Nucl. Sci. and Tech., Vol.11, No. 3 (2021), pp. 01-10

Nuclear Science and Technology

Journal homepage: https://jnst.vn/index.php/nst

Evaluation of neutron flux distribution in structural components and activation products in aluminum alloy at 13-2 channel of the DNRR

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Abstract: The estimation of radiological properties of activated structural components of a nuclear reactor due to irradiation of neutron produced by fission is a very important task for radiation safety and reasonable cost of dismantling and radioactive waste management in the decommissioning plan of the reactor. In this work, the calculation approach was carried out by using three-dimensional neutron transport model with the Monte Carlo code MCNP5 to evaluate neutron fluxes and reaction rates. The Bateman equation was solved with neutron absorption reactions (fission and capture) and disintegration by ORIGEN2 code to obtain the activity of materials in reactor structures. This paper presents the evaluation results of the neutron flux distribution and the radioactivity of long-lived key activation products such as ^{60}Co, ^{55}Fe, ^{59}Ni, ^{63}Ni, etc. isotopes in the structural components of the Dalat Nuclear Research Reactor (DNRR). The validation of calculation methodology of the two codes was implemented by comparing calculation results with measured neutron fluxes at irradiation positions in the reactor core as well as specific activities at the bottom part of the aluminum guiding tube at 13-2 channel, which has been removed from the reactor core about six years. The calculation results were in good agreement under 7% difference with the experimental neutron flux value of (6.05±0.52) × 10^{12} n/cm^2.s, and under 33% difference with the experimental specific activities of ^{60}Co isotope being 1.86×10^4, 9.99×10^4, and 1.28×10^5 Bq/g at the positions of -32.5, -17.5 and -2.1 cm (the centerline of the reactor core is at 0 cm), respectively, in the aluminum guiding tube of irradiation channel 13-2. The neutron flux distributions in other structural components such as the graphite reflector, thermal column, thermalizing column, concrete shielding, etc. of the reactor were also evaluated. The obtained calculation results and experimental data are very valuable for the development of a suitable decommissioning plan and a reasonable dismantling strategy for the DNRR.

Keywords: *Neutron flux, component activity, DNRR, MCNP5, ORIGEN2, HEU, LEU, decommissioning.*

I. INTRODUCTION

The DNRR with a nominal power of 500 kW was reconstructed and upgraded from the USA-made 250 kW pool-typed TRIGA Mark II reactor, with the highest thermal neutron flux of 2.10×10^{13} n/cm^2.s, light water cooled, moderated, and shielded. After the reconstructed and upgraded activities, since March 1984, the reactor has been officially put into operation for the purposes of radioisotope production, neutron activation analysis, fundamental and applied research, and training [1].

From the design stage of the DNRR, the selection of materials for the purpose of the activated radiation level is low or generating

https://doi.org/10.53747/nst.v11i3.364
Received 25 October 2021, accepted 21 December 2021

mainly short-lived isotopes was considered. However, for the impurities, although with very small amounts but they have long-life activation products such as ^{55}Fe, ^{60}Co, ^{63}Ni, ^{65}Zn, ^{152}Eu, etc., which are the main factors contributing to the radiation dose while conducting dismantling and waste management activities of the decommissioning process.

Following IAEA safety guidelines, planning for decommissioning is required to start at the design stage and to continue throughout the lifetime of a nuclear facility. A study to estimate the residual activity in the decommissioning waste of the FIR 1 research reactor of Finland has been published in [2] and in the case of this reactor was permanently shut down and the final decommissioning planning should be developed. Meanwhile, the main purpose of our study is to evaluate the radioactivity of long-lived key activation products in the structural components of the DNRR in order to plan for decommissioning during its lifetime, following the safety requirement of IAEA guidelines and international experiences [3]. For this purpose, the MCNP5 and ORIGEN2 computer codes were used to evaluate the neutron flux, reaction rates, and radioactivity accumulated in the structural components of the reactor. For experimental measurements, the structural components were sampled and analyzed to determine impurity elements, and the measured data were also used in calculations to determine the specific radioactivity in the structural components of the reactor. The neutron activation analysis methods was used to evaluate thermal neutron fluxes at the irradiation positions inside the reactor core and the aluminum guiding tube at the 13-2 irradiation channel as well.

