Development of Portable Beta Spectrometer for Sr-90 Activity Measurements in Field Conditions and Its Application in Rehabilitation Activities at RRC Kurchatov Institute

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ABSTRACT

A new method to measure the Sr-90 ground specific activity in situ was developed. It is based on the count-rates determination in selected energy ranges of two registered apparatus spectra: total $\beta + \gamma$ spectrum and γ spectrum. A numerical simulation of the detector performance defined these energy ranges used for calculation of activity. For implementation of the proposed method a portable instrument was developed and manufactured. Parameters of the instrument are the following: the range of measurement for a specific activity mode - ($60 – 3.0x10^6$) Bq/kg; the range for total activity countable mode ($0.5 – 2.0x10^4$) Bq; minimum measurable specific activity Sr-90 for samples containing natural radionuclides - 60 Bq/kg, minimum measurable activity for samples not containing NRN – 0.5 Bq.

INTRODUCTION

The operations on rehabilitation of site of the temporary radwaste repositories (TRWR) at RRC Kurchatov Institute are carried in the frame of the project "Rehabilitation" [1]. The main tasks of the project are removing of radwaste (RW) and liquidation of old temporary repositories.

The composition of RW in repositories is determined now mainly by radionuclides Cs-137, Co-60 and Sr-90. As a result of operations there is a contamination of surfaces of underlying hardware, special vehicles, building mechanisms, transport means, protective building constructions and ground.

For support of the radiological safety for staff executing operations and urban population living near to Kurchatov Institute site, all technological operations with radwaste and the liquidations of repositories are carried out with application of technologies and resources for aerosol-suppression. For preventing the spreading of contamination the protective polymeric coatings were sprayed on dusty surfaces. For decontamination processing of engineering and transport means on boundary of inspected territory there was constructed the post of special processing and decontamination.

All operations are carried out with continuous radiological control, control of activity of aerosols in air at working areas, measurement of the contaminations level of different surfaces by radioactive substances, periodic sampling of a ground for detailed inspection in laboratory conditions and control of radiation conditions at site in general. During realization of such operations the necessity of Sr-90 contamination measurement of surfaces and ground in field conditions became very important, since in first, laboratory analysis of samples for activity Sr-90 measurement is time consuming and laborious process, and, in second, sampling is not always possible from surfaces, on which a polymeric sheeting were sprayed.

The portable beta spectrometer intended for measurements in field or laboratory conditions of Sr-90 activity in ground, air strainers and in other samples at presence of man-made radionuclides Cs-137 and Co-60 in samples was developed and these instrument and method of measurements are described in following section of manuscript.

THE DESCRIPTION OF BETA SPECTROMETER

A mathematical model of the detector was developed to calculate the apparatus spectra of registered γ- and β-radiation. It is based on the Monte-Carlo simulation of γ- and β- particles interactions in the ground, filter and in the sensitive volume of the detector. The model was used to find optimum geometry of detector (thickness of scintillator and passive light-guide). When detector prototype was developed the model served for search of optimum working-energy ranges in the measured spectrum for calculation of source activity with best accuracies and account the background radiation.

