NUCET NUCLEAR ENERGY AND TECHNOLOGY

Nuclear Energy and Technology 5(2): 163–169
DOI 10.3897/nucet.5.36478

Research Article

Use of mathematical modeling to extend the scope of application for the procedure of measuring the mass of ^{235}U in solid radioactive waste*

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Academic editor: *Georgy Tikhomirov* ♦ **Received** 19 March 2019 ♦ **Accepted** 15 May 2019 ♦ **Published** 21 June 2019

Citation: Rykov NS, Bezhunov GM, Gorbachev VM (2019) Use of mathematical modeling to extend the scope of application for the procedure of measuring the mass of ^{235}U in solid radioactive waste 5(2): 163–169. https://doi.org/10.3897/nucet.5.36478

Abstract

The known dependence of absolute efficiency on energy and space for particular measurement conditions is used to determine the mass (activity) of ^{235}U in solid radioactive waste by gamma-spectrometric method. The ISOCS system makes it possible to avoid laborious and time-consuming calibration measurements using standard samples to obtain the absolute efficiency curve due to using the so-called characterized detector having a file with a set of efficiencies for various measurement geometries.

In many cases, the establishment of standard samples with parameters covering the ^{235}U mass measurement range in the variation intervals of influencing factors, including density, non-uniformity, isotopic composition, geometry, etc., is very expensive and, most often, not feasible. With regard for this, a computational and experimental approach is used based on results obtained by Monte Carlo method using the MCNP code with variation of the key influencing parameters in a broad range.

Calculations were performed for detector-recorded spectra of gamma quanta from casks containing waste differing in the density of the cask content (the density was calculated with regard for the uranium contained in waste) – from 0.016 to 1 g/cm^3, in the mass of uranium in waste – from 0.64 g to 2 kg, and in the matrix material – graphite, cellulose, quartz, cellulose with 20 % of iron dust.

Applicability boundaries have been defined for the developed procedure to measure uranium-containing waste in terms of the material matrix (~ 2.2 %) and its density (~ 10 %) and the contribution of the uranium mass uncertainty in the cask (5 % for nonporous matrices, 10 % for porous matrices) to the obtained result has been estimated.

Keywords

Nondestructive analysis of nuclear materials, solid radioactive waste, uranium mass, gamma-spectrometry, ISOCS system, absolute efficiency curve, Monte Carlo method, MCNP code, measurement procedure, measurement procedure range

* Russian text published: Izvestiya vuzov. Yadernaya Energetika (ISSN 0204-3327), 2018, n. 4, pp. 102–114.

Introduction

Inspection of casks with uranium-containing solid radioactive waste (SRW) has the purpose to determine the mass (activity) of ^{235}U in the casks. A high-resolution detection unit and the In Situ Object Counting Systems (ISOCS 2018) software (In Situ Object Counting System, In Situ Object Counting System (ISOCS)) from Canberra Industries, Inc. form a system which makes it possible to solve the tasks in hand for a reasonable time. The system efficiency calibration is based on the use of the mathematical tools implemented in this software. In accordance with the Measurement Traceability Law (Federal Law No. 102-FZ 2008), the respective measurement procedure (MP) requires to be developed and appropriately certified. For the investigations, samples of uranium with well-characterized isotopic composition and mass available at IPPE (the certificate data on the mass and isotopic composition of uranium in the samples were obtained by destructive methods) in the form of a uranyl nitrate solution were used to develop the MP for the ^{235}U mass (activity) in SRW with matrices of light-weight materials (cotton, rubber, debris, etc.). Plastic cells with a diameter of 20 mm and a wall thickness of 3 mm contained 8 ml of the uranyl nitrate solution with a uranium mass of 0.4 to 2 g. In total, 10 cells were used. The use of small plastic cells with the uranyl nitrate solution made it possible to minimize the gamma radiation self-shielding effect in the sample. In the process of real measurements, the spatial distribution of the samples used throughout the measured matrix and the measured object (SRW cask) rotation led to a high level of the nuclear material (NM) homogeneity achieved in the samples, as regards the recording of gamma radiation of the energy 185.7 keV.

