NUCET NUCLEAR ENERGY AND TECHNOLOGY

Nuclear Energy and Technology 4(4): 257–262
DOI 10.3897/nucet.4.31875

Research Article

On the necessity and the role of descriptors of neutron activated structural and shielding materials of nuclear installations for future decommissioning*

Boris K. Bylkin[1], Igor A. Engovatov[2], Alexey N. Kozhevnikov[1], Dmitry K. Sinyushin[3]

1 *National Research Center "Kurchatov Institute,1 Academician Kurchatov sq., Moscow, 123182 Russia*

2 *National Research Moscow State University of Civil Engineering, 26 Yaroslavskoe shosse, Moscow, 129337 Russia*

3 *Joint Stock Company "State Specialized Design Institute", 4 Sharikopodshipnikovskaya st., Moscow, 115088 Russia*

Corresponding author: *Igor A. Engovatov* (eng46@mail.ru)

Academic editor: *Yury Korovin* ♦ **Received** 5 September 2018 ♦ **Accepted** 20 November 2018 ♦ **Published** 13 December 2018

Citation: Bylkin BK, Engovatov IA, Kozhevnikov AN, Sinyushin DK (2018) On the necessity and the role of descriptors of neutron activated structural and shielding materials of nuclear installations for future decommissioning. Nuclear Energy and Technology 4(4): 257–262. https://doi.org/10.3897/nucet.4.31875

Abstract

Existing situation in nuclear industry is characterized with simultaneous development of the following two processes: design and construction of new generation of nuclear installations and decommissioning of installations of older generations.

Significant amounts of radioactive wastes generated during the decommissioning phase are determined both for the first and the second types of installations by the induced activity of neutron irradiated structural and shielding materials. Concentration of the so-called radioactivity-hazardous nuclides in primary building and construction materials is the most important characteristics determining the resulting levels of induced activity. Values of these concentrations for the same type of material extracted from different geological deposits may differ by one or two orders of magnitude. Information about concentrations of radiation-hazardous elements in radiation shielding materials is fragmented and, as a rule, unsuitable for practical application.

The purpose of the present study was to substantiate the necessity of compiling and recording the data on the concentrations of radioactivity-hazardous nuclides for building and structural materials for nuclear installations during the phases of design, operation and decommissioning.

Three types of shielding concrete compositions were selected for the investigation. Concentrations of radioactivity-hazardous nuclides were mainly obtained by neutron activation technique. Neutron transport calculations were performed in one-dimensional cylindrical geometry at the core mid-plane according to usual core-vessel-shielding model of modern VVER reactor unit including 2-m thick concrete shield. Both transport and activation calculations were undertaken using modules of SCALE system.

The obtained results allow estimating neutron-induced activation levels in the material as the function of irradiation time, amounts and categories of radioactive waste and their evolution during the decay time from 1 to 100 years. It was established that neutron-induced activity of shielding concrete strongly depends on the actual concentrations of radioactivity-hazardous nuclides in the concrete including 'trace' concentrations (other factors being the same). It was also shown that failure to take such concentrations into account may lead to the underestimation of neutron-induced activation levels and amounts of radioactive wastes and their category.

* Russian text published: Izvestiya vuzov. Yadernaya Energetika (ISSN 0204-3327), 2018, n. 3, pp. 41-48.

The obtained results confirmed the necessity of compiling and maintaining data records on the concentrations of radioactivity-hazardous nuclides for materials used in structural and shielding materials of nuclear installations. Proposals were formulated on the potential accumulation of information, composition and formatting of descriptors of chemical composition of shielding and structural materials of nuclear installations.

Keywords

Nuclear facilities; NPP unit; decommissioning; neutron induced activity; radioactive waste; radiation shielding concrete; descriptor

Reactor core equipment, structural and shielding materials beecome radioactive under the effects of neutrons generated by the nuclear reactor. During the phase of decommissioning nuclear installations such structures act as the main source of radioactive wastes (RWs). The fact that such RWs are not decontaminable constitutes their specific feature.

