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DECOMMISSIONING OF THE VVR-S RESEARCH REACTOR – RADIOLOGICAL CHARACTERIZATION OF THE REACTOR BLOCK

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Abstract. The paper describes the methodology used for the radiological characterization of the VVR-S reactor block that is the major contributor to the overall radioactive inventory of the reactor. It combines theoretical calculations and gamma spectrometry measurements of representative samples. Based on this methodology, the radionuclide inventory of the reactor block and the quantities of radioactive waste and materials which can be released from regulatory control were estimated. Finally, the methodology was validated by dose rate measurements and calculations using MicroShield code.

Key words: decommissioning, neutron activation, radiological characterization.

1. INTRODUCTION

The VVR-S nuclear reactor from Magurele is a research reactor with a maximum thermal power of 2 MW using distilled light water as moderator, coolant and reflector. The reactor was commissioned in 1957 and dedicated to nuclear physics research and radioisotopes production. Until 1984 the reactor was operated by nuclear fuel type EK-10. From 1984, this fuel was replaced by S-36, that was used until 1997 when the reactor was definitively shut-down. On average, the reactor was operated 5 days per week at full or variable power levels. During 40 years of operation, the VVR-S reactor produced 9.59 GWd.

The radiological characterization of the VVR-S research reactor is an important stage in its decommissioning [1, 2]. Neutron activated materials from the reactor block represent, by far, the major contribution to the overall radioactive inventory of the reactor. The reactor block is mainly composed of reactor vessels, experimental channels, reactor core, mobile thermal column, upper rotating lids, in-core fuelling mechanism and biological shield. All these components are made of four types of materials: aluminum, cast iron, concrete and graphite.

This paper describes the methodology used for the radiological characterization of the VVR-S reactor block. The methodology was validated for the key radionuclide ^{60}Co, in the case of reactor core and thermal column, by dose rate measurements and calculations using MicroShield code [3]. The radionuclide inventory of the reactor block was estimated using both measured and calculated results. The quantities of radioactive waste and slightly activated materials which can be released from regulatory control were also estimated. As the results show, more than 80 % of reactor block materials can be released because their radioactive content is below the clearance levels for unrestricted re-use.

2. INITIAL ESTIMATION OF THE RADIONUCLIDE INVENTORY

Calculations for determining the induced activity in the reactor block components were performed in the initial step of the decommissioning process [4] using dedicated computer codes. Thus, for the space-energy distribution of the neutron flux inside the reactor block the DORT code was used, which solved the neutron transport equation in *R-Z* geometry [5]. A cylindrical coordinate system was assumed by taking its center in the middle of the core and the angular coordinate (θ) as the angle between the axis of the horizontal channel no. 9 and the radial line (see Fig. 1). The DORT model of the geometry is illustrated in Fig. 2, while Fig. 3 illustrates the spatial distribution of the thermal neutron flux inside the reactor block as obtained by using the DORT code.

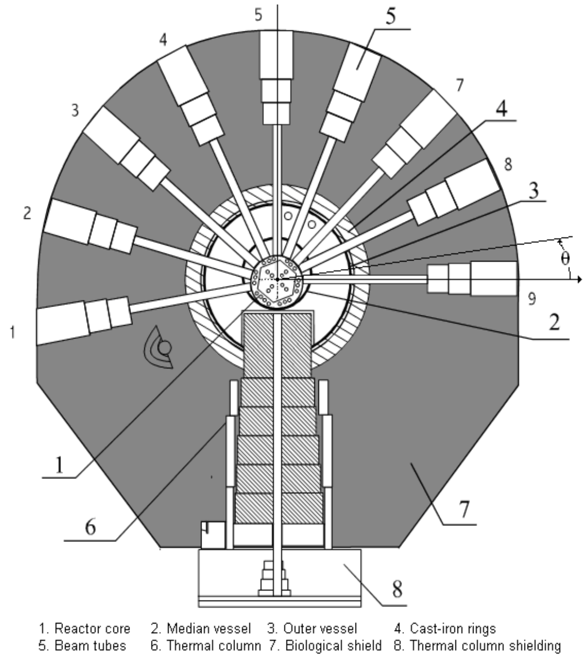

Fig. 1 – Horizontal cross section of the VVR-S reactor.

