Determination of ^{238}U Content by Gamma Radiation Emitting from ^{234m}Pa Radionuclide

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Abstract

Radionuclide ^{238}U is one of the most important radioactive elements that must be controlled in nuclear power engineering, geological exploration, control of radioactive contamination of soils and raw materials used in construction. The most optimal way to control ^{238}U is to use the ^{234m}Pa radionuclide, the activity of which, due to its short lifetime (≈ 1.2 min), is unambiguously related to the activity of ^{238}U even if the secular equilibrium is disturbed in the sample under study

Possibility of use of the ^{234m}Pa nuclide gamma radiation to determine ^{238}U with a scintillation detector in a medium containing natural radionuclides is investigated and demonstrated using the simplest examples. The proposed algorithm for determining of the ^{238}U content is based on the Monte Carlo simulation of the detector response to the radiation of the ^{234m}Pa radionuclide at its 1001 keV energy line and subsequent processing of the experimental spectrum, including the Wiener filtering of the signal. This method makes it possible to determine the content of ^{238}U in a continuous homogeneous medium while presence of natural radionuclides in it.

The algorithm for determining of ^{238}U content includes several main steps. Filtering based on the Wiener algorithm allows selecting a slowly changing part of the spectrum. Results of Monte Carlo simulations make it possible to determine the detection efficiency in a limited informative region of the spectrum, which includes, along with the 1001 keV peak from the ^{234m}Pa nuclide, which is a decay product of the radionuclide ^{234}Th, and the peak of an interfering radionuclide from the decay chain of ^{232}Th. This part of the spectrum does not contain any other lines of gamma radiation from natural radionuclides – decay products of both thorium and uranium chains. These two peaks in the spectral region under study can be separated from each other in a medium with a typical concentration of ^{234}Th.

Analysis of results of the activity of depleted uranium metal measuring in accordance with the proposed algorithm shows the possibility of determining of ^{238}U content with an uncertainty of 3–5 %.

Keywords: radionuclide ^{234m}Pa, Monte-Carlo simulation, experimental spectrum processing algorithm, depleted metallic uranium.

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Определение содержания ^{238}U по гамма-излучению ^{234m}Pa

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В атомной энергетике, в геологоразведке, при контроле радиоактивного загрязнения почв и сырья, используемого при строительстве, одним из важнейших радиоактивных элементов, который необходимо контролировать, является ^{238}U. Наиболее оптимально для контроля ^{238}U использовать радионуклид ^{234m}Pa, активность которого из-за малости времени его жизни (≈ 1,2 мин) однозначно связана с активностью ^{238}U даже при условии нарушения векового равновесия в исследуемом образце.

Исследована и продемонстрирована на простейших примерах возможность использования гамма-излучения нуклида ^{234m}Pa для определения ^{238}U с помощью сцинтилляционного детектора в среде, содержащей естественные радионуклиды. Предложенный алгоритм определения содержания ^{238}U основан на моделировании методом Монте-Карло отклика детектора на излучение радионуклида ^{234m}Pa на его монолинии 1001 кэВ и последующей обработке экспериментального спектра прибора, включающей винеровскую фильтрацию сигнала. Этот способ позволяет определить содержание ^{238}U в сплошной однородной среде при наличии в ней естественных радионуклидов.

Алгоритм определения содержания радионуклида включает в себя несколько основных этапов. Фильтрация на основе алгоритма Винера позволяет выделить медленно меняющуюся часть спектра. Результаты Монте-Карло моделирования дают возможность определить эффективность регистрации в ограниченном информативном участке спектра, включающем наряду с пиком 1001 кэВ от нуклида ^{234m}Pa, являющегося продуктом распада радионуклида ^{234}Th, и ближайший к нему пик мешающего радионуклида из цепочки распада ^{232}Th. Этот участок спектра по определению не содержит никаких других линии гамма-излучения от естественных радионуклидов – продуктов распада как ториевой, так и урановых цепочек. Указанные два пика на исследуемом участке спектра могут быть отделены друг от друга в среде с типичной концентрацией ^{234}Th.

Анализ результатов измерения активности обеднённого металлического урана в соответствии с предложенным алгоритмом показывает возможность определения содержания ^{238}U с погрешностью 3–5 %.

Ключевые слова: радионуклид ^{234m}Pa, моделирование методом Монте-Карло, алгоритм обработки экспериментального спектра, обеднённый металлический уран.