This paper presents the calculation results and experimental data of neutron flux distributions and the specific activity of long-lived key activation products such as ^{60}Co, ^{55}Fe, ^{63}Ni, ^{59}Ni, etc. isotopes in the aluminum guiding tube at irradiation channel 13-2. In addition, the calculation results of neutron flux distributions in the reactor core and in other structural components of the reactor are also given. Furthermore, validation of calculation method using two computer codes as MCNP5 and ORIGEN2 was also carried out to confirm fidelity in decommissioning calculation to estimate activities level of waste for the DNRR.

II. BRIEF DESCRIPTION OF THE REACTOR

The DNRR has a core in a cylindrical shape with a height of 60 cm and a diameter of 44.2 cm. The fuel assemblies (FAs), beryllium blocks, control rod guide tubes, and irradiation channels are fixed by two grid plate structures at the core bottom. The core is placed inside an aluminum tank and suspended by a supporting structure; the core bottom is above the tank bottom by a distance of 60 cm. The reactor has four neutron beam tubes, a thermal column, and a thermalizing column. The layout of the reactor and its main components are shown in Fig. 1 [1, 4].

The graphite reflector surrounding the core is a structure retained from the former TRIGA reactor. It consists primarily of a ring-shaped block of graphite having an approximate inside diameter of 45.7 cm, a radial thickness of 30.5 cm, and a height of 55.9 cm. Water is kept from contact with the graphite by encasing the entire reflector in a welded aluminum can [1, 4]. The aluminum alloy used in the structures such as the reactor pool tank, beam tubes, reflector, and thermal column cans kept from the former TRIGA reactor is the USA 6061 alloy, whereas the remaining structures and components are made of the SAV-1 alloy of the former Soviet Union. The chemical compositions of these alloys are given in Table I [1, 5].

Table I. Aluminum alloy compositions

Alloy	Cu	Cr	Mg	Si	Al	Impurities
6061 (wt%)	0.25	0.25	1	0.6	> 97.7	Ti, Fe, Mn, Zn
SAV-1 (wt%)	0.0058	-	0.48	0.8	> 98.5	Ti, Fe, B, Ni

The thermal column with dimensions of 1.2×1.2×1.6 m of the former TRIGA reactor remains unchanged. The column has waterproofed walls, made of aluminum and covered with boron. Graphite blocks with dimensions of 10.2×10.2×127 cm fulfill the volume of the column. The outer portion of the column is embedded in the concrete shielding, the inner portion is welded to the reactor tank and extends to the outer surface of the graphite reflector. In a vertical plane, the column extends approximately 33.0 cm above and below the graphite reflector and the centerline coincides with that of the core active height. The column door is made of heavy concrete. The concrete structures of the reactor have average density of 2.35 g/cm^3 and 3.5 g/cm^3 in particular around the thermal column [1, 4].