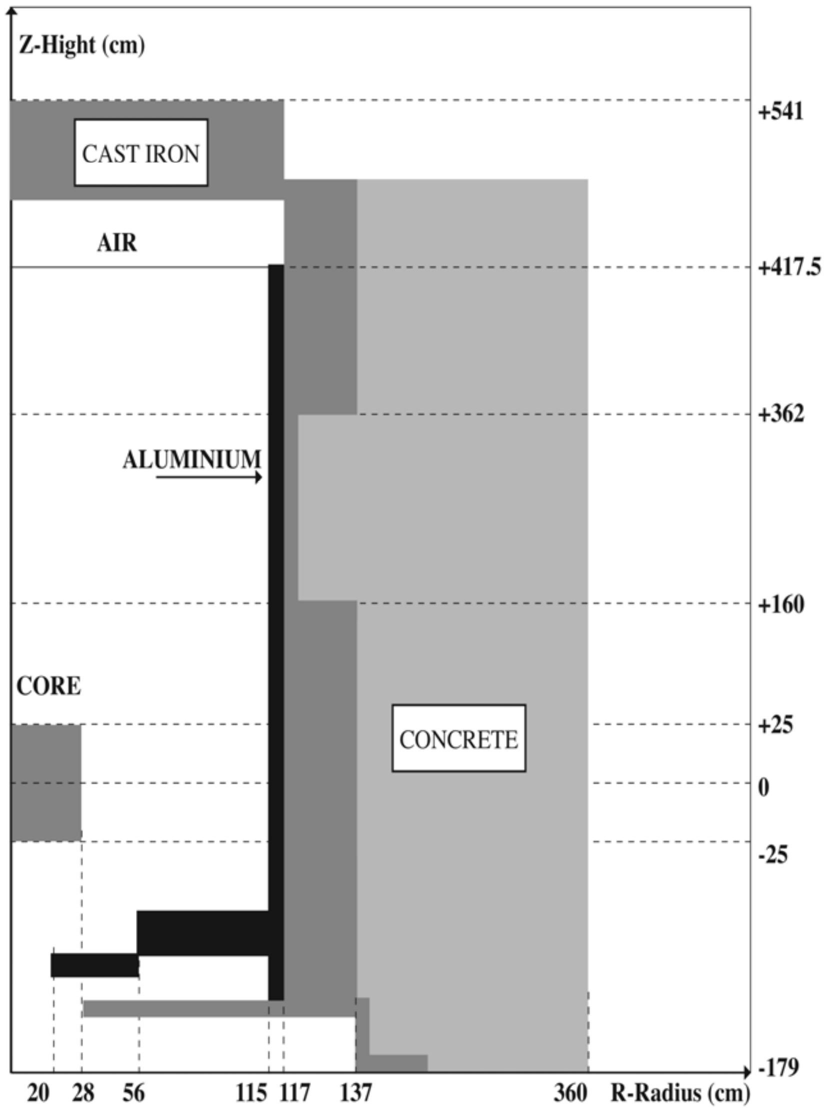

Fig. 2 – DORT model of the VVR-S reactor block.

Activation calculations were performed with ORIGEN code [6] using as inputs space-energy distribution of the neutron flux (previously calculated with the DORT code), nuclear and decay data, material composition (impurities) as well as the history of operation.

Although the theoretical calculations based on the DORT and the ORIGEN codes were affected by large errors due to: (*i*) inaccuracy of data on impurity concentrations; (*ii*) inability of the two dimensional DORT code to adequately treat the neutron transport in the complex geometry of the VVR-S reactor structures; (*iii*) the use by ORIGEN code of effective cross sections specific to LWR cores, we have obtained a set of conservative preliminary estimations by taking the maximal values of all impurity concentrations used in the calculations.

