PRE-DECOMMISSIONING STUDY FOCUSED ON WWER-440 REACTOR UNIT NO. 3 AT NOVOVORONEZH NPP EXPLORING THE RADIATION CHARACTERISTICS OF THE REACTOR VESSEL AND ITS INTERNALS

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The paper presents computational and experimental studies focused on the radiation characteristics of the reactor vessel and reactor internals performed to estimate the volume and the characteristics of activated intermediate- and high-level waste expected to be generated from the decommissioning of unit No. 3 at the Novovoronezh NPP. Two software products, namely, MCU-6 and Serpent were used in the calculations by the Monte Carlo method. To verify the calculation results these were compared with data on metal samples of the reactor internals and reactor vessel obtained from their laboratory studies.

Keywords: *induced activity, WWER-440, Monte Carlo method, spectrometry, verification, decommissioning, radioactive waste.*

Introduction

In terms of volume, the largest intermediate- and high-level waste (RW) inventory generated from nuclear power plant (NPP) decommissioning accounts for the reactor vessel and its internals activated in neutron fields. Detailed information about their radiation characteristics is required to arrange relevant operations in a consistent way, including dismantling, fragmentation of these structures and conditioning of the resulting RW. In 2019—2021, a series of efforts was implemented at the Novovoronezh NPP, namely, at its reactor units No. 1 and 2 undergoing decommissioning and the shutdown unit No. 3, to characterize the reactor vessels and their internals. As part of these efforts, the volumes of RW Class 2 expected from reactor unit (RU) decommissioning were estimated, as well as their suitability for storage in a temporary facility.

The article summarizes the calculated specific activities of radionuclides found in the reactor unit No. 3 of the Novovoronezh NPP and its elements comparing them with those obtained from laboratory studies of metal sampled from the baffle, reactor vessel and neutron measurement channel (NMC).

Unit No. 3 of the Novovoronezh NPP and its main characteristics

Unit No. 3 involves a pilot reactor unit V-179 of the WWER-440 type, i. e., it's a pressure vessel thermal-neutron reactor with chemically pure demineralized water used as moderator and coolant. The WWER-440 core comprises 349 hexagonal assemblies with some of them operated as working

elements of the reactor control and protection system. An encapsulated fuel bundle consists of 126 rod fuel elements installed into a triangular lattice, the core of which is made of UO_2 pellets with an initial enrichment of 3.3% [1].

First criticality was achieved in December 1971. After completing the design operational life of 30 years, its operational license was extended for another 15 years. On December 25, 2016, the reactor was shut down for decommissioning.

The following elements are exposed to neutron fluxes in WWER-440 reactor units: reactor vessel, protective tube unit, NMC, internal reactor shaft, removable core basket, thermal shield, shaft bottom, annular tank and bioshield concrete. The article considers the most activated areas of the reactor vessel, NMC and the removable basket provided with relevant calculations and laboratory studies of the radionuclide levels.

Calculation model

To calculate the induced RU activity, neutron transfer was modelled and the changes in the isotopic composition of RU elements prompted by irradiation were calculated using the MCU-6 [2], [3] and Serpent [4] software based on the Monte Carlo method.

This generally accepted approach complies with IAEA recommendations [8] and is also provided for by the national standard GOST R 59968-2021 [12].

The evolving isotopic composition was modelled with an account taken of only neutron reactions on nuclei, whereas other processes and their contribution, including those associated with material interactions with beta/gamma radiation, were considered insignificant and were not taken into account. According to thermal power curves, the entire operational time was divided into several intervals. The following equations have been solved sequentially for each interval:

- a homogeneous conditional-critical neutron transfer equation yielding the neutron multiplication coefficients, reaction rates and flux densities in the materials of the studied system;
- a system of isotope kinetics equations yielding the nuclide material composition.

A full-scale computational model representing the WWER-440 reactor unit No. 3 of the Novovoronezh NPP was built to simulate the neutron transfer assuming its core being loaded heterogeneously with fresh fuel not getting burned out during reactor operation. A graph representing actual reactor power levels during reactor operation was used.

