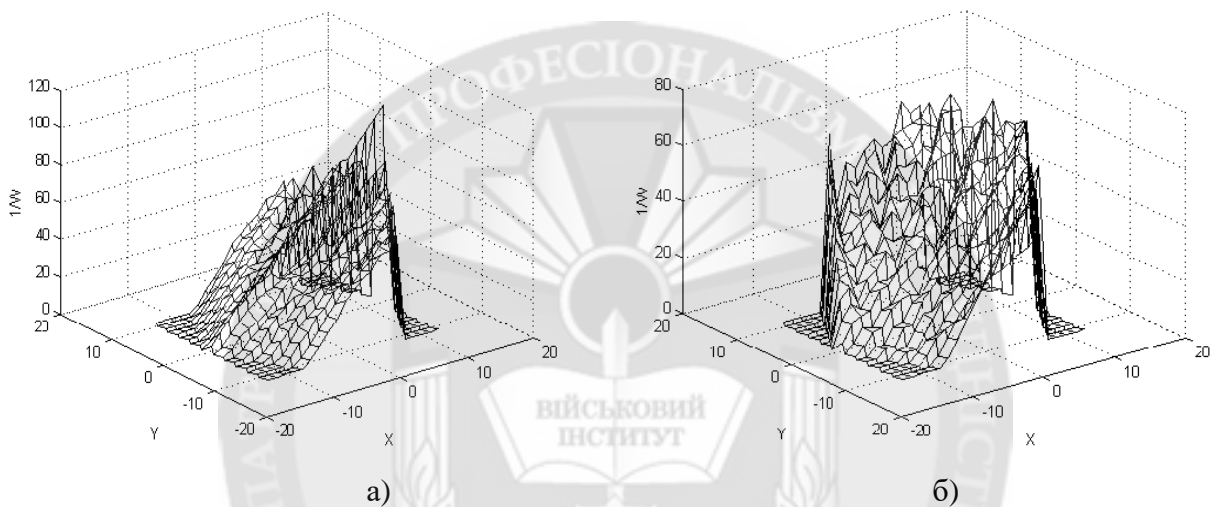


Fig. 7. Coefficients of fuel rod contributions (inverse values) for ^{235}U 185 keV (a – 0° , b – 30°)

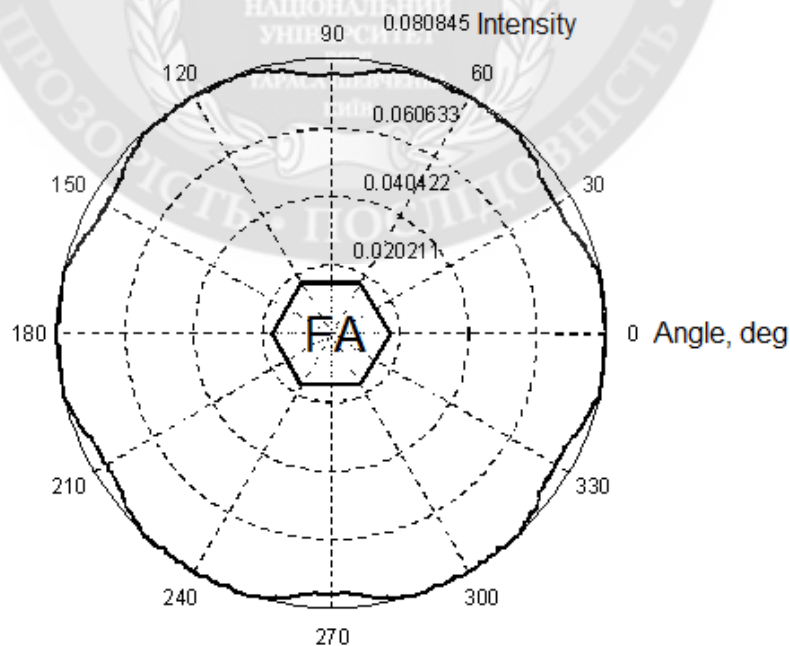


Fig.8. Angular distribution of the intensity of the gamma self-radiation of ^{238}U with an energy of 1001 keV (fresh FA)