Reactor vessel, internal equipment, metal structures, graphite cladding, concrete and reinforced concrete are the main activated structures and materials for NPP power units with different types of reactors and for research nuclear reactors.

The problem of activation of shielding materials, equipment and structural elements of nuclear installations is well enough investigated (Evans John et al. 1988, Engovatov et al. 2005, Engovatov et al. 1999, Nazarov et al. 1991, Bittner et al. 1984, Ser. No. WS-G-2.1 1999, Borisov et al. 1996, Bylkin et al. 2017, Bylkin and Engovatov 2014, Engovatov et al. 2014). Induced activity depends on the radiation and physical parameters of the nuclear installation including neutron flux density and neutron energy spectrum, chemical composition of the irradiated material, values of respective activation cross-sections, irradiation and decay time, etc.

With all other conditions remaining the same levels of induced activity will be determined by chemical elements present in the composition of structural and shielding materials as the main components (mass concentration in the material over 1%), impurities (with concentrations from 0.01% to 1%) and trace amounts (with concentrations below 0.01% mass).

It is known from reference sources (Evans John et al. 1988, Engovatov et al. 2005, Engovatov et al. 1999, Bittner et al.1984) that quantities of the so-called radioactivity-hazardous elements, on isotopes of which long-lived radionuclides are generated, are limited: among metals these include iron, cobalt, nickel; for graphite – carbon and lithium; for concrete of radiation shielding – lithium, iron, cobalt, europium, calcium, cesium and nickel.

Concentration in structural and shielding materials of such elements as europium, cobalt, cesium, nickel, lithium and carbon amounts to 10^{-2} – 10^{-6} mass per cent. Concentrations of calcium and iron reach several and several dozen mass per cent.

Spread of concentrations of impurity and trace elements is extremely wide not only between different forms of materials of the same type, but even among the samples of materials of the same form. Data on the spread of concentrations of radioactivity-hazardous chemical elements in structural and shielding materials of nuclear installations in Russia, USA and EU countries are presented in Table 1 (Evans John et al. 1988, Engovatov et al. 2005, Bittner et al. 1984, Bylkin et al. 2017).

Data on the concentrations of such elements in the Earth crust are also given in Table 1 as the explanatory information (Voytkevich et al. 1977).

Significant dispersion of concentrations of radioactivity-hazardous elements and incorrect determination of

Table 1. Range of variation of mass concentrations of activation-hazardous elements in concrete and steel compositions of reactor installations in different countries

Element	Concentration in Earth crust, $A_{e.c.}$, 10^{-4} mass%	Range of concentrations in the group of investigated materials ($A_{min} - A_{max}$), 10^{-4} %mass						
		Concrete compositions			Steels			
					Carbon steels		Stainless steels	
		RF	USA	EU	RF	USA	RF	USA
Eu	1.3	0.01–4.4	0.11-1.2	0.31-1.18	-	-	-	-
Co	18.0	0.07-174	1.1-31.0	2.3-20	30-120	115	167-875	229-2570
Fe	4.65	007-61.4	0.5-24.0	0.6-61.2	99	99	68-97	68-76.7
Cs	3.7	0.01-5.9	0.32-6.2	0.26-5.0	-	-	-	-
Ca	2.96	1.5-39	8.8-35	3.4-29	-	-	-	-
Ni	58	0.1-13.6	11.9-87.0	6.1-26.0	-	0.93	0.25-10	8.8-11

their contents in shielding and structural materials can lead to the underestimation of activation levels and, as the consequence, of volumes of radioactive wastes during decommissioning reactor installations (Engovatov et al. 2014, Engovatov 2011, Engovatov et al. 2015).

This conclusion is illustrated using the example of information on the activation of concrete radiation shielding of nuclear installations.

Calculations of levels of induced activity and volumes of activated RWs for radiation shielding concrete compositions were performed based on the data in (Engovatov et al. 2014, Engovatov et al. 2015, ORNL-RSICC C-650 1998, ONL-ESICC DLC-185 1999, NUREG/CR-200 1995) for different binding agents for minimum, medium and maximum concentrations of radioactivity-hazardous elements, numerical values for which were collected as the result of analysis of their concentrations in samples from different deposits in Russia, USA and EU countries.