Along with the accumulated neutron fluence, chemical composition of structural materials is seen as a most important parameter affecting the induced activity [8]. The reactor vessel is made of heat-resistant pearlitic steel 48TS [5], the in-reactor devices, including the NMC, are made of austenitic stainless steel 08Kh18N10T [6] and the annular tank is made of St3sp [7]. Table 1 presents the chemical material composition according to GOST and TU standards.

No reliable information is available on the cobalt concentrations in steels. Nevertheless, the induced activity at the operational stage is greatly contributed by ^{60}Co generated from reactor vessel and NMC activation. This nuclide is generated mainly due to radioactive neutron capture by the stable ^{59}Co isotope, which is present in structural RU materials as an admixture.

The lack of reliable information on the activated admixtures and their content contributes to considerably higher uncertainty in the calculated induced activities.

Information on the material composition of steels given in Table 1 was used as input data in the first iteration of calculations launched to evaluate the composition of the irradiated RU elements (systems).

To calculate the actual admixture content of activated elements in the RU materials, steel samples were taken from the reactor vessel, NMC and the baffle of Unit No. 3 to study their chemical composition. The irradiated samples have been analyzed and relevant findings were compared with those calculated at the first iteration. Based on this comparison, the initial isotopic composition was adjusted, which was required for a refining calculation. The Laboratory studies section below presents the detailed data obtained from the chemical analysis of the metal samples.

Table 1. Elemental composition of steels, %

STEEL GRADE	C	Si	Mn	Cr	Ni	Mo	V	P	S	Co	Cu
48TS	0.13–0.18	0.17–0.37	0.3–0.6	2.5–3.0	< 0.4	0.6–0.8	0.25–0.35	0.02	0.02	<0.025	–
08Kh18N10T	0.03–0.06	< 0.8	1.0–2.0	17.0–19.0	10.0–11.0	3.0	–	0.035	0.02	–	–
St3sp	0.14–0.22	0.15–0.30	0.40–0.65	0.3	0.3	–	–	0.05	0.05	–	0.3

The laboratory samples were studied to evaluate the admixture content. It was found that the mass fractions of elements in the base metal of the reactor vessel were scattered in patterns corresponding to the experimental method uncertainty. Further study showed that according to the admixture content, this material could be considered homogeneous.

Nevertheless, the scattering observed in the reactor vessel's weld turned out to be higher than the measurement uncertainty, which could be explained by the manufacturing method. However, this area constitutes only to a small fraction of the area irradiated in the reactor vessel. For this reason, this material was not taken into account in further assessment of the reactor vessel material and its activation.

Table 2. Mass fractions of chemical elements in steel 48TS constituting to the reactor vessel assumed in the calculations

Sample	Mass fraction of elements, %							
	V	Cr	Mn	Fe	Co	Ni	Cu	Mo
Mean value	0.295	2.850	0.438	95.417	0.012	0.183	0.142	0.685

Table 3 presents the chemical elements present in 08Kh18N10T steel and their mass fractions accounted for in the activation calculations focused on reactor internal devices.

Table 3. Mass fractions of chemical elements in 08Kh18N10T steel baffle and NMC assumed in the calculations

Sample	Mass fraction of elements, %								
	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Mo
Mean value	0.580	0.240	17.200	0.983	69.233	0.076	11.200	0.300	0.076

Laboratory studies

To verify the calculated radiation characteristics of the irradiated elements (systems) in Unit No. 3 of the Novovoronezh NPP, the accredited laboratory of the National Research Center Kurchatov Institute launched experimental studies focused on the induced activity and the chemical composition of the sampled activated reactor elements. The corresponding metal subjected to spectrometric and chemical analyzes was sampled from reactor vessel, core baffle and NMC case as part of materials science research.

This study has investigated the composition of two samples cut out from the base metal constituting to the irradiated part of the reactor vessel (samples 1 and 2, respectively), one sample from weld No. 4 of the reactor vessel (sample 3), three micro samples taken from the core baffle, as well as the NMC case material.