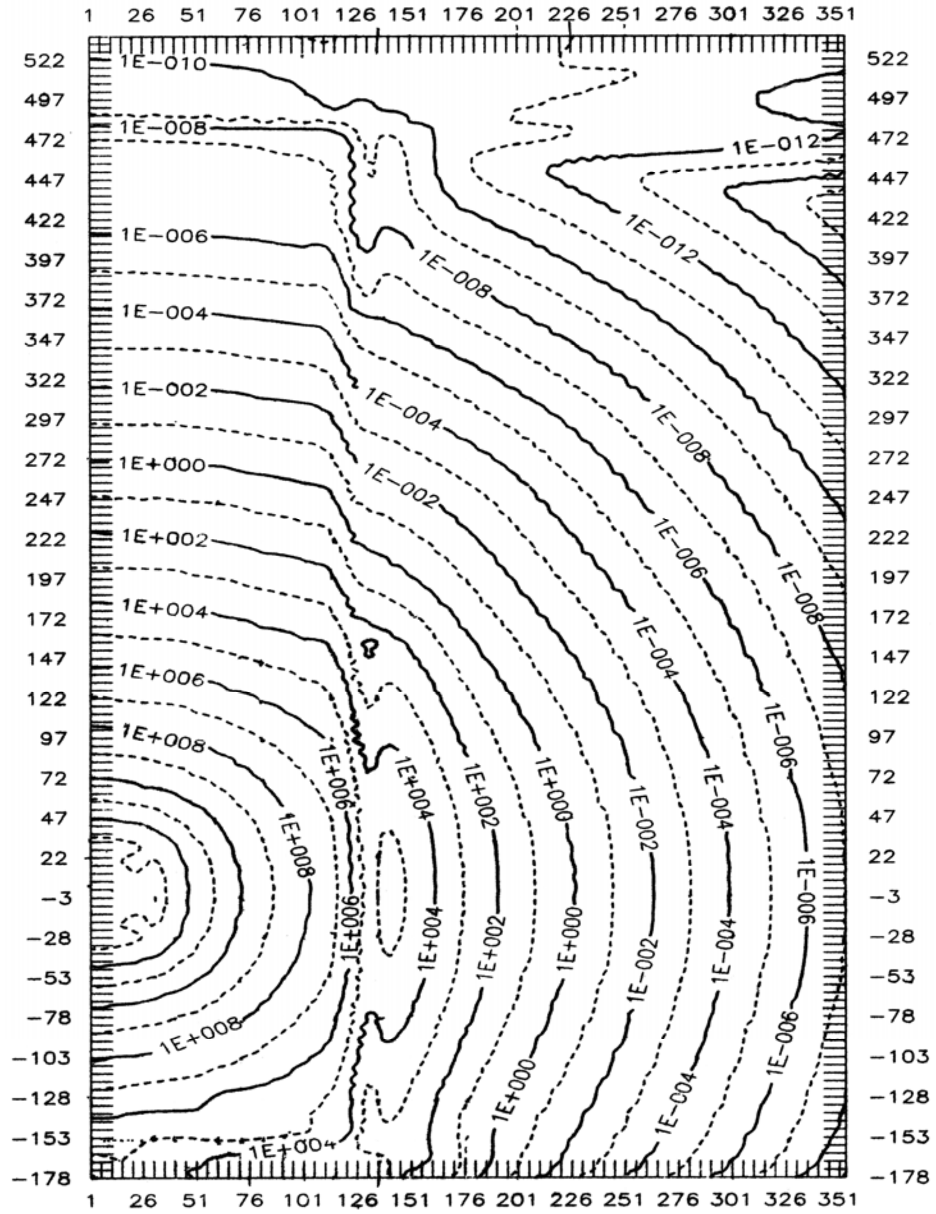

Fig. 3 – Spatial distribution of the thermal neutron flux obtained with DORT code.

3. ESTIMATING THE RADIONUCLIDE INVENTORY

3.1. MEASUREMENT RESULTS

In order to obtain a reasonable estimation of the radionuclide inventory, more samples were collected from the VVR-S reactor block and their specific activity was measured by gamma spectrometry whose final results concerning samples taken from reactor core, reactor base plate and thermal column, their coordinates as well as the corresponding thermal neutron fluxes are reproduced in Table 1.

