Preliminary Waste Characterization Measurements in FiR 1 TRIGA Research Reactor Decommissioning Project

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PRELIMINARY WASTE CHARACTERIZATION MEASUREMENTS IN FIR1 TRIGA RESEARCH REACTOR DECOMMISSIONING PROJECT

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ABSTRACT

The objective of the study has been to verify the calculated residual activity in the decommissioning waste of TRIGA Mark II type research reactor FiR 1 in Finland. The knowledge of radioactive inventory of irradiated materials is important in the planning of the decommissioning activities and is essential for predicting the radiological impact to personnel and environment. Measurements are performed for low active material samples from outer parts of the reactor. Methods include gamma spectrometric measurements, composition measurements with mass spectrometry, oxidation measurements of especially C-14 in graphite and full combustion measurements of lithium enriched shielding materials. Results are compared to estimates calculated with a combined Monte Carlo model of the reactor and a point-depletion code modelling the irradiation history. Decommissioning waste consists mainly of ordinary concrete, aluminium, steel and graphite parts. Only preliminary measurements of low active samples are reported so far, but the same methods will be used later for characterizing and classifying dismantling waste. Some discussion of characterization requirements and future sampling is also included.

Keywords: decommissioning, TRIGA research reactor, activity inventory, characterization

I. INTRODUCTION

FiR 1 is a 250 kW TRIGA Mark II open pool reactor from General Atomics that has been in operation in Espoo, Finland, for years 1962-2015. During its history FiR 1 has been used for training [1], scientific purposes [2], BNCT treatments [3] and isotope production [4].

The reactor is entirely above ground and is surrounded by a concrete shield structure, as shown in Figure 1. The reactor core and reflector assembly are located near the bottom of an aluminium tank 6.4 meters deep and 2.0 meters in diameter. Approximately 4.9 meters of demineralised water above the core provides vertical shielding. FiR 1 reactor has four beam tubes extending from the reflector assembly through the water and concrete to the outer face of the shield structure. These beam tubes have been used, for instance, to material research with neutrons during first decades of the FiR 1 operational history. The have been been plugged in the late 1980s'.
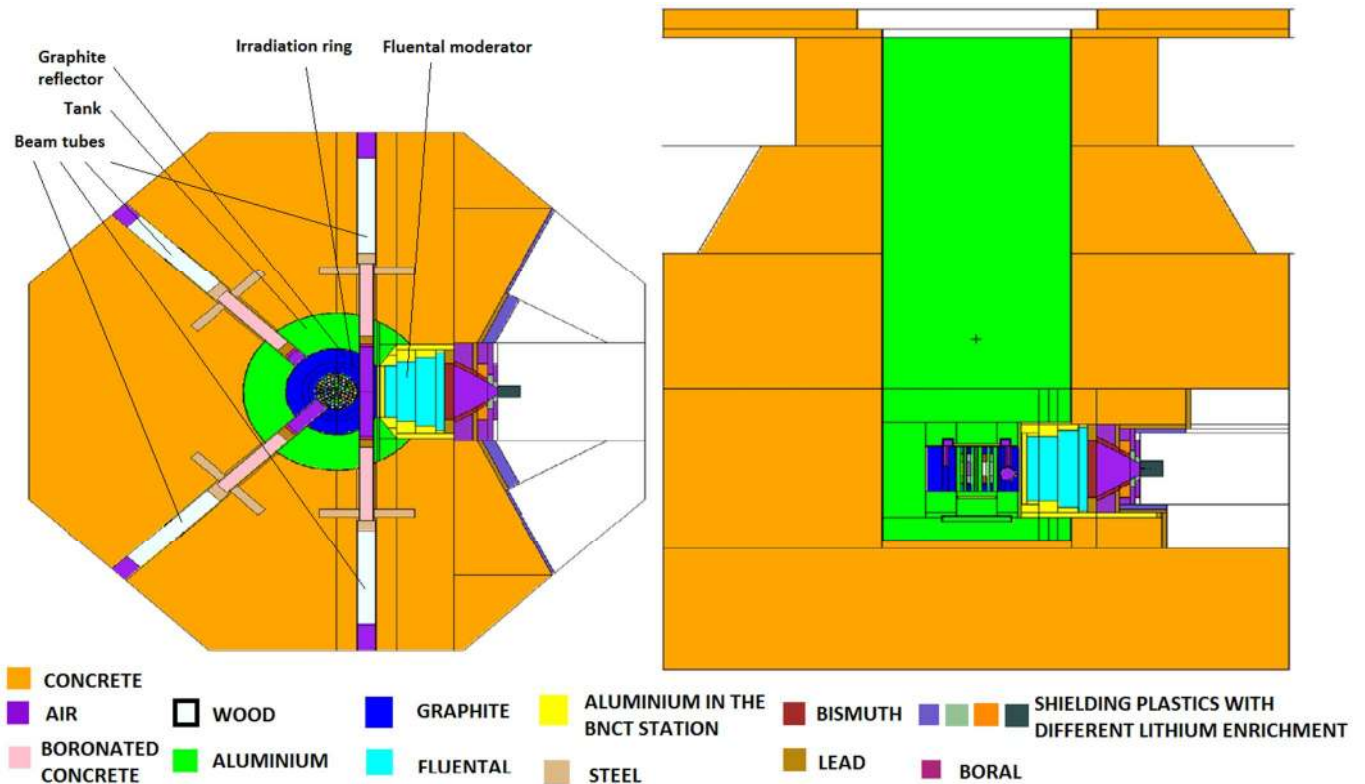
Figure 1: Horizontal and vertical cross sections of the FiR 1 reactor.

Originally, FiR 1 reactor has had a 1.2x1.2x1.7 meters graphite thermal column extending from the outer surface of the reflector assembly and penetrating the reactor tank and shield structure. In 1995-1996, the thermal column was replaced by an epithermal boron neutron capture therapy (BNCT) beam [5]. A BNCT irradiation room was also built of steel tube elements filled with density-optimized heavy concrete and a heavy steel framed lead door. Based on the earlier shielding design calculations [6], it can be assumed that these structures are not activated, and it is justified to exclude them from current waste measurements. The BNCT beam moderator and collimator structures consist of several different materials, the most important of which is Fluental neutron moderator used to shape the neutron energy spectrum to suitable epithermal energy range for BNCT. Bismuth, lead and lithiated polyethylene are also used for gamma and neutron shielding.

FiR 1 reactor will be decommissioned in the following few years. Decommissioning planning requires knowledge on the activation levels and amounts of radioactive decommissioning waste to design safe dismantling procedures. Knowledge on the activity inventories of different waste streams are also important for appropriate waste packaging and planning both the interim storage and the final disposal of the decommissioning waste.

Computational estimates of the activity inventories were completed in 2016 [7]. This paper presents initial measurement results to verify earlier computational estimates. First general requirements and official regulations are introduced, then results of material-wise measurements are presented and finally the utilization of these results e.g. in active waste clearance procedures and other future work is discussed.