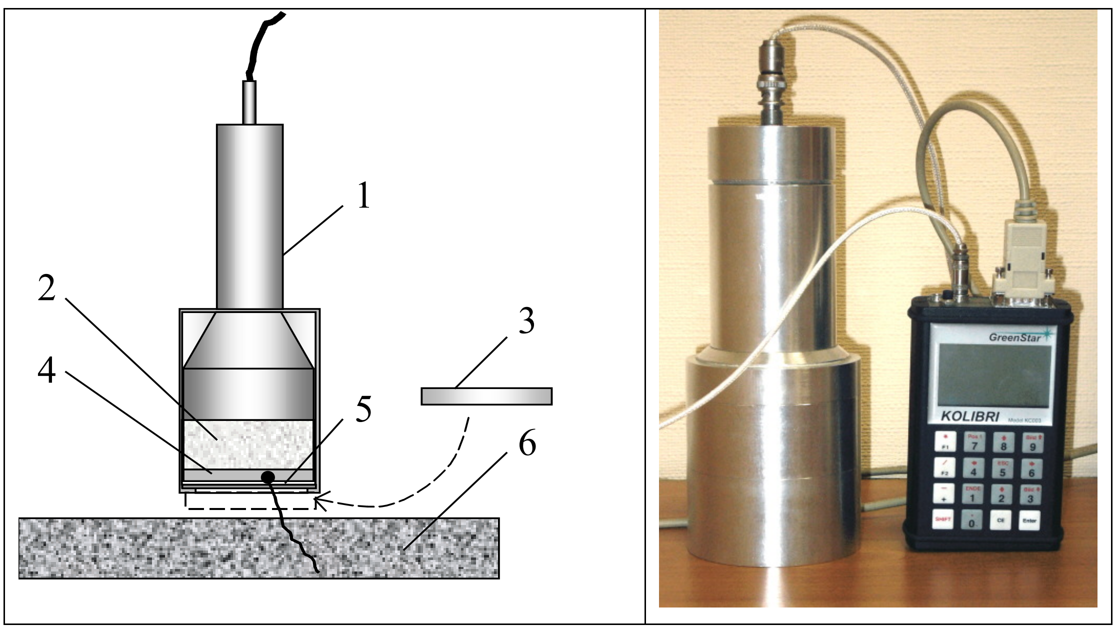

Fig. 1. Scheme of measurements and photo of the spectrometer

1. photo multiplier tube; 2. light-guide (inactive plastic); 3. β-particle (aluminum disk); 4. sensitive scintillating layer; 5. titanium foil; 6. contaminated ground

The scheme of the detector and photo of the instrument are shown in a Fig. 1. The gamma and beta radiation is registered by a plastic scintillating disk made of a slice of polystyrene (cylindrical disk of ∅80x3.5 mm) with sensitive area of ~43 cm^2. For equalization of detector sensitivity over its area (a photocathode of PMT has non-uniform light sensitivity) the optical light-guide made from a plastic (Plexiglas) is used. The light flash in a scintillator gives rise to a plenty of light quanta, which, passing through light-guide, get mixed up after multiple reflections from walls, losing the information on the birthplace of flash in a scintillating slice. Thus, the dependence of the value of a light signal on a location of β-particle interaction in the detector is eliminated, that improves energy resolution of the detector.

The plastic scintillator with thickness larger than 6 mm absorbs practically 100 % of Y-90 β-particles, uniformly distributed in ground. However thickness of a layer of a scintillator was selected equal to 3.5 mm. At such thickness of a scintillator the apparatus spectrum of Y-90 electrons is deformed in a high-energy part of the spectrum, being displaced on a energy scale to the area of smaller energies, therefore value of the right boundary of a working energy interval ΔE_e (selection of energy intervals is described in the next section) was accepted equal 2.0 MeV. As volume of scintillator is decreased (approximately by 42 %) the essential reduction of efficiency of registration of a γ-ray background has place.

The external surface of the detector is protected with a titanium foil of thickness 50 microns, which effect on a registered spectrum of β-radiation is very small. As a photo detector the photo multiplier tube of FEU-56 type is used. Energy resolution of the detector for E_β = 624 keV (energy of monoenergetic Cs-137 conversion electrons) is about 20 %.

For processing and storage of the spectrometric information the portable 2048-channel spectrum analyzer "Kolibri" is used [2]. At realization of field measurements the spectrometric information is archived with the help of the internal program of a spectrometer "Kolibri", and later is transmitted through a sequential data link RS-232 to the personal computer for further processing. Laboratory measurements with spectrometer "Kolibri" are carried out usually when spectrometer is connected via the serial interface (RS-232) to the personal computer, and the operation of spectrometer is controlled totally by the spectrometer-implementation program with user-friendly interface.