Table 1

Specific activity of samples taken from reactor core, base plate and thermal column

Sample material	Sampling point	Thermal neutron flux ($n\ cm^{-2}\ s^{-1}$)	Radionuclide	Specific activity* (Bq/g)
Aluminum	Reactor core (R=15 cm, Z=-25 cm, θ=0°)	8.0E+12	^{60}Co	(3.53±0.30)E+04
			^{154}Eu	(1.40±0.20)E+03
			^{155}Eu	(7.75±1.00)E+02
	Thermal column (R=43 cm, Z=0 cm, θ=-70°)	3.0E+12	^{60}Co	(1.55±0.20)E+04
			^{154}Eu	(1.50±0.20)E+03
	Thermal column (R=47 cm, Z=0 cm, θ=-60°)	1.5E+12	^{60}Co	(9.91±1.00)E+03
			^{154}Eu	(3.50±0.40)E+02
	Thermal column (R=53 cm, Z=0 cm, θ=-50°)	5.0E+11	^{60}Co	(5.10±0.50)E+03
			^{154}Eu	(3.60±0.40)E+02
Cast iron	Reactor base plate (R=67, Z=-170, θ=30°)	1.0E+03	^{60}Co	(0.25±0.10)E+00
Graphite	Thermal column (R=40 cm, Z=0 cm, θ=-90°)	5.0E+12	^{60}Co	(1.66±0.20)E+03
			^{154}Eu	(3.20±0.30)E+01
	Thermal column (R=44 cm, Z=5 cm, θ=-65°)	1.0E+12	^{60}Co	(5.60±0.60)E+02
			^{154}Eu	(0±0.10)E+00

*Reference data: 4.12.2010

At the same time, three horizontal core-drill samples were taken at different locations along the circumference of the biological shield. Thus, cylinders of approximately 5 cm in diameter and of lengths exceeding two meters were extracted. From the cylinders and beginning with the most activated side (sample no. 1), subsamples with lengths of 5 cm were cut and examined by gamma spectrometry. Table 2 reproduces the specific activity of each subsample as well as its coordinates.

Table 2

Specific activity of subsamples taken from the biological shield

Cylinders	Specific activity of subsample* (Bq/g)							
	No. 1 R∈(137÷142)		No. 2 R∈(147÷152)		No. 3 R∈(157÷162)		No. 4 R∈(167÷172)	
	^{60}Co	^{152}Eu	^{60}Co	^{152}Eu	^{60}Co	^{152}Eu	^{60}Co	^{152}Eu
Cylinder 1 R∈(137÷360) Z=0, θ=11°	16.0±2.0	12.0±1.0	9.2±1.0	4.7±0.5	2.2±0.2	1.2±0.1	0±0.1	0±0.1
Cylinder 2 R∈(137÷360), Z=0, θ=-30°	5.4±0.5	2.4±0.2	1.6±0.2	0.6±0.1	0.4±0.1	0±0.1	0±0.1	0±0.1
Cylinder 3 R∈(137÷360), Z=160, θ=11°	0±0.1	0±0.1	0±0.1	0±0.1	0±0.1	0±0.1	0±0.1	0±0.1

*Reference data: 4.12.2010

3.2. RADIONUCLIDE INVENTORY ESTIMATION USING MEASURED DATA AND THEORETICAL CALCULATIONS

In the case of easy-to-measure radionuclides (^{60}Co, ^{152}Eu, ^{154}Eu and ^{155}Eu), their specific activity was calculated by means of gamma spectrometric data and the neutron flux results obtained with DORT code. Thus, the specific activity $\Lambda(R,Z,\theta)$ for a given material in a point of coordinates (R,Z,θ) can be calculated with the following equation

$$\Lambda(R,Z,\theta)=\Lambda_{sp}\frac{\sigma_{th}\Phi_{th}(R,Z,\theta)+\sigma_{ep}\Phi_{ep}(R,Z,\theta)}{\sigma_{th}\Phi_{th}^{(sp)}+\sigma_{ep}\Phi_{ep}^{(sp)}}, \tag{1}$$

where σ_{th} and σ_{ep} are activation cross-sections (average values) for thermal and epithermal neutrons, $\Phi_{th}(R,Z,\theta)$ and $\Phi_{ep}(R,Z,\theta)$ are the thermal and epithermal neutron fluxes in the point (R,Z,θ), $\Phi_{th}^{(sp)}$ and $\Phi_{ep}^{(sp)}$ represent the thermal and epithermal neutron fluxes in the sampling point and Λ_{sp} is the specific activity (Bq/kg) of the sample. Note that Eq. 1 is valid only for homogenous samples (including trace elements).