II. INVENTORIES AND MEASUREMENT REQUIREMENTS

Activity inventories of FiR 1 structures and components have been estimated in Reference [7]. In principle, activity inventory calculations require modelling of neutron fluxes for all the reactor structures and components with appropriate neutron transport code and combining these neutron flux values to material compositions and operating history in a suitable point-depletion code. An illustration following IAEA recommendation [8] is presented in Figure 2.
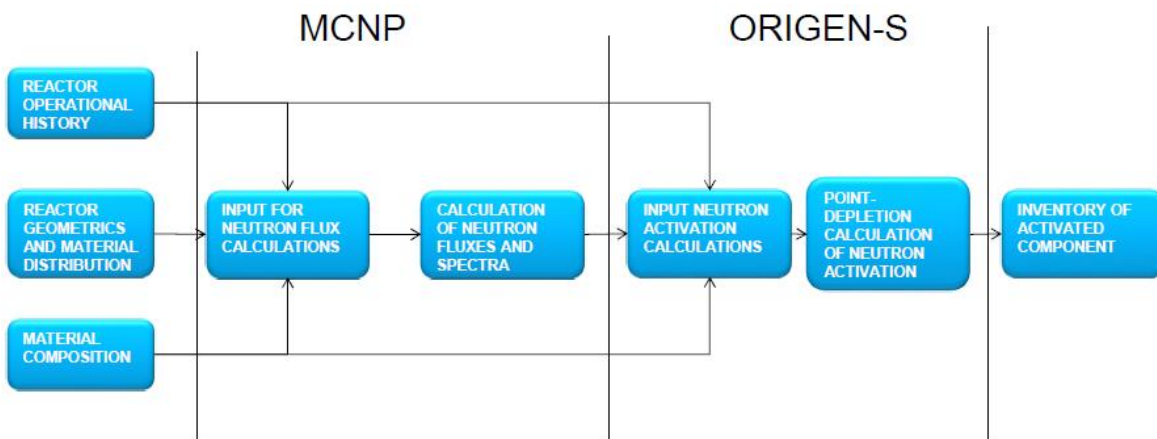
Figure 2: Calculation model.

Neutron fluence rates in FiR 1 reactor were calculated with the Monte Carlo code MCNP5 [9] using criticality eigenvalue search mode and cell-based tallies for all modelled structural components. Results per fission neutron were scaled to 250 kW nominal power and divided to three-group form as specified in ORIGEN-S manual. After calculating the fluxes in three-group form, they were used to model the decay chains of different nuclides in all the structures with the point-depletion code ORIGEN-S [10]. The irradiation history was modelled according to the reactor's actual operational history for years 1962-2015 taking into account all major reactor modifications (affecting the geometry and materials) during its history. After irradiation, a decay period was modelled to study the radionuclide inventory both during dismantling and final disposal. The total neutron flux was considered constant during all irradiations and zero in between them. Altogether, FiR 1 has generated around 11 500 MWh of thermal energy.

ORIGEN-S calculations were performed on component or structure base resulting in around 200 output files.

Technically FiR1 contains several types of aluminium and steel from different manufacturers. This taken into account in the component-wise inventory calculations and detailed results will be used during practical dismantling work. Material-wise total values are presented in Table I to give an overview of the material quantities in the project.

Table I: Material-wise total activities. Volumes are ideal quantities without taking into account packaging efficiency.

Material	Volume (m3)	Mass (tons)	Main isotopes	Total activity (TBq)
Concrete	25	61	H-3, Eu-152, Co-60, C-14	0.1
Graphite	2.6	4.4	H-3, C-14, Eu-152, Eu-154	0.46
Steel	0.4	3.5	Fe-55, Ni-63, Co-60	1.91
Aluminium	0.8	2.2	Zn-65, Co-60, Ni-63	0.39
Fluental	0.45	1.3	H-3	1.3
Other	10	2.7	H-3, C-14, Ni-63	0.4

The main sources of error in the used inventory calculation system are that diffusion of gaseous nuclides, e.g. H-3, cannot be taken into account. Another source of error is that if exact material specification are not available, initial compositions are partially based on assumptions. Because activity is usually due to small impurities in materials, even low concentrations of, e.g., cobalt, nickel, europium, or zinc may have a notable effect on total activities. This was taken into account by assuming conservatively high impurities, but results will be verified with systematic measurements before and during the dismantling work. Eventually the total activity and nuclide vector for each waste package must be known. Both gamma and beta active nuclides are relevant. Gamma activities are relatively easy and fast to measure, but measuring beta activities requires chemical separation and different techniques such as liquid scintillation counting (LSC), mass spectroscopy etc. According to Finnish regulatory guides for nuclear and radiation safety [11], a scaling matrix approach is used. This means that before actual dismantling, material samples of each (plausibly) activated material are collected and studied in detail. Later, when actual waste packages are studied, only fast non-destructive measurements (gamma spectrometry) are performed and the earlier data is used to scale the hard-to-measure beta activities assuming the same ratio between nuclide-wise activities as in the samples. The procedure is further illustrated in Figure 3.
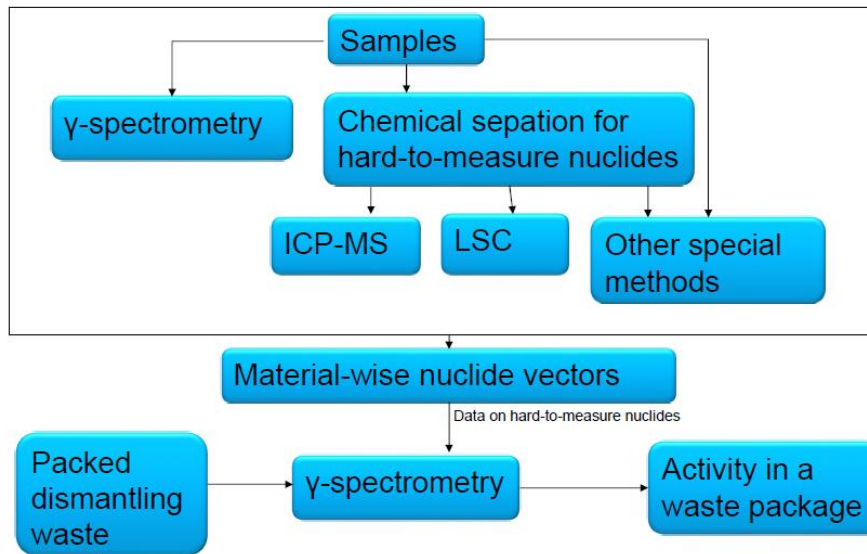
Figure 3: Measurement scheme

The gamma active nuclides that are used in scaling are called *key nuclides*. Identifying appropriate key nuclides requires considering their half-life, radiation energy, chemical challenges in the final disposal, toxicity etc. with a conservative approach, but also enabling the measurements to be performed later systematically for dozens of waste packages during dismantling. VTT uses In Situ Object Counting System (ISOCS) equipment by CANBERRA [12] for gamma spectrometric measurement. It is based on a liquid nitrogen cooled HPGe detector and spectroscopy software GENIE2000 [13]. Sourceless detector-specific calibrations are generated by using ISOCS software.

At the early stage of the project, the main restriction to sampling is that the structural integrity of the reactor must be maintained strictly. Consequently, this report mostly studies only low-active samples from the edges of the reactor. This helps to develop later a more detailed and logically optimized sampling and measurement plan.