Mass fraction of chemical elements was measured by the Elan DRC-e spectrometer (Perkin Elmer) based on the mass spectrometry with inductively coupled plasma, as well as by Avio 200 spectrometer from Perkin Elmer based on the atomic emission method with inductively coupled plasma. Tables 4 and 5 summarize the measurement results.

Trephines from baffles 1–3 were sampled at a distance of 1364 mm, 1654 mm, 364 mm, respectively, from the upper part of the baffle. NMC case was sampled at a distance of 700 mm from its bottom end.

Relative expanded uncertainty of the measurement results in the vessel steel samples accounted for 10 %, whereas in the baffle samples it amounted to 15 % (assuming a coverage coefficient of k = 2 corresponding to a confidence probability of 0.95).

Table 4. Mass fraction of chemical elements compared for different samples in the reactor vessel

Samples		Sample 1			Sample 2		Sample 3					
Height from main connector, mm		4,846	4,954	4,846	6,754	6,646	6,850					
Height from the core bottom, mm		2,314.8	2,206.8	2,314.8	401.1	509.1	304.6					
Distance from the inner surface, mm		17.3	63.3	126.5	82.7	128.1	16.9	24	53.8	78.6	98.1	133.2
Mass fraction of elements, %	Mo	0.7	0.71	0.66	0.67	0.66	0.71	0.54	0.55	0.44	0.45	0.42
	V	0.3	0.3	0.28	0.29	0.29	0.31	0.21	0.22	0.13	0.12	0.14
	Co	0.012	0.012	0.012	0.012	0.012	0.013	0.012	0.012	0.034	0.041	0.029
	Mn	0.44	0.45	0.42	0.43	0.43	0.46	0.97	1.02	0.93	0.96	0.86
	Ni	0.18	0.19	0.18	0.18	0.18	0.19	0.14	0.13	0.23	0.27	0.2
	Cu	0.14	0.14	0.14	0.14	0.14	0.15	0.12	0.13	0.15	0.17	0.14
	Cr	2.9	2.9	2.7	2.8	2.8	3	1.8	1.6	1.4	1.5	1.4
	Fe	95.4	95.3	95.6	95.5	95.5	95.2	96.2	96.3	96.7	96.4	96.8

Table 5. Measured mass fractions of chemical elements in baffle samples in the reactor vessel and the 11B NMC case

Baffle and NMC samples	Mass fraction of elements, %								
	Mo	V	Co	Mn	Ni	Cu	Cr	Ti	Fe
Sample 3	0.089	0.160	0.0227	1.06	10.6	0.28	17.6	0.58	69.5
Sample 2	0.070	0.280	0.0203	0.95	11.5	0.31	17.0	0.58	69.0
Sample 1	0.069	0.280	0.0204	0.94	11.5	0.31	17.0	0.58	69.2
NMC case	0.095	0.076	0.0760	1.30	10.6	0.16	16.6	0.45	70.6

The measured chemical composition of steel 08Kh18N10T samples was analyzed: the content of admixture elements in the baffle material, in particular cobalt, differed considerably from the one found in the NMC sample. This circumstance could be explained by baffle irradiation as part of the reactor unit since 1975, thus, causing massive cobalt burn up, whereas NMC irradiation time accounted for two years only. Since the cobalt content measured in the NMC sample turned out to be more than three times higher than the one found in the baffle material, further calculations conservatively assumed the initial cobalt content in all internal elements as equal to the one measured in the NMC sample.

The samples were analyzed based on semiconductor spectrometry of X-ray and gamma radiation, whereas the liquid scintillation spectrometry method was used to measure the beta radiation. These measurements revealed the specific activities of ^{54}Mn, ^{55}Fe, ^{59}Ni, ^{60}Co and ^{63}Ni isotopes.

^{54}Mn and ^{60}Co specific activities in the studied samples were measured using semiconductor gamma spectrometers from Ortec fitted with multichannel analyzers involving 16,000 channels and detectors made of high-purity germanium HPGe p-type GEM-10, as well as n-type GMX-10 of an extended range with an ultra-thin beryllium window. The measured counting sample activity accounted for the maximum expanded total uncertainty (with a coverage factor k=2) of 3.8 % and 3.2 % for ^{54}Mn and ^{60}Co respectively.

