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APPROACHES TO THE CHARACTERIZATION OF RW GENERATED DURING IMPLEMENTATION OF THE PROJECT «PRORYV»

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Methods of determining the content of difficult to measure radionuclides and fissile materials in radioactive waste that will be formed during the operation of the Pilot Demonstration Energy Complex (PDEC) created within the framework of the «Proryv» project are considered. Taking into account the planned characteristics of PDEC radioactive waste, the composition of the software and hardware complex for the PDEC radioactive waste characterization is proposed.

Keywords: *difficult to measure radionuclides, fissile materials, radioactive waste, non-destructive monitoring method, destructive monitoring method, computational method, nuclide vector.*

Relevance of the task

Pilot Demonstration Energy Complex (PDEC) demonstrating the nuclear fuel cycle closure is currently under construction at the site of Siberian Chemical Combine in the framework of "Proryv" project. PDEC includes a lead cooled fast reactor BREST-OD-300, a module for nuclear fuel fabrication and refabrication and a module for spent nuclear fuel reprocessing.

The site-centered nuclear fuel cycle of BREST-OD-300 provides nearly unlimited extension of the resource basis of nuclear power by inclusion of U-238 and recycled Pu in a mixed equilibrium uranium-plutonium fuel.

PDEC construction and start-up is planned in 4 stages. At the first stage the fabrication module and start-up batch of the complex of refabrication of dense mixed uranium-plutonium fuel are commissioned (MFR). At the second stage a nuclear power plant unit with BREST-OD-300 reactor is constructed and commissioned. The third stage includes construction of a spent nuclear fuel (SNF) reprocessing module, which would transfer reprocessing products to MFR, and radioactive waste (RW) management facilities. At the fourth stage MFR is converted to manufacture of nuclear fuel from BREST-OD-300 reactor SNF reprocessing products.

One of the most important elements affecting the safety and economics of PDEC functioning is management of RW generated. RW generation will start with commissioning of the first batch of PDEC. At the same time, the specific features of PDEC, which is a system of a reactor and on-site nuclear fuel cycle, leads to generation of a wide range of radioactive waste in terms of morphology, physical and chemical properties, radionuclide composition and specific activity.

According to the federal legislation [1, 2] the generated RW shall be brought into compliance with acceptance criteria and transferred for disposal to FSUE "National operator for management of RW" (NO RW). The RW being transferred shall have a

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passport describing the main parameters of the waste, including the radiation parameters determining the class of RW: radionuclide composition and level of specific activity. List of radionuclides subject to inclusion in the RW passport is determined in agreement with NO RW [3]. Though the procedure of determination and agreement of the list of radionuclides with NO RW is not defined, it is evident that the list shall include all radionuclides relevant from the point of view of long-term disposal facility safety.

Identification of radionuclides present in the RW requires the use of a variety of instruments and methods: the wider the list of controlled radionuclides, the more complex is the procedure of RW characterization.

A system of RW passportization shall be developed before the commissioning of PDEC and start of RW generation. The first stage of works shall be identification of radioactive waste characterization methods and justification of the requirements to the elements of RW passportization system.

List of radionuclides in PDEC RW

Operation of nuclear fuel MFR, lead-based liquid-metal cooled fast reactor, spent nuclear fuel reprocessing module will lead to generation of various RW flows, including fragments of spent fuel assemblies (SFA), waste of pyrochemical SNF reprocessing, vitrified high-level waste (HLW) produced by hydrometallurgical processes, filters of gas purification systems, cement compound produced in cementation of liquid radioactive waste, pressed and non-reprocessible solid radioactive waste (SRW) and other categories of waste ranging from high-level waste to very low level waste.

An approximate list of radionuclides present in RW of PDEC and subject to control in process of characterization is given Table 1. The list includes radionuclides that may be potentially present in PDEC RW flows and have half-life periods of over 100 days. Such a threshold value was chosen out of the fact that the activity of radionuclides with half-lives under 100 days will drop by more than 10^{11} times over the allowed intermediate storage time of 10 years, thus making all RW including high-level waste, non-radioactive waste with regards to the content of these radionuclides.

Requirements to the PDEC technology imply that presence of uranium isotopes and trans-uranium elements in RW flows shall be minimized. Therefore, there is a problem of accurate identification of the content of fissile materials in RW along with radionuclide activation products of construction materials ((Fe-55, Co-60, Ni-63, Nb-94 etc.) and nuclear fuel fission products (Sr-90; Tc-99; Ce-144; Cs-134; Cs-137 etc.).