In the case of aluminum, concrete and graphite components, Eq. 1 becomes

$$\Lambda(R,Z,\theta)\cong\Lambda_{sp}\frac{\Phi_{th}(R,Z,\theta)}{\Phi_{th}^{(sp)}} \tag{2}$$

as for these components $\Phi_{ep} / \Phi_{th} \cong \Phi_{ep}^{(sp)} / \Phi_{th}^{(sp)}$.

It should be noted that the specific activity for the cast iron ring was calculated by means of Eq. 1, and may have large errors because calculated values of $\Phi_{th}^{(sp)}$ and $\Phi_{ep}^{(sp)}$ are inaccurate.

The total activity Λ_T of a given component from the reactor block (composed of a given material) was computed by means of the equation:

$$\Lambda_T = \rho \iiint_V \Lambda\left(R, Z, \theta\right) \mathrm{d}V \,, \tag{3}$$

where V is the volume of the component and ρ is the density of the material.

To calculate the total activity, the following approximations were used: (*i*) the components of the reactor block are homogeneous (ρ=constant); (*ii*) the content and the concentration of impurities do not vary within the volume of components composed of the same material; (*iii*) the specific activity $\Lambda\left(R, Z, \theta\right)$ does not depend on the angular coordinate θ.

Therefore, materials in the close vicinity of the horizontal beam-tubes may have high activation levels due to neutron deflections but these anomalies were neglected.

In the case of hard-to-detect radionuclides, the specific and the total activity of reactor block components were calculated by means of the scaling factors method using ^{60}Co as the reference radionuclide. To estimate the scaling factors, theoretical calculations, gamma spectrometric results and data from the literature were used [7, 8, 9]. Thus calculated values of the scaling factors are shown in Table 3 for all materials of the reactor block.

Table 3

Values of the scaling factors

No.	Radionuclide	Scaling factors*			
		Aluminum	Cast iron	Concrete	Graphite
1	^{14}C	0.000	0.000	0.000	16.20
2	^{3}H	0.000	0.000	18.50	52.90
3	^{55}Fe	6.100	0.220	0.025	0.630
4	^{63}Ni	30.00	0.110	0.000	0.150
5	^{59}Ni	0.330	0.000	0.000	0.000

*Reference date: 4.12.2010

The final radionuclide inventory of the reactor block regarding both easy-to-measure and hard-to-detect nuclides is presented in Table 4.

Table 4

Radionuclide inventory of the reactor block

	Radionuclide	Activity* (Bq)			
		Aluminum m=4.8 Mg	Cast iron m=143 Mg	Concrete m=550 Mg	Graphite m=5.3 Mg
1	^{60}Co	1.10E+10	2.81E+09	3.50E+08	1.63E+08
2	^{152}Eu	0	0	2.52E+08	0
3	^{154}Eu	4.80E+08	0	0	3.13E+06
4	^{155}Eu	1.44E+08	0	0	0
5	^{14}C	0	0	0	2.65E+09
6	^{3}H	0	0	6.46E+09	8.62E+09
7	^{55}Fe	6.70E+10	6.30E+08	8.70E+06	1.03E+08
8	^{63}Ni	3.30E+11	3.00E+08	0	2.58E+07
9	^{59}Ni	3.63E+09	0	0	0
10	TOTAL (Bq)	4.12E+11	3.74E+09	7.07E+09	1.16E+10
11	TOTAL		4.34E+11 Bq	(11.7 Ci)	

*Reference date: 4.12.2010

4. RADIOACTIVE WASTE ESTIMATION

4.1. MODELING THE SPECIFIC ACTIVITY DISTRIBUTIONS

The use of simple analytical models for specific activity distributions is useful for the practical application of clearance regulations and radioactive waste classification. As a result, an exponential model was developed to calculate easily with sufficient accuracy the specific activity as a function of spatial coordinate in the biological shield, thermal column and cast-iron ring.