The procedure used to develop the MP (OST 95 10353-2007, OST 95 10289-2005, OST 95 10571-2002) suggests that there is no notable quantity of sizeable solid inclusions (of several mm and more) from a material with a high Z in the SRW matrix and that the requirement of the distribution uniformity of both the SRW density and the ^{235}U mass (activity) in the volume is fulfilled. In reality, inclusions are however present and their impact needs to be assessed. The software part of the ISOCS system includes the Ratio Tester, a subprogram for indirect assessment of this factor. To reduce the effects of the ^{235}U mass (activity) irregular distribution in the SRW matrix on the measurement results, the cask (a 120 liter drum for the purpose hereof) is rotated in the process of measurement.

The known dependence of absolute efficiency on energy and space for particular measurement conditions needs to be used to determine the ^{235}U mass (activity) in SRW by gamma-spectrometric method. The use of the ISOCS makes it possible to avoid laborious and time-consuming calibration measurements based on standard samples to obtain the absolute efficiency curve due to using the so-called characterized detector including a file with a set of efficiencies for various measurement geometries.

In many cases, the establishment of a set of certified objects (CO) with parameters covering the ^{235}U mass measurement range in the variation intervals of influencing factors, including density, non-uniformity, isotopic composition, geometry, etc., is very expensive and, most often, not feasible. For this reason, use of computational methods is evolving worldwide for the experiment modeling (Akhnazarova and Kafarov 1985, Laborie et al. 2002, Sima 1996). The application of these has been justified by numerous studies. A computational and experimental approach was adopted therefore for the MP development. In addition to the measurement results, calculated data obtained by Monte Carlo method using the MCNP code (MCNP 2017, 2018a, 2018b, Kolesov 2008, Korobeynikov 1994) were used with variation of the key influencing parameters in broad limits.

Single-type casks with model matrices of various densities (from 0.016 g/cm^3 to 0.708 g/cm^3) were used for measurements. For the ^{235}U mass range of 0.25 through 1.8 g (20 through 140 kBq), the measurements were based directly on models with COs. In the SRW ^{235}U mass range of 1.8 to 100 g (140 ÷ 8000 kBq), studies were performed computationally due to lack of representative standard samples.

Equipment used for measurements

The ISOCS system used for the studies had the following components:

- a mobile trolley with a detection unit positioning system;
- a Canberra coaxial-type high-resolution detection unit (pre-characterized by the manufacturer);
- a side shield and a set of collimators;
- an InSpector-2000 portable spectrometric station;
- a personal computer with the preinstalled Genie-2000 spectra acquisition and analysis software and the ISOCS Geometry Master program.

The measurement geometry implemented by the ISOCS system's software tools is shown in Fig. 1. The detection unit crystal has its end directed at the geometrical center of the drum's side surface. The distance from the drum surface to the detection unit cut is 33 cm as the detector sees the whole of the drum volume through the employed collimator. The drum with the SWR matrices (models) is a cylinder with a 1 mm thick steel wall, an inner diameter of 463 mm, and a height of 720 mm.

Gamma peaks in a range of 140 to 220 keV, which include the base ^{235}U gamma peaks, are used for the analysis using the Geometry Master program. An intensive and readily detectable gamma peak of 185.7 keV is used for the quantitative analysis of ^{235}U.

Sources from an SSGS set and/or uranium samples are used for the energy calibration of the gamma-spectrometer.