After measurements, with the help of special program running on PC, the processing of the spectrometric information, determination of Sr-90 specific activity and estimation of a statistical error of measurement is carried out.

The geometry of the measurement allows the determination of Sr-90 activity distribution in surface layers of ground. The detector has a low γ-efficiency. Nevertheless, the γ-rays still give the main contribution to the background spectrum because of the volumetric accumulation factor of γ-rays in ground (paths of β- particles in ground are a few millimeters, while the paths of photons can be tens of centimeters).

The filter (3 mm aluminum plate) is used for selection of the γ-ray component in the detected spectrum. The filter practically passes the photons of containing natural radionuclides (NRN) without loss and absorbs the β- particles completely. Thus, two measurements are carried out: with the filter (auxiliary measurement) and without the filter (main measurement). The β-component of the spectrum is obtained by subtraction of an auxiliary spectrum from the main spectrum. The typical spectra of γ- and β- components of NRN background calculated within the framework of model of spectrometer and measured in actual conditions with developed instrument are presented in a Fig. 2.

THE DEVELOPMENT OF FIELD METHOD OF SR-90 ACTIVITY MEASUREMENT

NRN and Cs-137 Background Account

For determination of Sr-90 activity from apparatus spectra the working energy range, which contains information on Sr-90(Y-90) radiation should be selected. The calculated and measured spectra (see Fig. 3) show that β-spectra of Sr-90 and NRN are intersected in wide energy range and at low level of Sr-90 activity the account of NRN background is very important.

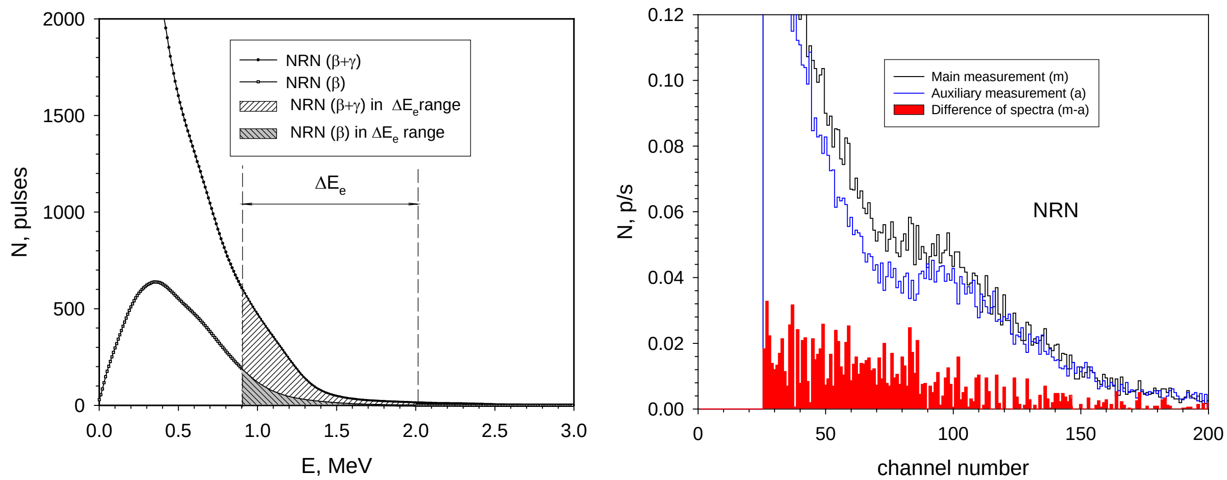

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Fig. 2. γ- and β-components of apparatus spectra of natural radionuclides (NRN)

A. simulated spectra, B. measured spectra, ΔE_e – working spectral energy range used for determination of Sr-90 specific activity.

In developed approach to NRN β - background account we use assumption (which is true for old contamination of the ground) that among man-made radionuclides, accompanying Sr-90 contamination, Cs-137 is dominant and consequently the γ-ray background with energy above 0.7 MeV is formed by NRN γ-radiation only [3,4]. Thus, to estimate the NRN β- background in total β-spectra one may use the correlation between the NRN component β- spectrum and their component γ – spectrum in the energy range above 0.7 MeV.

