NEUTRON ACTIVATION MEASUREMENTS IN RESEARCH REACTOR CONCRETE SHIELD

Tomaž Žagar, Matjaž Ravnik
"Jožef Stefan" Institute
Reactor Physics Division
Jamova 39, SI-1000 Ljubljana, Slovenia
Tomaz.Zagar@ijs.si, Matjaz.Ravnik@ijs.si

Matjaž Božič
Nuclear Power Plant Krško
Vrbina 12, SI-8270 Krško, Slovenia
Matjaz.Bozic@nek.si

ABSTRACT

The results of activation measurement inside TRIGA research reactor concrete shielding are given. Samples made of ordinary and barytes concrete together with gold and nickel foils were irradiated in the reactor body. Long-lived neutron-induced gamma-ray-emitting radioactive nuclides in the samples were measured with HPGe detector. The most active long-lived radioactive nuclides in ordinary concrete samples were found to be ^{60}Co and ^{152}Eu and in barytes concrete samples ^{60}Co, ^{152}Eu and ^{133}Ba. Measured activity density of all nuclides was found to decrease almost linearly with depth in logarithmic scale.

1   INTRODUCTION

Decommissioning and dismounting of nuclear research reactors after their service life is raising new questions about residual radioactivity present in the reactor shielding material ([1] and [2]). In a small pool type research reactor (like TRIGA) after defuelling and reactor core structure removal the most important remaining activated part is the biological shield of the reactor. Several studies of residual radioactivity in the concrete shields of accelerator facilities have appeared in open literature, but there is little information available on this subject for research reactors. Activation of concrete biological shield material depends on the concrete properties and neutron flux in the concrete shield. Our studies ([3] and [4]) show that for accurate prediction of activation experimental determination of actual activation rates in the concrete reactor body is needed.

To determine neutron activation inside the reactor concrete body a special concrete sample-holder for irradiation has been developed. In this sample-holder different samples can be irradiated at different concrete depths.

In the paper the neutron activation measurements in the biological shield of the TRIGA Mark II reactor in Ljubljana for four different irradiation conditions are presented. Experiments in the reactor body included two sets of gold and nickel foils irradiation and a set of barytes concrete samples and ordinary concrete samples irradiation. Barytes concrete sample was taken from barytes blocks around the reactor body. To compare and verify

experimental results, neutron flux in the biological shield was also calculated using TORT code [5]. Calculation model and method is described in more detail in a paper also presented at this conference [6]. Experimental results in this paper show good agreement with the calculated results.

2   EXPERIMENT DESCRIPTION

2.1   Irradiation

2.1.1 Irradiation Set-up and Equipment Description

All samples were irradiated in the horizontal experimental beam port number 4 of TRIGA Mark II research reactor at "Jožef Stefan" Institute in Ljubljana. Beam port number 4 is a radial beam port and extends from the reflector assembly through the water and concrete to the outer face of the shield structure. A special sample-holder was designed to fit in the beam port 4 to reproduce irradiation conditions inside the concrete reactor body structure. The sample-holder (Figure 1) is made of aluminum tube and is filled with concrete cylinders. Before the irradiation various samples (e.g. neutron activation foils) can be placed at eight different depths between the concrete cylinders. After irradiation and appropriate cooling time the sample-holder is removed from the beam port 4 and the irradiated samples are removed from the holder through the opening at the front end. During the irradiation the opening is closed with aluminum lid.
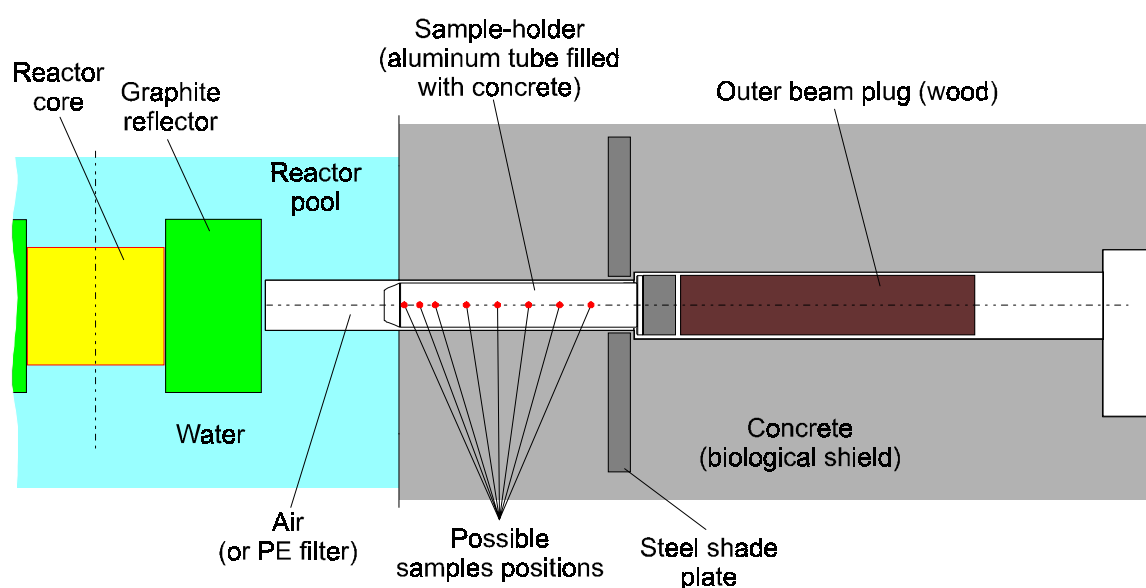
Figure 1: Sample-holder position during irradiation inside the horizontal experimental beam port number 4 of the TRIGA Mark II research reactor at "Jožef Stefan" Institute. Air region between graphite reflector and sample-holder can be filled with polyethylene (PE) filter.

