

Non-Destructive Measurements of the Characteristics of Radioactive Contamination of Near Surface Layers of Concrete and Ground with Collimated Spectrometric Detectors

V.N. Potapov, A.S. Danilovich, S.M. Ignatov, A.G. Volkovich,
O.P. Ivanov, V.E. Stepanov, V.G. Volkov
RRC Kurchatov Institute
1 Kurchatov Square, Moscow 123182
Russia

ABSTRACT

The remote radiometric method for estimation of the characteristics of concrete and ground contamination by gamma nuclides was investigated. The studies were carried out for possible realization of a method with two types of spectrometric detectors: 1) scintillator + PMT and 2) scintillator + photo diode. The measurements of Cs-137 contamination were considered, as most widespread gamma-contaminant. The mathematical model of the radiometric device was developed for studying of a method, optimization of device's parameters and search of all dependences necessary for method realization. The collimated radiometer with volume of the detector 20 cm³ has the limiting sensitivity - MMA = 0.5 μCi/m² (19 kBq/m²) at thickness of contaminated concrete layer 5 cm. The method can be realized for measurement of contamination of concrete and ground by other gamma radiating nuclides also.

INTRODUCTION

A sampling method is a traditional means of determining basic quantitative characteristics and parameters of distribution of radionuclides in a medium [1, 2]. In particular, this method is used to examine soil contaminated as a result of global or accidental fallout. In certain situations, however, the sampling method is hardly practicable. For example, concrete settlers are used for temporary storage of liquid radioactive waste (RW). In the course of the storage radionuclides enter the concrete. A similar situation may occur when solid RW is kept in rooms (storage vaults) with concrete walls and coatings. Upon discharge of the RW it is often necessary to remove an upper layer of concrete that contains radionuclides, therefore it is desirable to have preliminary information on thickness of a layer to be removed. If the sampling method is used for radionuclide content analysis in this case, additional special equipment shall be employed which makes the analysis quite labour-intensive.

There are field radiometric methods of determining characteristics of soil and bottom deposit contamination [3-6] that allow evaluating surface activity of radionuclides and thickness of the upper layer of soil that contains more than 80% of the total activity, and establishing some other distinctive features of radionuclide distribution in a medium. Radiometric methods may be applicable for evaluating concrete contamination characteristics as well. In these situations radiometric analysis methods are cheaper, faster and less labour-expensive.

The manuscript presents the development of a technique for measuring the surface activity of Cs-137 and Co-60 in concrete and evaluating their penetration depth with radiometric instruments. The main achievable parameters in realisation of the technique with different types of scintillating collimated detector are studied on bases of mathematical simulation of the instrumental spectra of the detectors. The results obtained are the bases for implementation of developed technique with particular types of detector with some fixed parameters.

The collimated detector with scintillator and photodiode readout was developed and used in measurements of contamination of different concrete surfaces. The results of measurements are presented and discussed.

THE TECHNIQUE OF RADIOMETRIC MEASUREMENTS OF CHARACTERISTICS OF CS-137 CONTAMINATION IN CONCRETE AND GROUND

The radiometric method of evaluating the depth of Cs-137 radionuclides penetration in concrete is implemented according to a measurement schematic presented in Fig. 1a. To put this measuring method into practice, a spectrometric detector with a lead shielding and a collimator is placed above the concrete surface. This schematic provides for two measurements – with a plug and without it, which allows excluding the effect of radiation that passed through the shielding and collimator sidewalls. Thus the use of the collimator and detector shielding ensures high spatial resolution of the method, i.e. allows obtaining information on characteristics of contamination with radionuclides for a limited concrete surface area.

Discussed below are two types (A and B) of the detector systems differing in collimator geometry parameters and detector characteristics. The basic distinction between the detector systems under consideration consists in their sensitivity, which primarily has to do with the difference in volumes of the detector scintillation crystals and the value of the collimator angular aperture. Besides, the type A detector is expected to be built as spherical scintillator CsI(Tl) + photodiode optical pair, and the type B detector uses a photomultiplier-based standard photodetector with a NaI(Tl) scintillation crystal.

At the development of technique of measurements the following actual parameters of detectors were used for simulation of two types of collimated detectors:

type A: the spherical scintillation crystal CsI(Tl) of volume 20 cm^3 ; the collimator angular aperture $\theta = 32^\circ$, the thickness of the shielding and collimator walls is 40 mm, and the thickness of the plug (shutter) is 30 mm .

type B: the scintillation crystal NaI(Tl) $\varnothing 3'' \times 3''$ + photomultiplier tube, the collimator angular aperture $\theta = 54^\circ$, the thickness of the shielding and collimator walls is ~ 30 mm, and the thickness of the plug (shutter) is 30 mm. Shielding collimators and plugs are made from lead (Pb) for both detectors.

The main objective of the measurement technique is to establish a method for determining surface activity and evaluating the Cs-137 penetration depth in the upper layer of concrete. It is proposed that several energy regions of the collimated detector spectrum be used to accomplish this objective. Sizes and locations of the energy regions, as well as relationships and dependences required for implementation of the measurement technique were defined using a mathematical model based on the Monte Carlo method. This mathematical model allows calculating spectrometric detector readings (instrumental spectra) that are fully consistent with the measurement schematic in Fig. 1a. The model takes into account all processes of interaction and transport of scattered and non-scattered radiation in concrete, shielding, collimator and the detector scintillation crystal. Fig. 1b shows measured and calculated instrumental spectra of the collimated spectrometric detector. The measurements were taken in laboratory conditions using specially manufactured plane Cs-137 sources for simulation contamination in concrete. The calculations performed using the model also corresponded to the experiment geometry, and are in close agreement with the measured data, which allows using the model for development and analysis of the measurement technique.