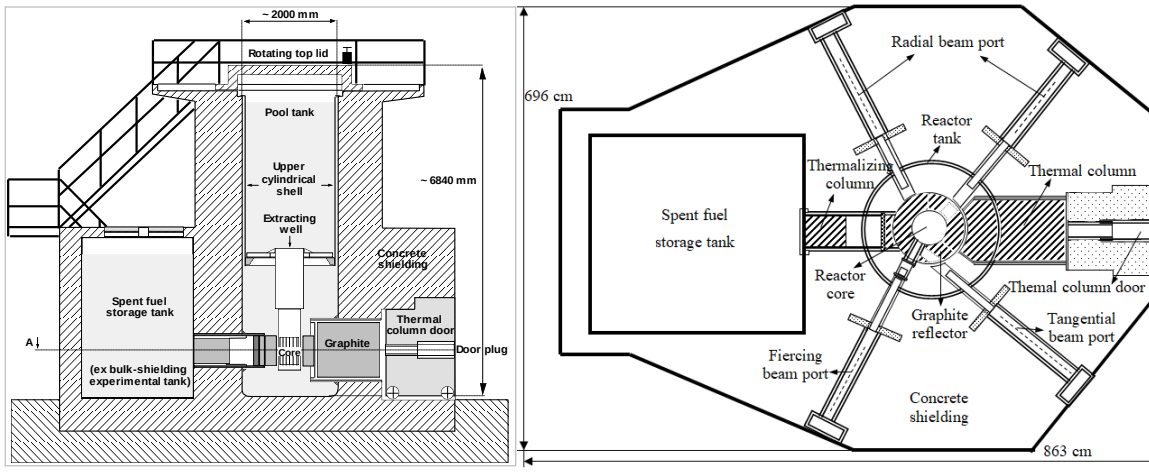

Fig. 1. Vertical (left) and horizontal (right) section views of the DNRR

Since the first start-up, the reactor core has been loaded with many different working configurations. The first working core configuration in 1984 after the reactor upgrading includes 89 high enriched uranium (HEU) fresh FAs with the central neutron trap, two safety rods, four shim rods, a regulating rod, two dry irradiation channels (at cell 7-1 and cell 13-2) and a wet irradiation channel (at cell 1-4). After the reactor core was fully converted from HEU to LEU fuel in 2011, the working core configuration has been loaded with 92 LEU FAs [1].

III. CALCULATION MODEL AND EXPERIMENTAL METHOD

A. Calculation model

The geometric structure of the DNRR remains to keep the structure of the TRIGA Mark II reactor especially having four horizontal beam tubes, which increases asymmetry and therefore, the neutron flux distribution varies significantly in its structures. To obtain the radioactivity in the structural components of the reactor, the material compositions, which generate long-lived activated products, were priorly selected for

calculating their specific activity inventory. The calculations to obtain the neutron energy spectrum, the density of neutron flux, the cross section or reaction rate of elements have been performed. Then, the neutron-activated product isotopes were calculated based on the historical operation of the reactor.

The MCNP5 computer code [6, 7] and ENDF/B-VII.1 library were used for calculation of the neutron flux distribution and the neutron spectrum at different locations in the structural materials of the reactor. To ensure the neutron fluxes error is less than 1% and the standard deviation of multiplication factor is smaller than 1.0E-05, in all calculation problems using the MCNP5 code the total particles are about 150 million including 20 source cycles to be skipped and 130 active cycles of tally accumulation. The calculation models for the DNRR in the MCNP5 code are shown in Fig. 2. The 3D calculation model for the entire reactor structures such as the reactor core, the aluminum components, the graphite reflector, the beam ports, the concrete shielding, etc. was basically described as very close to the real geometry of the DNRR. And the coordinate axis (0, 0, 0) is at the reactor core center.