The void region in the beam port 4 between graphite reflector and sample-holder is normally filled with air. Alternatively it can be filled with polyethylene (PE) filter designed to fit on the front end of the holder. PE filter is made of aluminum and filled with 30 cm long polyethylene cylinder. Polyethylene with high hydrogen concentration acts as a good neutron moderator and replaces neutron moderation in water between graphite assembly and concrete structure. Some characteristic dimensions and composition data for PE filter and sample-holder are given in Table 1.

Table 1: Sample-holder and polyethylene (PE) filter dimensions and material data.

Component	Dimension [cm]	Material	Density [g cm^{-3}]
Sample-holder			
Outer diameter	15,00 (+0,00, - 0,01)		
Total length	85,5		
Aluminum tube			
Inner diameter	13,0	Aluminum	2,70
Length	80,0	(ISO: AlMgSi 1)	
Concrete cylinders			
Diameter	12,95 (+0,00, - 0,10)	Concrete	2,25 (±0,05)
Length	10,0 and 5,0		
Aluminum tube lid			
Length	5,0	Aluminum	2,70
Polyethylene filter			
Outer diameter	15,00 (+0,00, - 0,01)		
Length attached to sample-holder	30,0		
Aluminum tube			
Inner diameter	13,0	Aluminum	2,70
Length	33,0	(ISO: AlMgSi 1)	
Polyethylene			
Diameter	12,90 (+0,00, - 0,01)	Polyethylene	0,95 (±0,03)
Length	30,0	$(CH_2)_n$	

2.1.2 Irradiation conditions

Altogether 32 samples in 4 groups were irradiated in beam port 4 from the beginning of December 2000 to the end of May 2001. Two groups of gold and nickel foils were irradiated for approximately one day at full reactor power at 250 kW without interruptions. The second set of gold and nickel foils was irradiated with PE filter in place.

The other two groups of samples consisted of concrete samples and were irradiated for approximately 100 hours at full reactor power. The maximum achievable uninterrupted irradiation length for TRIGA Mark II reactor at "Jožef Stefan" Institute is just 30 hours once per week and so 100 hours long irradiation was interrupted several times.

In all four cases after the end of irradiation the sample-holder was left in the beam port 4 for at least 24 hours to allow the short radioactive nuclides in the sample-holder material and gamma radiation from the core to decay. Detailed sample and irradiation conditions data are given in Table 2. Pure gold foils used in the experiments were in the form of small thin discs (8 mm in diameter and 50 μm thick). Gold wire was made of 0,1 % diluted gold in aluminum wire (1 mm diameter). Nickel wire was made of 99,982 % pure nickel in form of thin wire (0,75 mm in diameter). Barytes concrete sample was taken from barytes concrete shielding blocks around TRIGA Mark II reactor in Ljubljana. The ordinary concrete sample was prepared at Faculty of Civil and Geodetic Engineering, University of Ljubljana. Both types of concrete samples were crushed into powder and filled into small cylindrical vials for irradiation. Outer dimensions of the vial were 1.0 cm in diameter and 1.2 cm in height. Concrete samples that had not been exposed to neutron radiation were used to determine natural background gamma ray spectra. An empty irradiated vial was also measured to confirm that there are no long-lived radioactive nuclides present in the vial material. No special gamma ray peaks above detector background were found in the non-irradiated samples and in the irradiated empty vial. Sample compositions are described in more details in [4].