III. MATERIAL-WISE MEASUREMENTS

III.A. LEAD
Lead has been used as a shielding material in the BNCT station of FiR1 reactor. Very pure (e.g. almost antimony-free) lead material was originally chosen for BNCT station shielding. Lead has been

exposed to a neutron flux of around 10^8 n/s×cm^2 for years 1997-2015 and according to inventory calculations, it is expected to have a very low specific activity of less than 10 Bq/g. Main contributing nuclides are Ag-110M, Ag-108, Co-60 and Cr-51.

Samples of lead were collected from BNCT station shielding materials. Drilling holes were chosen such that the distance of the samples from the BNCT station neutron channel is the same thus representing redundant parallel samples. Drilling locations are presented in Figure 4 with red color. The figure also lists some of the main components in the BNCT station with numbers 1-10. They have been labelled in Table II.

Figure 4: Location of the lead drilling holes marked with red color.

Table II: Components in Figure 4.

Number in Figure 4.	Component
1	Boral plate
2	Aluminium
3	Fluental moderator
4	Lead shielding

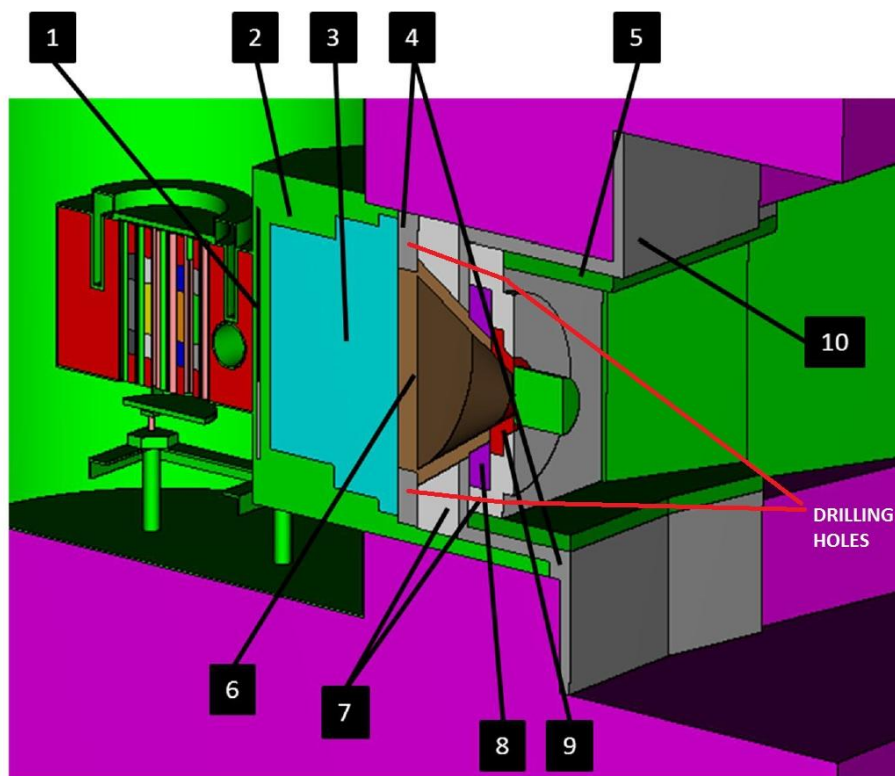| 5	Lithium carbonate plastic shielding
6	Bismuth plate and cone
7	Li-Nat RX215 plastic shielding
8	Li 3.5% enriched RX215 plastic shielding
9	Li 7% enriched plastic collimator
10	Steel encapsulation for lead shielding

The impurities of the drilled lead samples were measured with High Resolution sector field Inductively Coupled Plasma Mass Spectrometer (HR ICP-MS, Element 2, ThermoScientific). Lead samples, with mass less than 200 mg were purified in 1 M HNO_3 and ethanol (≥99.5 %) prior to the microwave assisted acid digestion. Samples were weighted before and after the purification and 6.2 - 9.6 mg mass loss was detected due to the purification of the samples. Samples were completely dissolved in HNO_3 and milliQ water (Millipore) solution (3 mL concentrated HNO_3 + 3 mL milliQ water) with microwave digestion unit (Milestone MLS 1200 Mega). Digested samples were diluted to 50 g with milliQ water. Further, for HR ICP-MS analysis samples were diluted 1/5, 1/10, 1/100 and 1/100000 with 1 % nitric acid solution. 10 µg/L of rhodium was added as internal standard to all samples.

Calibration curve and control samples were diluted from ICP-MS Multi-Element Solutions 2 and 4 (SPEX), and control samples from Semi-Quantitative Standard (SQS-1, AccuTraceTM) and Laboratory Performance Check Standard 1 (LPC-1, SPEX) solutions. 10 µg/L of Rh was used as an internal standard in all samples, background, calibration and control samples. The samples were injected through SeaSpray nebulizer (0.4 ml/min) and double pass spray chamber equipped with Peltier cooling unit. Aluminum sample cone and Nickel skimmer cone were used during the measurement. Final results and measurement relative standard deviations (RSD) are listed in Table III.

Table III: Measured impurities and relative standard deviations (RSD) of lead in the BNCT station.

Nuclide	w%	RSD (%)	Nuclide	ppm	RSD (%)	Nuclide	ppm	RSD (%)
Ag-107	2.59E-04	4.40	Fe-56	1.41E-04	5.40	Sb-121	9.31E-05	3.53
Cd-111	8.46E-07	24.00	Ni-60	1.20E-05	8.88	Cr-52	7.60E-07	15.88
Sn-118	1.64E-06	16.95	Cu-63	2.98E-04	5.33	Co-59	1.22E-07	34.75
Bi-209	1.23E-03	5.58	Zn-66	4.23E-05	8.83			
Ca-44	1.51E-03	4.80	As-75	9.46E-06	4.23			

Mn-55	1.57E-07	15.00	Se-77	2.31E-05	14.00			

Lead samples were measured at the end February 2017 using Canberra's ISOCS gammaspectrometer [12] with HPGe detector (BE2020), Inspector 2000 DSP analyzator and Genie 2000 v.3.4 gamma analysis software. The efficiency calibration were done with Geometry Composer-program (version 4.4). For gamma measurements the drilled lead samples (first corkscrew-like samples) were rotated to spiral-like discs to ease the efficiency calibration done with Geometry Composer-program. Measurement time was around 24 hours.

Results are listed in Table IV. Listed values are averages from four measured parallel samples.

Table IV: Comparison of measured and calculated gamma activities. Estimated uncertainties are given in parentheses.

Sample / specific activity	Ag-110M (Bq/g)
Average measured	0.81 (0.07)
Average calculated	0.93 (0.09)

Apart from silver-110M measured activities were smaller than minimum detectable activities. In addition, after a decay time of around 600 days some Ag-108M, Fe-55, Ni-63 and Zn-65 activities could be expected, but based on activation calculation, all these values are clearly below Finnish clearance levels. Consequently, lead in the BNCT station can very likely be cleared from regulatory control.

