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Activity Analysis of Primary Coolant in TRIGA MARK II Research Reactor

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ABSTRACT

Detailed in-situ gamma spectroscopy measurements of specific activity of short lived activation products in primary coolant in TRIGA MARK II reactor were carried out. Continuous on-line sampling system was set to sample short lived radionuclides in the water. An important contribution to elevated radiation levels during reactor operation comes from activation of short lived radionuclide N-16 in the primary coolant. Spatial concentration distribution in the pool of short lived radionuclides was determined during reactor operation. The measured values were compared to the results of Monte Carlo simulations. The activity measurements directly above the core were performed at different power levels of the reactor.

It was shown that short lived activation products can be used as tracers for the study of primary coolant flow. A linear relation between specific activity of N-16 and the power level was established which shows that N-16 can be used for continuous monitoring of the reactor power level.

1 INTRODUCTION

Extensive measurements of radionuclide concentrations in the primary cooling water were performed in the TRIGA MARK II reactor at Jožef Stefan Institute [1]. Research reactor can be operated in steady state mode (maximum 250 kW) or in a pulse mode (maximum 1000 MW). The cooling of the core is performed by natural convection of water. During the operation the water coolant flows through the reactor core and is exposed to an intense neutron flux (~ 1E+13 n $cm^{-2}s^{-1}$).

The activity induced in cooling water comes from radioisotopes produced in neutron reactions with water molecules, dissolved gases (Table 1) and impurities in water. The most important activation products are N-16, O-19, N-17, H-3, C-14 and Ar-41. Radioactive Ar-41 is produced in the reactor coolant water (in the water-dissolved Ar-40) and in the air voids near the core. These isotopes decay by emission of beta particles, gamma rays or neutrons. The reactor primary water contains small amounts of impurities as it is constantly being filtered. Hence the activation of impurities in the water presents a negligible activity compared to the induced activity of the water.

It was shown that short-lived activation product in water present approximately 14 % to total dose rate above the reactor pool [2], [3]. The main contributors are prompt gamma rays from fission and neutron capture.

Table 1: The most important radionuclides in the reactor coolant

Target isotope	Natural abundance (%)	Reaction	Activation product	$t_{1/2}$	Relevance
O-16	99.76	(n,p), fast neutron	N-16	7.13 s	6.129 MeV, 7.117 MeV gamma
O-17	0.04	(n,p), fast neutron	N-17	4.14 s	1 MeV neutron
O-18	0.20	(n,γ), thermal neutron	O-19	26.9 s	0.197 MeV, 1.357 MeV gamma
H-2	0.015	(n,γ), thermal neutron	H-3	12.3 y	low energy beta
N-14	99.63	(n,p), fast neutron	C-14	5500 y	low energy beta
Ar-40	99.6	(n,γ), thermal neutron	Ar-41	1.83 h	1.294 MeV gamma

Radiologicaly the most important is $^{16}O(n,p)^{16}N$ reaction and contributes to elevated radiation levels due to high energy gamma radiation. Due to very short half-life it is important only during reactor operation. Some contribution to the dose rate can be measured also from O-19 and Ar-41. Since H-3 and C-14 are not gamma emitters they do not cause external exposure. N-17 has a very low reaction rate and consequently its decay by neutron emission has a negligible contribution to external exposure.

2 PRIMARY COOLANT ACTIVITY MEASUREMENTS

Regular weekly activity measurements of the primary coolant are performed at research reactor. The main purpose is to detect any leakage of the fuel by looking for the presence of fission products. The collected samples are subsequently measured by HPGe spectrometer. In this way very short-lived radionuclides cannot be detected.

In this study we focused mostly on the activity measurements of short lived gamma emitters. For this purpose an on-line sampling system of primary coolant combined with in-situ gamma spectrometry was used. Direct activity measurements during reactor operation were performed on the reactor platform above the reactor pool.