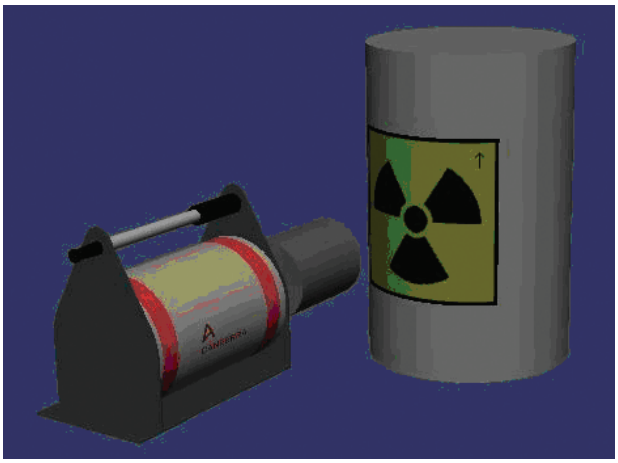

Figure 1. ISOCS measurement geometry

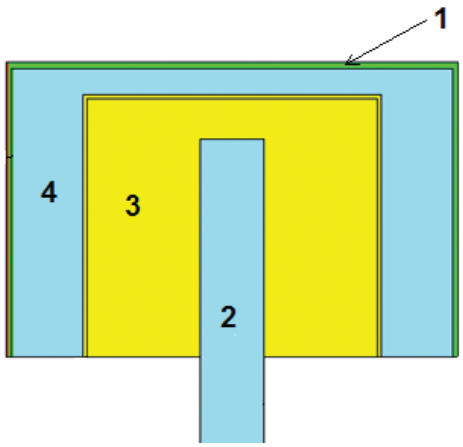

Figure 2. Cross-section of the detector's detection unit model: 1 – aluminum lid with a carbon window; 2 – wire leading channel to detector crystal; 3 – detector crystal; 4 – vacuum gap

Following an analysis of the uranium gamma spectrum, the Ratio Tester is used to determine, by analyzing the ^{235}U peaks of 143.8, 163.3, and 185.7 keV, the degree of the ^{235}U distribution homogeneity in the measured SRW cask.

For measurements, the SRW cask is placed on a platform equipped with a rotation device for the cask uniform rotation. The SRW cask weight was measured using a CAS floor scale of the DBII-150 type with a weighing range of 10 to 150 kg. The matrix density was determined from the weight measurements with the drum filled up.

Mathematical model of the detector

A mathematical model of the cask detector system was built for the MCNP code. It was assumed for easier calculations that there were no external radiation diffusers (walls). And the model included both the cask as such, which contained various materials simulating the potential waste matrix, and the detector's lead collimator similar to that used in measurements.

A cross-section of the selected HPGe detector model is presented in Fig. 2.

The design of the detector in the selected model reflects its characteristics known from the Canberra descriptions. Spectra of the ^{152}Eu source from a set of standard spectrometric gamma sources (SSGS) were measured to determine more accurately the geometrical parameters of the crystal and to confirm the validity of the detector model built. Two series of measurements were conducted:

- the sample was opposite the detector and on its axis (at a distance of 33 cm from the end);
- the sample had a shift of 20 cm relative to the axis perpendicular to the detector axis.

The experiment conditions were modeled in the MCNP code based on the built detector model. In the event of the ^{152}Eu measurement, the area of the 121.8, 344.3 and 778.9 keV peaks was measured as the ratio of the recorded peak area per one emitted quantum multiplied by the total number of gamma quanta emitted by the source for the measurement time. These peaks were selected due to having a high intensity (over 10 %). The lines of 121.8 and 344.2 keV were chosen since this energy range includes the base gamma lines representative of ^{235}U. The same lines were used to determine the crystal area. The 778.8 keV line was used for the model verification in a more high-energy interval and to determine the crystal thickness.

The calculated and measured spectra from an uranium CO with a mass of 134.6 g and a ^{235}U enrichment of 89.2 % (at a distance of 33 cm from the detector to the sample) for the three base gamma peaks of the ^{235}U isotope with the energies 143.8, 163.3, and 185.7 keV were compared to confirm the accuracy of the model built.

To select the germanium crystal diameter and height, their values were varied in a range of 60 to 65 mm (diameter) and 30 to 35 mm (thickness). Table 1 presents the compared results of the calculations and experiments with ^{152}Eu from the SSGS set and the uranium CO.