III.B. ALUMINUM

FiR1 reactor contains aluminum in the reactor tank, horizontal beam tubes and in most of the equipment inside the tank. Aluminum parts are illustrated in Figure 5.
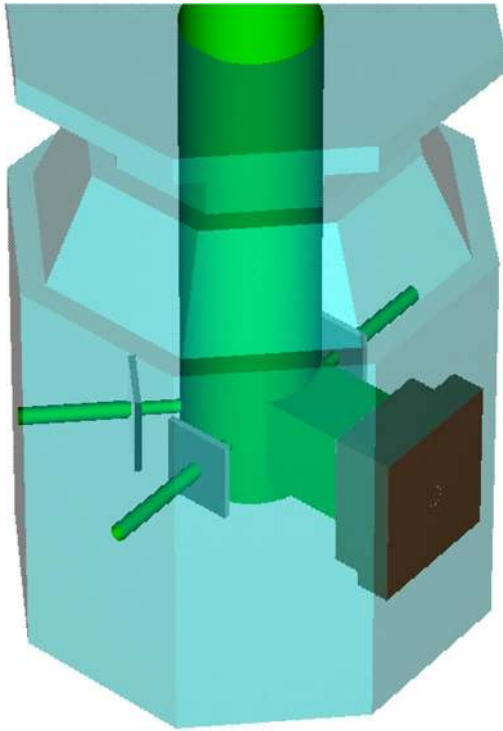
Figure 5: Aluminum parts in FiR1 reactor are illustrated in green color.

Only small impurities of e.g. cobalt, iron, nickel and zinc activate in aluminum. Specific activities are much lower than in steel parts, but all the aluminum structures near the core region are still active and need to be considered as low- and intermediate level nuclear waste. Nevertheless, typical power reactors seldom contain any aluminum parts and consequently the safety of final disposal of aluminum has not been studied as thoroughly. Chemical behavior of FiR1 aluminum in active waste final disposal conditions has been studied at VTT in Reference [14].

Aluminum was sampled from the inactive upper parts of the tank. According to the manufacturing specification, the aluminum should be of AlMg3 type. The tank, thermal column walls and beam tubes were manufactured in Finland by Alstrom company, whereas smaller special components (grid plates, tubes, etc.) were manufactured in US by General Atomics.

The approach was to measure the composition with inactive samples and irradiate a few of those to validate the computational activation mechanisms.

The composition of aluminum was measured with HR ICP-MS technique from three samples from the upper parts of the reactor tank. . Aluminum samples, with mass less than 300 mg were purified in 1 M HNO3 and ethanol (≥99.5 %) prior to the microwave assisted acid digestion. Samples were completely dissolved in aqua regia (5 mL) with microwave digestion unit. Digested samples were diluted to 100 g with milli-Q water. Further, for HR ICP-MS analysis samples were diluted 1/2, 1/10, 1/100 and 1/10000. 10µg/L of indium was added as internal standard to all samples.

Calibration curve and control samples were diluted from ICP-MS Multi-Element Solutions 2 and 4 (SPEX), and control samples from Semi-Quantitative Standard (SQS-1, AccuTraceTM) and Laboratory Performance Check Standard 1 (LPC-1, SPEX) solutions. 10 µg/L of In was used as an internal standard in all samples, background, calibration and control samples.

The samples were injected through SeaSpray nebulizer (0.4 ml/min) and double pass spray chamber equipped with Peltier cooling unit. Aluminum sample cone and nickel skimmer cone were used during the measurement. Results including measurements relative standard deviations (RSD) are presented in Table V. Results are average values from three parallel samples.

Table V: Measured aluminum composition [ppb] and measurement relative standard deviations (RSD).

Material	ppb	RSD (%)	Material	ppb	RSD (%)	Material	ppb	RSD (%)
Li-7	228	24	Pb-208	24025	1	Cr-52	2006694	1
B-11	2092	7	Bi-209	70	2	Mn-55	784529	1
Sr-88	44	10	Th-232	116	2	Fe-56	3805567	1
Zr-90	6308	1	U-238	1195	2	Co-59	900	3
Mo-95	3216	1	Na-23	1338	9	Ni-60	17184	1
Ag-107	188	1	Mg-24	11397907	1	Cu-63	1830652	1
Cd-111	555	1	Al-27	974157202	1	Zn-66	308425	2
Sn-118	8934	1	Si-28	4251635	12	Ga-69	84170	1
Cs-133	8	140	P-31	1206	6	K-39	276	2
Ba-137	415	5	S-32	1846	18	Ge-72	327	1
Ta-181	1	5	Ca-44	2313	4	As-75	94	17
W-182	1242	1	Sc-45	104	1	Se-77	536	235
Re-185	0	108	Ti-47	143215	2	Se-78	1122368	14
Tl-205	274	1	V-51	32603	1			

Samples of inactive aluminum from the upper parts of the reactor tank were irradiated in FiR1 reactor before shutdown in June 2015. Samples were irradiated in the irradiation ring around the core in a neutron flux of 2.32×10^{12} n/s×cm^2 and cadmium ratio of 2.02. Two of the samples were irradiated for four hours and one of them was irradiated for 14 hours.

Two of these activated samples were studied with ISOCS gammaspectrometry system [12] [15]. Used detector was BE2020 and the calibrations were done using ISOCS version 4.3. Small samples were measured as close as possible, top of the detector. Specific activities were also calculated with the same method as in [7] using the measured aluminum composition. The calculation contains still some uncertainties due to neglecting the shielding effect of the irradiation capsule and neutron input spectrum cannot be modelled exactly. An estimate for the calculation uncertainly is around ten percent. The results show that at the time of measurements, a specific activity of around 2.7 kBq/g is expected. Main contributing nuclides will be Cr-51, Fe-55, Zn-65, Mn-54, Fe-59, Ni59, Ni-63, Sc-46 and Co-60. Measured spectrum is illustrated in Figure 6. Uncertainties in the measurement arise mainly from separating the background from the measurement data and converting the cpm to Bq/g. Moreover, the material composition of the samples has been assumed to be homogenous, but in real life some variations always occur. Comparison between the measured and calculated gamma activities is presented in Table VI.
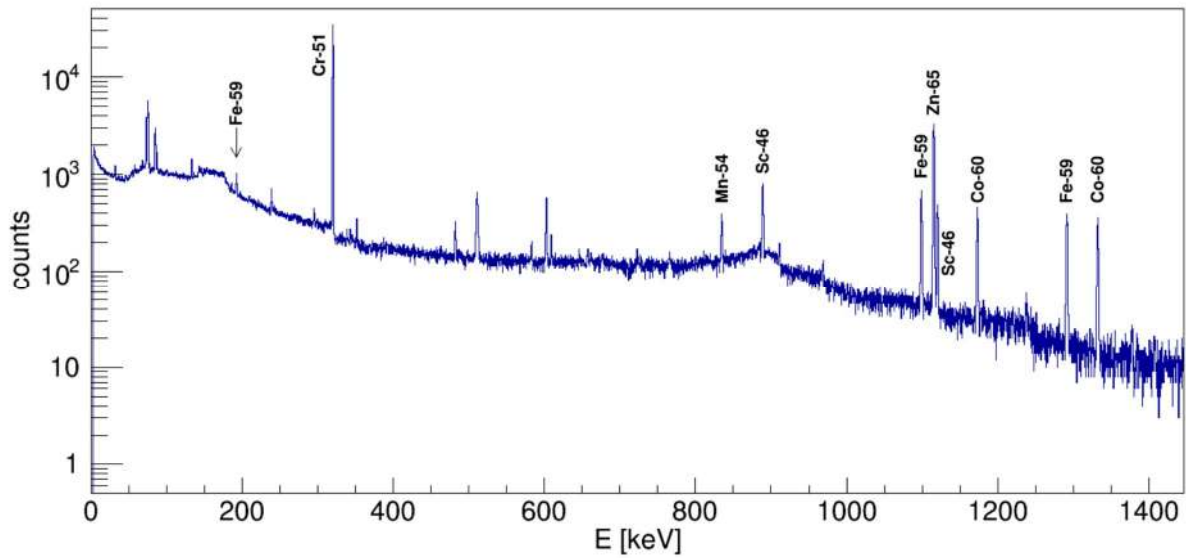
Figure 6: Measured gamma spectrum for aluminum.