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Introduction

In nuclear power engineering, in geological exploration, in the control of radioactive contamination of soils, one of the most important radioactive elements, the concentration of which has to be determined, is the isotope ^{238}U. A common method for determining of the ^{238}U content in such media is the use of scintillation or germanium detectors which measure the gamma activity of one or several daughter nuclides in selected representative samples [1–3]. The media to be investigated contain radioactive nuclides of various origins. Soils contain not only nuclides that are the decay product of ^{238}U, but also nuclides that have got into this soil area from other areas due to various transfer processes. The fallout of radioactive dust and the subsequent migration of radionuclides will also disrupt any connection between these nuclides and ^{238}U in the soil, although they are decay products in its chain. The same inconsistency can occur in ores and various other materials. Only one nuclide ^{234m}Pa will be uniquely associated with ^{238}U contained in the mentioned media, since its lifetime is ≈ 1.2 min, so the transfer processes can be neglected [2].

The proposed technique can also be used for the case when for the identification of a nuclide and the determination of its content several gamma lines are used. In this case the procedure described below is applied independently for each such line.

Materials and methods

Relationship between ^{238}U *and* ^{234m}Pa *activities*

To determine of ^{238}U amount in a medium at ^{234m}Pa, it is first of all necessary to relate the amount of uranium to the amount of ^{234m}Pa. For this, it is necessary to use the uranium decay scheme (Figure 1) [4].

From the diagram a system of equations describing the time evolution of nuclides of interest in this problem can be used:

$$\frac{dN_u}{dt} = -\lambda_u N_u;$$

$$\frac{dN_T}{dt} = \lambda_u N_u - \lambda_T N_T;$$

$$\frac{dN_p}{dt} = \lambda_T N_T - \lambda_p N_p,$$

where N_U, N_T, N_P – the number of uranium, thorium and protactinium nuclei, respectively; λ_U, λ_T, λ_P are decay constants.

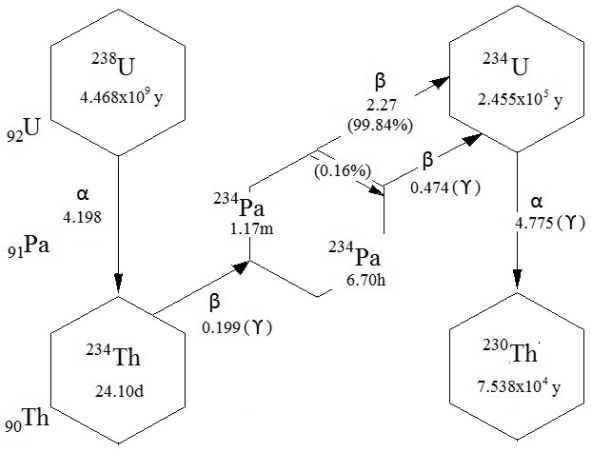

Figure 1 – Part of the ^{238}U decay scheme

These equations are easily integrated by the varying constants method and the solution for the relationship between the number of protactinium and uranium atoms is given by the equation:

$$N_p = \lambda_T \frac{\lambda_u N_0}{\lambda_T - \lambda_u}\left(\frac{e^{(-\lambda_u)t}}{(\lambda_p - \lambda_u)} - \frac{e^{(-\lambda_T)t}}{(\lambda_p - \lambda_T)}\right) =$$

$$= \lambda_T \frac{\lambda_u N_0}{\lambda_T - \lambda_u}\left(\frac{1}{(\lambda_p - \lambda_u)} - \frac{e^{-\lambda_T t}}{(\lambda_p - \lambda_T)}\right).$$

Taking into account that $\lambda_P >> \lambda_T >> \lambda_U$, and the lifetime of uranium is longer than the lifetime of thorium, we have:

$$N_p = \frac{\lambda_u N_0}{\lambda_p}; \quad N_u = \frac{T_u}{T_p} N_p.$$

It can be seen from the above relations that the relationship between the number of atoms of two nuclides does not depend on the time and concentration of other nuclides included in the ^{238}U decay chain, regardless of their origin in the medium.