On-line sampling was performed by pumping water from the pool with peristaltic pump with a constant flow rate and using silicon pipe (Figure 1). For the purpose of better efficiency and good geometry the activity of water was measured in the sampling vial connected to the pipe. As the pipe was 9 m long we took into account the activity correction factor for the decay of radionuclides when travelling from the sampling position to the vial. With this sampling arrangement it was possible to collect samples from almost any location inside the pool.

In-situ gamma spectrometry was performed with HPGe coaxial detector produced by Canberra (40 % relative efficiency). The detector was characterized by the manufacturer. The efficiency for sample geometry was calculated with ISOCS/LABSOCS software [4]. Because of elevated radiation levels at the measuring location the detector was shielded by use of ISOCS shield system with 50 mm Pb rings. Measurements and analysis were performed by GENIE 2000 software. The measuring energy range was extended up to 7000 keV in order to measure high energy gamma rays from N-16. In Figure 2 a typical gamma-ray spectrum of primary coolant during reactor operation is shown.

Figure 1: Sampling vial in front of the shielded HPGe detector. The glass vial was connected to silicon pipe. The water was pumped through the vial with the peristaltic pump.

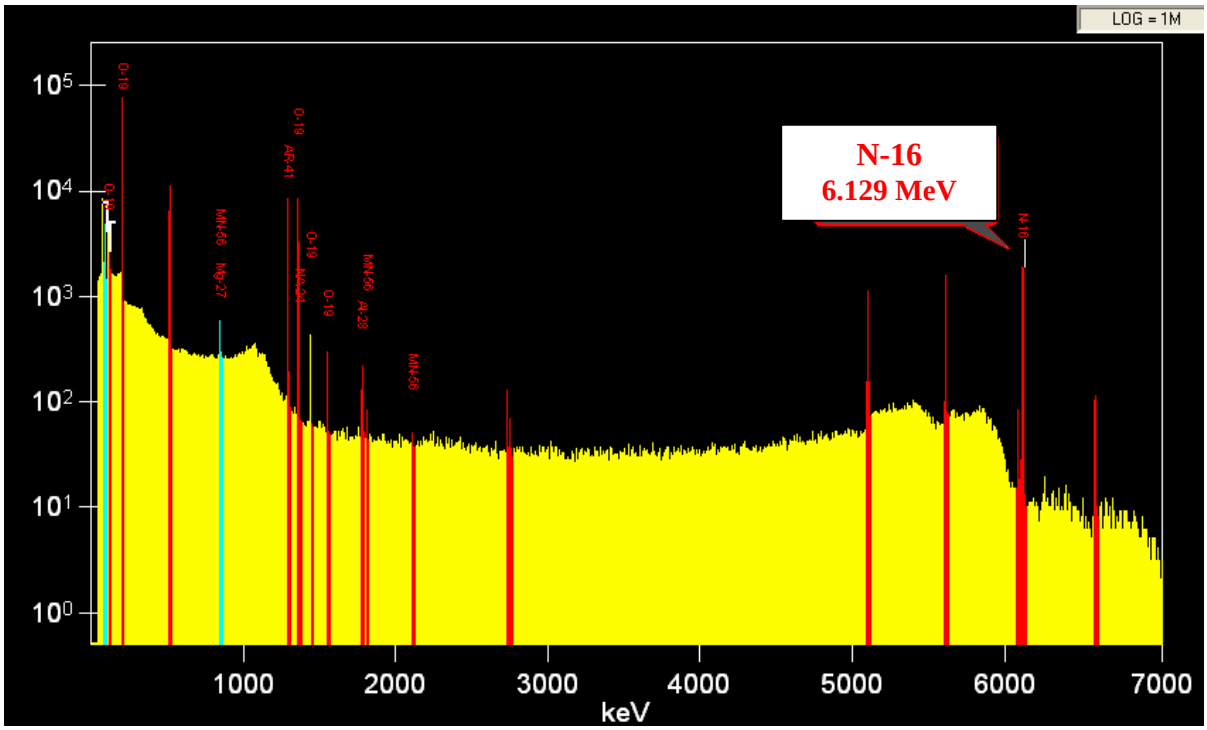

Figure 2: The gamma ray energy spectrum of primary coolant

2.1 Distribution of radionuclides in the reactor pool

Specific activity of short lived radionuclides was determined at 23 locations in the reactor pool (Figure 3). During the measurements the reactor was operating at constant power level of 250 kW. Each measurement lasted 1000 s so the average specific activity in this time interval was determined.