Information shown in Table 1 shows that only a part of radionuclides (such as Mn-54, Co-60, Nb-94, Cs-134, Cs-137) are gamma-emitters with

Table 1. Recommended list of radionuclides for characterization of PDEC RW

Item	Radionuclide	$T_{1/2}$, years	Main type of radioactivity
1	H-3	12.3	β^- (E_{max} = 18.6 keV)
2	C-14	$5.73 \cdot 10^3$	β^- (E_{max} = 156 keV)
3	Mn-54	0.85	γ (834.8 keV)
4	Fe-55	2.7	X (5.9 keV), X (6.5 keV)
5	Co-60	5.27	β^- (E_{max} = 318.2 keV), γ (1173.22 keV), γ (1332.51 keV)
6	Ni-59	$7.5 \cdot 10^4$	X (6.93 keV), X (7.65 keV)
7	Ni-63	100.1	β^- (E_{max} = 66.9 keV)
8	Sr-90	28.9	β^- (E_{max} = 546 keV)
9	Zr-93	$1.53 \cdot 10^6$	β^- (E_{max} = 90 keV)
10	Nb-94	$2.03 \cdot 10^4$	γ (702.6 keV), γ (871.1 keV)
11	Tc-99	$2.13 \cdot 10^5$	β^- (E_{max} = 293.5 keV)
12	Ru-106	1.01	β^- (E_{max} = 39.4 keV), γ (511.9 keV) γ (621.9 keV), γ (1050.4 keV),
13	Ag-110m	0.68	β^- (E_{max} = 83.7 keV), γ (657.8 keV) γ (884.7 keV), γ (937.5 keV),
14	Sb-125	2.77	γ (427.7 keV), γ (600.6 keV) γ (635.9 keV)
15	I-129	$1.57 \cdot 10^7$	β^- (E_{max} = 149 keV), X (29.8 keV), X (29.5 keV), X (33.6 keV), γ (39.6 keV)
16	Cs-134	2.06	γ (604.7 keV), γ (795.9 keV) γ (569.3 keV)
17	Cs-135	$2.3 \cdot 10^6$	β^- (E_{max} = 209 keV)
18	Cs-137	30.17	β^- (E_{max} =1173 keV), γ (661.6 keV),
19	Ce-144	0.78	γ (133.5 keV), γ (80.1 keV) γ (696.5 keV), γ (2185.7 keV)
20	Eu-154	8.8	β^- (E_{max} =1855 keV), γ (123.1 keV), γ (1274.5 keV), γ (723.3 keV)
21	Eu-155	4.96	β^- (E_{max} = 147.4 keV), γ (86.6 keV), γ (105.3 keV)
22	U-234	$2.5 \cdot 10^5$	α
23	U-235	$7.04 \cdot 10^8$	α; γ (143.8 keV); γ (185.7 keV);
24	U-238	$4.47 \cdot 10^9$	α; γ
25	Np-237	$2.14 \cdot 10^6$	α; γ (29.4 keV); γ (86.5 keV);
26	Pu-238	87.7	α
27	Pu-239	$2.41 \cdot 10^4$	α
28	Pu-240	6561	α
29	Pu-241	14.29	β^- (E_{max} = 20.78 keV)
30	Pu-242	$3.75 \cdot 10^5$	α
31	Am-241	432.6	α; γ (59.5 keV); γ (26.3 keV);
32	Cm-242	0.45	α
33	Cm-243	29.1	α
34	Cm-244	18.1	α

gamma-radiation energy and intensity sufficient for monitoring using gamma-spectrometric methods used in standard RW characterization techniques. The remaining radionuclides are either alpha-, beta- or X-ray emitters or their decay either does not lead to gamma emission or the intensity of the gamma-line has insufficient energy and/or intensity for reliable recording in process of non-destructive RW characterization against the background radiation of other radionuclides. Such radionuclides were named difficult-to-detect in international practice. At the same time, radionuclides readily detectable by gamma-spectrometry were named easy-to-detect.

Application of a number of methods, including instrumental and calculation methods is required to measure the complete list of radionuclides potentially present in PDEC RW.

Methods of radionuclides measurement in PDEC RW

Three groups of radionuclide measurement methods may be distinguished in RW:

- non-destructive methods (gamma-spectrometry, neutron methods, calorimetry);
- destructive test methods;
- calculation methods (based on radionuclide vector methodology or mathematical modeling methods).

National regulatory documents impose a number of requirements on the procedure of determination of radionuclides content in RW. First, the measurements need to be carried out using calibrated measurement instrumentation of an approved type. Second, the measurement methods used shall be certified by a certified organization. Third, both experimental (instrumental) and calculation measurement methods are eligible. At the same time, calculation methods shall be based on the results of preliminary direct and (or) indirect measurements of the controlled parameters [3, 4].