The measurement technique was developed on the assumption that radionuclides are found in the upper layer of concrete and their distributions are described by functions of the type $A(z) = A_s P(z)$, where

A_s is the total surface activity of the entire layer of radionuclides in the concrete ($[A_s] - \text{Bq/cm}^2$), and

$P(z)$ is the distribution density function ($\int_0^\infty P(z)dz = 1$).

In view of the specificity of the task involved (determined by conditions of contaminated concrete layer formation), only radionuclide distributions decreasing with depth were used when developing the measurement technique. The distribution patterns used were described by power- and exponential function profiles of radionuclide activity in concrete as a function of depth. These functions are the following:

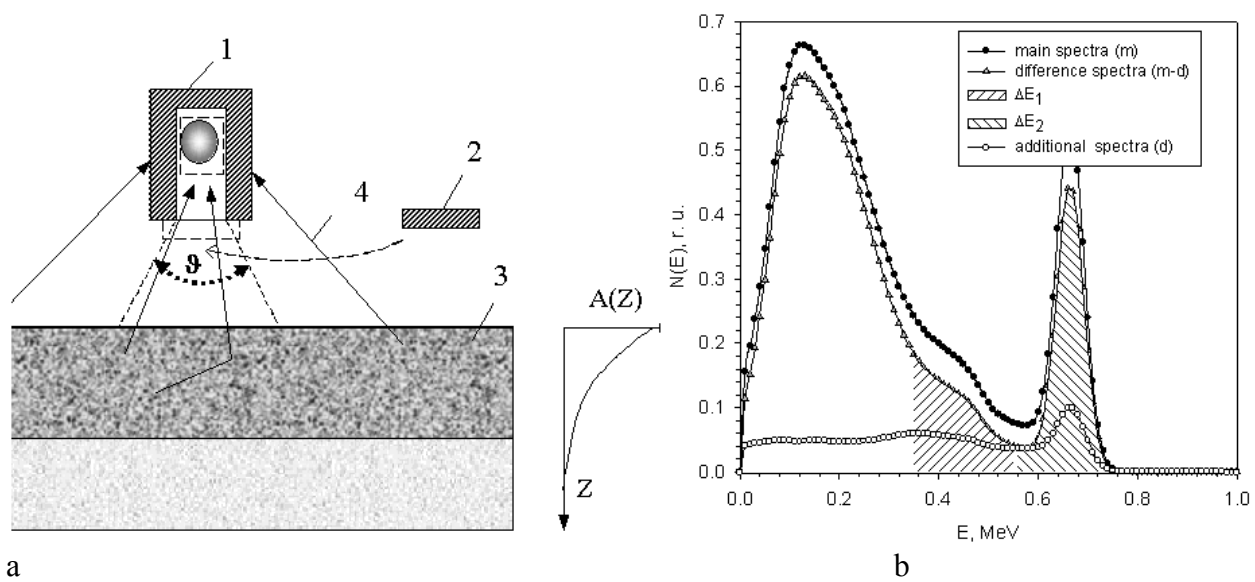


Fig. 1. a) The schematic of measurements with the collimated spectrometric detector. 1 – spectrometric detector with the shielding and collimator; 2 – collimator plug; 3 – contaminated layer with decreasing volume activity of Cs-137 in concrete; 4 – gamma quanta of scattered and non-scattered radiation recorded by the detector. b) Instrumental spectra of the main measurement (m), additional (background) measurement (d) and their difference (m-d). Shaded areas correspond to energy ranges ΔE_1 and ΔE_2 that are used in the measurement procedure.

$$P(z, Z_0) = \begin{cases} \frac{(n+1)}{Z_0} \left(1 - \frac{z}{Z_0}\right)^n, & 0 \leq z \leq Z_0, \\ 0, & z > Z_0 \end{cases}, \quad (\text{Eq. 1})$$

$$P(z, Z_0) = \frac{2.3026}{Z_0} \exp\left(-\frac{2.3026}{Z_0} z\right). \quad (\text{Eq. 2})$$

Here $A(z)$ defines the volume activity of radionuclides in a medium ($[A(z)] - \text{Bq/cm}^3$), Z_0 is the parameter that characterizes thickness of the concrete layer that contains radionuclides (for power-function distributions Z_0 corresponds to the thickness of the layer containing 100% of the overall activity,

and for the exponential distribution – 90%). For $n = 1$ power-function distributions is triangle. In two limit cases, when $n \rightarrow 0$ the distribution presents uniformly contaminated layer, and when $n \rightarrow \infty$ distribution presents thing plane source on surface.

If the distribution of the volume activity of radionuclides in depth $A(z)$ is defined, the following simple relationship can be used for description of distributions of their specific activities:

$$A_{\gamma 0}(z) = A(z)/\rho,$$

where ρ - concrete density ($[\rho]$ – kg/cm^3).

As noted above, implementation of the technique requires two measurements – the main one (m – without the plug), and the additional one (d – with the plug). The difference spectrum is for the most part formed only by those gamma quanta that passed through the detector collimator. To determine the surface activity and evaluate the depth of Cs-137 penetration in concrete, it is sufficient to use two energy ranges of the difference instrumental spectrum ΔE_1 and ΔE_2 (see Fig. 1b). The spectrum energy region ΔE_2 is selected so that it corresponds to the peak of the total absorption of radiation with the energy of 662 keV. The instrumental spectrum in the energy range ΔE_1 is determined by recording of both non-scattered Cs-137 radiation (Compton part of the spectrum) and Cs-137 radiation scattered in concrete.

