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ON THE DEVELOPMENT OF REQUIREMENTS FOR MEASUREMENT OF RADIATION CHARACTERISTICS OF SOLID RADIOACTIVE WASTE GENERATED DURING DECOMMISSIONING

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The paper summarizes the practical experience associated with characterization of radioactive waste emplaced into primary packaging gained during decommissioning of nuclear power facilities at JSC VNIINM site. The paper analyses and suggests some measures for arranging and implementing the measurement of solid radioactive waste (construction waste, plastic, fragments of equipment, personal protective equipment and soils) radiation characteristics.

Key words: *measurement requirements, measurement procedure, solid radioactive waste, radionuclide activity, decommissioning*

Nuclear decommissioning results in waste generation. Such waste is RW of different classes composed of various materials cleared from regulatory control [1, pp. 5—7]. Clearance from regulatory control suggests that materials or items containing radioactive substances are released from any further regulatory control under permitted practical activities [2, 3]. Waste are assigned to a particular category based on relevant measurements of their radiation characteristics. Correct measurements of radiation characteristics are believed to be of great importance since waste management costs depend substantially on waste category (industrial waste contaminated with technogenic radionuclides, VLLW, LLW, ILW, HLW) [4].

Papers [3, 5, 6] discuss the requirements associated with performing measurements of waste radiation characteristic and relevant procedures.

The study of the procedural framework has shown that almost no standardized procedures for measuring radionuclide specific activity using the methods of alpha, beta, gamma spectrometry and radiometry complying with modern requirements are available at nuclear sites [5]. Oftentimes, relevant requirements on quality control are missing or no measurement error characteristics depending on the subranges of the measured values are set forth. Moreover, no standard measurement procedures are being stipulated in the industry register. Whereas these should explain on the way the samples of different morphology should pre-treated before the measurements take place and the influence of environmental parameters (temperature, humidity, pressure) should be assessed during such measurements. This is viewed of particular importance when performing in situ measurements with non-destructive testing methods (for example, by applying mobile gamma-spectrometers) requiring no preliminary treatment of the samples.

Practice has shown that with no specific procedures set forth for radiation characteristics measurements associated with waste from nuclear decommissioning and regulations on the measurement process during nuclear decommissioning, errors can occur when assigning waste to a particular category based on the measurements performed.

The paper summarizes the experience of JSC VNIINM in dealing with the issues encountered when identifying radiation characteristics of packages with solid radioactive waste from nuclear decommissioning.

JSC VNIINM experience

JSC VNIINM was established at the dawn of the nuclear industry. Therefore, a number of facilities are available at the site that are to be decommissioned.

To date, decommissioning efforts were completed for VNIINM's Reagentnaya building and research unit B. In 2017, decommissioning of U-5 installation was started. These activities enabled to gain some knowledge and to test in practice some methods and techniques enabling the characterization of the generated SRW, namely, construction waste, plastic compound, equipment fragments, PPE, soils. ^{60}Co, ^{90}Sr, ^{134,137}Cs, ^{239,241}Pu, ^{234,235,238}U and ^{241}Am appeared to be the key radioactive contaminants.

To date, existing regulations provide no uniform requirements to the flow chart of activities performed during decommissioning of nuclear and radiation hazardous facilities in terms of measuring the radiation characteristics of generated SRW. At the same time, the retrieved solid radioactive waste transferred to disposal should meet general acceptance criteria for disposal set forth in federal norms and rules [7]. General RW acceptance criteria for disposal are established to ensure safe disposal of particular class RW and predefine the requirements considered as sufficient for waste transfer to the National Operator for RW management. It is considered impossible to determine the compliance of SRW characteristics with waste acceptance criteria without correct radiation characteristic measurements.

SRW radiation characteristic measurement involves the identification of waste radionuclide composition, as well as of specific and total activity of radionuclides contained therein. In keeping with IAEA recommendations [8], verification of the identified radiation characteristics should be based on a procedure involving: *in situ* measurements by means of material's nondestructive testing, laboratory measurements featuring representative samples, the use of properly derived radionuclide ratios (radionuclide vector method), adequate traceability of the material, including its origin.

Prior to decommissioning, measurement methods should be selected and suitable instruments should be chosen and calibrated based on the data discussing the opportunities for:

- grouping the materials under consideration so that they are as homogeneous as possible in terms of the material itself and its origin (and, thus, radionuclide composition and activity level);
- assessment of the radionuclide composition of the considered materials — sample evaluation taking into account all relevant information on the material's background and origin.

For primary waste from decommissioning, radioactive waste should be collected, analyzed and segregated into categories (in situ) according to waste physical, chemical, biological and radiological properties. These activities facilitate further treatment of the waste enhancing its efficiency, decreasing the amount of end waste amounts sent for storage and disposal. Overall strategy for segregation and separate collection of waste is set forth in regulations and depends on the current radioactive management system in place. Such system determines the main categories of waste, waste treatment, conditioning and disposal methods [8, page 29]. Oftentimes, decommissioning involves mixing of waste with different morphology during the package filing, resulting in significant difficulties impeding reliable measurement of RW radiation characteristics.