Table VI: Specific activities of the studied samples.

Nuclide	Half-life (days)	Measured activity (Bq/g)	Measurement uncertainty (%)	Calculated activity (Bq/g)	Calculation uncertainty (%)
Sc-46	83.79	5.46	2.9	7.4	10
Cr-51	27.7	699	4.7	639	10
Mn-54	312.2	1.80	7.9	4.1	10
Fe-59	44.5	13.5	2.9	20.7	10
Co-60	1925	5.24	3.0	18.4	10
Zn-65	243.9	81.1	3.0	133.9	10

Specific activities correspond to the values reasonably well and provide a basis for further aluminum waste characterization and classification. Apart from short living Cr-51, assumptions in the calculated values also result in a conservative estimate. However, Cr-51 is short living nuclide with a half life of only 27.7 days. Considering the half-lives and nuclide-wise clearance levels, Co-60 will be chosen as a key nuclide for aluminum in future waste measurements.

III.C. GRAPHITE MEASUREMENTS

FiR1 research reactor contains graphite both in the reflector around the core and in 1.2x1.2x1.74 m thermal column that was disassembled in 1995. Disassembled graphite is stored at VTT premises in Finland.
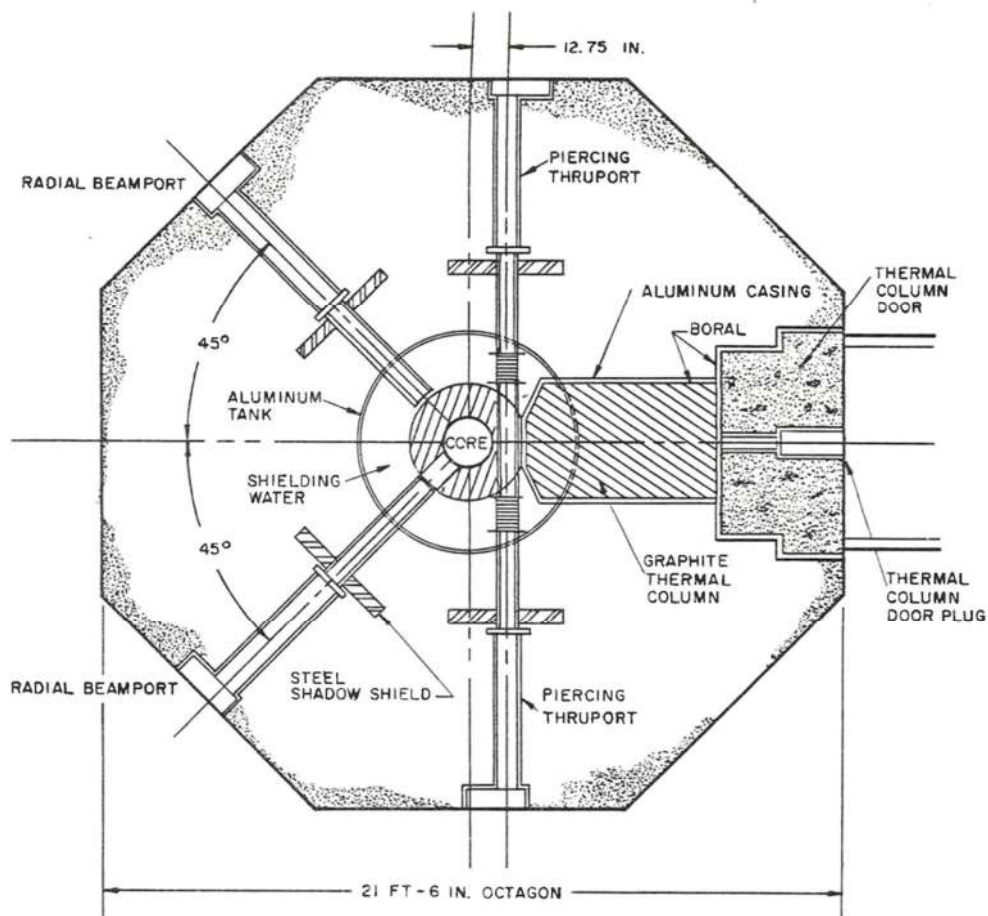
Figure 7: Graphite in FiR1 reactor is illustrated on the right side of the core [7].

Graphite is a porous material, and the most important activation mechanism in the material is the activation of nitrogen in the air of the porous holes into carbon-14 via reaction N-14(n,p)C-14. Carbon-14 is an especially important nuclide in waste disposal, because it spreads rapidly in the biosphere. Other important isotopes are e.g. Eu-152, Eu-154 and Co-60. Moreover, power reactors in Finland do not contain any active graphite, and thus it has not been studied as thoroughly. The chemical behavior of active graphite in final disposal conditions has also been studied at VTT in a separate report [14].

In this study, carbon-14 activity of cylindrical 1.0x1.0 cm samples from thermal column graphite have been measured both at VTT and at the University of Helsinki radioisotope laboratory in Viikki, Finland.