The spatial distribution of specific activity of the most important radionuclides is presented in the figures below (Figure 4 and Figure 5). The highest specific activity was measured directly above the reactor core (~5 cm above the top grid plate). The N-16 concentration ((7.7 ± 0,3) E+11 Bq/m^3) was at least two orders of magnitude higher then the activity of other radionuclides. The activated water travels towards the surface of the pool due to natural convection. Short lived radionuclides decay very rapidly and also mix with non

activated water. Less then 1 m above the core we observed a strong decrease of concentration. Similar distribution was seen with O-19 measurements (Figure 5). This can be explained by horizontal mixing of active and non active water above the core. At a depth of 1 m we also observed (especially with O-19) higher activity above reflector then above the core. This is an indication of horizontal movement of water 1m below the pool surface. We can also see that activity decreases with the depth at the side of the pool. This indicates the downward movement of water.

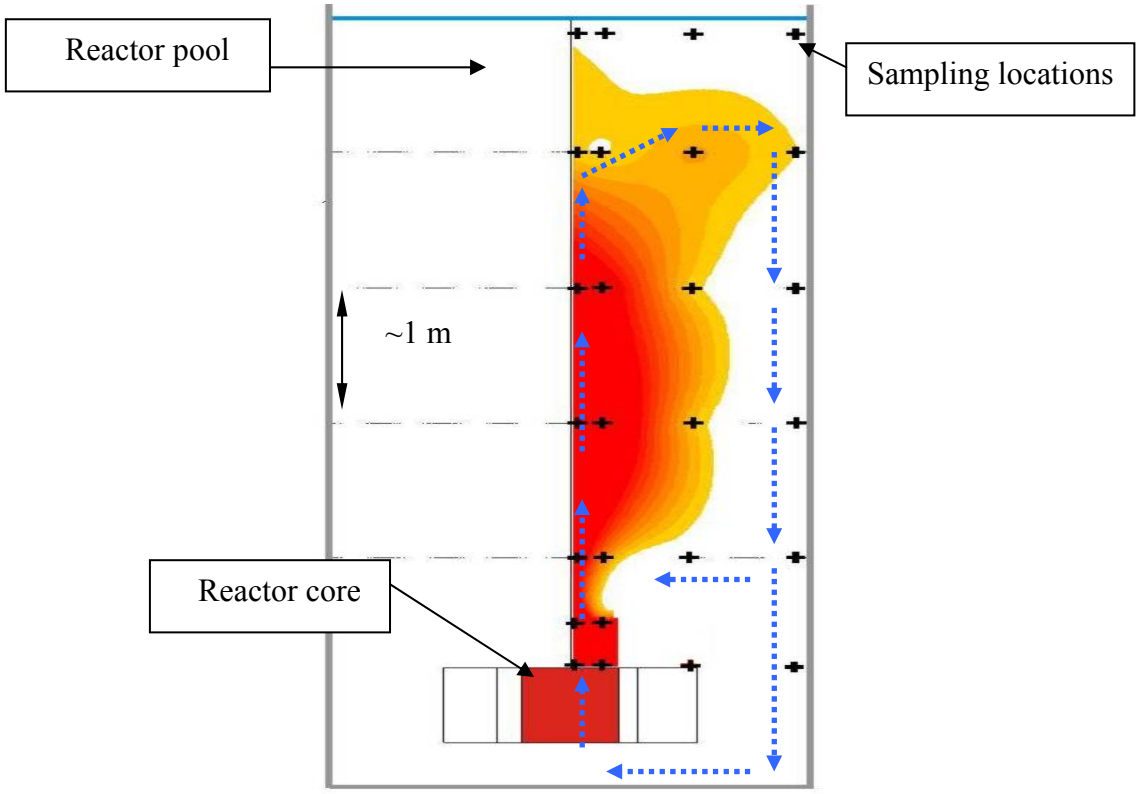

Figure 3: Vertical cross section of reactor pool and the sampling locations. Graphical presentation of specific activity distribution and the flow of water due to natural convection and mixing

2.2 Activity and power correlation

The specific activity of N-16 and O-19 was measured at different power levels of reactor operation. The water was sampled near the top grid plate of the core above the fuel elements. From Figure 6 one can see a linear relation between specific activity of N-16 and the total power. Similar linear relation was also obtained with O-19 measurements.