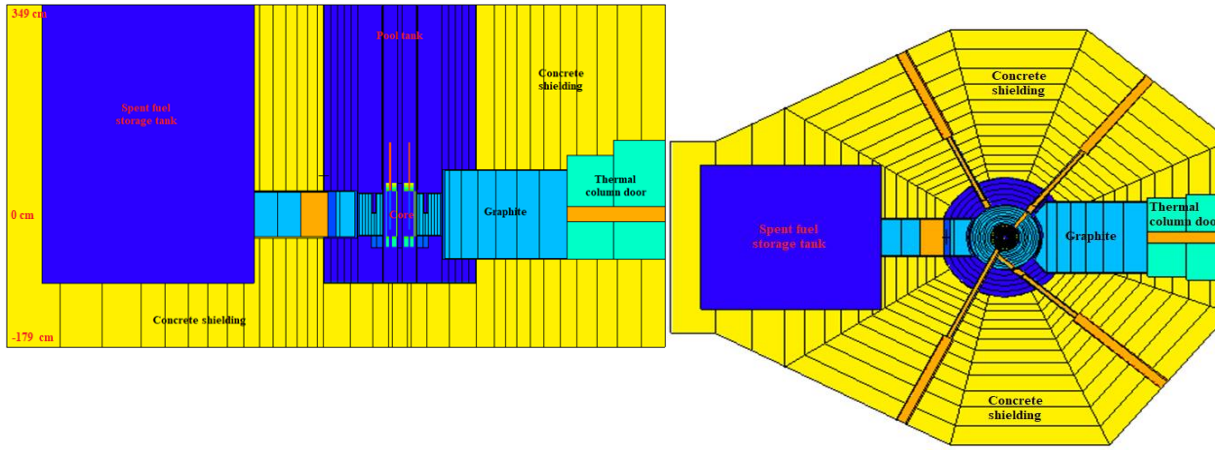

Fig. 2. The vertical (left) and horizontal (right) cross-section calculation models of the DNRR in the MCNP5 code (z axis in the left: from -179 cm to 349 cm; z = 0 cm at the reactor core center)

The ORIGEN2 code was used for calculating the activity of activated products in the structural materials of the reactor. The code has the ability to calculate the buildup, decay, and processing of radioactive materials [8]. The ORIGEN2 code is a revised version of ORIGEN and incorporates updates of the reactor models, cross sections, fission product yields, decay data, and decay photon data, as well as the source code.

Because the original libraries of the ORIGEN2 are not suitable to be applied for research reactors as the DNRR, the activation cross-sections of material compositions in the reactor have been determined by the MCNP5 code. The flowchart of the calculation scheme is shown in Fig. 3.

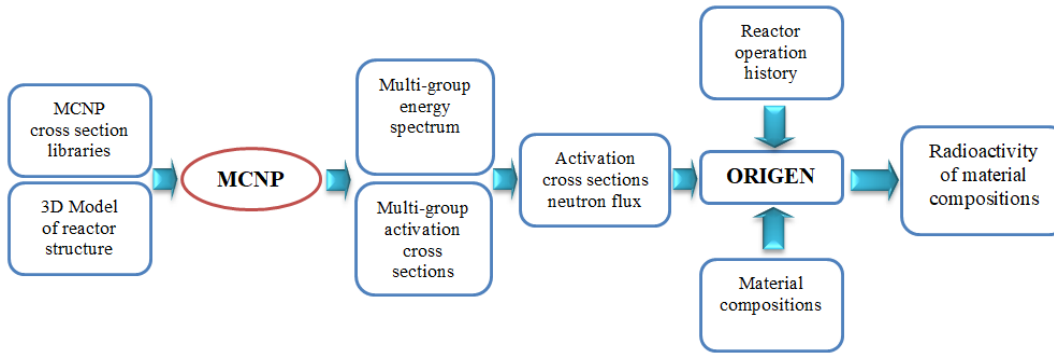

Fig. 3. Flowchart of calculation scheme

B. Experimental method

The neutron flux density is measured by the foil activation method. The reaction rate of a thin foil during activation is expressed by:

$$R = \Phi.\sigma_{act} \qquad (1)$$

where, Φ is neutron flux, σ_{act} is microscopic activation cross section.

The neutron flux distribution at the irradiation positions was obtained by using the foil activation method. The thermal neutron flux values were measured using the bare and cadmium covered gold foils [9]. The thermal neutron flux values were calculated by using the following equation:

$$\Phi_{th} = \frac{2.A.e^{-\lambda\tau}}{\sqrt{\pi}N_A\alpha\sigma_{0,act}G_{th}(1-e^{-\lambda T})}\sqrt{\frac{T_n}{T_0}}\left[\frac{A_b(T,\tau)}{m_b} - \frac{A_{Cd}(T,\tau)}{m_{Cd}}\right] \qquad (2)$$

where, m_b is mass of bare gold foil (g); m_{Cd} is mass of Cadmium covered gold foil (g); T is irradiation time (s); τ is cooling time after irradiation (s); t_m is real time measurement (s); $t_{m,eff}$ is effective time measurement (s); λ is decay constant of nuclide compound (s^{-1}); η is counting efficiency of detector; γ is gamma abundance factor; m is mass of foil (g); α is isotope enrichment; G_{th} is thermal neutron self-shielding factor; N_A is Avogadro constant; A is atomic number of isotope; G is ratio of isotope in foil; T_n is neutron temperature (K); T_0 is room temperature (293 K), A_b is activity of bare foil, and A_{Cd} is activity of cadmium covered foil.