Before processing the experimental results, it is necessary to simulate the measurement process and determine the main parameter of this method: $S_{0\gamma}$, it is the area under the Full Absorption Peak (FAP) of the gamma-radiation line, normalized to the one played gamma quantum. It should be noted that this parameter does not relate to a specific activity; it characterizes the average response to one played photon. Calculating the same value for the experiment, reduced to a unit of volume (mass) and time, we obtain from their ratio the number of protactinium atoms that underwent decay through a given channel

per unit of time and per unit of volume (n). Taking into account other decays, the total number of decayed protactinium atoms per unit volume and per unit time is $N_d = n/\eta$, η is the fraction of gamma decays of protactinium with $E_\gamma = 1.001$ MeV. Further, the reduction of the number of ^{238}U atoms is carried out according to the above formulas of the described decay kinetics.

For the algorithm to work correctly, it is necessary that the same hardware functions that characterize the Analog-to-Digital Converter of the equipment are used in both the simulation and the processing of the experiment. In our case, these are: the channel – energy dependence and the dependence of the line width on the energy of the registered gamma radiation.

Monte Carlo simulation

The purpose of the first stage of the proposed algorithm is to simulate the process of measuring and calculating the amplitude distribution of pulses of each radionuclide using the Monte Carlo method and determining $S_{0\gamma}$ [5].

The developed model of the detecting device (DD) used in the experiment in the measurement geometry of metallic uranium at a distance of 10 cm from the DD surface opposite the geometric center of the scintillation crystal is shown in Figure 2.

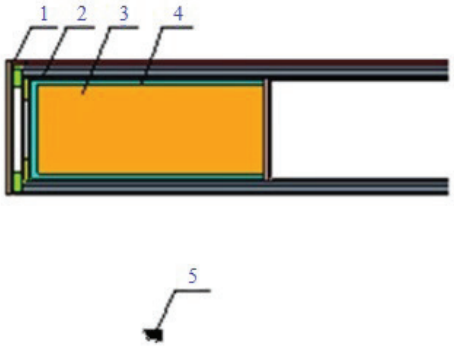

Figure 2 – The detecting device and source (metallic ^{238}U) used in Monte Carlo simulations: 1 – case of the detection device; 2 – probe; 3 – NaI(Tl) scintillation crystal; 4 – MgO reflector; 5 – source – metal ^{238}U (1/4 cylindrical column)

In Monte Carlo simulations, it is possible to take into account the energy dependence of the line width on the energy of the recorded radiation of a real spectrometer. To account for this dependence on energy E, the MCNP program uses a special formula for the energy dependence of the full line width at half maximum (*fwhm*):

$$fwhm_{mcnp}(E) = a + b\sqrt{E + c * E \wedge 2}, \quad (1)$$

where the coefficients a, b, c should be determined from the experimental data for a given detector by approximating them using formula (1). The coefficients $\{a, b, c\} = \{-0.00819936,\ 0.0704576,\ -0.0154056\}$ obtained as a result of approximating the experimental data for the NaI(Tl) detector with dimensions Ø63 × 160 mm. This *fwhm* were used to calculate the pulses height spectrum in the MCNP program.

The DD model was verified by comparing the theoretical and experimental spectra from a coin source with ^{137}Cs with known activity, placed at a distance of 10 cm from the lateral surface. The difference in the heights of the FAP line of gamma radiation with an energy of 0.662 MeV did not exceed 3 %.

As indicated earlier, the most representative ^{238}U radionuclide is ^{234m}Pa, and its preferred gamma-emitting line in terms of quantum efficiency is 1.001 MeV. Figure 3 shows the simulated pulses height spectrum of metallic ^{238}U (main picture). The result of Monte Carlo simulation of only ^{234m}Pa gamma radiation with an energy of 1.001 MeV and its *Math-background* (envelope of the slowly changing part of FAPs calculated by second-order polynomials [4]) are shown in small picture.