To measure ^{59}Ni, ^{55}Fe, ^{63}Ni activities, iron and nickel were separated from other elements found in the studied samples via multistage ion exchange chromatography.

To measure ^{59}Ni activity, "weightless" targets were manufactured and subsequently measured on a Canberra X-ray gamma spectrometer consisting of a multichannel LYNX 2.0 analyzer (Canberra) involving 32,000 channels and a detector made of highly pure germanium with an ultra-thin beryllium window GUL-0110. Counting sample activity was measured assuming the maximum expanded total uncertainty (given a coverage factor k of 2) of 8 %.

^{55}Fe, ^{63}Ni activities in the counting samples taken from solutions that had previously experienced radiochemical separation were measured by the liquid scintillation method using a Quantulus 1220 radiometer. The maximum expanded total uncertainty (with a coverage factor k=2) accounted for 4.5 % and 3.2 % for ^{55}Fe and ^{63}Ni respectively.

Verification of calculated induced activities

To verify the calculation model, the induced activities in the reactor vessel, NMC and baffle elements were calculated for the spots corresponding to the layout of the samples taken along the core height. A total of 12 samples have been studied: 1 from the NMC, 3 from the baffle and 8 from the reactor vessel.

Table 6 summarizes the generalized verification results presented as relative deviations of the calculated radionuclide specific activities from the experimental ones.

The compared computational and experimental data demonstrated that the calculation results provided adequate description of radionuclide distribution over the RU element volume in the intense neutron flux region and could be applied to calculate the volume of activated ILW and HLW generated from RU dismantling and fragmentation. The observed deviations were explained by possible variations in the mass content of activated isotopes in the reactor elements, the approximations of the calculation model used and the activity measurement errors.

Table 6. Deviations of calculated induced activities from the experimental findings

Sample		Calculation/ measurement ratio, rel. units				
		^{54}Mn	^{55}Fe	^{59}Ni	^{60}Co	^{63}Ni
Vessel	sample 1	0.71–2.06	1.11	1.16	1.04–1.42	0.99
	sample 2	1.03–1.1	1.38	1.01	0.92–1.36	0.76
	sample 3	0.88–0.97	0.6–1.25	0.64–1.17	0.41–1.7	0.89–1.86

End of ***Table 6***

Sample		Calculation/ measurement ratio, rel. units				
		^{54}Mn	^{55}Fe	^{59}Ni	^{60}Co	^{63}Ni
Baffle	sample 1	1.17	2.06	4.59	1.24	1.61
	sample 2	1.08	2.15	4.19	1.22	1.55
NMC		0,46	0.94	1.19	0.76	0.8

Estimated induced activity in the reactor vessel and NMC

To estimate the volumes of HLW and ILW generation from reactor vessel and NMC dismantling and fragmentation, the specific activities of activated RU elements were calculated in accordance with the following breakdown:

- lid flange — height-wise in two sections (80 cm each), the lid and bottom are separate zones, body shell — into 10 identical components (approximately 95 cm each), which were subsequently divided into 3 equally thick sections;
- basket — into ten zones;
- the upper part of the shaft shell — into five zones;
- the upper grate of the shaft bottom — into two zones;
- the middle part of the shaft bottom shell — into five zones;
- the lower part of the shaft bottom — into two zones;
- the bottom of the shaft bottom is a separate zone;
- the inner wall of the biological protection tank — into five zones height-wise;
- the bottom and the lid of the biological protection tank — into three annular zones;
- the middle part of the shaft — into five zones height-wise;
- heat shield — into five zones height-wise;
- the unit featuring protective tubes, heads and tails of fuel assemblies, the reactor shaft bottom and shield assemblies were assumed as homogeneous.

Figure 1 shows the distributed total specific activities of radionuclides along the height of the basket, shaft and the reactor vessel at the time of the RU final shutdown for decommissioning (December 25, 2016) on a logarithmic scale. The activity distribution height-wise along the vessel is presented for the inner layer closest to the core.