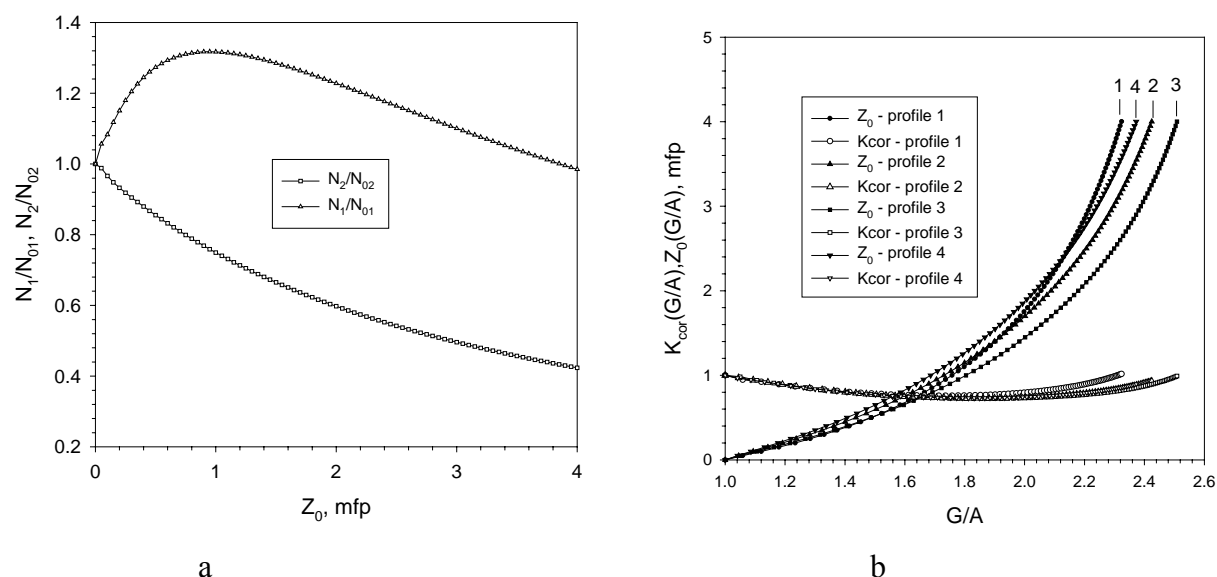


Fig. 2. The simulation of type A detector performance: a) relative values of count rates in the spectrum energy regions ΔE_1 and ΔE_2 versus parameter Z_0 for exponential depth profile of radionuclides penetration in concrete; b) correction factor K_{cor} and depth Z_0 versus G/A for different depth profiles: 1 – exponential depth profile; 2, 3 and 4 – power-function profiles with $n=1, 0.75$ and 1.25 correspondently

If Cs-137 has the unvaried depth distribution in a surface layer of concrete $P_c(z, Z_c)$, the count rate in the energy ranges mentioned above would be proportional to the surface activity, and with appropriate calibration of the instrument this characteristic of the concrete surface contamination could be easily determined. However, when Cs-137 enters into concrete and Z_0 is increased, then due to absorption of

gamma quanta in concrete, the count rate will depend on the depth of caesium penetration in the concrete. Fig. 2a shows how the count rates N_1 and N_2 will vary in the energy ranges ΔE_1 and ΔE_2 with the radionuclides penetration depth Z_0 when their surface activity is constant. The figure shows relative changes of the count rates, i.e., values N_1/N_{10} and N_2/N_{20} , where N_{10}, N_{20} - spectrometric detector count rate in respective energy regions ΔE_1 and ΔE_2 for the case of a thin surface contamination of concrete (not deepened radionuclides). As is seen from the figure, penetration of radionuclides into the interior of concrete results in a noticeable reduction of the count rate in the region of the total absorption peak due to absorption of non-scattered radiation.

However, with increasing depth of radionuclides penetration the count rate in the range ΔE_2 not only does not decrease but even increases up to the penetration depth $Z_0=1$ mfp, and only then begins to decrease slowly. This increase is due to gamma quanta scattered by small angles and recorded by the collimated detector. As radionuclides penetrate deeper, a scattering medium (concrete) appears between the detector and the radiation source, and quanta scattered toward the detector (by small angles) occur in this media. When the radionuclides penetration depth increases to 1 free path, the total number of such scattered quanta increases, and then (at $Z_0>1$) begins to decrease, now due to the decreasing volume density of radionuclides in the concrete. The latter circumstance (weak dependence of N_1 on the penetration depth) allows determining the surface activity of Cs-137 from the count rate in the range ΔE_2 . Since the count rate in the range ΔE_2 is proportional to surface activity of radionuclides and virtually independent of the penetration depth (within ~30% of the error of method), the mentioned characteristic of concrete surface contamination can be easily determined if the instrument is calibrated accordingly.

Since N_1 and N_2 vary in different manners with increasing depth of Cs-137 penetration, the ratio N_1/N_2 does not depend on the overall activity of radionuclides and depends solely on their penetration depth. The latter circumstance allows both evaluating the penetration depth and recovering the true surface activity when radionuclides enter concrete (adjust its value in view of the penetration depth).

Formally this procedure is implemented as follows. The instrument is calibrated by finding coefficients α and β for which the following condition is fulfilled:

$$\alpha N_{10} = \beta N_{20} = A_0, \quad (\text{Eq. 3})$$

where N_{10}, N_{20} are count rates in the spectrum energy regions ΔE_1 and ΔE_2 when taking measurements with a spectrometric detector placed above a plane film source in the standard position (see Fig. 1a); A_0 is the surface activity of the plane calibration Cs-137 source (in the subsequent discussion the values of the coefficients α and β are defined in terms of unit surface activity, e.g. $A_0 = 1 \mu\text{Ci}/\text{m}^2$).