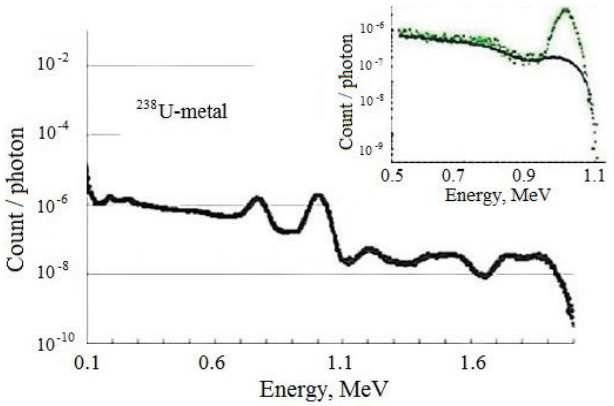

Figure 3 – The simulated pulses height spectra of metallic ^{238}U (main picture) and the result of Monte Carlo simulation of gamma radiation with energy of 1.001 MeV with its *Math-background* (insert)

It is find the difference between the model spectrum smoothed by the Wiener filter [5] and its *Math-background* in the energy region from 0.85 to 1.05 MeV. The resulting value is approximated by a Gaussian distribution and the area under this curve ($S_{0\gamma}$) is found, which in MCNP is automatically normalized to the total number of played stories. For the model of this experiment, $S_{0\gamma i} = 0.00001773$.

The Wiener filter is the optimal filter for the formation of the output signal $z(t)$ from the input signal $x(t)$ with the known form of the useful signal $s(t)$, which is contained in the input signal in the amount of noise. As a criterion for its optimization, the standard deviation of the signal $y(t)$ at the filter output from the given waveform $z(t)$ is used. In this case, such a signal was the sum of a slowly changing background and Gaussian peaks [6, 7].

Algorithm for determining the content of radionuclide in the object of measurement

Experimental spectrum processing primarily includes background spectrum subtraction, Wiener filtering, *Math-background* determination and subtraction. The processing block diagram is shown in Figure 4.

To understand the operation of the algorithm, an experiment was carried out in which a portable scintillation spectrometer was used, the Monte Carlo model of which is presented above.