Table 7 presents the calculated specific activity ranges for the radionuclides in the RU structural elements in accordance with the above breakdown.

Figures 2 and 3 represent the specific activity levels changing over a 1,000-year period for the elements constituting to the core level subjected to verification (removable basket and vessel shell).

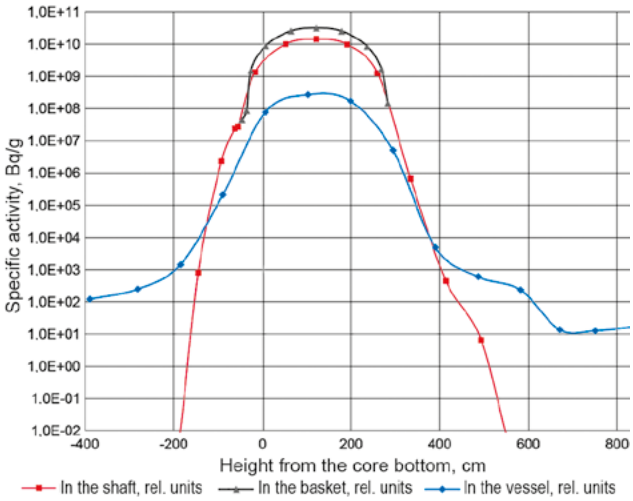

Figure 1. Distribution of total specific activities along the height of the reactor vessel, basket and shaft (logarithmic scale)

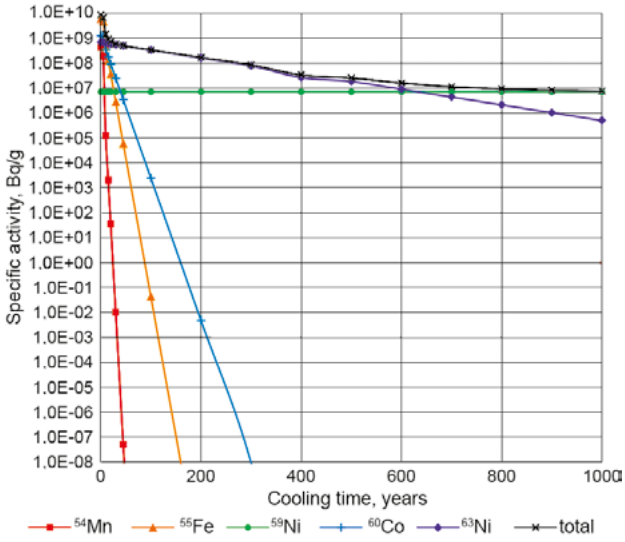

Figure 2. Fluctuating specific activity levels in the removable basket at the core level

Calculations showed that according to the total specific activity levels and relevant criteria [10], the shell material of the reactor vessel's internal section at the core level could be referred to the HLW category if cooled for 10 years, to the ILW category – if cooled for 10—400 years, to VLW category — if

Table 7. Calculated ranges of specific activities for radionuclides found in the structural RU elements in accordance with the calculation model breakdown