Table 2: Material and irradiation data for samples presented in this work. Effective irradiation time was calculated from total energy production recorded in reactor operation log file during each irradiation.

Sample	Depth [cm]	Sample material	Mass [mg]	Irradiation start and irradiation end	Total irradiation time	Effective irradiation time at 250 kW
1	60	Au foil	47,3	from: 18.12.2000 14:20 to: 19.12.2000 12:20	22 hours	22 hours
2	50		47,1			
3	40		46,5			
4	30		47,0			
5	20		47,5			
6	10	0,1 % Au wire and Ni wire	20,7 and 153			
7	5		13,0 and 107			
8	0		14,2 and 130			
B41	60	Barytes concrete	1173,5	from: 20.12.2000 11:22 to: 25.01.2001 11:50	36 days and 28 minutes	100 hours and 2 minutes (100,04 h)
B42	50		1156,2			
B43	40		1051,0			
B44	30		1170,5			
B45	20		1224,4			
B46	10		1187,9			
B47	5		1104,8			
B48	0		1165,4			
A41	60	Ordinary concrete	784	from: 05.02.2001 14:20 to: 23.02.2001 11:40	17 days, 21 hours and 20 minutes	100 hours and 11 minutes (100,19 h)
A42	50		735			
A43	40		768			
A44	30		771			
A45	20		776			
A46	10		763			
A47	5		762			
A48	0		768			
PE1	60	Au foil	47,3	from: 07.05.2001 13:01 to: 08.05.2001 13:01	24 hours	24 hours
PE2	50		47,1			
PE3	40		46,5			
PE4	30		47,0			
PE5	20		47,5			
PE6	10	0,1 % Au wire and Ni wire	20,7 and 162,4			
PE7	5		13,0 and 131,5			
PE8	0		14,2 and 115,5			

2.2 Sample Activity Measurements

Gamma activity of the irradiated samples was measured using a Princeton Gamma-Tech (PGT) HPGe detector type NIGC 26 with a FWHM resolution of 2,00 keV at 1,33 MeV, connected to a multi-channel pulse-height analyzer personal computer system based on The Nucleus Personal Computer Analyzer (PCA-II) card. The minimum detectable gamma ray energy of our detector configuration was 30 keV.

Radioisotopes were identified from the pulse-height spectrum by their gamma photo-peak energies and half-lifes. Their activities were determined from gamma photo-peak area and detection efficiencies at the photo-peak energy. The full-energy peak efficiency for point source geometry was measured using a set of three point source standards ([133]Ba, [152]Eu and [226]Ra calibrated on 28.05.1990) with 4 % accuracy. The full-energy peak efficiency for concrete sample geometry was measured using two concrete samples of the experimental

geometry with known activity. Activity of these two samples was determined by Department of Environmental Sciences at "Jožef Stefan" Institute.

From the measured count rate R_m we calculated the saturated activity A_∞ for all interesting nuclides in samples according to equation (1) below. Equation (1) is correct if we assume that the neutron flux does not vary during the exposure, and neglect any decrease in the number of target nuclei over the exposure time [7].

$$A_\infty = \lambda \, \Delta t \, \frac{e^{\lambda \, t_{dk}}}{(1 - e^{-\lambda \, \Delta t})(1 - e^{-\lambda \, t_{irr}})} \frac{R_m}{P_\gamma \varepsilon} \tag{1}$$

Here λ is the nuclide decay constant ($\lambda = 0.6931/t_{1/2}$), Δt is the overall count interval (real time), t_{dk} is cooling time from the end of irradiation to the start of the measurement and t_{irr} is the uninterrupted irradiation time. P_γ is the emission probability (or yield) of the specific gamma ray and ε is the detector efficiency for sample geometry at the measured gamma-ray energy.