Non-destructive radionuclide monitoring methods

Non-destructive gamma-spectrometric method has found wide use in RW characterization due to its quick response and absence of laboursome sampling and sample preparation procedures. The method implemented in standard RW characterization installations based on HPGe or scintillation detectors provides remote measurements of such radionuclides as Mn-54, Co-60, Nb-94, Cs-134, Cs-137, Ce-144, Eu-154 in PDEC RW packages. A number of radionuclides decaying with emission of fairly high gamma-radiation energy and/or intensity may also be detected in some RW flows. Mathematical modeling performed using "Gamma PPD professional" software for gamma-spectra of SRW packages planned to be generated after the commissioning of PDEC, showed that for specific RW flows gamma-spectrometry is capable of identifying the quantity of Np, AM, Pu, Cm isotopes in the RW based on characteristic gamma-lines. However, in most cases, the presence of other gamma-emitting radionuclides and location of RW inside protective containers shielding the radiation, lead to minimum detectable activity values for fissile radionuclides, which prevent reliable RW characterization and identification of the need for additional extraction of fissile materials (FM) from the monitored object.

Neutron methods based on recording neutrons emitted in spontaneous fission (fission caused by an external neutron source or alpha-capture reactions (α, n) are used for determining the quantities of FM (uranium, plutonium, curium isotopes, etc.) in RW.

Passive, active and active-passive neutron methods are distinguished. Passive methods are based on recording spontaneous fission neutrons. Therefore, they are capable of monitoring only the radionuclides susceptible to spontaneous fission. Spontaneous fission is mainly characteristic for even plutonium isotopes Pu-238, Pu-240, Pu-242. For most practical applications the spontaneous fission rates of U-235, U-238 and odd Pu isotopes are too low to detect. Passive methods are widely used to monitor the even plutonium isotopes in samples of various geometry and dimensions.

However, interpretation of results of such measurements requires a priori information on the radionuclide composition and the ratios of fissile materials in RW, as these methods do not distinguish contributions of specific radioisotopes.

Active neutron methods are based on irradiation of the controlled objects by neutrons from an external source. External neutrons cause induced fission of fissile materials, which is recorded by counting the neutron coincidences. Induced fission caused by external neutron source may be used to control U-235 isotopes and odd plutonium isotopes (Pu-239, Pu-241). This task requires using a source of neutrons with energy lower than the fission threshold for even-even isotopes (U-238, Pu-240)

Experimental verification of the possibility of detecting fissile materials in PDEC RW was carried out in the framework of the research. RW simulants were manufactured for the study, comprising of waste of various morphology characteristic for PDEC (metal waste, rags, crushed glass, crumbled concrete, bricks) packed inside standard 200-liter drums and containing radionuclide uranium, curium and plutonium sources. The fissile materials were monitored using a modified passive method of fast coincidences of neutrons with analysis of the time spectrum.

The following values were obtained for the minimum detectable masses per 200 l drum in the experiment — 200—300 mg for plutonium, 1—2 g for uranium and 0.5 μg for curium. The obtained values corresponded to the values found in literature, which generally range from 0.1 g to several hundreds of

grams. The paper [5] by FSUE RIA reported that the active method FM quantity measurement provides highly sensitive recording of FM responses in sealed containers. For example, the following minimum detected masses values were found for 8—15 minutes of measurements of a 70-liter container with various matrix fillings with density ranging from 0.32 to 1.6 g/cm^3 and neutron generator flux of about $5 \cdot 10^8$ neutron/s: (3 ± 1) mg for graphite matrices and (32 ± 11) mg for iron matrices.

Active neutron methods and recording of delayed neutrons are planned for further research and development works on construction of a software and hardware system for FM detection in process of PDEC RW characterization. It is planned that the system will include a neutron generator based on d-t reaction with neutron generation of at least 10^9 s^{-1} and He-3 based neutron detectors.

Detection of fissile materials would be based on recording delayed neutrons over a single or repeated 10 second interval after irradiation of the controlled object by fast and thermal neutrons over 20—40 seconds.

Calorimetric methods are widely used for passive non-destructive analysis of nuclear materials. Calorimetric method is based on activity assessment derived from measurement of heat produced in process of radionuclides decay.

The advantages of calorimetry are that the measurement results are independent of sample geometry and morphology, or distribution of fissile materials inside the sample. However, calorimetry methods are unable to provide information on sample activity if no a priori information on radionuclide composition and ratios between specific activities of individual radionuclides is available. Therefore, this method may be recommended for characterization of PDEC RW only as an element of additional verification of correctness of measurements and calculations.

There are alternative non-destructive methods, which have not yet found wide use, e.g. the method based on irradiation of controlled objects by intense fluxes of high-energy gamma-quanta causing nuclei photo-fission processes, followed by recording of fission neutrons using neutron methods.

Destructive radionuclide monitoring methods

Destructive test methods are based on sampling of a representative quantity of materials from the RW package being characterized, followed by preparation of counting samples using various radionuclide separation methods, and measurement of specific radionuclide activities in the samples.