Assume that N_e is the count rate in a working energy range ΔE_e of the NRN β- background spectrum. It is formed by radiation of K-40 (count rate N^K_e), Th-232 (N^{Th}_e) and U-238 (N^U_e). N^{Th}_e and N^U_e include the radiation of all affiliated decays, which are in equilibrium with them. Thus $N_e = N^K_e + N^{Th}_e + N^U_e$.

If to denote by N^K_γ , N^{Th}_γ and N^U_γ the count rate of the photons, caused by radiation of the appropriate radionuclides for an energy range ΔE_γ in the spectrum of the NRN γ-background, the ratio will be given by

$N^K_e = \sigma^K N^K_\gamma$, $N^{Th}_e = \sigma^{Th} N^{Th}_\gamma$ and $N^U_e = \sigma^U N^U_\gamma$,

where σ^I (I =K, Th, U) are the proportionality factors between the specified values.

It is possible to find an energy range ΔE_γ for which

$$\sigma^U = \sigma^{Th} = \sigma^* \qquad \text{(Eq.1)}$$

In this case one can write

$N_e = \sigma^* (N^K_\gamma + N^U_\gamma) + \sigma^U N^U_\gamma$ or $N_e = \sigma^* (N^K_\gamma + N^{Th}_\gamma + N^U_\gamma) + (\sigma^U - \sigma^*) N^U_\gamma = \sigma^* N_\gamma + \Delta\sigma N^U_\gamma$,

where N_γ is the total count rate of the γ-ray component in the chosen range ΔE_γ.

If we assume that $N^U_\gamma = \langle N^U_\gamma \rangle + \Delta N^U_\gamma$, where $\langle N^U_\gamma \rangle$ is the count rate in the range ΔE_γ for U-238 specific activity in ground, which corresponds to the world average value of 26 Bq/kg, the following equation may be written:

$$N_e = \sigma^* N_\gamma + \Delta\sigma(\langle N^U_\gamma \rangle + N^U_\gamma) \approx \sigma^* N_\gamma + \Delta\sigma \langle N^U_\gamma \rangle$$

The error of this approximation does not exceed 12% for all soil types with the maximum deviations of the U-238 contents from the average world parameters. Thus, the determination of the Sr-90 (Y-90) specific activity A_{Sr} is possible as realization of the following computing procedure:

$$A_{Sr} = \{[N_{e\gamma}(\Delta E_e) - N_\gamma(\Delta E_e)] - [\sigma^* N_\gamma(\Delta E_\gamma) + \Delta\sigma \langle N^U_\gamma \rangle]\} \times C ,$$

where $N_{e\gamma}(\Delta E_e)$ and $N_\gamma(\Delta E_e)$] are the count rates in the working energy range ΔE_e of the spectrum measured for the main and auxiliary measurements, $N_\gamma(\Delta E_\gamma)$ is the count rate in the range ΔE_γ of the auxiliary spectrum (the ΔE_γ is selected so that (Eq. 1) is valid), C the calibration factor, which is equal to the reverse value of the count rate of β- particles for unit Sr-90 (Y-90) specific activity in the ground.