We calculated appropriate reaction rates ($\sigma_{act} \, \phi_0$) for production of measured radioactive nuclides in gold and nickel samples on the basis of measured saturated activity A_∞ according to equation (2) presented below. Here N_0 is appropriate target nuclide number in the sample.

$$A_\infty = N_0 \, \sigma_{act} \, \phi_0 \text{ or exactly } A_\infty = N_0 \, \phi_0 \int \sigma_{act}(E) \, \chi(E) \, dE, \text{ if } \int \chi(E) \, dE = 1. \tag{2}$$

Both conditions (constant neutron flux during exposition and negligible target nuclide burnup) for equation (1) were satisfied for gold and nickel foils irradiation. In the case of concrete samples the irradiation was interrupted several times and also neutron flux was changed several times. Equation (1) was still used to calculate the saturated activity of the long-lived nuclides in the concrete samples; we only replaced the total irradiation time with the effective irradiation time at 250 kW. Error introduced with this simplification was estimated using isotope generation and depletion code ORIGEN2.1 [8]. Using ORIGEN2.1 code we calculated activation of seven long-lived nuclides found in barytes concrete samples for two different irradiation procedures. In the first case the irradiation was performed at realistic irradiation conditions identical to conditions during irradiation in beam port 4. For the second case a constant flux irradiation for effective uninterrupted time was calculated and compared to the realistic case. The differences between real irradiation conditions and uninterrupted effective irradiation conditions were found to be smaller than 1 % for long-lived nuclides whit half-lives longer than 5 years. Hence also the errors introduced through the use of eq. (1) are smaller than 1 % for these nuclides. Results of ORIGEN2.1 calculation are given in Table 3 and presented in Figure 2

Table 3: ORIGEN2.1 calculation results for activation of seven long-lived nuclides found in barytes concrete samples calculated for two different irradiation conditions. First case is constant flux irradiation at full power for 100,04 hours *effective* irradiation time. Second case is *realistic* irradiation with interruptions in total length of 36 days and 28 minutes. Table shows calculated neutron activation products concentrations in [pg/g].

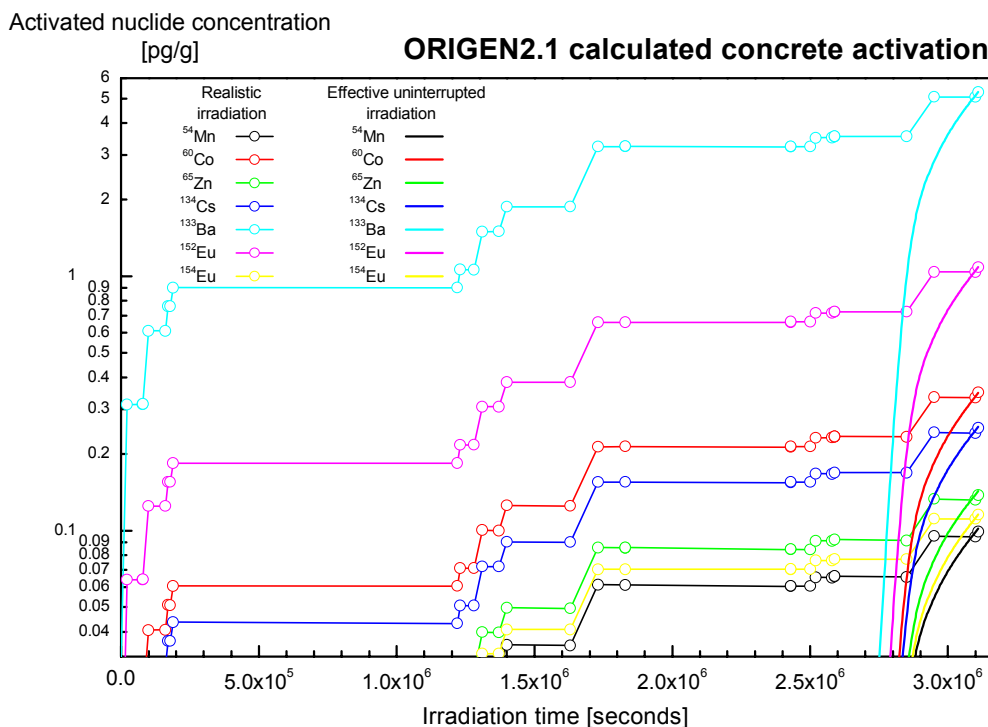| Nuclide	^{65}Zn	^{54}Mn	^{134}Cs	^{60}Co	^{154}Eu	^{133}Ba	^{152}Eu
$t_{1/2}$	244,3 d	312,5 d	2,06 y	5,27 y	8,60 y	10,52 y	13,32 y
Effective irradiation (uninterrupted)	0,1432	0,1019	0,2565	0,3476	0,1159	5,3090	1,0854
Realistic irradiation (interrupted several times)	0,1381	0,0990	0,2533	0,3492	0,1155	5,2960	1,0832
Difference	3,7 %	2,9 %	1,3 %	0,46 %	0,29 %	0,25 %	0,20 %

Figure 2: Comparison of activation for effective uninterrupted and realistic irradiation conditions calculated using ORIGEN2.1 code (see Table 3 caption for details).