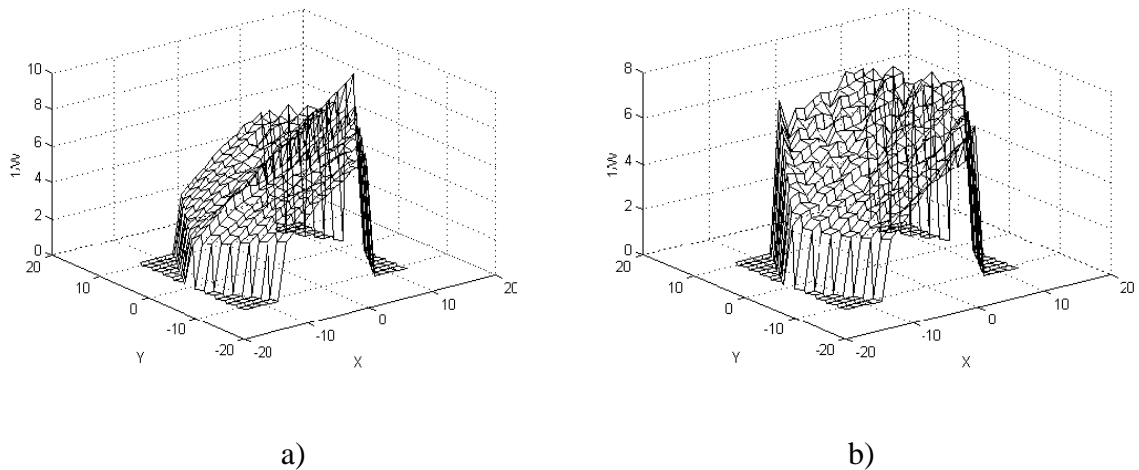


Fig. 9. Coefficients of fuel element contributions (inverse values) for ^{238}U 1001 keV (a - 0° , b - 30°)

Quantitative analysis of the contribution coefficients w_{mn} for both analytical lines of ^{235}U , ^{238}U isotopes made it possible to determine the following: with an allowable unevenness of contribution coefficient of 40%, placing the detector in opposite to the midpoint of the fuel assembly allows coverage of the entire external row of fuel rods and 4 central fuel elements of the second row for 185 keV ^{235}U energy, and almost completely two fuel rods close to the detector for energy 1001 keV ^{238}U . Considering the homogeneity of fresh nuclear fuel inside the fuel assembly (except for the first row), this is quite sufficient to estimate the initial enrichment. The number of detectors should be determined experimentally, depending on the required accuracy of measurements.

The above experimental spectra and simulation results show that the developed technical facilities to control the burn-out of spent nuclear fuel can be successfully used to determine the initial enrichment of fresh nuclear fuel.

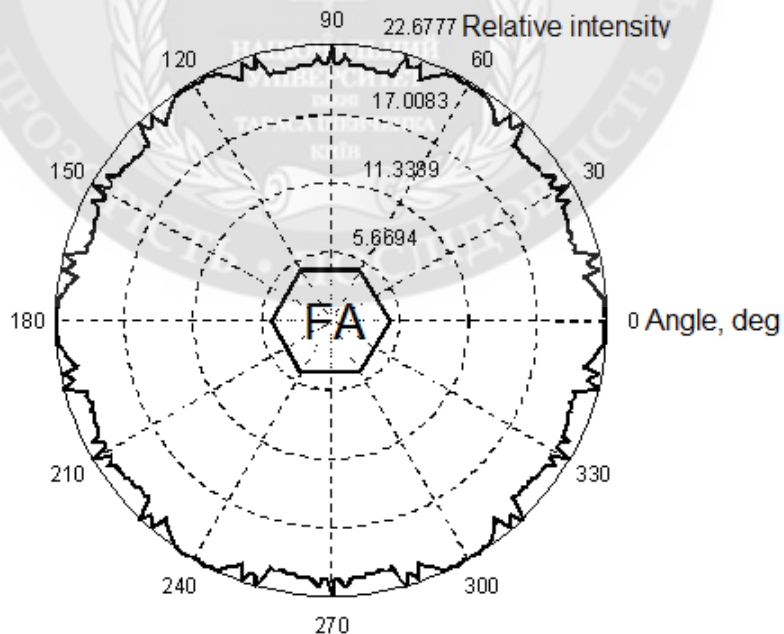


Fig.10. The ratio of the measured intensities of ^{238}U 1001 keV and ^{235}U 185 keV depending on the detector location angle