To measure waste activity, appropriate measuring tools should be available, proper skills in applying relevant methods of direct and indirect measurements, sampling or sample preparation are required. This is viewed of special importance when decontaminated facilities, equipment or materials are recategorized as non-radioactive waste: the lower are the control levels, more comlex are the measurements, whereas, very low activity level of potentially clean or cleared materials should be demonstrated by relevant measurements.

Direct activity measurements suggest the use of various detectors of ionizing radiation located in the immediate vicinity of the measured item. Indirect activity measurements involve the identification of non-fixed or weakly fixed contaminant content with a smear taken from a surface (contaminant content is identified by direct measurements indicated by an instrument) [8, p. 30].

Reliable measurement of SRW radiation characteristics directly inside waste packages (containers) is possible only for gamma emitting radionuclides with high gamma ray energies, as low energy gamma radiation, alpha and beta radiation is almost completely absorbed by container walls and waste material.

Difficulties associated with SRW categorization by radionuclide specific activity in packages (containers) are mainly due to the following:

- poor accuracy (increased uncertainty) in determining the efficiency of detecting gamma-quanta of various energy when calculating radionuclides' specific activity due to uneven distribution of SRW and radionuclides in a package;

- high value of the minimum detected activity of radionuclides with low energy gamma radiation (for example, ^{241}Am Eγ = 59.5 keV) due to absorption of radiation by container walls and waste materials;
- presence of hardly detectable radionuclides. The decay of such radionuclides proceeds with gamma radiation emission the intensity of which is too low to enable its recording by non-destructive measurement methods (for example, $^{239-241}$Pu, ^{234}U, ^{90}Sr);
- errors at segregation and packaging stages resulting in SRW packages of mixed morphology;
- absence of unique requirements on relevant work flows implemented during decommissioning in terms of measuring radiation characteristics of generated SRW.

Preliminary categorization of SRW generated from decommissioning of U-5 facility was performed by measuring waste radiation characteristics and evaluation of gamma spectra emitted by SRW in large containers (3.1 m^3, steel wall thickness 5 mm). To reduce the heterogeneity effect of radionuclide activity distribution in SRW, measurements were carried out using a portable gamma-spectrometer at each side of the container with further averaging of the results obtained. Efficiency of gamma-quanta registration required to calculate the specific activity was evaluated only depending on the container parameters and the density of waste materials without taking into account the morphological composition. As the result, minimum detectable activity (MDA) of individual radionuclides exceeded 10—100 times their minimum significant activity (MSSA) (for example, based on MDA measurements for ^{241}Am — over 10^5 Bq/kg) due to the absorption of X-ray and gamma-radiation emitted by radionuclides at the container walls and in waste material. SRW categorization under such conditions proved to be impossible.

It should be noted that in the industry-wide technique registry and federal information fund only one formal technique is documented enabling to measure the specific activity of gamma-emitting radionuclides immediately inside SRW packages. Moreover, this technique is applicable to packages with a volume of no more than 400 liters. Methods for SRW radiation characteristic measuring for larger containers are not available.

The following measure were implemented to ensure accurate measurements and decrease the MDA during the identification of radionuclide specific activity:

- the volume of measured SRW packages was decreased to 200 liters;
- library of radionuclides was adjusted, based on the expected nature of SRW contamination;
- to reduce the effect of heterogeneity of radionuclide activity distribution in SRW the, measurements were performed at a rotating table;
- increased was the time of gamma-emitting radionuclides spectrum widening.

These efforts enabled to measure the specific activities of the main gamma-emitting radionuclides, ^{137}Cs and ^{241}Am with MDA level being not higher than the MSSA one and to categorize SRW packages.

It should be noted that U-5 facility was designed to test the technological process associated with extraction and purification of plutonium fission products resulted from uranium irradiated in nuclear reactors. Thus, structural materials of the building and installation, as well as the laboratory equipment was also contaminated with hardly detectable isotopes of uranium, plutonium and ^{90}Sr.

Relevant measurements of specific activity for such radionuclides is possible only by applying laboratory methods of destructive control and/or the method of “radionuclide vector”.

Most of the existing methods enabling to determine the activity of hardly detectable alpha- and beta-emitting radionuclides are based on the use of labor-intensive techniques with radiochemical isotope extraction, electro-precipitation of isotopes on targets for alpha-emitters and subsequent radiometric and/or spectrometry measurements requiring on top of all highly qualified staff.