3   RESULTS

Due to the large amount of experimental data, the complete results of measurements are presented only in figures in this work.

3.1   Gold and Nickel Foil Activation Results

Measured reaction rates for ^{197}Au(n,γ)^{198}Au, ^{64}Ni(n,γ)^{65}Ni and ^{58}Ni(n,p)^{58}Co reactions in gold and nickel foils in eight different concrete depths are presented in Figure 3. Note that the measured saturated activity per parent nuclide number in the sample is according to eq. (2) identical to the appropriate reaction rate ($\sigma_{act} \phi_0$). All measured reaction rates decrease linearly with depth in logarithmic scale. Measured ^{198}Au reaction rate on the concrete surface is approximately 63 times ^{65}Ni reaction rate. This is in a good agreement to the ratio between thermal cross sections (0,0253 eV) of respective reactions. Cross section for ^{197}Au(n,γ) is 98,8 barns [9], for ^{64}Ni(n,γ) is 1,52 barns and the ratio between these two is 65.

The measured reaction rates for ^{197}Au(n,γ)^{198}Au and ^{58}Ni(n,p)^{58}Co reactions in 8 different concrete depths behind PE filter are presented in Figure 4. Measured ^{198}Au reaction rate on the concrete surface behind PE filter is found to be 45 times smaller than the same reaction rate without PE filter. The fitted potential functions in form presented in equation (3) for all measured reaction rates are also presented in Figures 3 and 4.

$$\frac{A_\infty}{N_0} = a_0 * 10^{-k \cdot d} \tag{3}$$

Here a_0 is saturated activity per parent nuclide on the concrete surface, k is fitted factor and d is concrete depth in centimeters.

Saturated Activity per
Parent Nuclide Number [Bq/barn]
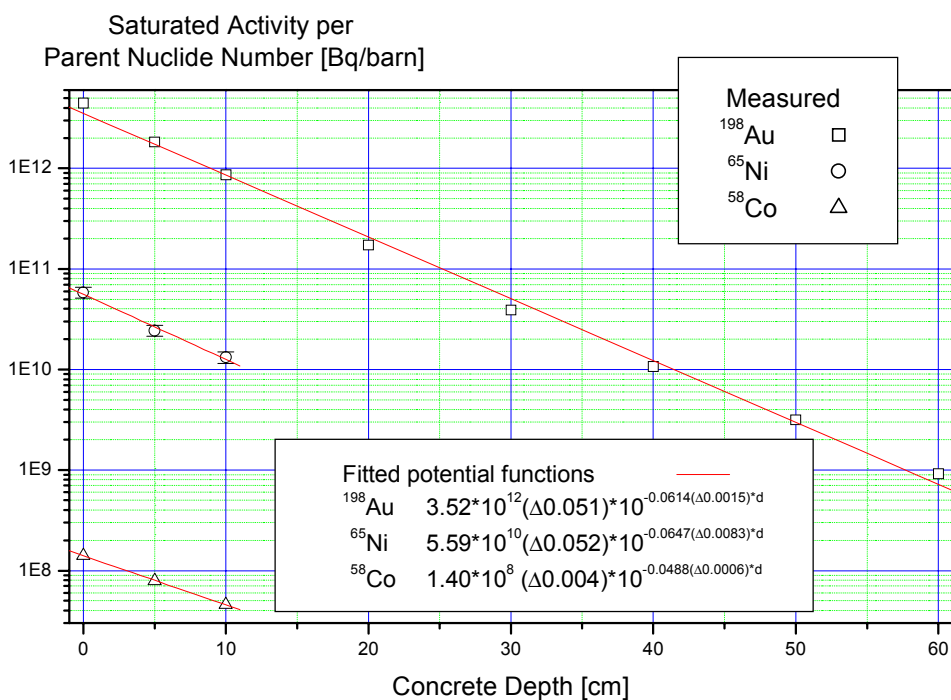

Figure 3: Measured saturated activity per parent nuclide number for gold and nickel foils
irradiated in beam port 4 for 22 hours.