For measurements in actual conditions when caesium is some way or other distributed in depth in concrete, the surface activity of caesium G^* (adjusted value) can be determined from the following relationship:

$$G^* = GK_{cor}(G/A), \quad (\text{Eq. 4})$$

where $G = \alpha N_1$, $A = \beta N_2$. $K_{cor}(G/A)$ is correction factor that takes into account variation of the count rate N_1 as caesium penetrates in concrete. As noted above, the degree of penetration (thickness of the caesium layer in concrete) determines the value of the ratio N_1/N_2 (or up to the constant G/A),

therefore the correction factor K_{cor} may be a function of G/A which eventually allows recovering the true surface activity of caesium without a priori information on the penetration depth.

Three most common distribution patterns – exponential, triangular and uniform triangular – were considered. As seen from the figure, the correction factor depends only slightly on the form of the function of caesium distribution in depth $P(Z, Z_0)$. Since information on the pattern of caesium distribution in depth is not available when taking spectrometric measurements of the surface activity, one of the K_{cor} plots in Fig. 2b can be chosen as a universal function. This will obviously involve an error of method (according to our estimates, no greater than 15%) that will have effect primarily at caesium penetration in concrete deeper than 10 cm (>2 free paths). The distributions considered are most typical ones (in view of the decreasing pattern of Cs-137 distributions in concrete) for which a universal correction function can be chosen. Such a universal function for the correction factor can also be used for an arbitrary combination of these distributions.

The Cs-137 penetration depth in concrete is evaluated in a similar manner. To this end, the following relationship is established by calculations:

$$Z_0 = f(G/A), \quad (\text{Eq. 5})$$

and further used to determine the penetration depth parameter Z_0 . The penetration depth parameter Z_0 is expressed in terms of mean free paths (mfp) of gamma quanta with the energy of 662 keV. The following expression can be used to convert the value of this parameter into [cm]:

$$Z_0[\text{cm}] = 5.5 \times \frac{\rho_0}{\rho} \times Z_0[\text{mfp}], \text{ where } \rho_0 = 2.35 \text{ g/cm}^3, \rho - \text{concrete density.}$$

Fig. 2b shows these plots for various patterns of Cs-137 distribution in concrete. As is seen from the figure, these plots also depend only slightly on the pattern of radionuclides distribution in depth, therefore, a plot that corresponds to the most probable distribution can be chosen as a universal function.

Only one point remains unclear: what energy ranges ΔE_1 and ΔE_2 shall be used in this technique? While for ΔE_2 this question has a single answer (location and size of the range ΔE_2 are determined by the detector energy resolution in the 662 keV line), further studies are required for ΔE_1 . For example, for a detector with the energy resolution of $\sim 8\%$, the range $\Delta E_2 = [560 - 750]$ keV, and the bounds of the range ΔE_1 shall be defined such that the measurement error for sought-for quantities is minimum.

The relative error ($\delta_z \equiv \frac{\Delta Z_0}{Z_0}$) of the penetration depth Z_0 measurement is determined from the formula:

$$\delta_z \times \sqrt{A_s t} = \left(\frac{Y}{f} \frac{df}{dY} \right) \times \sqrt{\left(\frac{\Delta G}{G} \right)^2 + \left(\frac{\Delta A}{A} \right)^2}, \quad (\text{Eq. 6})$$

where A_s is the surface activity of Cs-137, t is the exposure time for the main and additional spectra measurements, $Y \equiv G/A$, and f is the function defining relationship (Eq. 5),

$$\frac{\Delta G}{G} = \frac{(N_{1m} + N_{1d})}{(N_{1m} - N_{1d})^2}, \quad (\text{Eq. 7})$$

$$\frac{\Delta A}{A} = \frac{(N_{2m} + N_{2d})}{(N_{2m} - N_{2d})^2}. \quad (\text{Eq. 8})$$

Here $N_{1,m,d}$ and $N_{2,m,d}$ are count rates in the energy ranges ΔE_1 and ΔE_2 of the main (m) and additional (d) spectra, respectively.

According to (Eq. 4), the relative error of measuring the surface activity $\delta_G \equiv \frac{\Delta G^*}{G^*}$ will be defined by the following formula:

$$\delta_G \times \sqrt{A_5 t} = \sqrt{\left(1 + \frac{Y}{K_{cor}} \frac{dK_{cor}}{dY}\right)^2 \times \left(\frac{\Delta G}{G}\right)^2 + \left(\frac{Y}{K_{cor}} \frac{dK_{cor}}{dY}\right)^2 \times \left(\frac{\Delta A}{A}\right)^2}, \quad (\text{Eq. 9})$$

where $\frac{\Delta G}{G}$ and $\frac{\Delta A}{A}$ are also found from (Eq.7) and (Eq.8).

The plots δ_G and δ_Z as a function of left bound E_1 of the energy range $\Delta E_1 = [E_1 - 560]$ keV were calculated from formulae (Eq. 6) - (Eq.9). The numerical differentiation ($\frac{df}{dY}$ and $\frac{dK_{cor}}{dY}$) was performed using Lagrange's interpolation formulae [7].

The minimum values of δ_G and δ_Z for the type A detector correspond to $E_1 = 360$ keV and $E_1 = 320$ keV, respectively. For the type B detector, the minimum values correspond to $E_1 = 360$ keV and $E_1 = 340$ keV. Since the minimum values of these two plots are not pronounced, their mean value, i.e. $E_1^{opt} = 340$ keV, can be chosen as the optimum value of E_1 .

For the optimum value of E_1 , there were calculated universal curves of G^* and Z_0 measurement errors (Fig. 3) that can be used for evaluation of exposure time required to achieve the prescribed accuracy of measurement of these quantities.