The following input data were accepted in this case:

- neutron fluxes and spectrum were taken in accordance with the data in (Engovatov et al. 2014, Engovatov et al. 2015);
- Radiation shielding materials: serpentinite concrete, granite concrete and hematite concrete, i.e. concrete-based materials widely applied and already used earlier for radiation shielding of NPP and research reactors;
- Irradiation time equal to 50 years;
- Decay time equal to 1, 25, 50, 100 and 1000 years.

Some results of calculation studies are presented in Figs. 1 – 3 and in Tables 2 and 3.

Distributions of total specific activity of main long-lived radionuclides determining the induced activity of serpentinite concrete for minimum, medium and maximum concentration of activation-hazardous elements are presented for the decay time from 1 year to 1000 years (see Fig. 1). The obtained data correspond to of the subsurface layer of concrete positioned in the vicinity of the reactor vessel.

The data presented in Fig. 1 allow formulating the following conclusions:

- Difference in the value of total activity can reach one order of magnitude for different decay times;
- For different decay times radioactive wastes within the interval of decay time from one year to 1000 years will refer to different categories (Decree of the RF Government No. 1069 of October 19 2012) in accordance with Table 2 depending on the accepted concentrations of activation-hazardous elements for concrete compositions with different binding agents.

Distributions of total specific activity of main long-lived radionuclides over the thickness of granite concrete

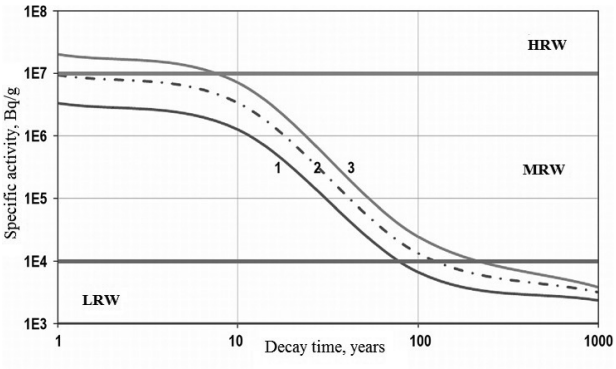

Figure 1. Distribution of total specific activity of main long-lived radionuclides for serpentinite concrete for minimum (1), medium (2) and maximum (3) concentrations of activation-hazardous elements for decay time from 1 year to 1000 years

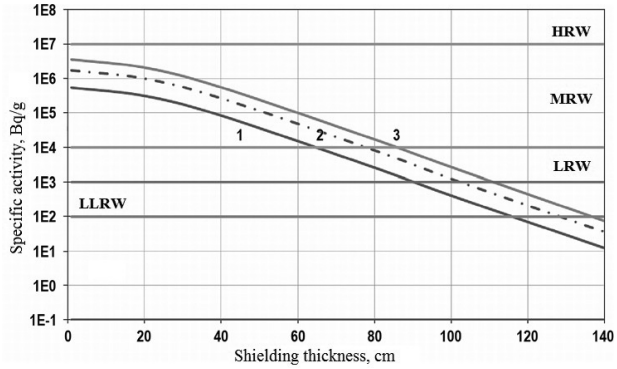

Figure 2. Distribution of total specific activity along the shielding thickness for minimum (1), medium (2) and maximum (3) concentrations of radioactivity-hazardous elements for irradiation time equal to 50 years and decay time equal to 10 years