The value δS is taken from the results of processing a spectrum (actually acquired or modeled) by the Gamma Spectra Acquisition and Analysis program of the Genie-2000 package (Genie 2018). It can be seen from the table that the modeling approach used provides for data that agree with the experiment in terms of the recorded peak areas in a broad range of energies for the selected dimensions of the detector with a diameter of Ø61 mm and a thickness of 31.9 mm.

Mathematical model of the measurement system

A variant was used to build the calculation model for spectra from waste-containing casks with variation of influencing factors which makes it possible to optmize the calculation as to the rate of acquiring the required statistics for the recorded pulses in the spectra obtained.

Table 1. Peak areas obtained as the result of the calculations and the experiment

Energy, keV	S_m	δS_m, %	S_c	δS_c, %	$(1 - S_c / S_m) \cdot 100$
			^{152}Eu, center		
121.8	16155	0.83	16335	0.85	–1.11
344.2	6458	1.29	6524	1.28	–1.02
778.8	1422	2.83	1384	2.82	2.67
			^{152}Eum, shift		
121.8	11635	0.985	10789	0.79	7.27
344.2	4481	1.55	4555	1.23	–1.65
778.8	1000	3.44	1018	2.82	–1.80
			Standard uranium sample		
143.8	29945	0.63	29850	0.62	0.32
163.3	16188	0.88	16092	0.88	0.59
185.7	195800	0.23	197341	0.23	–0.79

Note. S_m – S measured; S_c – S calculated

To expand the calculation statistics and to simulate the cask rotation effect, the detector was assumed to be a ring around the cask (Fig. 3) at a distance of 33 cm from the cask's outer surface in the axially central region. The ring had the germanium height of 6.1 cm and the thickness of 3.19 cm in an aluminum shell in accordance with the dimensions of the real detector as determined by measurements using the SSGS gamma sources. With such geometry, all detector parameters were modeled in accordance with the experiment and the efficiency (effective area of the detector) turned out to be 96 times as high as in the experiment. Further on, this efficiency factor was taken into account in comparing the absolute values of the model calculation and experiment results. The ring's upper and lower layers in the model were of lead and simulated the collimator.

The cask was simulated as a cylinder with a 1 mm thick steel wall in which different waste types with a content of different NM quantities were modeled. Both radially and axially, the sample had the form of the real cask used in the experiment.

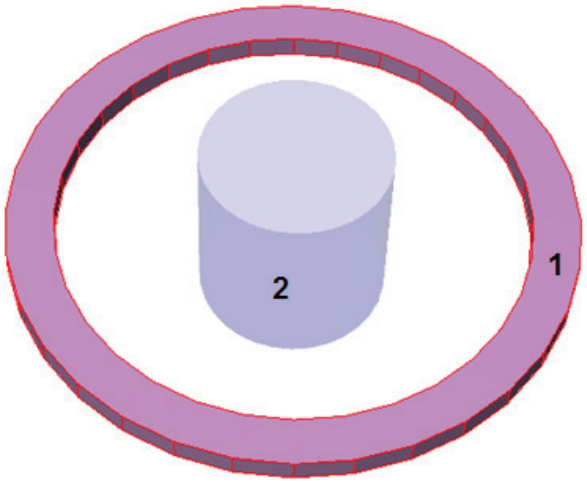

Figure 3. Computational geometry in modeling of spectra from waste-containing casks: 1 – ring-shaped detector with a lead collimator; 2 – cask with waste

The number of the selected gamma quanta events was $2 \cdot 10^7$ to $4 \cdot 10^7$, and the number of recorded pulses in the total absorption peak with the energy 185.7 keV was ~ 10^5. The number of the events selected varied depending on the density of the matrix of interest and the uranium mass in the cask. The yields of gamma quanta for ^{235}U were taken from (Table of Isotopes 2018, Doug Reilly et al. 1996, XCOM 2018, Nuclide Navigator Version 2018).