The developed equipment makes it possible to determine the enrichment of nuclear fuel when it is transferred by a reloading machine to the reactor core without additional time. The introduction

of such a system provides a qualitatively new level of monitoring of the state of nuclear fuel at all stages of its life cycle at nuclear power plants. In particular, there will be an automatic analysis of the initial enrichment of nuclear fuel, a burn-out analysis for each campaign based on the results of measurements during the overload, a control of the fuel cladding tightness during the overload, and an analysis of burn-out during shipment to dry storage. In addition, close integration with the control system of the overload machine ensures automatic maintenance of a database on the history of nuclear fuel, i.e. fulfillment of the IAEA requirements for nuclear safeguards. Continuous accounting of the burn-out of each fuel assembly is also the main source of information to refine the methods of nuclear-physical calculations of the reactor core.

REFERENCES:

1. [Reilly D. Passive Nondestructive Assay of Nuclear Materials / D. Reilly, Norbert Ensslh, Hastings Smith. Nuclear Regulatory Commission. – 1991. – 700 p.]
2. Soltys I., Yacout A. M., McKnigh R. D. Application of Control Charts and Isotope Correlations to Spent Fuel Measurements at FCF / 40th Annual Meeting of the Institute of Nuclear Material Management. July 25-29, 1999, Phoenix. – 7 p.
3. Neutron and gamma-ray energy reconstruction for characterization of special nuclear material / Shaun D. Clarke , Michael C. Hamel, Angela Di Fulvio, Sara A. Pozzi // Nuclear Engineering and Technology – 2017 – Vol. 49, No. 6 – P. 1354-1357 – DOI:10.1016/j.net.2017.06.005
4. A Method to Estimate the Atomic Number and Mass Thickness of Intervening Materials in Uranium and Plutonium Gamma-Ray Spectroscopy Measurements / Michael Streicher, Steven Brown, Yuefeng Zhu, David Goodman, and Zhong He // Nuclear Science IEEE Transactions – 2016 – Vol. 63, No. 5 – P. 2639-2648 – DOI: 10.1109/TNS.2016.2606763
5. Special Nuclear Material Characterization Using Digital 3-D Position Sensitive CdZnTe Detectors and High Purity Germanium Spectrometers / Michael Streicher, Steven Brown, Yuefeng Zhu, David Goodman, Zhong He // Nuclear Science IEEE Transactions – 2016 – Vol. 63, No. 5 – P. 2649-2656 – DOI: 10.1109/TNS.2016.2593631
6. Signatures and Methods for the Automated Nondestructive Assay of UF₆ Cylinders at Uranium Enrichment Plants / L. Eric Smith, Emily K. Mace, Alex C. Misner, and Mark W. Shaver // IEEE Transactions on Nuclear Science – 2010 – Vol. 57, No. 4 – P. 2247 - 2253
7. Fast Neutron Source Localization Using Digital Pixelated 3-D Position-Sensitive CdZnTe Detectors // David Goodman, Michael Streicher, Yuefeng Zhu, Steven Brown and Zhong He, // Nuclear Science IEEE Transactions – 2017 – Vol. 64, No. 9, – P. 2531 - 2535
8. Oleinik S. G., M. V. Maksimov, O V. Maslov, 2002, Determination of the burnup of spent nuclear fuel during reloading –, Atomic Energy, Vol. 92, No. 4, – P. 296-300.
9. Burnup Measurement of Spent Fuel Assembly by CZT-based Gamma-Ray Spectroscopy for Input Nuclear Material Accountability of Pyroprocessing / Hee Seo, Jong-Myeong Oh, Hee-Sung Shin, Ho-Dong Kim, Seung Kyu Lee, Hyun Su Lee, and Se-Hwan Park // Nuclear Science IEEE Transactions – 2014 – Vol. 61, No. 4 – P. 2169 - 2174
10. Oleinik S. G., V. A. Boltenev, O V. Maslov, Passive computer γ -tomography of nuclear fuel, – Atomic Energy. – 2005 – Vol. 98, No. 3, – P. 227-229.
11. Mokritsky V.A. Automated system for determining the burnup depth of spent nuclear fuel. V.A. Mokritsky, O.V. Maslov, O.V. Banzak // Technology and design in electronic equipment. - 2014. – No. 5 – 6. – P. 63 – 71.
12. Mokritsky V.A. Method for determining the burnup depth of spent nuclear fuel (SNF) / V.A. Mokritsky, O.V. Maslov, O.V. Banzak. – Collection of Scientific Papers of the Military Institute of Taras Shevchenko National University of Kiev. – Kiev, 2014. – No. 45. – P.79 – 83.