It should be noted that the f and K_{cor} curves obtained are not universal and to a great extent depend on the shape and sizes of the detector shielding, collimator and scintillation crystal. The analysis presented herein was performed for two particular cases of geometric parameters of detector systems with different sensitivity.

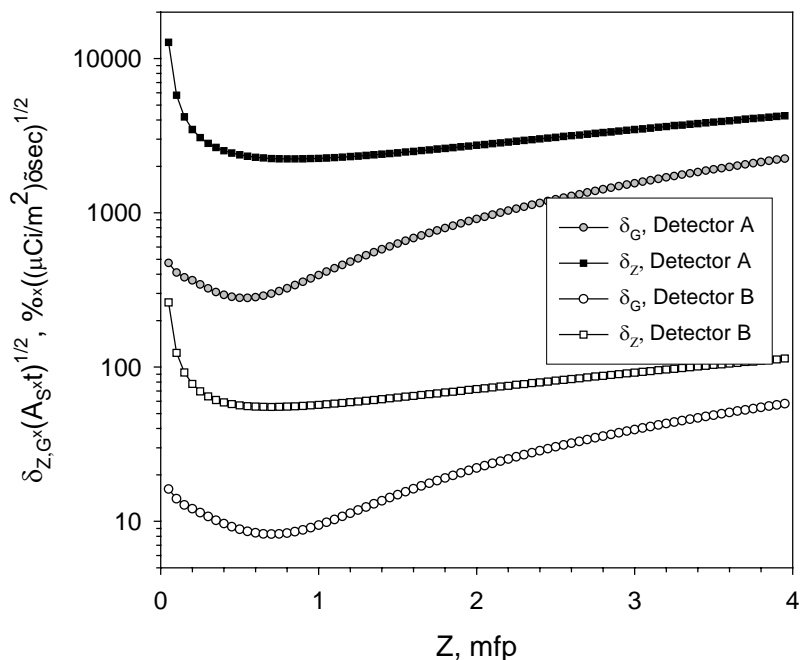


Fig.3. Universal curves for evaluation of a possible error (with the confidence level of 0.95) of G^* and Z_0 measurements shown as a function of thickness of the layer of Cs-137 radionuclides in concrete. Detectors of type A and B.

Consideration of the Effect of Natural Radionuclides Radiation

When concrete has a low content of Cs-137, natural radionuclides (NRN) radiation may have a noticeable effect on measurement results. Fig. 4 shows instrumental spectra of radiation from NRN and Cs-137 with the surface activity of $1.0 \mu\text{Ci}/\text{m}^2$ ($\sim 40 \text{ kBq}/\text{m}^2$). Specific activities of NRN corresponded to their typical content in concrete. As seen from the figures, even at rather high level of surface activity of technogenic Cs-137 radionuclides, the NRN radiation has a significant effect. (According Russian regulations the activities of NRN in concrete may 3- 5 times larger.)

Radiation from such natural radionuclides as K-40, U-238, Th-232 and their daughter decay products can be taken into account in two ways.

The first approach uses preliminary measurements of the NRN background spectrum taken for a sample of clean concrete, and takes into account the NRN contribution by subtracting respective background values in the spectrum operating energy ranges ΔE_1 and ΔE_2 from subsequent measurements.

The second approach is based on the use of an additional energy range ΔE_3 in the high-energy spectral region where the Cs-137 radiation does not make its contribution.

A drawback of the first approach is that it is not universal. For a different grade of concrete, NRN composition may change markedly which will result in an error of measurement method. A merit of this approach is a possibility to measure the background spectrum in advance with good statistic (long exposure time), which will allow taking into account the effect of NRN with a minimum error.

The second approach, while more universal as it takes into account changes in composition of NRN in concrete, involves a certain error, both systematic and statistical, in doing so.

If the first approach is evident from the procedural point of view, implementation of the second one calls for clarifications. Thus to exclude the effect of NRN on measurement results it is proposed to use the instrumental spectrum energy range ΔE_3 (see Fig. 4). This energy region of the spectrum is formed through recording of NRN radiation only, and is determined by three components, i.e. radiation from K-40 (count rate N_3^K), U-238 (count rate N_3^U) and Th-232 (N_3^{Th}). N_3^U and N_3^{Th} include radiation from all daughter decay products of U-238 and Th-232 being in equilibrium with the latter.

Let us designate the count rate of background radiation in the energy ranges ΔE_1 and ΔE_2 as N_{b1} and N_{b2} .

Now we consider two energy ranges ΔE_1 and ΔE_3 for which the following relationships between the background radiation count rates hold true:

$$N_{b1}^K = \sigma_2^K N_3^K, \quad N_{b1}^{Th} = \sigma_1^{Th} N_3^{Th}, \quad N_{b1}^U = \sigma_1^U N_3^U, \quad (\text{Eq.10})$$

where σ_1^i are respective proportionality factors.

It is desirable that the position and width of the energy region ΔE_3 be chosen such as to fulfill the following condition:

$$\sigma_1^K = \sigma_1^{Th} = \sigma_1^U = \sigma_1^* \quad (\text{Eq.11})$$

In this case

$$N_{b1} = \sigma_1^K N_3^K + \sigma_1^{Th} N_3^{Th} + \sigma_1^U N_3^U = \sigma_1^* (N_3^K + N_3^{Th} + N_3^U) = \sigma_1^* N_3 \quad (\text{Eq.12})$$

If there exists a spectrum energy range for which condition (Eq.11) is met, consideration of the background constituent N_{b1} reduces to determination of factor σ_1^* . If condition (Eq.11) is fulfilled, the value of this factor will not depend on variations in the ratios between K-40, U-238 and Th-232 activities in concrete.