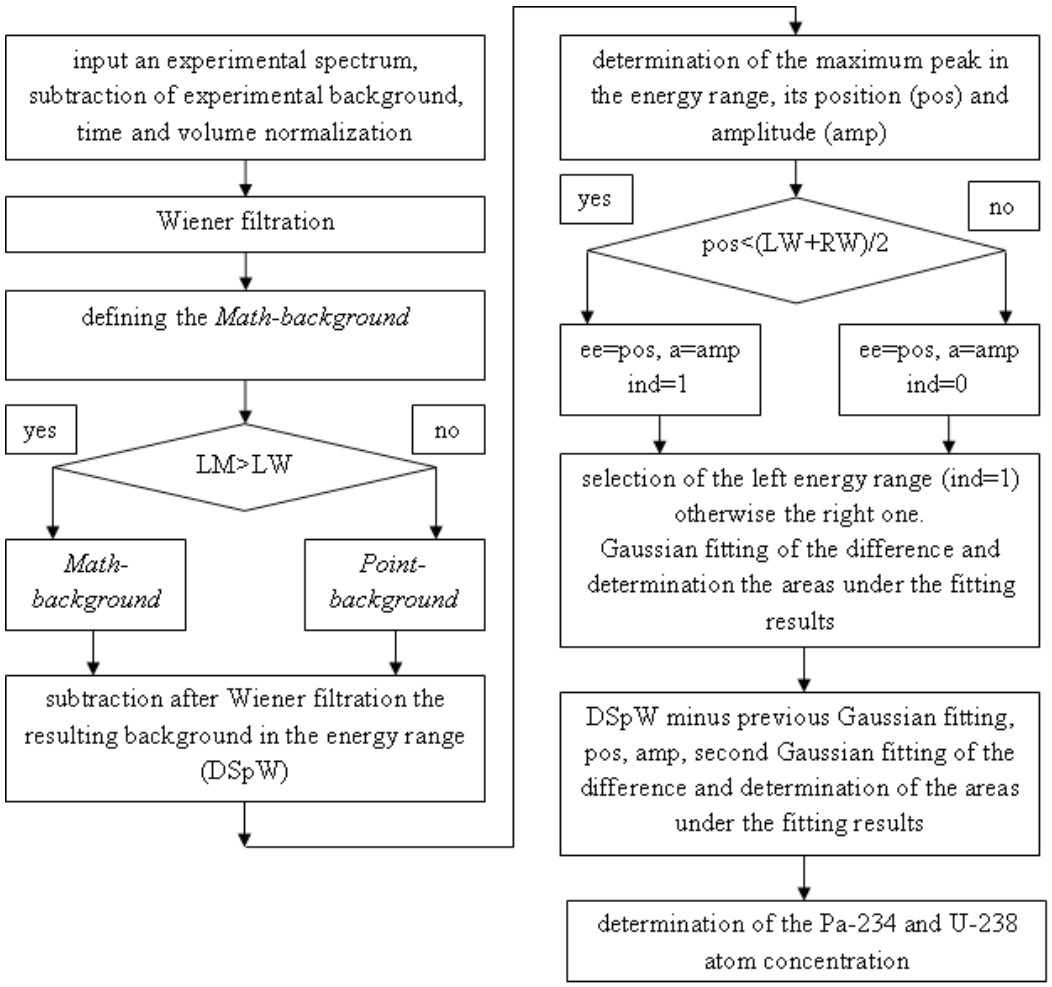

Figure 4 – Block diagram of the algorithm: LM – position of the left minimum of Math-background; LW – left border of the window; RW – right. DSpW is normalized spectrum minus Math-background in the energy range. Point-background is a smooth curve passing through the minima of the spectrum transformed by Wiener filtering

Experimental research

To test the proposed algorithm, an experiment was carried out using a DD based on a NaI(Tl) scintillation crystal with dimensions of Ø63×160 mm and a small volumetric cylindrical source of metallic ^{238}U (depleted metallic uranium) located at a distance of 10 cm (Figure 5).

The source with a density of 19.8 g/cm^3 consists of: 0.0023 % of ^{234}U with an activity of 10894 Bq/sample; 0.4054 % of ^{235}U with 667 Bq/sample and 99.5923 % of ^{238}U with 25485 Bq/sample. Figure 6 shows the experimental Gross spectrum measured over a time of 7200 s in the measurement geometry according to Figure 5, the background spectrum BKG and

the difference between the experimental and background spectrum – the experimental pulses height spectrum Net.

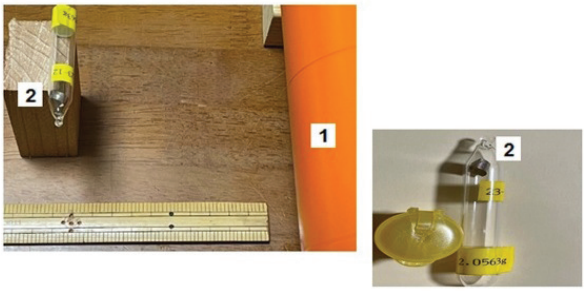

Figure 5 – Geometry of experiment with depleted metallic uranium source: 1 – the detection device; 2 – depleted metallic uranium source

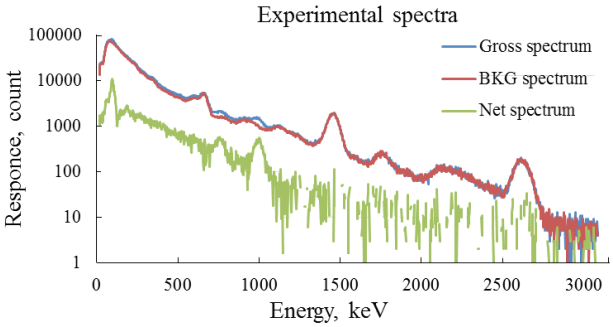

Figure 6 – The experimental spectra

The experimental pulses height spectrum Net was obtained by channel-by-channel subtraction of the background spectrum from the experimental one, measured when the source was located opposite to the annular mark applied to the lateral surface of the DD, which corresponds to the geometric center of the NaI(Tl) crystal.

Approbation of the algorithm on the example of the experimental spectrum of metallic ^{238}U

Approbation of the algorithm is presented using the example of processing the experimental Gross spectrum. The step of subtracting the background spectrum is excluded from the algorithm due to its insignificant contribution to the result of calculating the activity in this case. According to the algorithm, the experimental Gross spectrum was transformed into the pulses height spectrum using filtering according to the Wiener algorithm.

The most important step in this algorithm is the definition and subtraction of *Math-background*. The algorithm provides two variants:

1. Using an algorithm from the Wolfram Mathematica [8]. In the work we use the "EstimatedBackground" function without a parameter from "Mathematics", which allows to build a smoothly varying function passing through the minimums of the spectrum (see Figure 7).

2. Draw a curve passing through the background minima, determined by the algorithm from the Wolfram Mathematica. Often the minima of this background are located farther in energy than the gamma-radiation line of interest, and then the first option is used.