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**АНАЛІЗ ЗАСТОСУВАННЯ CDZnTE ДЕТЕКТОРІВ ДЛЯ КОНТРОЛЮ ПОЧАТКОВОГО
ЗБАГАЧЕННЯ СВІЖОГО ЯДЕРНОГО ПАЛИВА ПРИ ПЕРЕВАНТАЖЕННІ**

Задача визначення початкового збагачення ядерного палива (ЯП) на АЕС в реальному часі є актуальною для як економічних показників експлуатації енергоблоку так і для ядерної безпеки. Запропоновано для оцінки початкового збагачення свіжого ЯП в тепловиділяючих збірках (ТВЗ) використати вимірювальну систему на базі CdZnTe- детекторів. Оцінка початкового збагачення здійснюється по власному гамма-випромінюванню ізотопів урану ЯП. Особливістю системи є застосування технології пасивної томографії для врахування самопоглинання випромінювання в ТВЗ. Пропонований метод оцінки початкового збагачення підтверджується експериментальними вимірами і результатами моделювання. Показано, що розроблена система дозволяє визначати збагачення ЯП з урахуванням його розподілу за об'ємом ТВЗ. Отримані результати підтвердили можливість використання запропонованих технічних рішень для створення портативних систем контролю спеціальних ядерних матеріалів в польових умовах. Рішення вказаної задачі має важливе значення для національної безпеки.

Ключові слова: CdZnTe детектори, пасивна томографія, спектрометрія гамма-випромінювання, контроль ядерних матеріалів

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**АНАЛІЗ ПРИМЕНЕНИЯ CDZnTE ДЕТЕКТОРОВ ДЛЯ КОНТРОЛЯ НАЧАЛЬНОГО
ОБОГАЩЕНИЯ СВЕЖЕГО ЯДЕРНОГО ТОПЛИВА ПРИ ПЕРЕГРУЗКЕ**

Задача определения начального обогащения ядерного топлива (ЯТ) на АЭС в реальном времени является актуальной для как экономических показателей эксплуатации энергоблока, так и для всех составляющих показателей ядерной. Предлагается использовать для оценки начального обогащения свежего ЯТ в тепловыделяющих сборках (ТВС) измерительную систему на базе CdZnTe-детекторов. Оценка начального обогащения осуществляется по собственному гамма-излучению изотопов урана свежего ЯТ. Особенностью системы является использование технологии пассивной томографии для учета самопоглощения излучения в ТВС. Предлагаемый метод оценки начального обогащения подтверждается экспериментальными измерениями и результатами моделирования. Показано, что разработанная система позволяет определять обогащение ЯТ с учетом его профилирования по объему ТВС. Полученные результаты подтвердили возможность использования предложенных технических решений для создания портативных систем контроля специальных ядерных материалов в полевых условиях. Решение указанной задачи имеет важное значение для национальной безопасности.

Ключевые слова: CdZnTe детекторы, пассивная томография, спектрометрия гамма-излучения, контроль ядерных материалов