For a given radionuclide, $\Lambda(R,Z)$ of the biological shield and the cast iron ring, can be approximated with the following exponential function [10]

$$\Lambda(R,Z)=\Lambda_0 \exp\left[-\alpha(R-R_0)-\beta Z^2\right], \quad (4)$$

where $\Lambda_0=\Lambda(R_0,0)$ Bq/g, R_0=137 cm, α and β are parameters of the exponential distribution. In the case of thermal column, a new coordinate system was assumed by taking its origin in the center of the circular front face of the thermal column. In this case, we have

$$\Lambda(r,z)=\Lambda_0 \exp\left(-\beta r^2-\alpha z\right) \quad (5)$$

where $\Lambda_0 = \Lambda(0,0)$ Bq/g, α and β are parameters of the exponential distribution and Z axis of the system is identical with the symmetry axis of the thermal column. In Eq. 4 and 5, Λ_0 depends on the angular coordinate. To simplify calculations, a single conservative value for Λ_0 was used in each equation for all values of the angular coordinate.

The parameters α and β of exponential functions were determined by least-square fitting by using both calculated and measured values of the specific activity. Values of these parameters are given in Table 5.

Table 5

Values of the parameters α and β

Values of the parameter α (cm^{-1})			Values of the parameter β (cm^{-2})		
Biological shield	Thermal column	Cast-iron ring	Biological shield	Thermal column	Cast-iron ring
8.0E-02	5.0E-02	15.0E-02	5.0E-04	1.1E-03	5.0E-04

For the components constructed from aluminum, the modeling of the specific activity distributions is not useful because there are too many components. In this case, the specific activity distributions were calculated for each component without using an analytical model.

4.2. CLEARANCE OF MATERIALS AND RADIOACTIVE WASTE ESTIMATION

We consider that a large quantities of radioactive waste will be produced during decommissioning and dismantling of the VVR-S reactor. Therefore it is of a great importance, for the planning of the decommissioning process, to estimate the quantities of radioactive waste and slightly activated materials from the reactor block that can be released from regulatory control.

In Romania, clearance levels for unconditional release are stated in the NDR-02 norm [11]. According to NDR-02, the clearance criterion can be defined as:

$$\sum_{i=1}^{n} \frac{\Lambda_i}{CL_i} < 1, \tag{6}$$

where *n* is the number of radionuclides in the material, Λ_i and CL_i are the specific activity and clearance level corresponding to the *i*-th nuclide.

In Table 6 are shown the clearance levels for all radionuclides included in the inventory of the reactor block.

Table 6

Clearance levels of radionuclides from the reactor block

Clearance levels (Bq/g)								
^{60}Co	^{152}Eu	^{154}Eu	^{155}Eu	^{3}H	^{14}C	^{55}Fe	^{63}Ni	^{59}Ni
1	7	5	30	200	20	30	70	200

Based on Eq. 6 and by using the specific activity distributions, it is not difficult to estimate the quantities of radioactive waste. Hence, the numerical values of estimated quantities (Mg) for both radioactive waste and materials for unconditional release are reported in Table 7.

Table 7

Radioactive waste estimation

Material	Mass (Mg)	Radioactive waste* (Mg)	Materials for free release* (Mg)	Waste/Mass* (%)
Aluminum	4.8	1.8	3.0	37.5
Cast iron	143.0	65.0	78.0	45.5
Concrete	550.0	50.0	500.0	9.0
Graphite	5.3	3.5	1.8	66.0
Total	703.1	120.3	582.8	17.1

*Reference date: 4.12.2010

5. VALIDATING THE ACTIVITY CALCULATIONS BY DOSE RATE MEASUREMENTS

In the case of the reactor core and thermal column, the results of the specific activity estimation for the key radionuclide ^{60}Co were validated by dose rate measurements. The contribution to the dose rate of ^{154}Eu and ^{155}Eu was neglected because is smaller than 5%.

Regarding the thermal column, it is worth mentioning that it is made of 5 graphite discs placed on a mobile truck. The dose-rate of the first graphite disc was only calculated and measured because this disc contains more than 95% from the total activity of the thermal column. Fig. 4 shows the geometry for dose rate measurements containing the measuring points.

By using the MicroShield code, the dose rates were also calculated for each measuring points, final results being reproduced in Table 8. As both computed an measured data agree within an interval of 10%, we can consider this agreement as a validation for the methodology used for the radionuclide inventory estimation.