N-16 is produced by fast neutrons via (n,p) reaction, which has a threshold energy of 9 MeV. Measuring N-16 provides information on the fast neutron flux and is approximately proportional to the average power density at the measuring location. N-16 specific activity measurements can be used for monitoring fission rate and total reactor power level. On the other hand O-19 specific activity gives us information on the thermal neutron flux since it is produced with radiative capture reaction (n,γ) mainly induced by thermal neutrons .

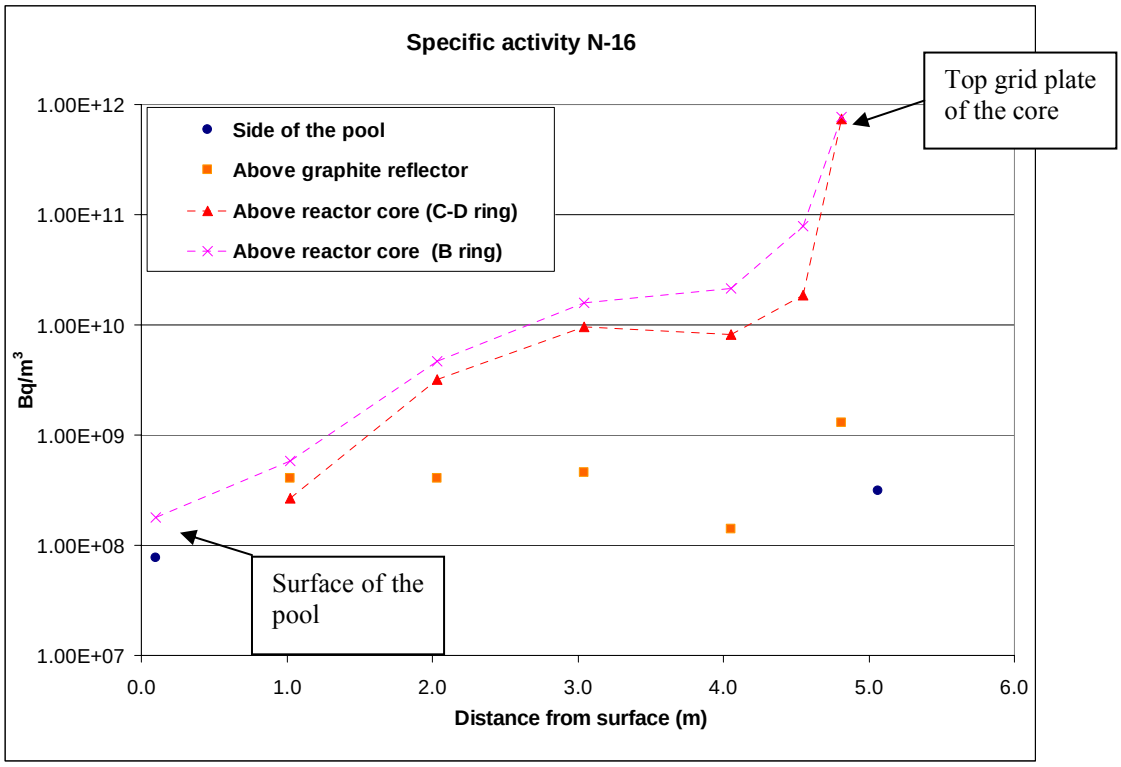

Figure 4: Distribution of N-16 specific activity in the reactor pool. Reactor was operating at 250 kW