The activity at time □ after finishing the irradiation [10]:

$$A_m\lambda(t_a,\tau) = A_i(1-e^{-\lambda t_a}) \qquad (3)$$

where, A_m is measured activity, A_i is actual activity at start of the measurement.

IV. RESULTS AND DISCUSSION

A. Validation of the computer codes and calculation method

The validation of the calculation model and experimental method was carried out by comparing the calculation results and experimental data as shown in Table II. The calculation values of the maximum thermal flux (E < 0.625 eV) at the reactor irradiation channels by using the MCNP5 code are in very good agreement with experimental data and the maximum difference is less than 7%.

Table II. Thermal neutron flux in the irradiation channels of the DNRR

Irradiation channel	Thermal neutron flux (n/cm^2.s)		Difference (%)
	Calculation	Experiment	
Neutron trap	2.21 x 10^{13}	(2.10 ± 0.13) x 10^{13}	5.2
Channel 7-1	6.70 x 10^{12}	(6.28 ± 0.58) x 10^{12}	6.7
Channel 13-2	6.47 x 10^{12}	(6.05 ± 0.52) x 10^{12}	6.9

To obtain impurity radioactivity in the aluminum components, which are the main structures of the DNRR, some samples were taken from three different positions (-32.5, -17.5, and -2.1 cm) of the former 6061 aluminum guiding tube of irradiation channel 13-2 as shown in Fig. 4. Specific activity measurements of the samples were performed by using HPGe gamma spectrometer (Canberra GC5019 type) and the main long-lived activated product being ^{60}Co isotope with its specific activities are shown in Table III. The error of specific activity of ^{60}Co, ^{63}Ni, ^{55}Fe, and ^{59}Ni isotopes is less than 3% from evaluation of the ORIGEN2.1 code.

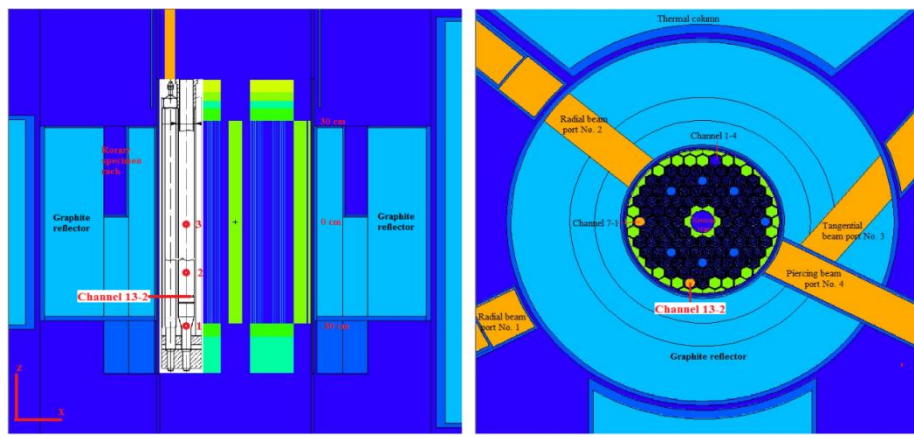

Fig. 4. Vertical (left) and horizontal (right) models of the reactor core

Table III. Activity of ^{60}Co isotope in the guiding tube of irradiation channel 13-2

Samples	Position in aluminum tube (cm)	^{60}Co specific activity (Bq/g)		Difference (%)
		Calculation	Experiment	
1	-32.5	2.77×10^4	$(1.86\pm0.17)\times10^4$	33
2	-17.5	1.35×10^5	$(9.99\pm0.19)\times10^4$	26
3	-2.1	1.71×10^5	$(1.28\pm0.16)\times10^5$	25

The neutron flux and energy spectrum in the different locations of the structural components, as well as the activation cross-sections of interested compositions were also determined by using the MCNP5 code. The calculated results of neutron flux in the aluminum guiding tube of the irradiation channel 13-2 are shown in Fig. 5, in which

values at positions of -32.5, -17.5, and -2.1 cm are corresponding with the values of the aluminum samples were taken for the radioactivity measurement shown in Table III.