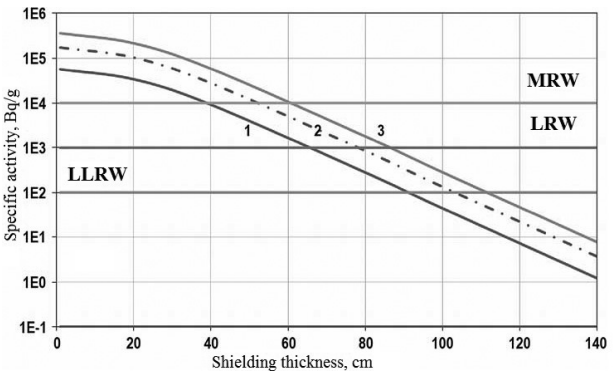

Figure 3. Distribution of total specific activity along the shielding thickness for minimum (1), medium (2) and maximum (3) concentrations of radioactivity-hazardous elements for irradiation time equal to 50 years and decay time equal to 50 years

radiation shielding are presented in Figs. 2 and 3 for minimum, medium and maximum concentrations of activation-hazardous elements for decay times equal to 10 years and 50 years, respectively.

Thus, thickness of activated layer of concrete radiation shielding will constitute significant value during the pha-

Table 2. Categories of RWs within the interval of decay time from one year to 1000 years for concretecompositions with different binding agents for minimum (Min), medium (Med) and Maximum (Max) concentrations of activation-hazardous elements

Aggregate (density, kg/m³)	Min				Med				Max			
	Category of wastes				Category of wastes				Category of wastes			
	HRW	MRW	LRW	LLRW	HRW	MRW	LRW	LLRW	HRW	MRW	LRW	LLRW
Serpentinite (2061.7)	-	Up to 100 years	Up to 1000 years	-	-	Up to 150 years	Up to 1000 years	-	Up to 10 years	Up to 300 years	Up to 1000 years	-
Granite (2145.6)	-	Up to 60 years	Up to 700 years	Up to 1000 years	-	Up to 180 years	Up to 1000 years	-	-	Up to 400 years	Up to 1000 years	-
Hematite (3527.8)	Up to 5 years	Up to 90 years	Up to 1000 years	-	Up to 8 years	Up to 150 years	Up to 1000 years	-	Up to 10 years	Up to 200 years	Up to 1000 years	-

Table 3. Distribution of RWs by categories along the thickness for different concentrations of activation-hazardous elements if eadiation sheilding concretes

Aggregate (density, kg/m³)	Decay time *t*, years	Min				Med				Max			
		Category of wastes				Category of wastes				Category of wastes			
		Thickness of radioactive layer, cm	MRW	LRW	LLRW	Thickness of radioactive layer, cm	MRW	LRW	LLRW	Thickness of radioactive layer, cm	MRW	LRW	LLRW
Serpentinite (2061.7)	10	90	50	20	20	100	60	20	20	110	60	20	30
	50	70	30	20	20	80	40	20	20	90	45	25	20
Granite (2145.6)	10	110	60	30	120	30	70	40	20	140	90	20	30
	50	90	40	20	30	110	50	30	30	120	60	30	30
Hematite (3527.8)	10	100	60	20	20	105	65	20	20	110	60	20	30
	50	70	30	20	20	80	40	20	20	85	40	25	20

se of decommissioning for different decay times. Notably, from the viewpoint of classification activated layer will refer to different categories of radwastes (Decree of the RF Government No. 1069 of October 19 2012).

Calculated data for total specific activity of activated concretes of radiation shielding are presented in Table 3 for different decay times and thicknesses of radioactive layer, which will determine volumes of RWs belonging to different categories.

The obtained results are important specifically for the phase of NPP decommissioning because of the generation of large quantities of non-decontaminable RWs (Bylkin et al. 2011, Bylkin et al. 2016, Engovatov and Sinyushin 2017) the cost of final disposal of which is extremely high and significantly differs for different categories of RWs according to the levels of specific activity (Ivanov et al. 2015).

For enhancing reliability of determination of activity levels and volums of radioactive wastes by the moment of decommissioning of operated NPP it is necessary to know for each specific case the chemical (element) composition of all structural and shielding materials in the reactor space subjected to irradiation with neutron fluxes.