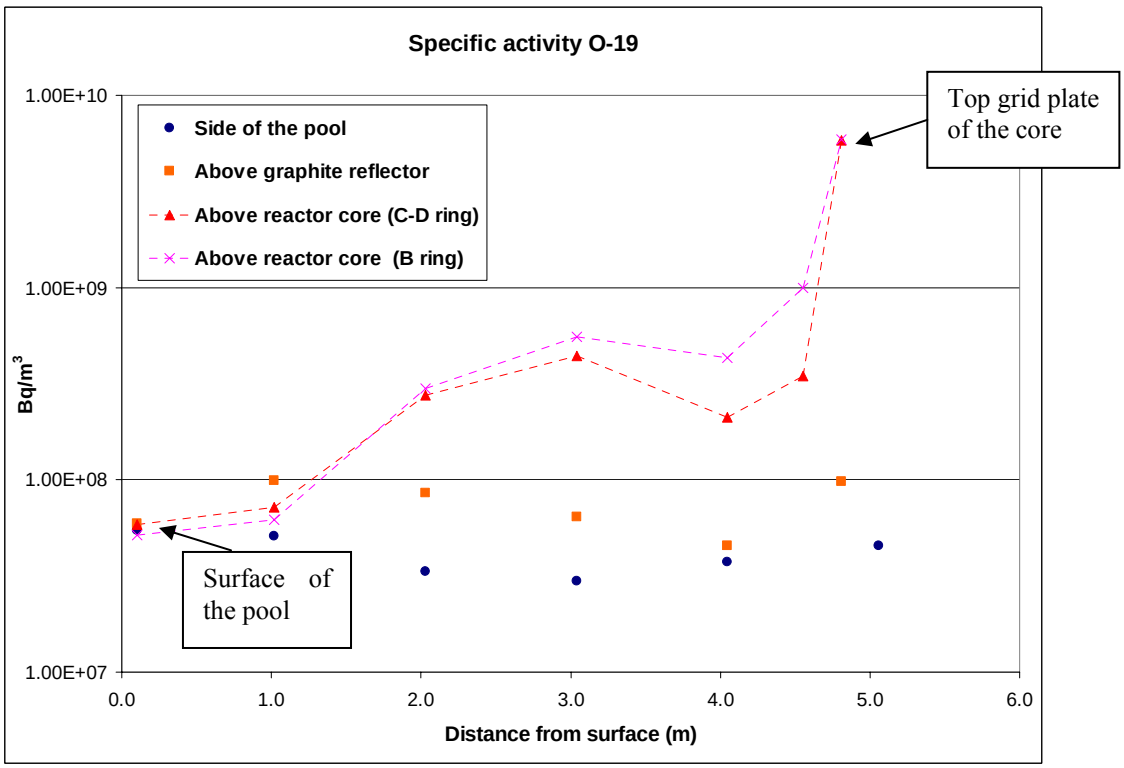

Figure 5: Distribution of O-19 specific activity in the reactor pool. Reactor was operating at 250 kW