Nuclides that emit only low energy beta radiation, such as tritium and carbon-14, are hard to measure from graphite with normal liquid scintillation counting, because of the poor solubility of the material in scintillation liquid. However, these problems are not present if the sample is oxidized to volatile oxides measuring the activity from their oxides is much easier. VTT measured the graphite samples with Junitek Oxidizer (Junitek co, Finland) device at the University of Helsinki isotope laboratory in Viikki, Finland. Samples of 100 mg were studied. Junitek Oxidizer is based on dry oxidation method and uses pure oxygen to fully burn the sample to water and carbon dioxide at a temperature of 960°C. CO_2 reacts with the amine in the reaction column to form carbamate. The released gases are cooled. Water and air rinsing through the reaction column and collect carbon-14 to liquid scintillation counter bottle. The operating principle is illustrated in Figure 8 and final results with estimates of measurement errors are listed in Table VII. Measurement errors arise partially from collecting the gasses and from performing the LSC measurement.
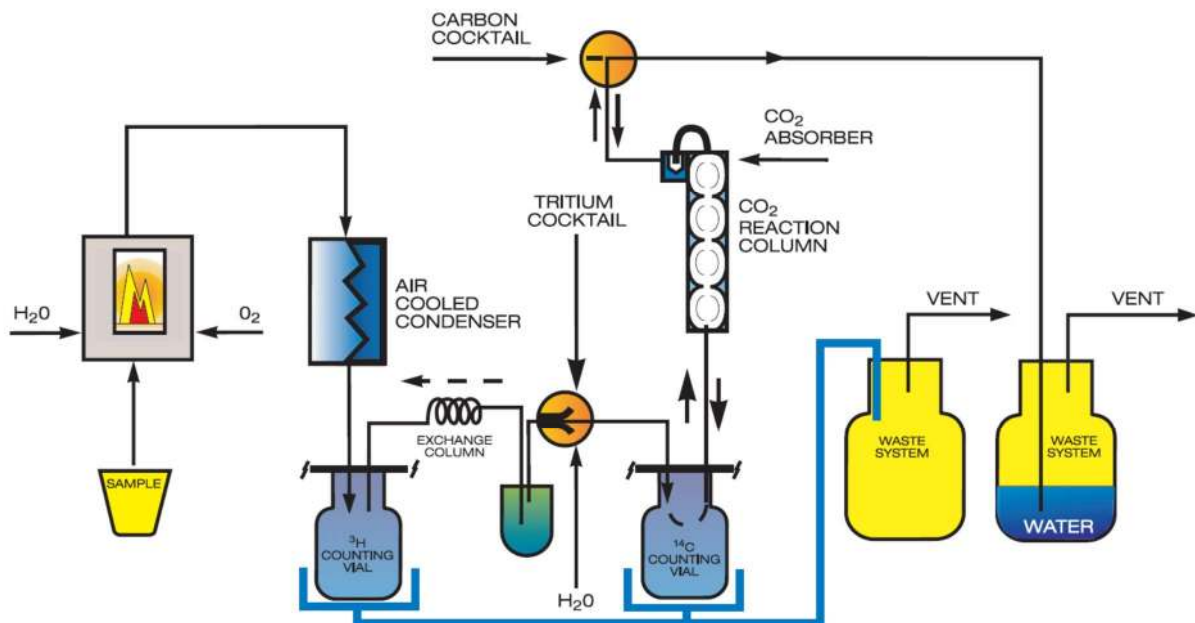

Figure 8: Operating principle of oxidizer device.

Table VII: Measured carbon-14 activities from samples 1–8.

Sample	1	2	3	4	5	6	7	8	Average (kBq/g)
kBq/g	0.293	0.115	0.001	0.023	1.660	0.241	0.100	0.393	0.353
Estimated measurement error (%)	4.9	5.1	23.6	6.6	4.5	4.8	5.2	4.7	

Activities in the thermal column graphite have been calculated earlier [7]. Calculated activity profile of C-14 in the graphite is presented in Figure 9.
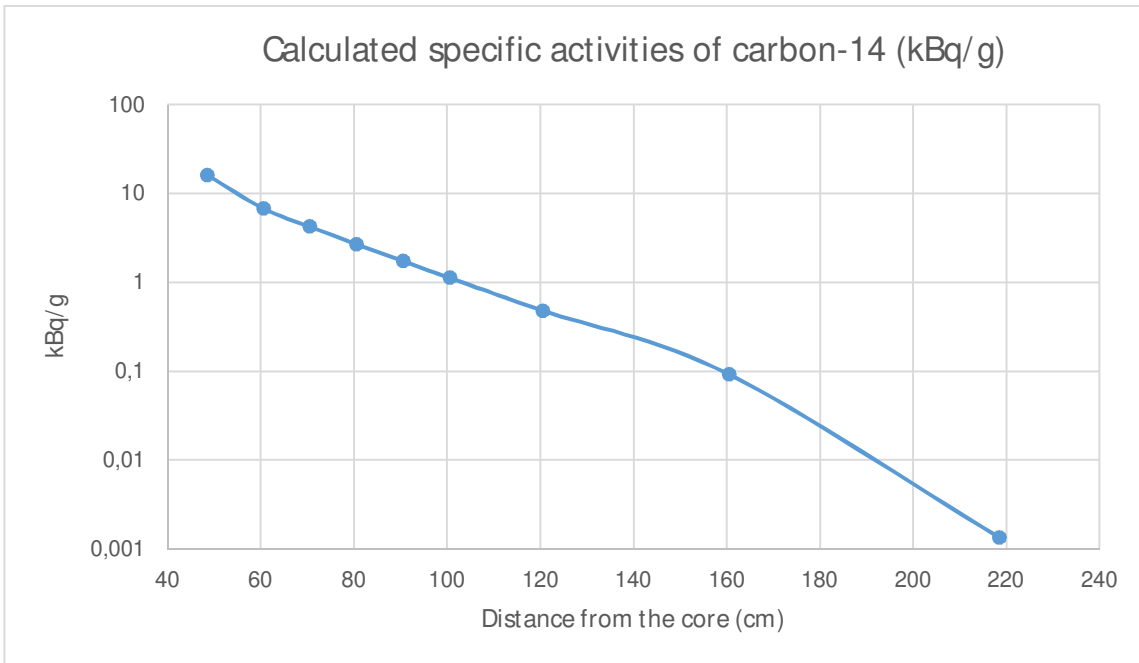
Figure 9: Calculated specific activities of C-14 in thermal column graphite as the function of distance from the core.

Unfortunately exact location of samples in the old thermal column was not saved, but based on dose rate measurements, samples were collected from graphite blocks around the middle of the thermal column. Comparing the measured and calculated specific activities, a reasonable correspondence is seen. However, other radionuclide from same graphite samples need to be studied in the future. This includes especially chemical dissolving and measuring exact chlorine and europium fractions.

III.D. LITHIUM ENRICHED PLASTICS

FiR1 contains lithium enriched plastic that has been used as a neutron shielding material in the BNCT irradiation room. It is a mixture of lithium carbonate (Li_2CO_3) and polyethylene paraffin. Shielding plastics is estimated to contain some tritium due to thermal neutron absorption of Li-6. No other significant activities are expected. Nevertheless, a large part of the tritium may have released from the porous material, and total activities may be close to clearance limits. Plastic samples were studied by full combustion method at Horia Hulubei National Institute for Physics and Nuclear Engineering (IFIN-HH).

The latest inventory calculations model [7] of the decommissioning waste does not contain a detailed model of neutron spectrum inside the BNCT irradiation room, but earlier estimates indicate that fluxes can be at highest around 10^6 neutrons/sec×cm^2. However, it is expected that the fluxes can vary a lot inside the room. With the assumed neutron flux, computational estimates show that specific tritium activities in the shielding plastic can be at highest around a few hundred Bq/g.

Studied samples were drilled from six locations around the room as illustrated in Figure 10. Points 3 and 4 are from two adjacent plastic plates on top of each other. Sample number 3 was the innermost one. Two parallel samples of all location were prepared.
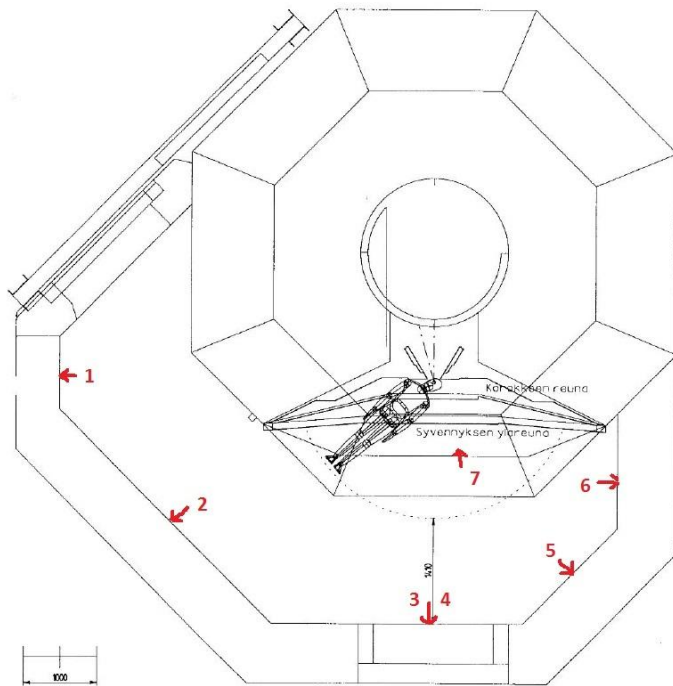
Figure 10: Locations of drilling holes.