Structure element	Specific activity of a radionuclide, Bq/g					Total specific activity, Bq/g
	^{54}Mn	^{55}Fe	^{59}Ni	^{60}Co	^{63}Ni	
Reactor vessel						
Flange	$0.0–5.86\cdot10^{-1}$	6.78–84.5	$6.88\cdot10^{-5}–1.11\cdot10^{-3}$	$3.71\cdot10^{-2}–8.88$	$4.89\cdot10^{-3}–9.52\cdot10^{-2}$	7.53–94.0
Lid	$2.46\cdot10^{-3}$	4.97	$7.29\cdot10^{-5}$	1.61	$4.85\cdot10^{-3}$	6.59
Bottom	$4.81\cdot10^{-1}$	56.4	$8.10\cdot10^{-4}$	2.28	$7.74\cdot10^{-2}$	59.2
Shell, inner part	$1.04\cdot10^{-1}–8.17\cdot10^{6}$	$105–1.34\cdot10^{8}$	$1.53\cdot10^{-3}–2.03\cdot10^{3}$	$7.94–5.08\cdot10^{6}$	$1.46\cdot10^{-1}–2.08\cdot10^{5}$	$115–1.48\cdot10^{8}$
Shell, middle part	$8.20\cdot10^{-1}–3.78\cdot10^{6}$	$90.2–1.91\cdot10^{7}$	$1.05\cdot10^{-3}–267$	$4.81–1.10\cdot10^{6}$	$8.23\cdot10^{-2}–2.49\cdot10^{4}$	$104–2.40\cdot10^{7}$
Shell, outer part	$2.91–1.65\cdot10^{6}$	$232–8.41\cdot10^{6}$	$3.22\cdot10^{-3}–114$	$19.4–5.39\cdot10^{5}$	$2.91\cdot10^{-1}–1.01\cdot10^{4}$	$254–1.06\cdot10^{7}$
Removable basket	$2.76\cdot10^{5}–4.28\cdot10^{8}$	$7.25\cdot10^{6}–6.16\cdot10^{9}$	$7.13\cdot10^{3}–7.08\cdot10^{6}$	$5.76\cdot10^{6}–1.27\cdot10^{9}$	$4.82\cdot10^{5}–6.79\cdot10^{8}$	$1.38\cdot10^{7}–8.54\cdot10^{9}$
Shaft						
Shell in the upper part	$0.0–5.83\cdot10^{3}$	$0.0–1.26\cdot10^{5}$	0.0–164	$0.0–4.86\cdot10^{4}$	$0.0–1.66\cdot10^{4}$	$0.0–1.97\cdot10^{5}$
Middle part	$1.37\cdot10^{7}–1.69\cdot10^{8}$	$2.28\cdot10^{8}–2.68\cdot10^{9}$	$2.85\cdot10^{5}–3.33\cdot10^{6}$	$1.03\cdot10^{8}–1.09\cdot10^{9}$	$2.79\cdot10^{7}–3.21\cdot10^{8}$	$3.73\cdot10^{8}–4.26\cdot10^{9}$
Upper part of the bottom	$1.47\cdot10^{4}–5.75\cdot10^{4}$	$4.15\cdot10^{6}–4.20\cdot10^{6}$	$4.46\cdot10^{3}–5.27\cdot10^{3}$	$3.7\cdot10^{6}–5.99\cdot10^{6}$	$3.10\cdot10^{5}–4.89\cdot10^{5}$	$8.35\cdot10^{6}–1.06\cdot10^{7}$
Bottom shell	0.0–664	$0.0–4.44\cdot10^{5}$	0.0–597	$0.0–1.91\cdot10^{5}$	$0.0–6.20\cdot10^{4}$	$0.0–6.98\cdot10^{5}$
Heat shield	$2.48\cdot10^{7}–5.91\cdot10^{7}$	$6.34\cdot10^{8}–1.54\cdot10^{9}$	$8.02\cdot10^{5}–1.95\cdot10^{6}$	$2.56\cdot10^{8}–6.02\cdot10^{8}$	$8.01\cdot10^{7}–1.94\cdot10^{8}$	$9.96\cdot10^{8}–2.40\cdot10^{9}$
Shield assemblies	$2.3\cdot10^{8}$	$3.55\cdot10^{9}$	$1.13\cdot10^{6}$	$1.13\cdot10^{9}$	$1.24\cdot10^{8}$	$5.03\cdot10^{9}$