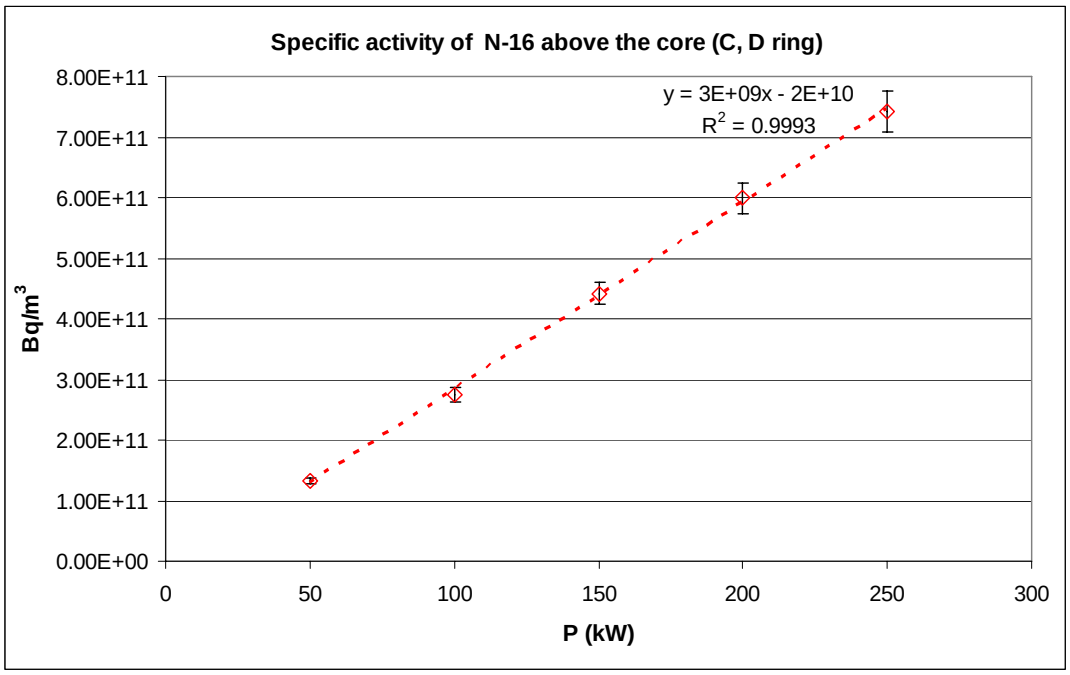

Figure 6: Dependence of specific activity of N-16 on the reactor power level

3 PRIMARY COOLANT ACTIVITY CALCULATION

Measured specific activities above the top grid plate of the core of N-16 and O-19 were compared to calculated values. Both short lived activation products are produced from neutron activation of oxygen in the water. For calculations information on the neutron flux $\phi_n(\vec{r},E)$ in the water channels in the core was needed. In general, the reaction rates $F(\vec{r})$ are calculated from equation:

$$F(\vec{r}) = \int_0^\infty N_0 \sigma(E) \phi(\vec{r},E) dE \qquad (1)$$

N_0 is number density of target elements and $\sigma(E)$ is the microscopic cross section. Reaction rates were calculated for $^{16}O(n,p)^{16}N$ and $^{18}O(n,\gamma)^{19}O$ reactions. Computing was done with Monte Carlo simulation (MCNP) based on the existing model of TRIGA MARK II core [5].

The water channel was divided into 5 regions from bottom to top grid plate (Figure 7). For each region an average reaction rate F_i was calculated. It was presumed that the water is exposed only in the channels between both grid plates an all other contributions are negligible.

For short lived radionuclides we can presume that nonactivated water enters the water channel at the bottom grid. Activity of the water on top of each region can be expressed as:

$$A_{R1} = F_{R1}(1 - e^{-\lambda \cdot t_{R1}}) \qquad (2)$$

$$A_{R2} = A_{R1} e^{-\lambda \cdot t_{R2}} + F_{R2}(1 - e^{-\lambda \cdot t_{R2}}) \qquad (3)$$

and for any region:

$$A_{Rn} = A_{Rn-1} e^{-\lambda \cdot t_{Rn}} + F_{Rn}(1 - e^{-\lambda \cdot t_{Rn}}) \quad (4)$$

A_{Rn-1} is the activity on top of the previous region. The duration of exposure was calculated based on the information on the known water velocity in the channels (11.6 cm/s) [3], [6]. The results of specific activities are presented in tables 2 and 3. The values for the last top region were compared to the measured results above the top grid plate. As can be seen from the last column, showing the ratios between the calculations and measurements, the agreement between the calculated and measured values is quite good.