However, no energy range ΔE_3 exists for which condition (Eq.11) would be completely fulfilled, but it is possible to find an energy range such that

$$\sigma_1^K \neq \sigma_1^{Th} = \sigma_1^U = \sigma_1^*. \quad (\text{Eq.13})$$

In this case we can write

$$N_{b1} = \sigma_1^* (N_3^{Th} + N_3^U) + \sigma_1^K N_3^K$$

or

$$N_{b1} = \sigma_1^* (N_3^K + N_3^{Th} + N_3^U) + (\sigma_1^K - \sigma_1^*) N_3^K = \sigma_1^* N_3 + \Delta\sigma_1 N_3^K$$

If we assume that $N_3^K = \langle N_3^K \rangle + \Delta N_3^K$, where $\langle N_3^K \rangle$ is the count rate in the spectrum energy range ΔE_3 from K-40 with the specific activity corresponding to mean statistical value for all grades of concrete, then

$$N_{b1} = \sigma_1^* N_3 + \Delta\sigma(\langle N_3^K \rangle + \Delta N_3^K) \approx \sigma_1^* N_3 + \Delta\sigma_1 \langle N_3^K \rangle \quad (\text{Eq.14})$$

Analysis of expression (Eq.14) has shown that this approximation provides a good estimate of N_{b1} with an error that does not exceed 10% for grades of concrete with K-40 content ranging between 100 and 500 Bq/kg.

Similar expressions can be written for the energy ranges ΔE_2 and ΔE_3 as well.

$$N_{b2} = \sigma_2^* N_3 + \Delta\sigma_2(\langle N_3^K \rangle + \Delta N_3^K) \approx \sigma_2^* N_3 + \Delta\sigma_2 \langle N_3^K \rangle. \quad (\text{Eq.15})$$

Thus relationships (Eq.14) and (Eq.15) allow evaluating the contribution of the background radiation to the spectrum energy regions ΔE_1 and ΔE_2 by the count rate in the range ΔE_3 that is beyond the region where the Cs-137 radiation has effect.

As results of the calculations has demonstrated, a spectrum energy region for which condition (Eq.13) is fulfilled shall fall in the range $\Delta E_3 = [750 - 1260]$ keV and $\Delta E_3 = [760 - 1340]$ keV for type A and B detectors, respectively.

The possibility to take into account the background radiation allows improving sensitivity of the method of determining the Cs-137 penetration depth and content at low levels of concrete contamination.

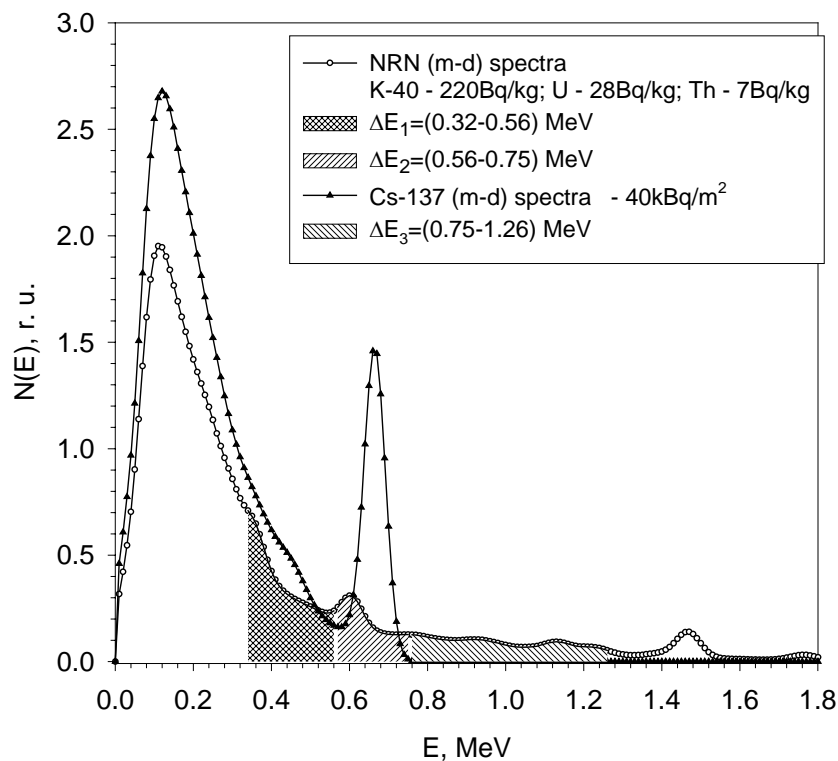
Relative measurement errors taking into account the effect of the NRN radiation are calculated by the same formulae (Eq.6) and (Eq.9) with the only difference that

$$\frac{\Delta G}{G} = \frac{(N_{1m} + N_{1d}) + [\sigma_1^{*2} (N_{3m} + N_{3d}) + (N_{1m}^{NRN} + N_{1d}^{NRN})] / A_S}{(N_{1m} - N_{1d})^2}, \quad (\text{Eq.16})$$

$$\frac{\Delta A}{A} = \frac{(N_{2m} + N_{2d}) + [\sigma_2^{*2} (N_{3m} + N_{3d}) + (N_{2m}^{NRN} + N_{2d}^{NRN})] / A_S}{(N_{2m} - N_{2d})^2}. \quad (\text{Eq.17})$$

Here A_S - is the surface activity of Cs-137. N_{jm}^{NRN} и N_{jd}^{NRN} are the count rates of NRN in spectrum energy regions ΔE_j ($j = 1,2$) for the main (m) and additional (d) measurements.

The simulation have shown that the NRN radiation influence on the accuracy of determining the values being measured depends on the concrete contamination level. The NRN radiation shall be taken into account at Cs-137 surface activity levels below 10 $\mu\text{Ci}/\text{m}^2$ (400 kBq/m²) as it has a significant impact on the measurement accuracy.