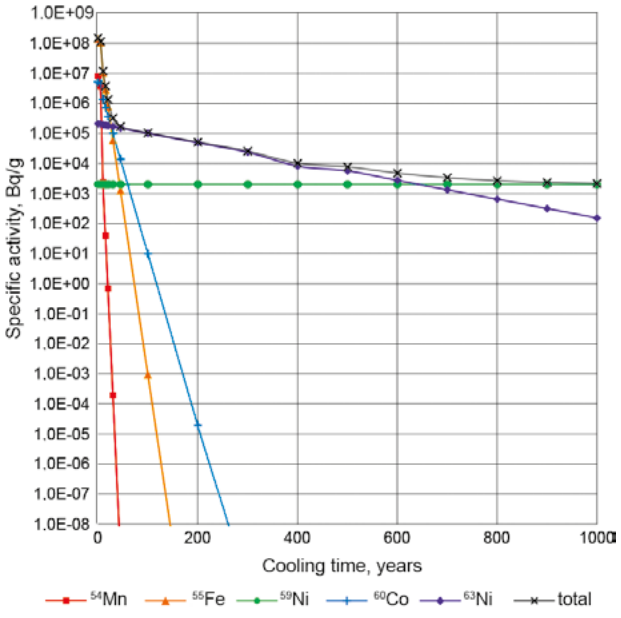

Figure 3. Fluctuating specific activity levels in the vessel shell at the core level

cooled from 400 and up to 1,000 years. Within the first two decades of cooling, the short-lived radionuclide ^{55}Fe is expected to provide the greatest contribution to the activity level (up to 92%), i. e., $1.04\cdot10^{8}$ Bq/g after 1 year of cooling. Over a period of 30 years, ^{60}Co is supposed to provide a large contribution (up to 30%) with its specific activity amounting to $4.45\cdot10^{6}$ Bq/g upon 1 year of cooling. After 30 years, the specific activity levels are expected to be largely contributed by the pure beta emitter ^{63}Ni ($1.67\cdot10^{5}$ Bq/g). On January 1, 2024, new RW categorization criteria established by the Decree of the Government of the Russian Federation of October 29, 2022 No. 1929 came into force [11]. Considering these new criteria, upon being cooled for 450 years, the shell material constituting to the inner part of the reactor vessel at the core level would be referred to RW Class 3, and then subsequently to RW Class 4.

In terms of the total specific activity level, the removable basket and its material at the core level would correspond to the HLW category if cooled for up to 700 years, to the ILW category if cooled from 700 years to 1,000 years. During the first 5 years, the activity would be decisively contributed by the short-lived ^{55}Fe radionuclide (up to 72%) with a specific activity of $4.77\cdot10^{9}$ Bq/g after 1 year of cooling. The activity in the first 15 years of cooling would be greatly contributed by ^{60}Co (up to 24%) with its specific activity reaching $1.11\cdot10^{9}$ Bq/g after

1 year of cooling. After 20 years of cooling, it would be mainly contributed by ^{63}Ni, which is a pure beta emitter, with a specific activity of $5.88 \cdot 10^8$ Bq/g. Taking into account the new RW classification criteria [11], after being cooled for over 1,000 years, the removable basket material would correspond to RW Class 2.

Conclusions

Calculation and experimental studies were performed to explore the radiation characteristics of the activated RU elements in Unit No. 3 of the NVNPP, which involved a full-scale RU modelling in MCU-6 and Serpent software implementing the Monte Carlo method. Under the experimental studies, the reactor vessel, baffle and NMC were sampled to identify the chemical composition and measure the ^{60}Co, ^{54}Mn, ^{55}Fe, ^{59}Ni, ^{63}Ni specific activities.

The studies revealed good agreement between the results obtained for the radionuclides in the RU elements and the experimental spectrometric data.

Studies showed that the total induced activity of structural elements was greatly contributed by radionuclides formed from the activation of chemical elements seen as undeclared admixtures found in the applied structural materials (not considered in the composition of steels in accordance with GOST and TU [5], [6]). Particular attention should be paid to cobalt causing ^{60}Co generation due to the radiation capture reaction. At the initial decommissioning stages, its specific activity in the reactor vessel shell made of steel 48TS and a removable basket made of steel 08Kh8N10T may range from 4% to 13% and from 15 % to 42 % respectively considering the total activity at a mass content of ^{59}Co in steels accounting for 0.012 % and 0.076 % respectively.

The specific activities calculated for different radionuclides showing the activity distribution over the RU element volume can be further used to estimate the amount and the characteristics of the activated RW generated from Unit No. 3 decommissioning at the Novovoronezh NPP, as well as in the development of RW Class 2 and 3 container designs.

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