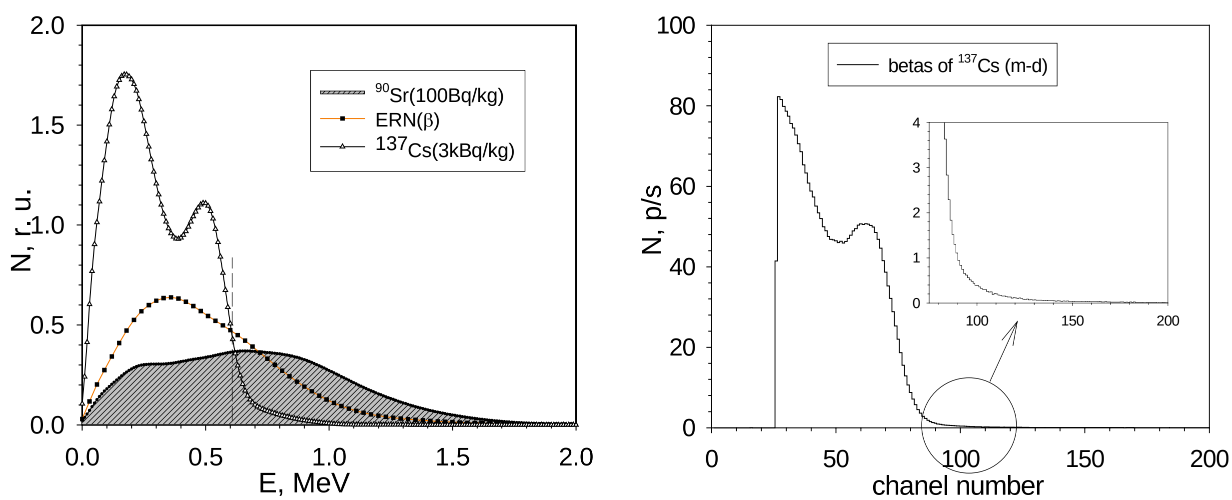

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Fig. 3. A. simulation of different components of β-radiation apparatus spectra of Cs-137, Sr-90 and NRN. B. experimental apparatus spectra of Cs-137 β-radiation

Main man-made γ- radiating radionuclides, presented at KI TRWR site and other places with old radioactive contamination are Cs-137 and Co-60. The radionuclide Cs-137 is β-emitter also. The information on apparatus β- spectra of Cs-137, Sr-90 and NRN, calculated by a Monte-Carlo method and obtained by measurements with developed instrument is represented in Fig. 3. As it is visible from this figure, because of availability of high-energy β-particles produced in Cs-137 decay (E_{max}=1.17 MeV, with an output per one decay ~5.6 %), the value of the left boundary of a working energy range should at least be higher than 0.6 MeV.

Using minimization of statistical error of measurement of Sr-90 activity at the presence of Cs-137 radiation the working energy interval was found as $\Delta E_e \in [0.9 \div 2.0]$ MeV. The energy interval for account of NRN ΔE_γ was selected as $\Delta E_\gamma \in [1.09 \div 2.0]$ MeV. For this interval the condition (1) is fulfilled.

The Account of Co-60 Background Radiation

It is necessary to point out that the beta radiation of Co-60 does not influence practically on measurement of Sr-90, since, in main, the energy of emitted by Co-60 β-particles is less than 0.3 MeV. The gamma rays of Co-60 influence on measurements in the same way as well as gamma radiation of Cs-137, but it happens in the range of energies of an apparatus spectrum below energy ~1.2 MeV. This energy corresponds the Compton edge of gamma quanta with energy of 1.33 MeV with account of its dispersion. Thus, the Compton edge overlaps on a working area of a spectrum, in which the Sr-90 activity is calculated. As at determination of Sr-90 activity two measurements main - without a filter and auxiliary - with a filter are carried out, formally, gamma background will be taken into account.

However at high activity Co-60 background components will increase considerably statistical noise that will be resulted in sharp increase of an error of Sr-90 determination. One of possible ways of exception of such situation is a shift of the left boundary of a working energy interval up to a level 1.2 MeV. The working area ΔE_e is reduced in this case, that also results in increase of a statistical measuring error. Therefore this transition should be carried out only in the event that the statistical measuring error at the expense of effect of irradiation Co-60 will be higher than a measuring error at the expense of offset of boundary of a working interval ΔE_e.

The experimental researches of effect of Co-60 radiation on results of Sr-90 measurement were carried out in laboratory conditions. The instrument was located above a cuvette with Sr-90 water solution of volumetric activity 1395±194 Bq/liter. Under the cuvette the point Co-60 source was allocated at miscellaneous positions.