Failure to account in the calculations for the concentrations of separate trace elements or their incorrect determination in shielding materials results in the underestimation of the volume of radioactive wastes during decommissioning reactor installations. This will negatively affect the costs of their final disposal, will lead to the miscalculation of volumes of radioactive wastes and to the underestimation of radiation impact on personnel, populations and the environment.

Analysis of design documentation for new nuclear installations demonstrates that selection of structural and shielding materials for reactor installations operated until the current moment and those finally decommissioned is performed during the phase of designing without in-depth analysis and determination of their chemical compositi-

ons for defining concentrations of radioactivity-hazardous impurity or trace elements in their chemical composition.

Necessity to estmate the volumes of radioactive wastes due to activation, their classification and forecasting their variation depending on the decay time arises during decommissioning and preparation to decommissioning of nuclear installations (mainly power units of NPPs and research nuclear reactors).

Unfortunately, practically no information about concentrations of radioactivity-hazardous elements in radiation shielding materials (at the level of impurities or trace amounts) is available and so far there is even no understanding from where such information must be obtained and maintained.

Conviction on the necessity of development and practical implementation of descriptors of chemical compositions of structural and shielding materials has long ago taken shape among experts in protection from ionizing radiation.

Descriptor which must be included in the list of design documentation must contain, besides the characteristics already required at present, the following components:

- Macrochemical compositions of structural and shielding materials for calculating transfer of different types of radiation and estimating radiation resistance;
- Microchemical compositions of structural and shielding materials for calculating their activation characteristics with indication of concentrations of impurity and trace radioactivity-hazardous elements;
- Methodologies and uncertainties of determination of concentrations of chemical elements.

Such information can be obtained for materials of reactor vessel and metal structures using respective witness samples.

For shieding concretes witness samples are not used at present. Therefore, it is necessary to implement complex studies for obtaining experimental information about concentrations of radioactivity-hazardous elements in structural and shielding materials and in raw materials for their preparation. Results of these studies must be included as initial information in the data bank of activation characteristics of shielding concrete compositions of nuclear installations (Bylkin et al. 2018).

Thus, availability of the descriptor will dictate already during the phase of design of new nuclear power inistallations application of such chemical compositions of structural and shielding materials which would allow generating in the course of decommissioning these nuclear installations minimal volumes of radioactive wastes, which will reduce the costs of the conditioning and subsequent disposal (burial) and will enhance competitiveness of the nuclear power complex in modern market conditions.