Destructive monitoring gives most accurate measurements of radionuclide activities in RW. However, due to the complexity and duration of destructive monitoring procedures these methods cannot be recommended for industrial-scale RW characterization. Field of application of destructive methods:

- process monitoring of various fluids in process of PDEC operation, including SNF reprocessing and fuel refabrication,
- detailed selective analysis of radionuclide composition and specific radionuclide activities in RW flows in order to determine stable conservative radionuclide ratios (radionuclide vectors) and their further application as a part of radionuclide vector in RW characterization.

Procedure which can be suggested for PDEC includes continuous specification of radionuclide vectors identified in RW studies based on intermediate results of destructive measurements of process fluids and calculation of activities of difficult-to-detect radionuclides in characterized conditioned RW based on gamma-spectrometry results.

Destructive radionuclide measurements in PDEC RW shall include alpha-spectrometric (for Np-237, U-235, U-238, Pu-238, Pu-239, Pu-240, Am-241, Cm-243, Cm-244), beta-radiometric (for Sr-90, Tc-99 etc.), gamma- and x-ray-spectrometric (for Ni-59, Co-60, Nb- 94, I-129, Cs-137, U-235, Np-237, Am-241 etc.), liquid scintillation spectrometric (for H-3, C-14, Cl-36, Ni-63, Sr-90, Tc-99, Pu- 241 etc.) methods.

Mass-spectrometry may also be applicable (for Tc-99, I-129, U-235, U-238 etc.).

Measurements of the content of alpha-emitting RW in characterized RW following radiochemical separation traditionally include alpha-spectrometric methods based on semiconductor ion-implanted detector. The drawback of the method is that it is labour-intensive and requires long preparation of counting samples.

Standard proportional counters may be used to determine the content of beta-radiation nuclides in the samples after their radiochemical separation.

Alpha-spectrometry and beta-radiometry may be replaced by liquid-scintillation spectrometry (LSS). Liquid scintillation detection method is characterized by highly effective recording of nearly all types of radiation, comparative simplicity and quickness of counting sample preparation and a theoretical possibility of simultaneous detection of alpha-, beta-, and gamma-emitting radionuclides in complex mixtures. These advantages suggest that the method may become invaluable for a range of radio analytical tasks. The drawback of LSS method is its low resolution and, thus, inability to separate alpha-emitters with similar energies.

Liquid scintillation counters equipped with specialized software for decryption of continuous spectra may work in spectrometric mode, which in some cases will provide complete information on radionuclide composition of the samples or, in other cases, reliable confirmation that thresholds for specific radionuclides have not been exceeded.

Calculation radionuclide monitoring methods

Calculation methods include identification of radionuclide activities based on mathematical

modeling of the processes of radionuclide generation and spreading in process chains leading to generation of specific types of RW. Mathematical modeling method may be recommended for waste that includes core elements contaminated by radionuclides as a result of irradiation in neutron flux, such as SFA fragments, in-vessel equipment, neutron measurement channels, etc. Modeling method is very sensitive to the quality of input data used as model parameters and cannot fully replace the need for instrumental RW monitoring.

The method may be used to determine the ratios between specific activities of difficult-to-detect and easy-to-detect radionuclides (radionuclide vectors).

An approach based on the method of radionuclide vectors may be used to determine the content of difficult-to-detect radionuclides in RW. The methodology is widely used in international practice of RW characterization [7, 8]. The content of difficult-to-detect radionuclides in process of RW characterization is measured based on the radionuclide vector methodology using the pre-determined vectors and measured activities of reference easy-to-detect radionuclides.

Requirements to the PDEC RW characterization system

It is evident that RW batch characterization based on destructive methods is not feasible due to considerable time and financial costs. The basic characterization procedure for conditioned RW shall be performed using gamma-spectrometric systems designed for RW characterization in standard containers. Content of difficult-to-detect radionuclides in the characterized RW, including FM, shall be determined by calculation radionuclide vector method based on gamma-spectrometry results. Confirmation monitoring of FM content shall be based on neutron methods.

Implementation of radionuclide vector methodology requires carrying out detailed spectrometric study of RW flows based on destructive methods and alpha-, liquid scintillation, x-ray and gamma-spectrometry.

The study may be performed by third-party laboratory or duly equipped own laboratory.

Conclusions

The paper considers RW parameters for PDEC. List of radionuclides to be monitored in process of PDEC RW characterization was drawn up based on information about the RW. Possible methods of radionuclides monitoring in process of RW characterization have been considered.

It was demonstrated that the software and hardware system for PDEC RW characterization shall include the following components to ensure reliable measurements and characterization of waste:

- RW characterization installations based on gamma-spectrometers;
- neutron measurement system.

It is recommended to implement monitoring of difficult-to-detect radionuclides based on radionuclide vector methodology.

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