fact that RAO conservation is environmentally friendly and economically reasonable in geological surrounding. It was stated that for preventing from self-sustaining stored energy release from CCUR core in first place 2–3 graphite upper bushes of operating cells should be removed as well as the complete set of bushes from CM3 and OM cells. Composition and way of preparing compositions on the basis of Tomsk clays was determined. They provide the following properties:

- high adsorptivity with regard to different radioactive nuclides;
- conservation of properties during several hundreds of years;
- stable behavior of construction materials in the filler environment;
- sufficient bearing resistance.

The question on adsorptivity of these clays is still an open question. The Institute of Physical Chemistry of RAS is engaged in its solution. As a result of this work the preservative for RAO storage and stopped reactors will be selected.

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GAMMA-SPECTROMETRIC CONTROL METHOD OF ACTIVITY AND NUCLIDE COMPOSITION OF LOW-ACTIVE SOLID RADIOACTIVE WASTE

P.M. Gavrilov, A.G. Kokhomskiy, K.M. Izmestiev, I.N. Seelev, M.E. Silaev*

Siberian Chemical Combine, Seversk, Tomsk region
*Tomsk Polytechnic University
E-mail: silaev@k21.phtd.tpu.ru

The gamma-spectrometric control method of low-active solid radioactive waste, based on direct measurement of activity and nuclide composition has been developed. The measurements were carried out in the geometry of standard steel container of 200 l. volume, where low-active wastes were placed. To take into account distribution non-homogeneities of solid waste over the geometry measured a special rotating platform was used, the low-active radioactive wastes being placed on it. Metrological certification was performed and the main errors of this method for 95 % of confidence probability were determined.

Introduction

For enterprises of atomic industry increase in safety processes of radioactive waste treatment is one of the perspective ways of development. The crucial moment in solving the safety problems is development and introduction of modern methods in radiation survey permitting for determination of basic characteristics (activity and radioactive nuclide composition) at all stages of treatment [1, 2].

At present radiation survey of solid radioactive waste (SRW) of low and middle activity produced in the process of operation at most atomic enterprises is performed by measurement of dosage rate and value of surface radioactive contamination. As a rule, metrologically certified techniques of SRW activity and nuclide composition considering distribution non-homogeneities of solid waste activity over the measured geometry is absent.

The purpose of the present work is to develop gamma-spectrometric control method of low-active SRW activity and nuclide composition including the corresponding methodological and metrological equipment.

Gamma-spectrometric control method of activity and nuclide composition of low-active solid radioactive waste

The method suggested is based on direct measuring the activity and nuclide composition of waste by gamma-spectrometer with extended uncertainty (at confidence probability P=0,95) from 30 to 60 % in the energy range from 80 to 1500 keV and SRW activity range from 10^6 Bc/kg and higher.

Measurements are performed in the geometry of standard steel cylindrical container of 200 l volume for long-term storage (burial) of low-active waste. Container with radioactive waste (RAW) is placed directly before semiconductor detector of gamma-radiation put into lead sheath with collimating device (Fig. 1). The distance from container with SRW to detector is 1 m, in this case statistic uncertainty of countable sample measurement does not exceed 40 %. Collimator in lead sheath is made in such a way that visibility angle spans the whole container. The centre axis of collimator is at the level of effective centre in gamma-radiation detector. To take into consideration non-homogeneities of waste distribution over the measured geometry rotating platform on which the container with RAW is placed is suggested to be used. Gamma-radiation spectrum is read at rotation of platform.

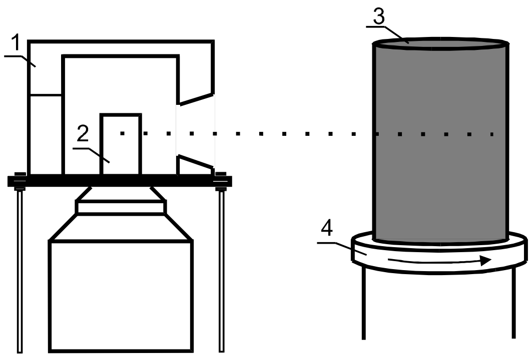

Fig. 1. *Bench of radiation control of low-active SRW activity: 1) lead sheath with collimator; 2) Ge-detector; 3) standard steel container with waste; 4) electromechanical rotator*

The essence of the measurement method consists in registering and subsequent processing of instrument spectrum of SRW calculation sample gamma-radiation by spectrometric complex. Measurements are carried out by gamma-spectrometer of «Green Star» production with semiconductor germanium detector [3]. The peculiarity of the suggested gamma-spectrometric measurement method is that dependence of gamma-radiation registration efficiency on its energy is determined at two fixed distances from detector in «point» geometry on the basis of instrument spectra of standard gamma-radiation sources. Hereafter, using calculation model (the Monte-Carlo statistical method) recalculation of gamma-radiation registration efficiency is made depending on its energy for «container with SRW» geometry (volume source including absorption), in this case geometrical parameters of container, density and composition of radioactive wastes are used in calculation [3]. The Monte-Carlo method allows for mathematically exact construction of model for transfer of gamma-quantum from the source to detector volume of different form and calculation for registration probability of gamma-radiation photons of different energies in detector.