Fig. 4 presents a comparison of the spectra obtained experimentally and computationally for a cask with a cellulose matrix of the density 0.708 g/cm^3 containing 0.611 g of ^{235}U. In the calculation, the uranium was assumed to be distributed uniformly in the matrix with a ^{235}U enrichment of 6.51 wt. %, while, for the experiment, uranium with an enrichment of 6.51 wt. % was assumed to be distributed quasi-uniformly within the matrix volume and had the form of bubbles with the uranyl nitrate solution. The number of energy channels in the experiment and in the calculation was 8192, and the detector energy resolution in the calculation was the same as the resolution of the spectrometer used in measurements.

To make the analysis more convenient, data are presented in absolute units (the number of pulses in the energy channel per 1 g of ^{235}U per second). It can be seen from the available data that the base gamma lines in the energy range of interest coincide in terms of energy and area. Slightly excessive calculated gamma line intensities, as compared with the measured ones, are explained by the radiation self-shielding in uranyl nitrate solutions. After this effect was taken into account with an allowance introduced for the self-shielding in solutions in the measured spectra, the divergence of the 185.7 keV peak recording intensity in the calculation and in the experiment did not exceed 10 % for all considered densities of the cask-contained waste.

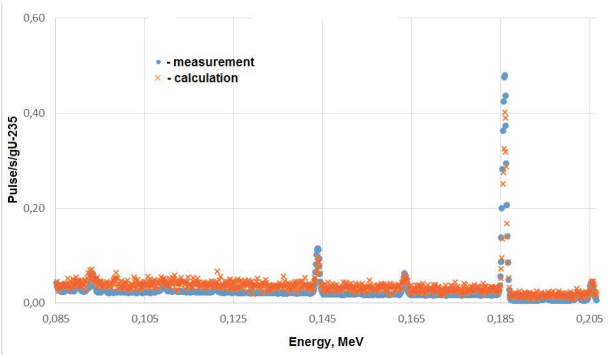

Figure 4. Comparison of the measured and calculated gamma quanta spectra from cellulose-containing cask. The waste material density is 0.708 g/cm^3

Therefore, the program used and the selected model make it possible to model correctly the gamma radiation spectra measurement, using a high-resolution detector, on real complex waste-simulating objects for a reasonable computational time.

Computational study into the influence of various factors on measurement results

Detector-recorded gamma quanta spectra were calculated from a cask containing waste differing in

- the density of the cask content (the density was calculated with regard for the uranium contained in waste) – from 0.016 to 1 g/cm^3;
- the uranium mass in waste – from 0.64 g to 2 kg;
- the matrix material – graphite, cellulose, quartz, cellulose with 20 % of iron dust.

The influence the variation of the factors above have on the determined intensity of the peak with the energy 185.7 keV and the measurement procedure application area were determined. The results are presented in the diagrams and tables below. The ^{235}U peak intensity (pulse/s/g) was calculated by summing up the pulses in the peak region with the background component deduced. Since the code yields the relative value of pulse/event, a coefficient was introduced that was equal to the yield of quanta of the considered line per 1 g of ^{235}U per second divided by the share of the gamma quanta with the energy 185.7 keV in the initially selected spectrum (depends on the isotopic composition of uranium and tabulated values of the yields of gamma quanta lines for the isotopes taken into account) and by the coefficient that allows for the ratio of the ring detector efficiency in the calculation to the efficiency of the real detector in the experiment. This makes it possible to simplify the analysis of the influence of factors and to compare the calculation with the experiment.

Assessment of the influence of the waste cask content density

Fig. 5 and Table 2 present the calculation and measurement results for the intensities of counts in the 185.7 keV peak with the background deduction for waste in the form of cellulose containing 0.313 g of ^{235}U (experiment, 4.81 g of uranium with an enrichment of 6.51 wt. %) and 1 g of ^{235}U (calculation, 20 g of uranium with an enrichment of 5 wt. %). The waste material density varied in a range of 0.016 to 1 g/cm^3. The measurement results were adjusted for the radiation self-absorption effect in samples in the form of a uranyl nitrate solution. The values were reduced to the absolute quantity of pulse/s/g of ^{235}U.