It is shown in Figure 7 the pulses height spectrum after Wiener filter and *Math-background* according to the algorithm from the Wolfram Mathematica. Since the left minimum of this background lies to the right of the line (1.001 MeV), the first variant is chosen.

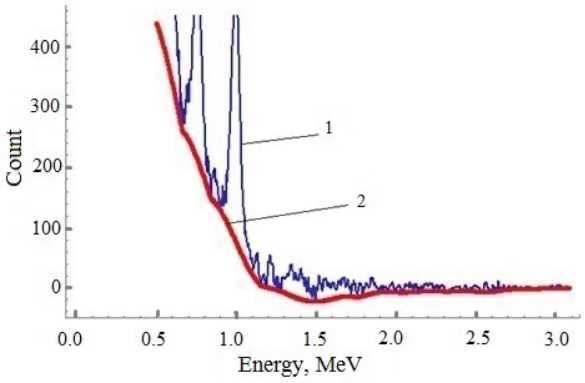

Figure 7 – Transformed pulses height spectrum according to the Wiener algorithm (1) and Math-background (2) (the first variant of the estimation)

Further consideration will be carried out only in the energy region from 0.85 to 1.05 MeV where is situated the protactinium's FAP with energy E = 1.001 MeV. The region is not quite symmetrical about this line (1.001 MeV), the center of the window is shifted to the left. The ^{234m}Pa peak on a spectrum is shifted to the left due to the presence of Compton scattering of gamma quantum from high-energy photons. In the analysis, the initial region is split by the midpoint into two windows, left and right one. If the main maximum of the region is in the left window, it means that the ^{232}Th peak is prevails. The Gaussian approximation of this peak is carried out, the difference between the experimental spectrum and

the obtained approximation in the full region is determined. Then the contribution of Pa and its peak and the Gaussian approximation of the latter are found by the points of this difference. This approach allows you to separate these peaks, even if they overlap significantly. If the main maximum in the full window is in the left part of the window, processing is starting from the right part, and then the mentioned nuclides are swapped. Now Gaussian fitting of the protactinium peak is performed, subtraction of the fitting result from the experimental spectrum and approximation of the Gaussian difference for the ^{232}Th peak.

The result of this processing is illustrated in Figure 8. In this case, the main maximum is in the right half of the energy window and corresponds to the contribution of ^{234m}Pa (the Gaussian approximation of this peak is curve 1), in the left half of the window is the thorium peak, curve 2 obtained after approximating the difference between the experimental curve and the curves 1.

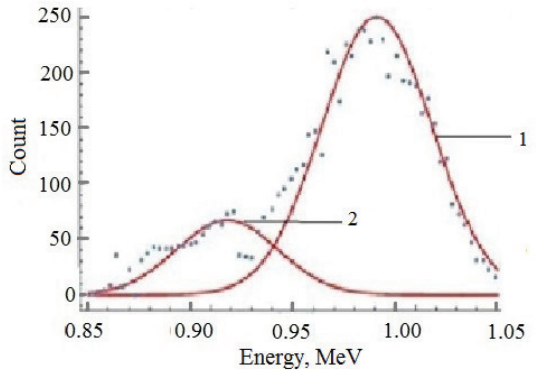

Figure 8 – Counting in the energy window of the useful signal minus background (points), curve 1 is Gaussian approximation of ^{234m}Pa peak, curve 2 is Gaussian approximation of ^{232}Th peak

Next, the area under the Gaussian approximation of ^{234m}Pa with an energy of 1.001 MeV ($S_{1\gamma}$) is determined. Then the ratio $S_{1\gamma}/S_{0\gamma i}$ determines the number of ^{234m}Pa nuclei decayed in a source of volume V during the experiment time t.

Reduced to a unit of time and volume, the number of decayed ^{234m}Pa atoms is given by the equation:

$$n = \frac{S_{1\gamma}}{S_{0\gamma}} \frac{1}{Vt},$$

where V is the volume of the source, cm^3; t is the experiment time, s; n is the number of ^{234m}Pa atoms decaying in one second in one cubic centimeter and emitting a gamma quantum with an energy of 1.001 MeV.

Then we have the following equations for the total decay rate of ^{234m}Pa and the number of protactinium atoms:

$$N_p = n/(\eta_\gamma \lambda_p) = n\, T^{Pa}_{1/2}/(\ln 2 * \eta_\gamma),$$

where η_γ = 0.0084 is the probability of the branch of the gamma decay of ^{234m}Pa in the decay of ^{234}Pa; N_P is the number of ^{234}Pa atoms in 1 cm^3.