Before measuring the container with wastes is weighted and placed on the platform rotating around vertical axis for measuring. With the help of gamma-spectrometer the counting rate for discrete energy of SRW gamma-radiation is measured in the specified energy range. Measurements are made in «container with SRW» geometry at rotation of platform. Identification and calculation of specific activity of waste gamma-radiation being in the container is performed by special software using measured counting rates at the total absorption peaks, definite efficiency of spectrometer registration and value of waste mass.

As a basic method of analysis decomposition of experimental gamma-radiation spectrum into models of nuclide spectra included in working list is applied in the programme [3]. The suggested method of analysis has a number of advantages over traditional searching for peaks with subsequent identification. Firstly, it is a high stability in nuclide identification having many lines, secondly, it is reliable determination of nuclides not having separate lines, but only included in multiplets, and thirdly, it is high sensitivity in determination of minimal activities, since the procedure of searching for peaks is practically excluded in its ordinary sense. At the same time the approach suggested imposes increased requirements on accuracy of energy calibration and completeness of nuclide working list.

Metrological certification of gamma-spectrometric method and analysis of certification results

To check the reliability of activity and nuclide composition determination of SRW low-active gamma-radioactive nuclides experimentally by means of gamma-spectrometer by the method suggested as well as to state the boundaries of uncertainties (errors) metrological certification was performed. Uncertainty of gamma-radiating nuclide activity of low-active SRW is to be measured in geometry of steel container (200 l).

The method of metrological certification consists in the following:

1. sample sources of special purpose (SSSP) have been prepared in «container» geometry with different distribution over the container geometry of certified sources on the basis of radioactive nuclides Eu-152 and Ba-133 with activity of the order of $4\cdot10^6$ Bc, with various values for density of matrix waste material;
2. by means of gamma-spectrometer the counting rate for discrete energies of SSSP gamma-radiation is measured in «container» geometry; measurements are made at rotation of electromechanical rotator;
3. with the help of software identification and calculation of gamma-radiation activity for sample sources of special purpose in «container» geometry and minimal detected activity determined by spectrometric complex is performed.

For certification three types of sample sources of special purpose (SSSP) have been prepared in «container» geometry made by certified radiation sources:

1. without absorbing material (air) in unconfined space of container (successively ranging carton layers and point sources are placed in the container);
2. with absorbing material (water is poured into the container, but the point sources are fixed in cartridges fastened on the wire; distribution of point sources within the container is performed by load and floater (cartridges) shift within the container;
3. with absorbing material of different packed density (non-radiant material of solid wastes (sawing, cloths, film, carton, elastron, metal plates, tubes etc) and point sources are places in the container successively; the number of alternate layers of non-radiant solid waste and point sample source material is so that the packed density and container weight correspond to typical packed density and container weight with SRW used at Siberian Chemical Combine). Several sample sources of special purpose have been made in «container» geometry with different packed density.

Measurement of counting rate for discrete energies of sample source gamma-radiation of special purpose in «container» geometry is carried out for each geometrical distribution of sources and density of the material placed within the container.

The processing of the results in certifying measurements consists in comparison of nameplate values of radioactive nuclides activity of sample sources used for SSSP preparation in «container» geometry (from certificates on tests of sources used) with average values of measured radioactive nuclide activity in sample sources of special purpose in «container» geometry, in this case compliance of the condition is tested [4, 5]:

$$\left|\overline{A_0} - A_0\right| \le \sqrt{U(P)^2 + \delta(A)^2}, \qquad (1)$$

where $\overline{A_0}$ is the average value of sample source activity of special purpose in «container» geometry obtained as a result of calculation, Bc; A_0 is the value of sample source activity (from the certificates on source checking), Bc; $U(P)$ is the extended uncertainty of calculation for sample source activity of special purpose in «container» geometry with confidence probability 95 %, Bc; $\delta(A)$ is the error in value of sample source activity (from the certificates of source check), Bc.

The results of comparison of nameplate values of radioactive nuclides activity of sample sources with average values of measured radioactive nuclide activity in «container» geometry are presented in Tables 1–3.

Analysis of errors in determination of radioactive nuclide activity in SSSP is performed by means of data presented in Table 4.

According to the results of investigation it follows that application of container rotation with SRW for measuring the spectrum of gamma-radiation qualitatively improves the accuracy of spectrometric measurements in case of nonuniform distribution of radiation sources over its space and is a necessary condition of their performance at the necessary level of quality.