It can be seen from Fig. 4 and Table 2 that the calculation and measurement results coincide for the entire range of the material densities in the limits of 10 %. It is evident that the absorption of radiation with the energy 185.7 keV in the uranium as such may be neglected with such small quantities of uranium in a 120 liter cask if the uranium is in a nonlocalized form. It can be also seen that the recording intensity of gamma quanta with the energy 185.7 keV decrease significantly as the matrix density is increased.

The residual bias (RB) of the calculation and experiment agreement, which is equal to ~ 10 %, needs to be accepted for all density ranges within the calculation range during the MP development.

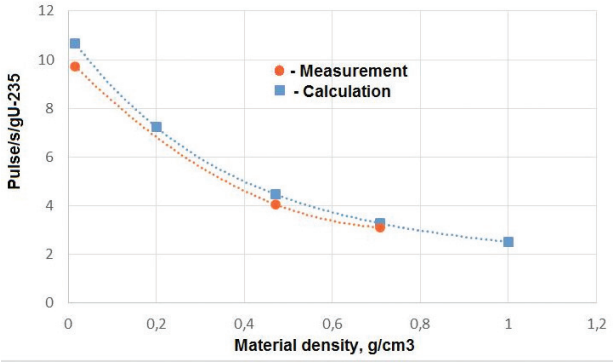

Figure 5. Dependence of the count intensity in the 185.7 keV peak on the density of the waste material in the form of cellulose with a small content of uranium (~1 g of ^{235}U)

Table 2. Count intensity in the 185.7 keV peak with the background deduction for different cellulose densities in the cask (experiment: 0.313 g of ^{235}U, 4.81 g of uranium; calculation: 1 g of ^{235}U, 20 g of uranium)

Waste material density, g/cm^3	Count intensity in 185.7 keV peak		
	Calculation	Experiment	Difference (%)
0.016	10.67	9.72	0.95 (9.77)
0.2	7.219	–	–
0.471	4.46	4.056	0.404 (9.96)
0.708	3.279	3.107	0.172 (5.54)
1	2.511	–	–

Assessment of the matrix type influence

The matrix of waste may have different elemental compositions containing, among other things, segregated fractions like rags, gloves, concrete, debris, etc. The presence of metal chips, grit or moisture is not excluded as well. In most cases, the elemental composition of the matrix depends on elements with the atomic number up to that of calcium but, occasionally, there are admixtures of elements in a range from *Z* to iron.

Calculations were performed for waste in the form of cellulose ($C_6H_{10}O_5$), quartz (SiO_2), graphite, and a mixture of 80 % of cellulose and 20 % of iron to assess the influence of the matrix type (elemental composition). The density for all waste types was 0.708 g/cm^3 with which the effect from the difference in the elemental composition has the greatest possible range presented in measurements. The results are given in Table 3.

It can be seen from the table that the chemical composition of the matrix in the range of the atomic numbers of elements in a range from carbon to iron influences slightly the intensity of the count in the 185.7 keV peak (~ 2 % for the tested matrixes as compared with the cellulose matrix). This is explained by the fact that the mass attenuation factor for the 185.7 keV line depends weakly on the atomic number of the element in the considered range of matrices and the attenuation of gamma radiation will be defined practically by the density of the material contained in the cask. Based on the data obtained, the value of the RB from the matrix type was assumed to be equal to ~ 2.2 %.

At the same time, an increase in the density of the matrix with uranium homogeneously distributed throughout the cask volume will lead to a further attenuation of the line due to the high atomic number of uranium and, accordingly, to a comparatively greater mass attenuation factor. It can be seen from Table 3 that the recording intensity for the 1185.7 keV line decreases by about 4.6 % as the uranium mass in the cask is increased to 500 g. The effect is practically the same for the cellulose matrix and the matrix with 20 % of iron. Since initially, prior to the measurements, the uranium mass in the cask was not known, such effect will produce a systematic drift (downward bias) if the quantity of uranium is large.