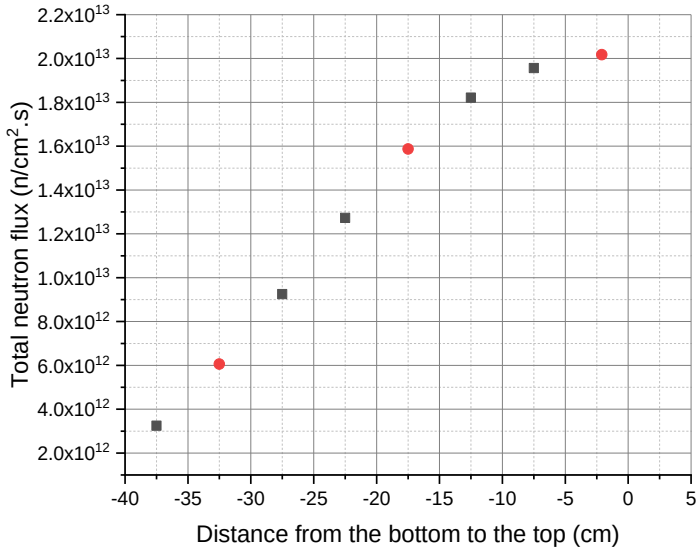

Fig. 5. The calculation results of neutron flux distributions in axial direction of channel 13-2 aluminum guiding tube

Based on the calculation results of activation cross-sections and one group neutron flux in the structural components in combination with the historical operation data of the DNRR [11], the specific radioactivities in the structural components such as irradiation channel 13-2 were determined by ORIGEN2 code. The obtained calculation and experimental results of the ^{60}Co isotope specific activity in the guiding tube of the channel 13-2 are presented in Table III, in which, the calculation values have maximum difference lower than 33% compared to experimental data. The main reason for the difference between calculation results and experimental data is 6061 aluminum alloy having only a very small and irregular impurity of ^{59}Co isotope according to each manufactured batch. In the data published by the IAEA and US laboratories for the ^{59}Co isotope impurity in 6061 aluminum alloy, the ^{59}Co isotope impurity is very variable and often estimated to be less than a few ppm without a specific value [12]. The calculation results performed in this work are mainly predictive but having good enough reliability and fidelity in decommissioning calculation.

Table IV and Fig. 6 show the calculated radioactivity of some long-life isotopes in the channel 13-2 aluminum guiding tube. The main isotopes of the activated materials are ^{60}Co, ^{63}Ni, ^{55}Fe, and ^{59}Ni, in which, the highest specific activity is of the ^{60}Co isotope. The specific activities of ^{55}Fe and ^{59}Ni isotopes are very low compared to those of ^{60}Co isotope. In the case of ^{63}Ni, although its activity is relatively high and its half-life is long because ^{63}Ni isotope mainly generates beta-rays with low energy of 0.067 MeV that is not so dangerous in the case of radioactive waste management in decommissioning.

Table IV. Calculation results of activity of long-life isotopes in the guiding tube of irradiation channel 13-2

Isotope	Half-life (year)	Specific activity (Bq/g) vs. decay time (year)			
		1	3	6	10
^{55}Fe	2.73	2.18×10^{2}	1.28×10^{2}	6.20×10^{1}	1.98×10^{1}
^{60}Co	5.27	3.17×10^{5}	2.43×10^{5}	1.710×10^{5}	9.70×10^{4}
^{59}Ni	5.80×10^{1}	5.80×10^{1}	5.80×10^{1}	5.80×10^{1}	5.80×10^{1}
^{63}Ni	100.1	8.50×10^{3}	8.37×10^{3}	8.21×10^{3}	7.95×10^{3}