Parallel samples of drilled lithium-enriched plastics were sent to IFIN-HH institute to be studied with full combustion oxidation method. The measurement system consisted of a pressurised oxygen tube and two heated tube furnaces that were used for sample oxidation and catalytic oxidation of produced gases at temperature of around 750°C. Gases were collected to water, which was then measured with liquid scintillation counting method. The measurement setup is illustrated in Figure 11.
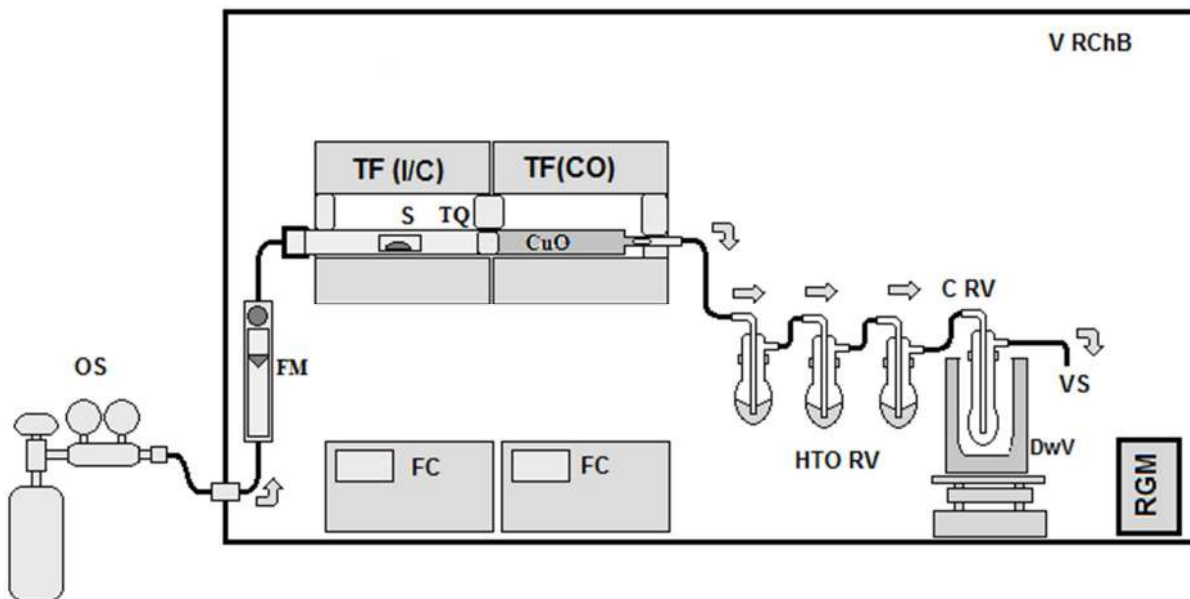
Figure 11: Schematic view of the oxidizer with retention of resulted HTO. QS- Oxygen supply; FM-Flow-meter; TF (I/C)- Tube furnace (Incineration/Calcination); FC- Furnace controllers: TF (CO)-Tube Furnace (Catalytic Oxidation); TQ- Quartz tube; S- Sample; CuO - CuO wire oxidation bed; HTO RV - HTO Collector with 3 retention vials; C RV- Cryogenic retention vial, DwV –Dewar vessel RGM- Radioactive gas monitor, V RChB - ventilated Radiochemical Box

Studied drill cores were cut into disks and smaller pieces to be able to study the inner portions of the material, since the assumption was that tritium may have diffused from the outer parts of the samples. Eventually only the three inner cubic pieces were measured. The cutting protocol is presented in Figure 12.

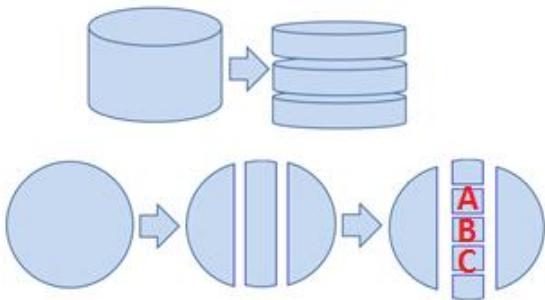Figure 12: Cutting of the drill cores.

Samples were oxidized by gradually increasing the temperature to 750°C in around 120 minutes (see Figure 13) with oxygen flow rate of 4 liters/min.
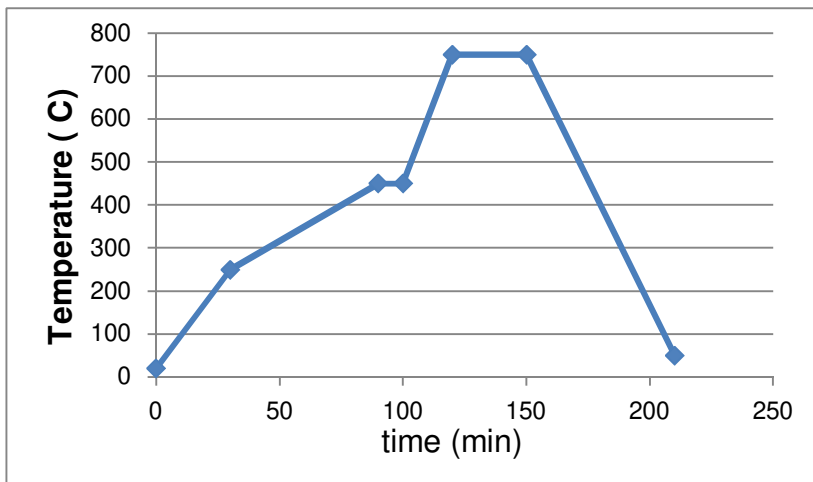

Figure 13: Temperature/time diagram applied for polyethylene/Li$_2$CO$_3$ composite oxidation

15 ml of tritiated water was collected from three retention vials to be measured in with liquid scintillation counting technique.

Specific activities were obtained by incineration of samples, tritiated water retaining and determination of the activities at Liquid Scintillation Counter. Obtained specific activities are listed in Table VIII.

Table VIII: Measured H-3 specific activities. Sample numbers refer to drilling locations in Figure 10.