Higher efficiency of RW characterization containing hardly detectable alpha and beta-emitting radionuclides is only possible by joint application of destructive and non-destructive control methods [9]. At the same time, sampling, pre-treatment and analysis of samples is carried out in the course of a special study enabling to identify relevant correlations between measured radionuclides activities (including alpha- and beta-emitters). Whereas, routine RW control is performed by non-destructive methods using the established correlation ratios or dependencies. This RW radiation monitoring approach was called “radionuclide vector method” (nuclide-vector) or “scaling-factor” method. The radionuclide vector method is used to characterize radioactive waste in the majority of most nuclear developed countries and is discussed in IAEA NW-T-18 [10] and ISO 21238: 2007 [11].

Its application significantly reduces the labor input for the measurements associated with extensive studies, but requires preliminary laboratory measurements to identify the specific activity of alpha, beta and gamma emitting radionuclides, which is necessary to select reference radionuclides and calculate correlation coefficients.

JS-spectrometry suggesting the use of purpose developed software to process radionuclide spectra is viewed as a modern laboratory destructive control method enabling quick analysis of hardly detectable radionuclides (alpha- and beta-emitting). This method suggests "screening" requiring no radiochemical pre-treatment of the sample or requiring their minimal simplified pre-treatment. This method applied jointly with semiconductor gamma spectrometry allows to address this issue [12—14].

Suggested solutions and conclusions

Considering all the abovementioned facts, the need for introducing appropriate regulations for radiation characteristics measurements appears to be quite evident. It seems true both for the measurements engaging SRW generated from nuclear legacy decommissioning and the development of unified requirements associated with the radiation characteristics measurements for SRW in packages (containers) of various size. The latter one will enable the issuance of appropriate certificates following relevant non-destructive and laboratory control. With the requirements to the measurements introduced in form of standard procedures, the probability of making correct decisions on SRW categorization (industrial waste contaminated with technogenic radionuclides, VLLW, LLW, ILW) is believed to be higher. Availability of such a regulation system will enable more accurate cost evaluation associated with decommissioning activities in terms of waste characterization.

Based on the knowledge gained during radiation characteristics measurements performed for SRW packages during U-5 facility decommissioning and evaluation of the flow chart framework, a regulation for SRW radiation characteristics measurements is proposed to be developed. Such framework would involve some unified requirements to the flow chart of activities performed under nuclear decommissioning as part of SRW radiation characteristics measurements. Another task proposed to be addressed is the development of standard procedures to be followed during the measurement of radionuclide specific activity.

These are believed to be of a great need considering the fact that according to the estimates, until 2022 nuclear decommissioning activities performed at VNIINM site alone (URZ № 2, 9, U-5 facility, building A, E, Zh and KhDM) will generate over 3,000 m^3 of SRW of various morphology (construction waste, plastic compounds, fragments of equipment, PPE, soils) contaminated with technogenic radionuclides including those deemed as hard detectable ones. According to similar estimates performed for other enterprises, the amount of SRW generation from nuclear decommissioning will amount to some hundred thousand cubic meters.

Regulation and standard procedures are developed based on the evaluation of literature sources, legal framework in field of atomic energy use, international and Russian RW management practices. The following aspects will be accounted for during their development:

- required characteristics of measuring equipment based on the initial data on physical parameters, SRW radionuclide composition and specific activity, parameters of the package;
- impact of the measured item parameters (geometry, density, composition), environment (temperature, humidity, pressure) and measurement time on the metrological characteristics of the measurement results (error, minimum detectable activity, accuracy of radionuclide identification) both for in situ measurements and for the laboratory methods.

Standard procedures proposed for development include those associated with the measurement of radionuclide specific activity by *in situ* and laboratory methods accounting for sampling procedures (sampling sites, amount, volume and size of the material fragment, the area of the smear surface) as it comes to SRW in form of construction waste, plastic compounds, equipment fragments, PPE, soils:

- gamma-emitting radionuclides in samples of SRW and SRW held in packages and standard containers having a volume of up to 3.1 m^3,
- uranium, plutonium, and strontium isotopes by the JS-spectrometry method in SRW samples,
- hard-to-detect radionuclides (^{90}Sr, $^{234,238}U$, $^{239,241}Pu$) in SRW identified by means of "radionuclide vector" method.

The standard procedures shall comply with the State Corporation Rosatom's standard OST 95 10351 General requirements to measurement procedures [6], order of the State Corporation Rosatom of October 31, 2013 № 1/10-NPA [5].

Development of the abovementioned regulation will enable to establish unified requirement to conditions and methods applied to measure the radiation characteristics of SRW (present in form of construction waste, plastic compounds, equipment elements, PPE, soils) generated from nuclear decommissioning activities. Application of standard procedures will enable to increase the probability of making correct decisions on SRW categorization.

Provisions of the documents developed can be extended to other facilities subject to decommissioning including the development of a unified approach to the characterization process for SRW generated from nuclear decommissioning.

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