4

Fig. 4. Instrumental difference (*m-d*) spectra of NRN and Cs-137 at the surface activity of 40 kBq/m² (~1.0 μCi/m²). Detector of type A.

There exists a notion of a minimum measurable activity (MMA) [8] that is used as a basis for setting an activity level determined by a measuring tool (instrument) with the accuracy of 50% and confidence level of 0.95 in the exposure time of 1 hour. The MMA for type A collimated detector has been calculated. The MMA value depends on the depth of caesium penetration in concrete, so these data are listed in Table I. for different values of the exponential penetration depth parameter Z_0 .

Table I. Cs-137 MMA Dependence on Penetration Depth. The Type A Detector

Z_0 , mfp	MMA, μCi/m ² (kBq/m ²)
1	0.5 (19)
2	1.2 (44)
3	2.5 (93)
4	4.0 (148)

For the type B detector, the MMA values formally lie below the level of 0.1 μCi/m². However measurements with the surface activity below 0.1 μCi/m² cannot be implemented due to the effect of the systematic error of method of evaluating the NRN background constituent of the instrumental spectra. The NRN background at these Cs-137 activity levels is comparable with or even higher than the useful signal. The above data on the NRN effect on measurement results include just the statistical component of

the NRN background measurement error. Therefore, the MMA values could not be estimated for the type B detector in a reliable procedure. But it is better than 10 kBq/m².

The Peculiarities of the Technique Application for Estimation of Co-60 Contamination

The radiometric method of estimation of Co-60 contamination characteristics in concrete is based on the same measurement scheme and spectra processing as for Cs-137. The same two types of the collimator detectors (type A and B) and the same depth profiles of Co-60 contamination in concrete, described by (Eq.1) and (Eq.2) are considered. The depth contamination parameter Z_0 in mentioned profiles will be measured in mean free path of gamma-quanta of 662 keV energy (both for Cs-137 and for Co-60) to have the same unity of depth.

For determination of surface activity and penetration depth of Co-60 in concrete two new energy ranges ΔE_1 and ΔE_2 in difference instrumental spectra are selected. The energy range ΔE_2 is selected to include both peaks of total absorption at 1173 and 1332 keV. The bounds of range ΔE_1 are chosen from condition of minimization of errors of measured values.

Then consideration follows the same general approach as for Cs-137.

Surface activity and the penetration depth of Co-60 in concrete are estimated with relationships similar for (Eq.4) and (Eq.5) for Cs-137:

$$G^* = GK_{cor}(G/A), \quad Z_0 = f(G/A).$$

Functions $K_{cor}(G/A)$ and $Z_0 = f(G/A)$ for different depth profiles of cobalt in concrete $P(z, Z_0)$ were calculated. The qualitative character of these relations is the same as for caesium.

The energy range ΔE_1 (and namely the position of its left bound) is determined by minimization of statistical error of measurement of G^* and Z_0 values, i.e. their relative measurement errors δ_G and δ_Z . Minimal values of δ_G and δ_Z (for both types of detectors) are reached at $E_1 = 360$ keV.

The possible positions of energy ranges for account radiation of natural radionuclides and their daughters are intersected with positions of energy ranges necessary for measurements of Co-60 contamination parameters. So estimation and account of NRN influence may be done only by preliminary measurements of background NRN spectra of clean concrete sample and by subtraction of this spectra in working energy ranges of ΔE_1 and ΔE_2 .

The calculation of relative errors of measurements with account of NRN radiation were carried out with (Eq.6, Eq.9) and (Eq.16, Eq.17). The results of calculation reveal that as in Cs-137 case the effect of the NRN radiation shall be taken into account at surface activity levels below 10 $\mu\text{Ci}/\text{m}^2$ (400 kBq/m²) as it has a significant impact on the measurement accuracy.

The MMA for type A collimated detector are listed in Table II for different values of the exponential penetration depth parameter Z_0 .

Table II. Co-60 MMA Dependence on Penetration Depth. The Type A Detector

Z_0 , mfp	MMA, $\mu\text{Ci}/\text{m}^2$ (kBq/m ²)
1	0.25 (9)
2	0.6 (22)

3	1.1 (41)
4	2.2 (82)

At simultaneous presence of two nuclides Cs-137 and Co-60 in concrete, the modification of measurements methodic is necessary. It is obvious that left boundary of ΔE_1 energy range for Co-60 must be out of spectral area, where the input from Cs-137 radiation has place, i.e. $E_{1_{Co}} \geq 0.72$ keV. So this range must lies right from photo peak 662 keV. Such replacement of the boundary will resulted in increase of statistical errors of measurements of Co-60 contamination parameters by factor 1.5-1.8. The obtained results of measured Co-60 contamination must then be used for estimation of Co-60 radiation background in spectra areas including Cs-137 radiation. Thus, net counting rates for two caesium energy ranges will be calculated and Cs-137 contamination parameters will be estimated. These values will be estimated with larger error then in case of presence of Cs-137 only. The account on NRN radiation may be carried out on the preliminary measured spectra of clean concrete sample of same type in advance.

RESULTS OF MEASUREMENTS

The developed measuring technique was implemented with detector of type A. Detector presented collimated spectrometric probe based on spherical CsI(Tl) scintillator and Si PIN photodiode. The dimensions of detector are practically the same as were used in simulation, energy resolution for energy 662 keV is better 7%. Measured spectra were saved in memory of portable spectroanalyzer and after processing with developed software running on PC the characteristics of contamination were obtained.

Laboratory tests were carried out with plane calibrated Cs-137 and Co-60 sources and which were putted between steel sheets, that simulates presents of radionuclide in volume of concrete. These test have given coincidence of actual and measured values of surface activity and penetration depth.