References

- Bittner A, Jungwirth D, Bernard M, Gerland L, Brambilla G, Fitzpatrick J (1984) Concepts Aimed at Minimizing the Activation and Contamination of Concrete. Decommissioning of Nuclear Power Plants. Proc. of the European Conference held in Luxemburg, 22-24 May 1984. Springer Netherlands: 371-388. https://doi.org/10.1007/978-94-009-5628-5_32
- Borisov SE, Kryukov AP, Mashkovich VP, Neretin VA (1996) Two-dimensional studies of induced activity in MIFI IRT materials during decommissioning and reconstruction. Atomnaya energiya [Atomic Energy], 81(4): 277-281. [in Russian]
- Bylkin B, Kozhevnikov A, Engovatov I, Sinyushin D (2017) Induced activity of radiation shielding before NPP decommissioning. Yadernaya i radiatsionnaya bezopasnost, 3(85): 1-14. [in Russian]
- Bylkin BK, Davydova GB, Shurbenko EA (2011) Radioactive Wastes at Dismantling of Reactor Installations. Atomnaya energiya [Atomic energy], 110(3): 171-172. [in Russian]
- Bylkin BK, Engovatov IA (2014) Decommissioning of Reactor Installations. Monograph. Moscow. Moscow State University of Civil Engineering Publ., 228 pp. [in Russian]
- Bylkin BK, Engovatov IA, Kozhevnikov AN, Sinyushin DK (2018) Data bank on activation characteristics of radiation shielding concrete of nuclear plants. Vestnik MGSU, 13(2): 213-218. [in Russian] https://doi.org/10.22227/1997-0935.2018.2.213-221
- Bylkin BK, Kozhevnikov AN, Engovatov IA, Sinyushin DK (2016) Radioactivity Category Determination for Radiation-Protection Concrete in Nuclear Facilities Undergoing Decommissioning. Atomnaya energiya [Atomic energy], 121(5): 298-301 [in Russian]
- Decree of the RF Government No. 1069 of October 19 (2012) On Criteria of Referring Solid, Liquid and Gaseous Wastes to "Radioactive Wastes Category; Criteria of Referring Radioactive Wastes to "Special Radioactive Wastes" Category and to "Disposed Radioactive Wastes" Category; and Criteria for Classification of "Disposed Radioactive Wastes". ["Collected Legislation of the Russian Federation", no. 44, Art. 6017; 09.02.2015, No. 6, Art. 974].
- Engovatov IA (2011) Volume of radioactive waste and activation radiation shielding of nuclear installations. Vestnik MSGU, 8: 325-330. [in Russian]
- Engovatov IA, Bylkin BK, Kozhevnikov AN (2014) Optimization of the Compositions of Concrete for Radiation protection of NPP. In: Proc. Of the IX International Scientific-Technical Conference "Safety, Efficiency and Economics of Nuclear Engineering (ISTC-2014)". Moscow. AO "ENIC" Publ.: 305-309 [in Russian]
- Engovatov IA, Kozhevnikov AN, Bylkin BK (2015) Selecting concrete for Radiation Protection for New-Generation NPP. Atomnaya Energiya [Atomic Energy], 118 (6): 350-354. [in Russian].
- Engovatov IA, Mashkovich VP, Orlov YuV, Pologikh BG, Khlopkin NS, Tsypin SG (2005) Radiation Safety Assurance: Decommissioning Nuclear Reactors at Civil and Military Installations. Monograph. Arlington, VA 22201 USA, 208 pp.

- Engovatov IA, Mashkovich VP, Orlov YuV, Pologikh BG, Khlopkin NS, Tsypin SG (1999) Radiation Safety at Decommissioning of Civil and Military Installations. ISTC Project No. 465-97. Under the editorship of Khlopkin N.S. Moscow. RAIMS Publ., 300 pp.
- Engovatov IA, Sinyushin DK (2017) Minimization of radioactive wastes in decommissioning of new generation nuclear power plants. Vestnik BSTU, 10: 45-51. [in Russian]
- Evans John C, Lepel Elwood A, Sundens Ronald W, Thomas Charles W, Robertson David E (1988) Long-lived activation products in Light-Water Reactor Construction Materials: Implication for Decommissioning. Radioactive Waste Management and the Nuclear Fuel Cycle, 11(1): 1-39.
- Ivanov E, Korotkov A, Pyrtsov I (2015) Radionuclide vector. Rosenergoatom, 1: 42-45. [in Russian]
- Nazarov VM, Frontyasyeva MV, Baboshin NG, Engovatov IA, Lavdansky PA, Stefanov NI (1991) Activation studies of concrete binding agent ingredients used for nuclear radiation shielding. Kernenergie, 34: 7-8.
- NUREG/CR-200 (1995) Rev. 5. SCALE: A Modular Code System for Performing Standardized Computer Analyses for Licensing Evaluation.
- ONL-ESICC DLC-185 (1999) Bugle-96: Coupled 47 Neutron, 20 Gamma-Ray Group Cross Section Library derived from ENDF/B-VI for LWR Shielding and Pressure Vessel Dosimetry Applications.
- ORNL-RSICC C-650 (1998) DOORS 3.2: One-, Two- and Three Dimensional Discrete Ordinates Neutron/Photon Transport Code System.
- Ser. No. WS-G-2.1 (1999) IAEA Safety Standards. Decommissioning of Nuclear Power Plants and Research Reactors. Vienna, 41 pp.
- Voytkevich GA, Miroshnikov AE, Povarennykh AS, Prokhorov VG (1977) Quick reference guide to geochemistry. Moscow. Nedra Publ., 180 pp. [in Russian]