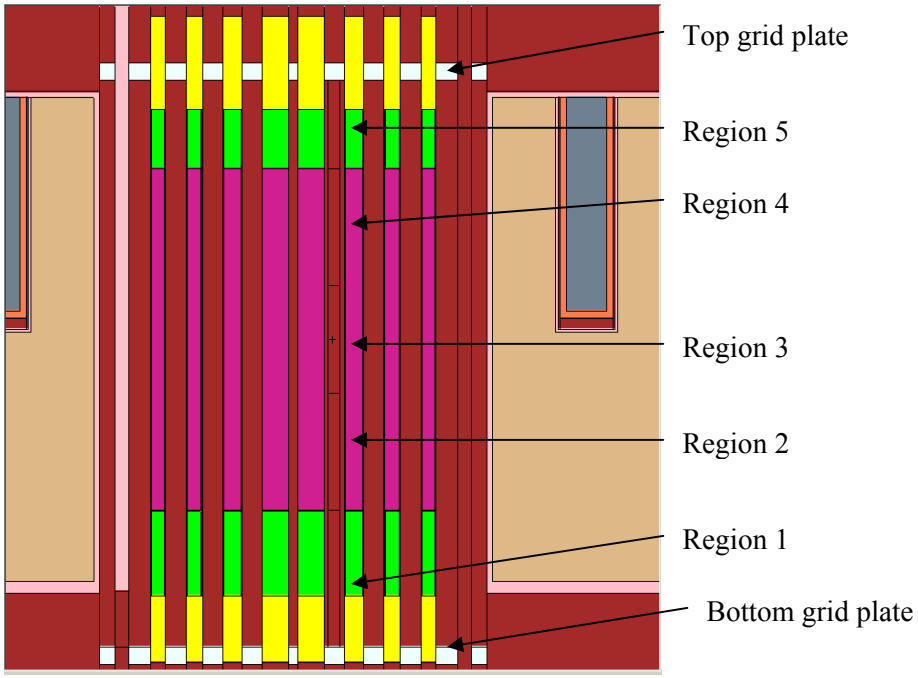

Figure 7: Vertical cross section of the core. Water channel between the fuel elements was divided into 5 regions

Table 2: Calculated and measured specific activities of N-16

B1-B2 position	Region 1	Region 2	Region 3	Region 4	**Region 5**	**Measured (top grid plate)**	R
Activity (Bq/m^3)	7,46E+10	4,79E+11	9,45E+11	1,18E+12	**1,14E+12**	**7,65E+11**	1,5
C3-D3 position	Region 1	Region 2	Region 3	Region 4	**Region 5**	**Measured (top grid plate)**	R
Activity (Bq/m^3)	6,34E+10	3,86E+11	7,71E+11	1,01E+12	**9,68E+11**	**7,42E+11**	1,3

Table 3: Calculated and measured specific activities of O-19

B1-B2 position	Region 1	Region 2	Region 3	Region 4	**Region 5**	**Measured (top grid plate)**	R
Activity (Bq/m3)	5,75E+08	2,18E+09	4,10E+09	5,40E+09	**5,62E+09**	**5,90E+09**	0,95
C3-D3 position	Region 1	Region 2	Region 3	Region 4	**Region 5**	**Measured (top grid plate)**	R
Activity (Bq/m3)	5,03E+08	1,85E+09	3,47E+09	4,58E+09	**4,77E+09**	**5,83E+09**	0,82

4 CONCLUSION

Detailed gamma spectroscopy measurements of specific activity of short lived activation products in primary coolant in TRIGA MARK II reactor were carried out. Continuous on-line sampling system was set up in such a way to sample activated water from almost any location in the reactor pool. According to the expectation the most important radionuclide is N-16 which half life is 7.14 s and decays by emission of high energy gamma rays. Other less important radionuclides identified were O-19, Ar-41 and some radionuclides from activation of impurities in water.

Spatial concentration distribution in the pool of short lived radionuclides was determined during reactor operation. It was shown that short lived activation products can be used as tracers for the study of primary coolant flow.

The activity measurements directly above the core were performed at different power levels of the reactor. A linear relation between specific activity of N-16 and the power was established which shows that N-16 can be used for continuous monitoring of the reactor power level.

Theoretical model of TRIGA reactor core based on Monte Carlo simulations (MCNP) was used to calculate the reaction rates for production of short lived N-16 and O-19. The calculated activities were compared to the measured values and there was a very good agreement.

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