Saturated Activity per
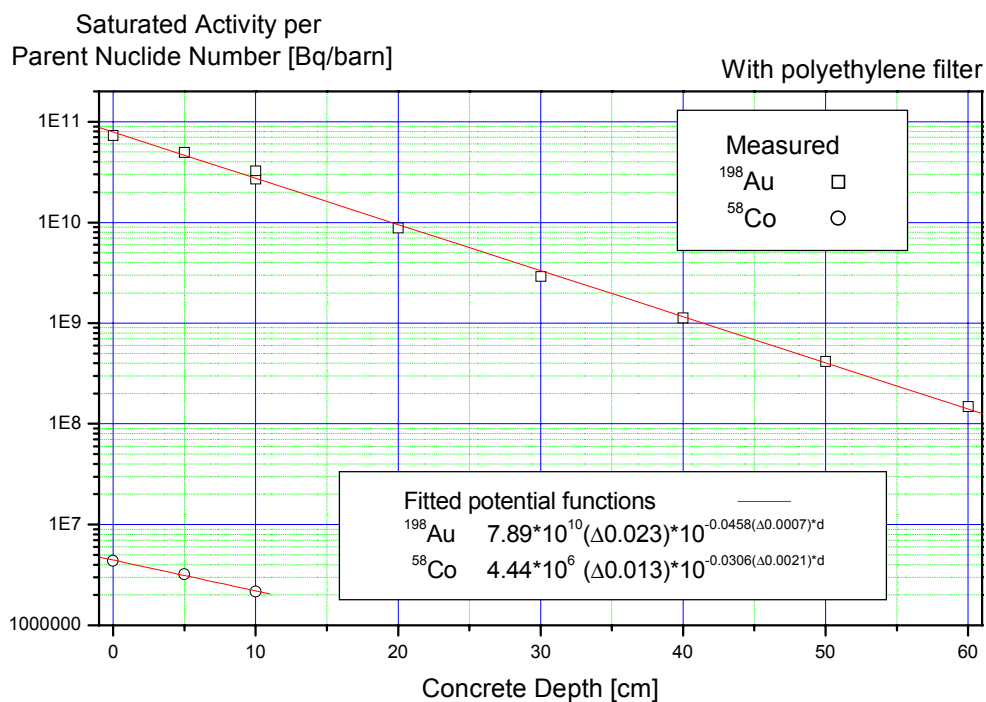Parent Nuclide Number [Bq/barn]

Figure 4: Measured saturated activity per parent nuclide number for gold and nickel foils
irradiated in beam port 4 behind PE filter for 24 hours.

Preliminary comparison between measured [198]Au reaction rates in beam port 4 and reaction rates calculated with three-dimensional discrete ordinates neutron/photon transport code TORT [5] show good agreement. Experimentally determined ratio between [198]Au reaction rates on concrete surface and in 60 cm depth is approximately 4800. The corresponding calculated ratio is 3340. Reactor body barytes concrete in this model had 5 % water content and concrete density was 3,7 g/cm^3. Increasing only the water content in the model to 6,25 % increased calculated ratio to 4800. This value of water content is still within the experimentally determined water content in the barytes concrete.