The obtained experimental results are presented in the table I.

Table I. Experimental Test of an Account of Co-60 Background Radiation

N of measurements	A_{Sr}, Bq/kg	Measurements with Co-60	
		A_{Sr}, Bq/kg (I)	A_{Sr}, Bq/kg (II)
1	1320±70	1360±290	1260±190
2	1420±70	1380±290	1330±190
3	1430±70	990±290	1180±190
4	1380±70	1140±290	1210±190

The first column of measured data presents the values obtained without Co-60 source, the second and third - presented measurements for different layouts of Co-60 source. As it is visible from the table, the effect of Co-60 irradiation was become apparent firstly in change measuring error, which has increased in 3÷4 times.

If the studied sample has a non-uniform distribution of gamma radiating Co-60 radionuclides through t its volume, the case is possible, when the local Co-60 source is on a surface of a researched sample near the edges of the detector. In this case γ components of Co-60 in spectra of main and auxiliary measurements will differ essentially (because of strong weakening of radiation in a filter plate at oblique falling of gamma quanta registered by the detector), that will reduce in a noticeable methodical error of measurement Sr-90.

Besides this, the radionuclide Co-60 is also beta emitter. With a noticeable output per decay for radionuclides Co-60 the interest represents beta particles with maximum energy 0.318 MeV (~100 %) and 1.49 MeV (0.12 %). The beta particles with energy 0.318 MeV have almost 100 % probability of output per decay, but at the working energy ranges used in the developed technique, they do not produce any effect on measurements. The beta particles with energy 1.49 MeV (their output per decay is 0.12 %) are not essential only for small activities of sources, and when the Co-60 source activity is high in relation to Sr-90 activity ($A_{Co}/A_{Sr} > 10^3$), their effect cannot be neglected.

In experiments with a point source Co-60, located close to a surface of the detector, the considerable proportion of a difference spectrum hits in working power area, on which the rating of activity Sr-90 is carried out. To exclude this uncompensated portion of the spectrum, it is necessary also to shift the left boundary of a working energy interval up to value 1.2 MeV.

In spite of the fact that such situation arises extremely seldom, the procedure of its registration is incorporated in an algorithm for processing of observed data, which select the working area in an apparatus spectrum automatically. It is carried out as follows. The specific activity Sr-90 is evaluated for two energy intervals - ΔE_e=[0.9÷2.0] MeV and $\Delta E'_e$=[1.2÷2.0] MeV. If the values of the obtained Sr-90 specific activities coincide in limits of statistical measuring errors, the first interval is selected as the working interval, otherwise - second. The incongruity of values Sr-90 on these intervals just also is stipulated by effect of beta radiation and uncompensated gamma radiation of Co-60.

The calculations have shown, that the value of minimum measurable activity (MMA) [5] of strontium by developed method is 60 Bq/kg in case of absence of other man-made radionuclides in sample and is ~75 Bq/kg - at availability in ground only of Cs-137 radionuclide with a specific activity up to 100 kBq/kg. The presence of Co-60 radionuclide of high activity in ground (that is rare, but possible case) results in increase of MMA value approximately in 2 times up to ~160 Bq/kg. The MMA evaluation was conducted also experimentally by measurements in laboratory and field conditions and has shown good correspondence to calculated values. Parameters of the instrument are the following: the range of measurement for a specific activity mode - (60 – $3.0x10^6$) Bq/kg; the range for total activity countable mode (0.5 – $2.0x10^4$) Bq; minimum measurable specific activity Sr-90 for samples containing natural radionuclides – 60 Bq/kg, minimum measurable activity for samples not containing NRN – 0.5 Bq.