Table 3. Count intensities in the 185.7 keV peak with the background deduction for different waste matrices and different uranium masses in the cask (the waste material density is 0.708 g/cm^3)

Uranium mass, g	Matrix composition			
	Cellulose $C_6H_{10}O_5$	Graphite C	Quartz SiO_2	80% of cellulose + 20% of iron
	Count intensity in the 185.7 keV peak			
1	3.367	–	–	3.301
500	3.22	3.292	3.272	3.15
	Difference from cellulose, %			
500	–	–0.072 (–2.2)	–0.052 (–1.6)	0.07 (2.2)

Assessment of the influence of the uranium mass in the cask

Computational modeling makes it possible to investigate any range of uranium masses in the cask, while measurement capabilities are often greatly limited by small dimensions of uranium samples with a "zero" mass or by large dimensions of samples with a small uranium density to exclude intensive self-absorption of gamma radiation in the local region of the sample as such. Calculations were performed for the uranium masses in the cask in a range of 0.64 g to 2 kg, with the statistical calculation error practically not depending on the uranium mass in the sample due to the calculation peculiarities when the statistics of the counts in the peak is defined just by the share of the considered line yield relative to all lines in the initial spectrum during modeling.

Table 4 presents the results of calculating the intensity of the count in the 185.7 keV peak with the background deduction for waste in the form of cellulose of a varying density with different uranium masses.

The table data show that the self-absorption of the 185.7 keV line in uranium, the uranium mass range in the cask being up to 100 g, may be neglected if there are no localized regions with a large density of uranium (pieces or nonporous lumps). And the larger the density of the matrix is as such, the smaller the relative influence of absorption in uranium is. With a uranium mass of 500 g, a downward bias is observed in the intensity of the 185.7 keV peak, normalized per 1 g of ^{235}U, in the amount of ~ 5 % for nonporous matrices and ~ 10 % for a porous matrix (0.016 g/cm^3). With a uranium mass of 1 kg, the downward bias reaches 11 % for nonporous matrices and ~ 20 % for porous matrices. This effect needs to be taken into account during measurements, specifically for waste with low-enriched uranium, when the total uranium mass in the cask is much larger than the measured mass of ^{235}U. At the same time, where the mass of ^{235}U obtained by measurements does not exceed several grams, the uranium enrichment may be neglected and the potential uncertainty from the unknown uranium enrichment (total uranium mass uncertainty) can be introduced to within the confidential boundaries of the relative RB.

Table 4. Calculated intensities of the count in the 185.7 keV peak with the background deduction for different waste densities and different uranium masses in the cask

Uranium mass in cask, g	Waste material density, g/cm^3		
	0.016	0.471	0.708
	Intensity of count in 185.7 keV peak		
0.64	–	–	3.28
1	10.81	4.47	3.279
5	–	4.466	–
10	10.8	–	–
20	–	4.464	–
50	–	4.449	–
100	10.59	4.46	3.252
500	9.79	4.233	3.139
1000	–	4.017	–
2000	–	3.63	–

Conclusions

The method proposed to calculate uranium-containing waste models makes it possible to estimate the contribution of errors from variation of influencing factors in a broad range with any required step which cannot be done traditionally using a CO.

An algorithm for building an efficient calculation model has been demonstrated. The adequacy of the model and the feasibility of describing correctly real waste types, while obtaining calculated gamma spectra that agree well with real measurements, have been proved by comparing the calculation results with the data of measuring uranium-containing samples. An example has been shown of optimizing the computational model by representing a detector in the form of a ring to simulate the sample rotation about its axis.

Based on the calculation results, with regard for direct measurement data, the contribution of the uncertainty of influencing factors to the error has been estimated for particular measured objects. The use of computational methods has made it possible to extend the measurement range of the ^{235}U mass in SWR (1.8 to 100 g) in the developed MP.

Boundaries have been defined for the developed procedure to measure uranium-containing waste from the material matrix (~ 2.2 %) and its density (~ 10 %), and the contribution of the uncertainty of the uranium mass in the cask to the obtained result has been estimated (5 % for nonporous matrices, 10 % for porous matrices).

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