3.2   Measured Long-lived Radioactivity in Concrete Samples

Measured saturated activity density for seven interesting long-lived neutron activation products in concrete samples for eight different concrete depths are presented on Figure 5 for ordinary concrete samples and on Figure 6 for barytes concrete samples. Radioactive isotopes of europium, cobalt, cesium and zinc are produced in thermal and resonance neutron absorption (n,γ) reactions from trace elements present in raw materials used in concrete production. All these four trace elements can be found in almost any type of concrete [4]. On the other hand, long-lived radioactive isotope of barium ([133]Ba) is produced in neutron absorption (n,γ) reaction from barium, the most abundant chemical element in barytes concrete. Radioactive isotope of manganese [54]Mn is primarily a product of fast neutron reaction (n,p) on iron isotope [54]Fe. Iron is also present in almost any concrete type.
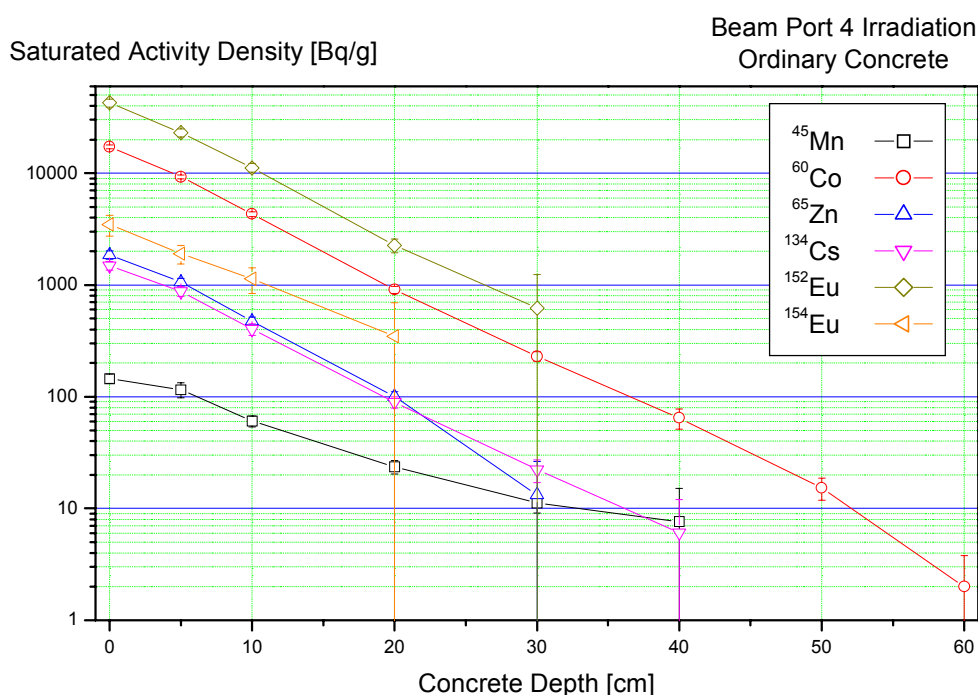

Figure 5: Measured saturated activity density of six interesting long-lived nuclides found in ordinary concrete samples. Samples were irradiated in beam port 4 for 100,19 hours.

Both ordinary and barytes concrete samples were irradiated in beam port 4 without PE filter. We can expect somewhat lower activities at the same concrete depths in the reactor body outside the beam port where reactor body is protected with water layer. We can treat

results presented in Figures 5 and 6 as a conservative estimate for activities in reactor body irradiated for very long time (e.g. full reactor operation lifetime).

If we use the most strict activity criteria for radioactive material [10] (10 Bq/g), then in our case TRIGA Mark II reactor body made of barytes concrete should be treated as radioactive waste to the depth of approximately 80 cm. We see that only radioactive nuclides [133]Ba, [152]Eu and ^{60}Co will contribute significant amounts of radioactivity in deeper layers of barytes concrete shielding.

Under the same conditions TRIGA Mark II reactor body made of ordinary concrete should be treated as radioactive waste to depth of 60 cm. We see that only radioactive nuclides [152]Eu and ^{60}Co will contribute significant amounts of radioactivity in deeper layers of ordinary concrete shielding. Note that this analysis is valid only if concrete shield would have approximately the same density as actual barytes concrete shield (3,7 g/cm^3).
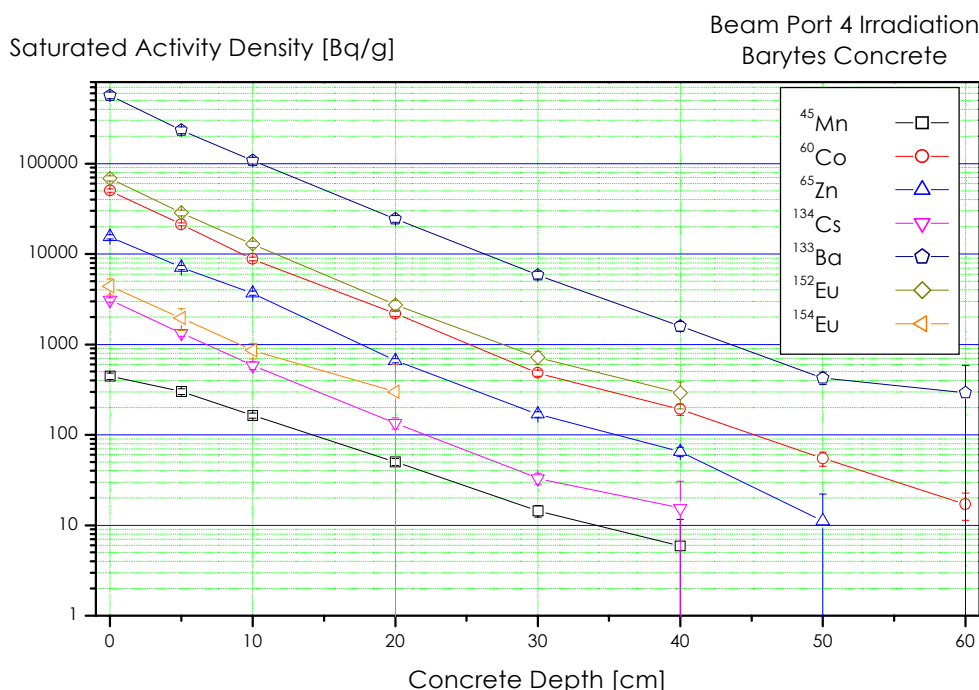