Knowing this number makes it possible to determine the number of ^{238}U atoms by the formula following from consideration of the kinetics of the decay chain of this nucleus, up to and including the decay of the ^{234}Pa nucleus. With sufficient precision:

$$N_u = \frac{T^{\mathrm{U}}_{1/2}}{T^{\mathrm{Pa}}_{1/2}} N_p,$$

where N_U is the number of U atoms in 1 cm^3; $T^{\mathrm{U}}_{1/2}$, $T^{\mathrm{Pa}}_{1/2}$ are the half-lives of U and Pa, respectively.

Applied to the above experiment, the final processing is as follows.

The number of protactinium gamma quanta emitted from 1 g of uranium per 1 s was determined (t_{ex} = 7200 s, m_U = 2.0563 g).

$$n_\gamma^{m\mathrm{Pa}} = \frac{S_{1\gamma}}{S_{0\gamma}} \frac{1}{t_{ex} m_\mathrm{U}},$$

where $T^{\mathrm{Pa}}_{1/2}$ = 1.2×60 s (half-life of ^{234m}Pa); $n_\gamma^{m\mathrm{Pa}}$ = 0.84 % is the fraction of ^{234m}Pa decays along the gamma channel.

Then, as follows from the solution of the equations of kinetics of ^{238}U decay, the number of U atoms in 1 g of metal is equal to:

$$n\mathrm{U}^{238} = n_a{}^{\mathrm{Pa}}/(T_{1/2}{}^{\mathrm{Pa}})\, T_{1/2}{}^{\mathrm{U}}.$$

The metal is practically pure ^{238}U, therefore, to estimate the accuracy, we calculate the number of uranium atoms in 1 g-atom of the metal, for this the value obtained above must be multiplied by 238 (as result we have 5.73×10^{23}) and compared with the Avogadro number (6.022×10^{23}). The accuracy of the match is 4.8 %.

The activity of this uranium sample, reduced to 1 g, is easily estimated by the formula:

$$I = \frac{\mathrm{Log}2}{T^{\mathrm{U}}_{1/2}} n\mathrm{U}^{238}.$$

And in this case it was 11924 Bq/g, the deviation of the method in relation to the measured value

(25500 Bq per sample weighing 2.0563 g) was 3.84 %.

Uncertainty of describing method

The accuracy of the method is determined by the error in calculating the ratio $S_{1\gamma}/S_{0\gamma}$. It is clear that the error $S_{0\gamma}$ is determined by the specified statistical error in the Monte Carlo simulation and, accordingly, the relative error in this case is expressed as $1/N_p^{1/2}$, where N_p is the number of events recorded in the FAP region $E = 1.001$ MeV.

The relative error of $S_{1\gamma}$ can be represented as follows $(N^{1/2}_{ex+ph} + N^{1/2}_{ph})/(N_{ex+ph} - N_{ph})$, where N_{ex+ph} is the number of pulses in the FAP region $E = 1.001$ MeV of the experimental spectrum without subtraction of background pulses; N_{ph} is the number of pulses in the same energy range of the background spectrum.

The relative error of the ratio $S_{1\gamma}/S_{0\gamma}$ is then given by the sum of the above errors and is determined by the expression:

$$\frac{\sqrt{N_{ex+ph}} + \sqrt{N_{ph}}}{N_{ex+ph} - N_{ph}} + \frac{\sqrt{N_p}}{N_p}.$$

Further actions with the value $S_{1\gamma}/S_{0\gamma}$ for determining the amount of ^{238}U are associated only with the use of constants, which are known values and, therefore, do not affect the accuracy.

Conclusion

The paper considers a simple and reliable method for determination of ^{238}U which can be applied for the case when uranium is in various media. The method is based on measuring of a scintillation detector response to the ^{234m}Pa (1.001 MeV) gamma line.

An algorithm for processing and the necessary Monte Carlo simulation are proposed which makes it possible to determine the ^{238}U content in a medium or sample from experimental data. The operation of the algorithm is illustrated by the experiment example with a piece of metallic uranium. It is shown that the determination accuracy of the ^{238}U content in this case is 3–5 %.

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