SR-90 SPECIFIC ACTIVITY MEASUREMENTS IN GROUND AT RADWASTE REPOSITORIES SITE OF KI

Two types of the objects are considered for measurements. First type, objects, in which attenuation of beta particles beam is negligible - small air strainers, smears from the contaminated surfaces, and other thin objects. Thus outcome of measurement is the gross activity. The second type of objects are the objects with volumetric contamination, about which it is possible to assume, that their contamination is homogeneous throughout the object or, at least, on several free paths for beta particles near the object surface. Such objects are the stratums of ground, water solutions or dredges. In this case a measured value is the specific activity of nuclide in surface layer (or in object, if the supposition about homogeneous contamination is reasonable). The measurements of contaminated ground at waste repository site are one of important application of developed instrument.

The activities on liquidation of old RW repositories are carried out on territory of RRC Kurchatov Institute site. The main part of these repositories represents concrete trenches in ground filled with solid WR. After extract of the RW from repositories the measurements of residual contamination of their bottom and walls are carried out. For example, with the help of the developed instrument the part of bottom of repository #6 was inspected. This part had a ground basement without a concrete coating. The dimensions of area of realization of measurements were 4 x 10 m. The observed data of Sr-90 specific activity distribution in surface layer of a ground at the bottom repository are shown in a Fig. 4. The radiometric survey was carried out on a rectangular grid with step 1 x 2.5 m. The exposure time was selected to be equal 180 seconds. As it is visible from the figure, the levels of Sr-90 specific activity achieve in some isolated points of the considerable values (up to ~10^5 Bq/kg).

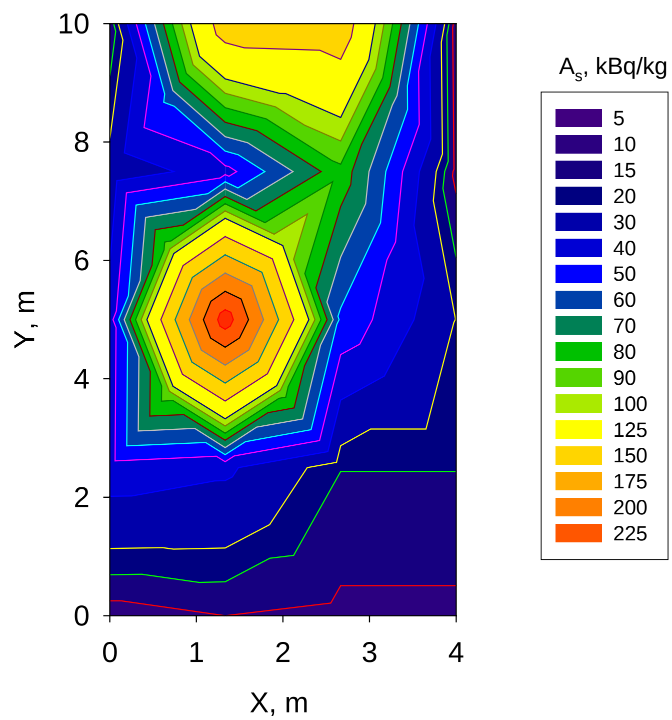

Fig. 4. The distribution of Sr-90 specific activity in the ground at the bottom of repository number 6

CONCLUSIONS

Thus, as a result of the theoretical analysis and laboratory researches the method of measurement and the instrument for measurement of Sr-90 specific activity in ground, both in field conditions and in laboratory have been developed. The measurement technique is based on usage of a method of a radiometry beta and gamma radiations of radioactive nuclides.

The measurements are carried out directly with object of investigation (ground, contaminated surfaces of structures and mechanisms) without usage of any shielding.

Parameters of the instrument are the following: the range of measurement for a specific activity mode - (60 - 3.0×10^6) Bq/kg; the effective range of measurable activity in counting model (0.5 - 2.0×10^4) Bq; minimum measurable specific activity (MMA) of Sr-90 for ground samples containing natural radionuclides (NRN) - 60 Bq/kg, minimum measurable activity for containing samples not containing NRN - 0.5 Bq. Is shown, that availability the gamma and beta man-made radionuclides in studied samples restricts MMA of developed method and this value is not less than ~75 Bq/kg.

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