Field measurements were made for estimation of Cs-137 and Co-60 contamination in concrete walls and bottom of repository, in separate concrete blocks and panels. These data of measurements are presented in Tables III. and IV.

Table III. Measurement of Cs-137 Contamination in Concrete Floor of Repository

N	$A_{Cs}, \mu\text{Ci}/\text{m}^2$	$Z_{Cs}, \text{mfp} (5.5\text{cm})$	EDR, mR/h
1	758.5	0.026	2.4
2	592.6	0.248	3.0
3	568.7	0.235	3.0
4	752.2	0.083	2.8
5	682.5	0.189	2.3
6	1193.5	0.328	2.9
7	940.8	0.0	2.7
8	654.3	0.142	2.4
9	604.7	0.427	2.8
10	1196.8	0.044	3.2
	$\langle A \rangle = 794.5$		

The field measurements were not checked up by direct measurement of the characteristics of contamination – executed analysis of nuclide distribution in depth, for example, with the sampling method. But these results have indirect quantitative and qualitative confirmation.

So, the measured doze rates corresponds to the doze rates calculated on the basis of measured values of surface activity for small depth of penetration for a case of contaminated bottom of repository (Table III.).

At measurement of contamination of concrete plates (Table IV.) only points 1 and 2, in which the large depth of penetration of contamination in concrete is appreciated, are located in area of cracks on a surface of plates, that qualitatively confirms the obtained results. The data on pollution Co-60 are inconsistent and have the large errors. It is, probably, connected to the large background irradiation of the detector through shielding by other sources of contamination, which are placed at a site. At a small collimation angle of the detector and, hence, small concerning the background useful signal of the detector it results in increase of errors of measurements.

Table IV. Measurement of Cs-137 and Co-60 Contamination in Concrete Panels

N	A _{Cs} , μCi/m ²	Z _{Cs} , mfp	A _{Co} , μCi/m ²	Z _{Co} ,mfp
1	0.571E+01	1.69	0.259E+00	0.00
2	0.175E+02	0.73	0.710E+01	4.00
3	0.356E+03	0.00	0.191E+02	4.00
4	0.451E+02	0.00	0.206E+00	0.00
5	0.579E+02	0.01	0.113E+02	4.00
6	0.108E+03	0.00	0.110E+02	4.00
7	0.457E+02	0.16	0.000E+00	0.00

Thus results of field measurements reveals that thickness of lateral shielding should be increased and narrow collimation angle should be enlarged to decrease influence of side background from contaminated walls of repository and other sources on statistical error of measurements of Co-60 contamination.

CONCLUSION

The following results on development of non-destructive measuring technique of the characteristics of radioactive contamination of near surface layers of concrete and ground were obtained and presented:

- A mathematical model of a radiometric instruments was developed and used in studying of a measurement technique and in optimization of its parameters, and all relationships required for the model implementation were obtained.
- The technique for measurement of characteristics of Cs-137 and Co-60 contamination in concrete was tried out. Based on results of processing instrumental radiation spectra recorded by a collimated scintillation detectors, this technique allows:
 - measuring the surface activity of Cs-137 and Co-60 in concrete and ground;
 - evaluating the Cs-137 and Co-60 penetration depth.
- The minimum and maximum measurable activity of these radionuclides in concrete was evaluated for radiometric measurements with collimated detectors of different volumes and sizes.
- One collimated detector system was manufactured for implementation and test of developed technique.

REFERENCES

1. Yu.F. Rodionov and N.P. Yashin, *VANT, ser: Nuclear-Physical Investigations (theory and experiment)* 12, (2), (1989), c.50
2. S.K. Lisin, N.P. Yashin, Yu.F. Rodionov, N.D. Smolov et. al. *VANT, ser: Nuclear-Physical Investigations (theory and experiment)* 13, (2), (1990), c.142
3. A.V. Chesnokov, V.I. Fedin, A.P. Govorun, O.P. Ivanov, V.I. Liksonov, V.N. Potapov, S.V. Smirnov, S.B. Shcherbak, L.I. Urutskoev. Collimated Detector Technique for Measuring a Cs-137 Deposit in Soil under a Clean Protected Layer. *Applied Radiation and Isotopes*, Vol. 48, No. 9, pp. 1265-1272, 1997.
4. A.P. Govorun, V.I. Liksonov, V.N. Potapov, L.I. Urutskoev, A.V. Chesnokov, S.B. Scherbak. The Method of Determining Contamination Density and Evaluation of the Depth of Cs-137 Penetration in Soil. *Atomnaya Energia*, Vol. 78, No.3, March 1995, pp. 199-204.
5. A.V. Chesnokov, A.P. Govorun, V.N. Fedin, O.P. Ivanov, V.I. Liksonov, V.N. Potapov, S.B. Shcherbak, S.V. Smirnov, L.I. Urutskoev, Method and device to measure Cs-137 soil contamination in-situ // *Nuclear Instruments and Methods in Physics Research. Section A*. Vol. 420, Nos. 1-2, pp. 336-344, 1999.
6. V.N. Potapov, S.M. Ignatov, V.M. Chirkin, V.G. Linnik. The Radiometric Method of Measuring Activity of Cs-137 Radionuclides in Bottom Deposits Using a Water Immersion // *Atomnaya Energia*, 2001, Issue 3, Vol. 9, pp. 216-222.
7. G. Korn, T. Korn. *Handbook of Mathematics for Research Workers and Engineers*. Moscow, Nauka Publishing House, 1970, 720 pages.
8. E.I. Grogoriev, E.K. Stepanov, V.I. Fominykh. Minimum Measurable Activity. The Notion and Quantities Used in Radiometry. - *ANRI*, 1994, No. 3, pp. 10-12.