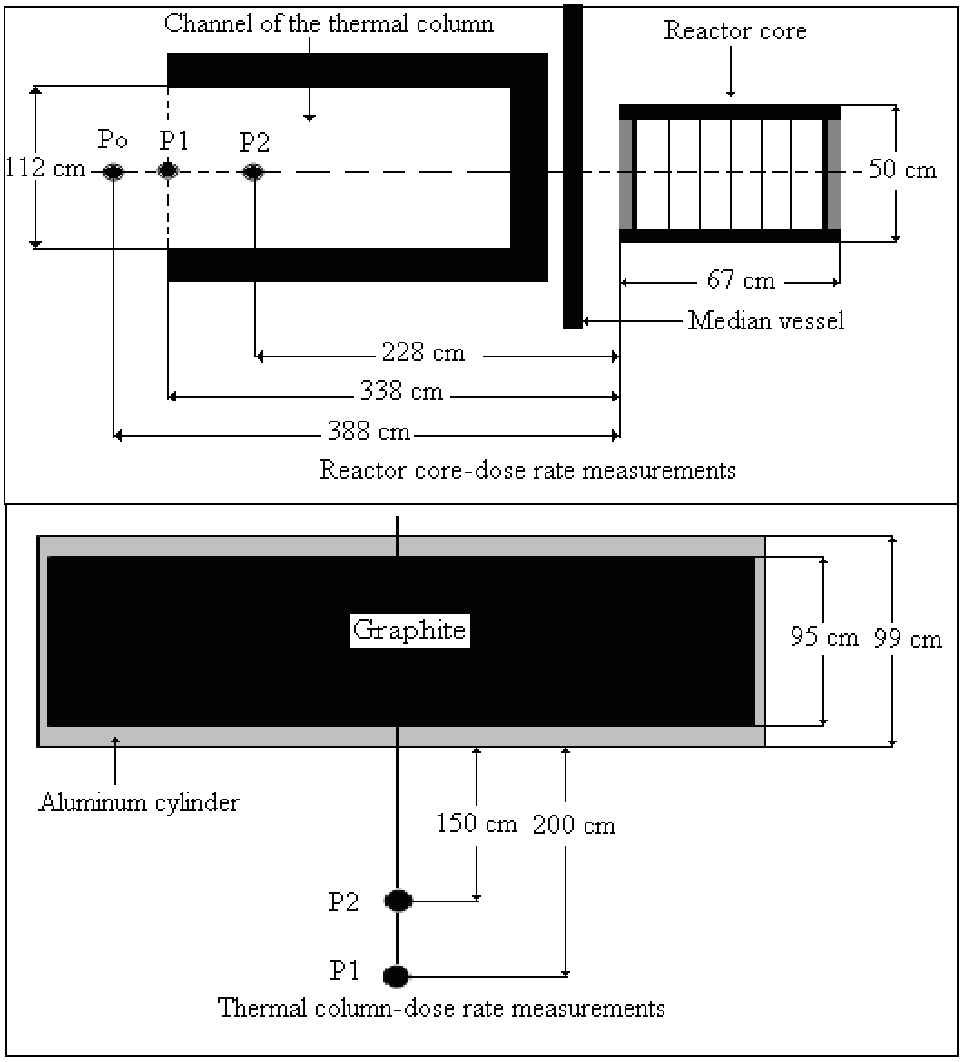

Fig. 4 – Geometry of dose rate measurements.

Table 8

Computed (C) and measured (M) values of dose rates

Dose-rate (mSv/h)									
Reactor core						Thermal column			
Po		P1		P2		P1		P2	
M	C	M	C	M	C	M	C	M	C
0.460	0.440	0.550	0.580	1.000	1.110	0.016	0.015	0.033	0.030

6. CONCLUSIONS

The paper describes the methodology used for the radiological characterization of the VVR-S nuclear reactor block that combines the results of theoretical calculations and gamma spectrometric measurements. The methodology was validated for the key radionuclide ^{60}Co, in the case of reactor core and thermal column, by dose rate measurements and calculations using MicroShield code.

Based on this methodology, the radionuclide inventory of the reactor block and the quantities of radioactive waste and materials that can be released from regulatory control were estimated. These estimates show that more than 80 % of reactor block materials can be released because their radioactive content is below the clearance levels for unrestricted re-use.

In summary, the radiological characterization of the reactor block provides useful results for both planning and performing the decommissioning activities of the VVR-S research reactor.

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