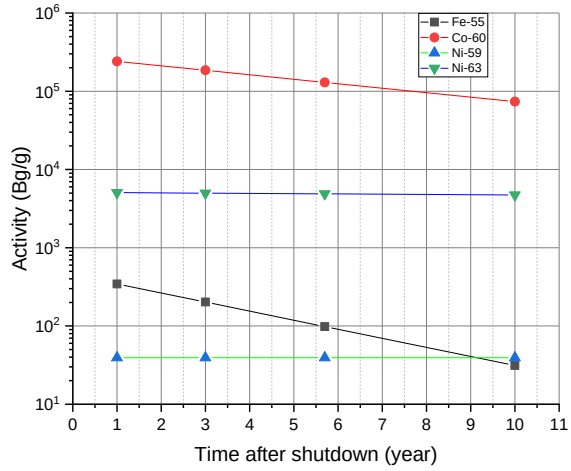

Fig. 6. The calculation specific activity of long-lived isotopes in the channel 13-2 aluminum guiding tube

B. Calculation of neutron flux in the structural components

The calculation of neutron flux distributions in all structural components at a nominal power of 500 kW of the reactor has been implemented using the MCNP5 code. The tasks are important in the first step for estimating specific activities of structural materials inside the reactor core and in the next step for evaluating accumulated activities of other structural materials of the reactor. Tables V, VI, and VII present the calculation results of one group of neutron flux distributions in the structural components including the graphite reflector, the thermal column, and the concrete shielding, respectively. Fig. 7 illustrates those calculated results in the horizontal and vertical views of the reactor.

Table V. The calculated results of neutron flux (n/cm^2.s) distribution in the graphite reflector

z axis (cm)	Distance from the reactor core (cm)				
	24	32	39	47	51
25.0	3.74×10^{12}	(*)	2.56×10^{12}	1.06×10^{11}	9.03×10^{11}
7.5	5.89×10^{12}	4.36×10^{12}	3.20×10^{12}	1.73×10^{12}	1.29×10^{12}
-7.5	6.19×10^{12}	4.37×10^{12}	4.24×10^{12}	2.10×10^{12}	1.54×10^{12}
-25.0	4.65×10^{12}	3.86×10^{12}	2.80×10^{12}	1.57×10^{12}	1.16×10^{12}

(*) Rotary specimen rack in the graphite reflector

Table VI. The calculated results of neutron flux (n/cm^2.s) distribution in the thermal column

z axis (cm)	Distance from the reactor core (cm)				
	92	114	140	168	201
40	4.52×10^{10}	2.13×10^{10}	8.30×10^{9}	2.75×10^{9}	6.54×10^{8}
10	1.23×10^{11}	5.24×10^{10}	1.92×10^{10}	6.19×10^{9}	1.39×10^{9}
-10	1.40×10^{11}	5.60×10^{10}	2.00×10^{10}	6.34×10^{9}	1.47×10^{9}
-40	5.78×10^{10}	2.50×10^{10}	9.14×10^{9}	2.91×10^{9}	6.77×10^{8}

Table VII. The calculated results of neutron flux (n/cm^2.s) distribution in the concrete shielding at beam tube 4

z axis (cm)	Distance from the reactor core (cm)					
	105	146	180	221	271	332
205	6.08×10^{4}	2.21×10^{4}	2.54×10^{3}	3.86×10^{2}	8.68×10^{1}	3.58×10^{1}
175	5.54×10^{5}	1.64×10^{5}	4.49×10^{4}	1.07×10^{4}	3.91×10^{3}	1.15×10^{3}
150	4.61×10^{6}	2.56×10^{6}	8.38×10^{5}	2.17×10^{5}	9.92×10^{4}	2.83×10^{4}
135	6.28×10^{7}	3.62×10^{7}	1.13×10^{7}	2.96×10^{6}	1.52×10^{6}	4.45×10^{5}
20	6.55×10^{8}	3.33×10^{8}	9.38×10^{7}	2.40×10^{7}	1.32×10^{7}	3.94×10^{6}
10	4.79×10^{9}	1.73×10^{9}	4.50×10^{8}	1.06×10^{8}	5.97×10^{7}	1.83×10^{7}
-5	Beam tube 4					
-15	1.51×10^{10}	4.87×10^{9}	1.19×10^{9}	2.10×10^{8}	1.19×10^{8}	3.72×10^{7}
-35	3.48×10^{9}	1.33×10^{9}	3.44×10^{8}	8.26×10^{7}	4.77×10^{7}	1.42×10^{7}
-55	2.75×10^{8}	1.55×10^{8}	4.45×10^{7}	1.13×10^{7}	6.45×10^{6}	1.81×10^{6}
-85	1.59×10^{7}	1.01×10^{7}	3.25×10^{6}	8.40×10^{5}	4.32×10^{5}	1.17×10^{5}
-120	1.30×10^{6}	4.75×10^{5}	1.47×10^{5}	3.95×10^{4}	1.70×10^{4}	4.41×10^{3}