Figure 6: Measured saturated activity density of seven interesting long-lived nuclides found in barytes concrete samples. Samples were irradiated in beam port 4 for 100,04 hours.

4   CONCLUSIONS

Results of neutron activation measurements in the beam port 4 using special sample-holder are presented in this work. Measured reaction rates in gold and nickel foils were used to estimate neutron flux in the reactor body. These experimental results can also be used as a benchmark results for full reactor body neutron transport calculations. For accurate neutron flux measurement further irradiation of activation foils will be needed.

From concrete samples irradiated in the sample-holder measurements, we calculated saturated activities of the long-lived nuclides in deeper layers of concrete reactor body. From these results we can conservatively estimate that activity concentrations in the reactor body will be below 10 Bq/g in layers deeper than 1 m. We concluded from measurements that in the ordinary concrete the residual long-lived radioactivity is predominantly due to trace

elements presence. These isotopes are ^{60}Co and ^{152}Eu. All these isotopes are produced from thermal neutron capture reactions. The residual long-lived gamma radioactivity in barytes concrete is predominantly due to ^{133}Ba, which is produced in thermal neutron capture reaction from barium, which is a basic element in barytes concrete. Besides ^{133}Ba in barytes concrete also ^{152}Eu and ^{60}Co nuclides can still be detected in deeper layers of concrete reactor body.

These experimental results and experiences gained through this work represent a good basis for further experimental work in this research field.

REFERENCES

[1] D. R. Parry, M. R. England, A. Ward, D. Green, "Decommissioning of the ICI TRIGA Mark 1 reactor", Transactions of Research reactor fuel management (RRFM) 2000 conference, Colmar, France, March 19-21, European Nuclear Society, 2000.

[2] E. W. Abelquist, A. Huda, S. State, J. Takahashi, "Decommissioning of a University Research Reactor", *Health Physics*, **67**, 1994, pp.80-87.

[3] T. Žagar, M. Ravnik, "Measurement of Neutron Activation in Concrete Samples", Proc. Int. Conf. Nuclear Energy in Central Europe 2000, Bled, Slovenija, 11-14 September, Nuclear Society of Slovenia, 2000.

[4] T. Žagar, M. Ravnik, "Determination of long-lived neutron activation products in reactor shielding concrete samples" accepted for publication in *Nuc. Technol.*, 2001.

[5] W. A. Rhoades, D. B. Simpson, *The TORT Three-Dimensional Discrete Ordinates Neutron/Photon Transport Code*, ORNL/TM-13221, Oak Ridge National Laboratory, USA, 1997.

[6] M. Božič, T. Žagar, M. Ravnik, "Calculation of Neutron Fluxes in Biological Shield of the TRIGA Mark II Reactor", to be presented on Int. Conf. Nuclear Energy in Central Europe 2001, Portorož, Slovenija, September 10-13, Nuclear Society of Slovenia, 2001.

[7] G. F. Knoll, *Radiation detection and measurement*, John Wiley & Sons, New York, USA, 1979, pp.765-771.

[8] A. G. Croft; *A User's Manual for the ORIGEN2 Computer Code*, ORNL/TM-7175, Oak Ridge National Laboratory, Oak Ridge, Tennessee, USA, 1980

[9] Joint evaluated file data library (JEF-2.2 evaluated data library), Organization for Economic Cooperation and Development / Nuclear Energy Agency Data Bank, Issy-les-Moulineaux, France, 1997.

[10] Basic safety standards for the protection of the health of workers and the general public against the dangers arising from ionizing radiation, Council Directive 96/29/EURATOM, Brussels, EU, 1996.