***Table 1**.* *Difference in values of radioactive nuclides being in container at 1 m distance from detector (without absorbing material)*

Nuclide	$\frac{\left\|A_0-\overline{A_0}\right\|}{A_0}$, %	$\frac{U_A}{\overline{A_0}}$, %	Geometry of source distribution in the container
Eu-152	0,39	22,93	In geometrical centre
Ba-133	5,90	22,94	
Eu-152	2,84	23,30	At the wall in the middle of the height
Ba-133	5,35	23,32	
Eu-152	10,87	23,38	In line in the middle of the height
Ba-133	9,39	23,19	
Eu-152	3,28	23,21	In horizontal plane in the middle of the height
Ba-133	4,80	23,01	
Eu-152	3,28	23,21	In horizontal plane in the middle of the height
Ba-133	4,80	23,01	
Eu-152	0,12	22,97	On the bottom from one of sides
Ba-133	0,79	23,02	
Eu-152	23,45	23,10	On the bottom from the side most removed from detector (without rotation)
Ba-133	20,94	23,17	
Eu-152	36,05	23,21	On the bottom from the nearest side from detector (without rotation)
Ba-133	39,92	23,05	
Eu-152	10,18	23,11	Sources on the bottom of container in horizontal plane
Ba-133	13,90	23,09	

Note: $U_A = \sqrt{U(P)^2 + \delta(A)^2}$

***Table 2**.* *Difference in values of radioactive nuclides uniformly distributed in container at 1m distance from detector (container is filled with water)*

Nuclide	$\frac{\left\|A_0-\overline{A_0}\right\|}{A_0}$, %	$\frac{U_A}{\overline{A_0}}$, %	Geometry of source distribution in the container
Eu-152	22,88	32,93	Lower than the middle
Ba-133	20,06	33,66	
Eu-152	25,51	31,89	Over the whole space
Ba-133	6,92	33,44	
Eu-152	28,35	32,50	Over the space (diameter of the container 40 sm)
Ba-133	30,63	36,87	
Eu-152	12,72	31,79	Over the whole space (5 sm lower the upper edge)
Ba-133	23,61	32,59	
Eu-152	46,41	41,31	Over the whole space (5 sm lower the upper edge, without rotation)
Ba-133	30,88	41,34	

According to the results of metrological certification for methods of measurements «Control of gamma-radiating nuclides of low-active SRW at SCC» the certificate of metrological attestation with extended uncertainty from 30 to 60 % (at confidence probability P=0,95) in the energy range from 80 to 1500 keV and the activity range to 10^6 Bc/kg. The certificate was given by State Standard Department of Russia «VNIIFTRI». The suggested gamma-spectrometric method of activity and nuclide composition measurements of low-active SRW was introduced at the reactor of «Siberian chemical combine» and adopted as an analytical technique «Control of gamma-radiating nuclides of low-active SRW at SCC». The technique is consistent with State Standard of Russia of **«VNIIFTRI».**

Table 3. *Differences in values of radioactive nuclide activity being in container at 1m distance from detector (container is filled with absorber of 0,1...0,2 g/sm³ density; the sources are distributed uniformly over the container space)*

Nuclide	$\frac{\lvert A_0-\overline{A_0}\rvert}{A_0}$, %	$\frac{U_A}{\overline{A_0}}$, %	Order number of measurement cycle
Eu-152	5,13	31,78	1
Ba-133	11,42	31,83	
Eu-152	3,08	31,59	2
Ba-133	12,36	31,66	
Eu-152	6,00	31,58	3
Ba-133	10,38	31,65	
Eu-152	5,97	31,55	4
Ba-133	9,10	31,68	
Eu-152	4,63	31,55	5
Ba-133	10,03	31,65	
Eu-152	7,25	31,45	6
Ba-133	12,42	31,61	
Eu-152	9,98	31,69	7
Ba-133	36,20	31,73	
Eu-152	9,68	31,68	8 (without rotation)
Ba-133	33,00	31,62	

Conclusion

The gamma-spectrometric method of control permitting for high degree of accuracy in determination of nuclide composition and calculation of specific and absolute activity of low-active SRW formed at operation and removing the nuclear installation from the service directly has been developed and introduced.

It was experimentally proved that application of calculation sample rotation (container with radioactive wastes) allows us to improve the accuracy of spectrometric analyses and is their necessary condition.

Table 4. *Errors of results in determination of radioactive nuclides activity in SSSP*

Условия проведения измерений		$\frac{\lvert A_0-\overline{A_0}\rvert}{A_0}$, not more %	$\frac{U_A}{\overline{A_0}}$, not more %
Container without filler	with rotation	14	24
	without rotation	40*	24
Container poured with water	with rotation	31	37
	without rotation	47*	42
Container with wastes (sawing, pieces of metal and etc)	with rotation	13	32
	without rotation	37*	32

**error in determination of activity exceeds the requirements for analysis*

The calculated algorithm (the Monte-Carlo statistic method) realised in the suggested gamma-spectrometric method permits for calculation of solid radioactive waste specific activity placed in steel container of 200l volume (non-homogeneous volume source including radiation self-absorption) with extended uncertainty fitting in the range from 30 to 60 %.

Instrumental-methodological developments make possible to treat low-active solid radioactive wastes at Siberian chemical combine according to requirements of existing State standards and rules in the sphere of atomic energy.

The developed gamma-spectrometric method of analysis may be used for control of solid radioactive waste at other radiation-dangerous objects.

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