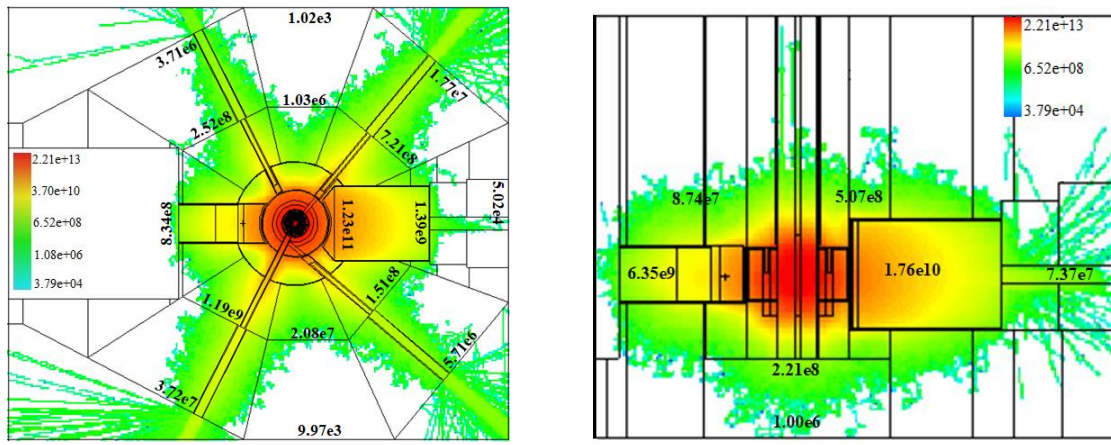

Fig. 7. The neutron flux (n/cm^2.s) distributions in the horizontal (left) and vertical (right) views of the DNRR [2, 7]

The spatial distribution of neutron flux from the center to outside shielding concrete of the reactor was obtained from MCNP5 code to meet the requirements for evaluation of the specific activity of structural materials. The results show that the high neutron flux distributions are located around the reactor tank within 1 m in shielding concrete, thermal and

thermalizing columns from 1×10^6 to 1×10^{11} n/cm^2.s. Outside four beam tubes and other positions in shielding concrete, the neutron fluxes only reach values from 1×10^3 to 1×10^4 n/cm^2.s.

V. CONCLUSION

The obtained calculation results and experimental data provide a general view of the neutron fluxes distribution from inside the reactor core to the biological shielding and along four beam tubes of the DNRR. In which, the highest neutron flux of 2.21×10^{13} n/cm^2.s is at the core center, 1.76×10^{10} n/cm^2.s in the thermal column, 6.35×10^9 n/cm^2.s in the thermalizing column and 2.21×10^8 n/cm^2.s in the outer side of the biological shielding. The specific activity and the neutron flux in the 6061 aluminum guiding tube at 13-2 channel are also provided and the discrepancy between calculation and experimental data is smaller than 33% and 7%, respectively. The main specific activity of long-lived activation products is of the ^{60}Co isotope with the highest value of 1.28×10^5 Bq/g in the position of -2.1 cm from core center. The experimental data also show that the specific activities of other long-life isotopes, such as ^{55}Fe, ^{59}Ni, are relatively low if the structural components will not be activated about six years before decommissioning and dismantling actions.

Even though the calculation performed in this work is mainly predictive, the obtained results are important and have enough reliability to provide data for formulating the decommissioning plan of the DNRR. The main expected study in the next step should be systematic measurement and calculation of the activity inventory in all structural components of the DNRR.

ACKNOWLEDGEMENTS

This research is funded by the Ministry of Science and Technology (MOST) of Vietnam under grant number ĐTCB.05/20/VNCHN, signed on 02/01/2020.

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