Sample	Activity determined with LSC (Bq)	Activity-Background	Sample mass (g)	df	As (Bq/g)
1A	6.658	6.0626	0.4977	15	190.618
1B	13.077	12.4818	0.5184	15	359.461
1C	2.105	1.5098	0.5084	15	58.996
Average					203.025
Standard deviation					150.616
2A	25.714	25.1194	0.5089	15	720.044
2B	37.738	37.1426	0.5154	15	1043.385
2C	17.635	17.0402	0.5273	15	476.582
Average					746.670
Standard deviation					284.338
3A	11.138	10.5434	0.5065	15	313.371
3B	21.758	21.1634	0.5179	15	598.682
3C	9.726	9.1314	0.5219	15	265.570
Average					392.541
Standard deviation					180.116
4A	12.434	11.8386	0.5277	15	335.757
4B	17.147	16.5522	0.5180	15	471.714
4C	9.632	9.037	0.5141	15	266.983
Average					358.151
Standard deviation					104.186
5A	17.890	17.2946	0.5170	15	519.041
5B	32.140	31.545	0.5160	15	934.302
5C	21.637	21.042	0.5170	15	627.766
Average					659.018
Standard deviation					204.573
6A	10.079	9.4842	0.5181	15	277.222
6B	15.502	14.9074	0.5748	15	384.324
6C	13.090	12.4946	0.5238	15	356.103
Average					339.216
Standard deviation					55.512
7A	31.849	31.254	0.5200	15	872.785
7B	32.637	32.042	0.5142	15	904.468
7C	27.993	27.398	0.5068	15	787.096
Average					854.783

Standard deviation	60.721

Shielding plastics may have possibly re-use as shielding material in other institutes in Finland. Studied results indicate that their activity in relatively low and consequently from the point of radiation safety, there is no obstacle for re-use.

III.E. CONCRETE

The aluminum tank in FiR1 reactor is surrounded by 1.5 meter thick layer of ordinary concrete, which serves as a biological shield. According to computational estimates in Reference [7], most of the concrete is inactive and activity is limited only close to the reactor core region about one meter from the surface of the tank and 0.5 meters around the beam tubes. Main radionuclides are Eu-152, Eu-154, Co-60, Ca-45, H-3, Ni-63, Ni-59, and Fe-55. Neutron fluxes inducing the activity are illustrated in Figure 14.

Concrete has been studied at VTT by measuring its composition from three drill cores from the inactive outer part of the reactor and by measuring an active older drill core from the thermal column cavity.
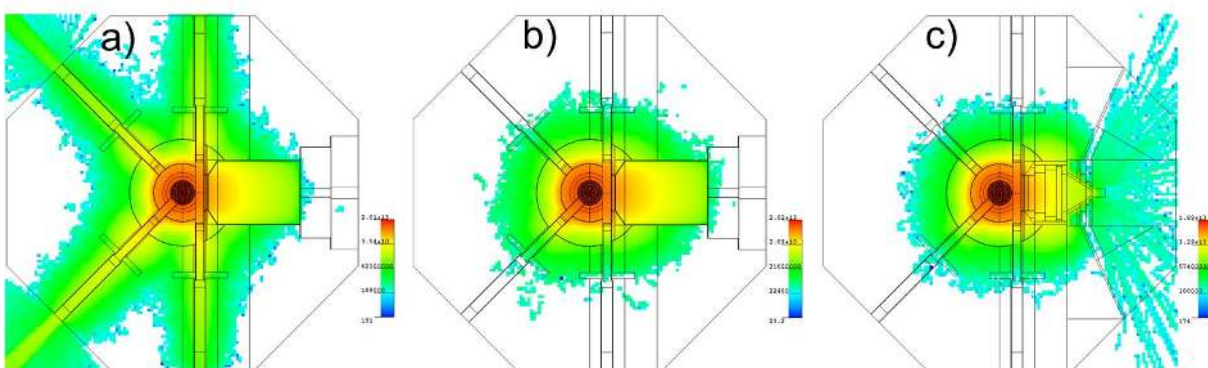

Figure 14: MCNP simulated total neutron fluence rates a) with thermal column and open beam ports, b) with thermal column and plugged beam ports, and c) with BNCT beam. [7]

Three drilling samples were taken from the outermost inactive part of the reactor to determine the detailed composition of the material. Direction of drilling is illustrated in Figure 15.
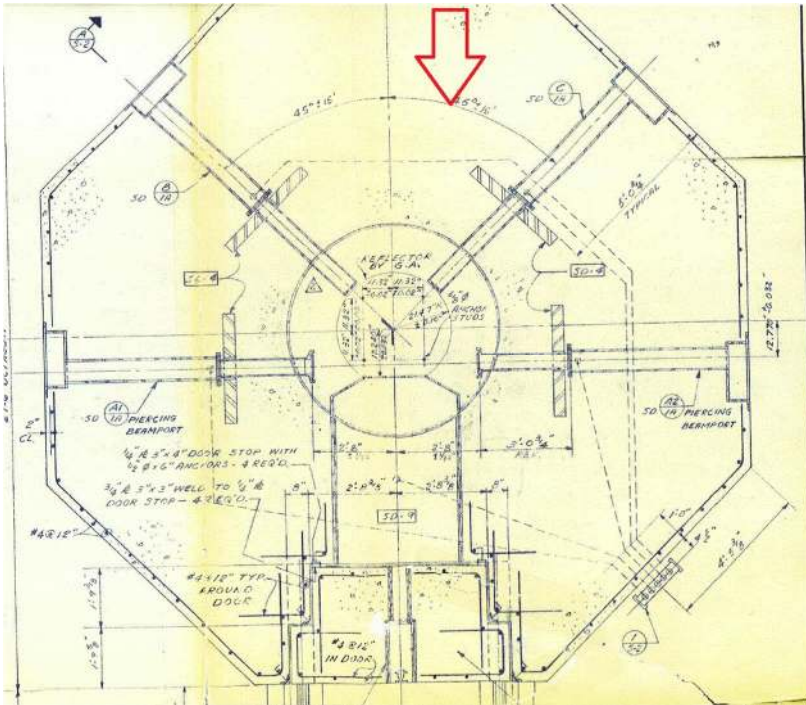
Figure 15: Direction of drilling.

Sample compositions were measured with inductively coupled optical emission spectrometry method by VTT Expert Services. Results are listed in Table IX.

Table IX: Measured concrete composition.

Sample# / element	1 (ppm)	2 (ppm)	3 (ppm)	Measurement uncertainties (ppm)
Al	70300	67900	68050	100
B	100	100	100	100
Ba	860	890	840	20
C	1730	1835	2165	80
Ca	91000	79000	95000	100
Cl	55	52	59	20
Co	12	13	13	5
Cs	1.9	1.7	1.7	100
Eu	2.1	2.0	2.2	0.1
Fe	23000	21000	23000	100
H	4290	4150	4430	8
K	35300	36450	33650	500
Li	36	27	39	10
Mg	9000	7700	8500	200
Mn	350	310	370	10
N	200	200	200	0.04
Na	19000	20000	19000	100
Ni	50	50	50	50
P	500	500	500	500
S	3200	2400	2900	200
Si	258500	258500	258500	500
Sm	8.3	8.0	13	0.1
Ti	1380	1210	1370	100
U	3.0	3.4	4.0	1

Inventory calculations of concrete activity are based on conservative assumptions, namely choosing the maximum impurities of activating nuclides.

During the construction of the BNCT station in the 1990's parts of biological shield concrete were removed by diamond drilling. The drilling location is illustrated in Figure 16. The most active drill core was studied with gamma spectrometry. The computational specific activity for this sample was around 10 Bq/g.
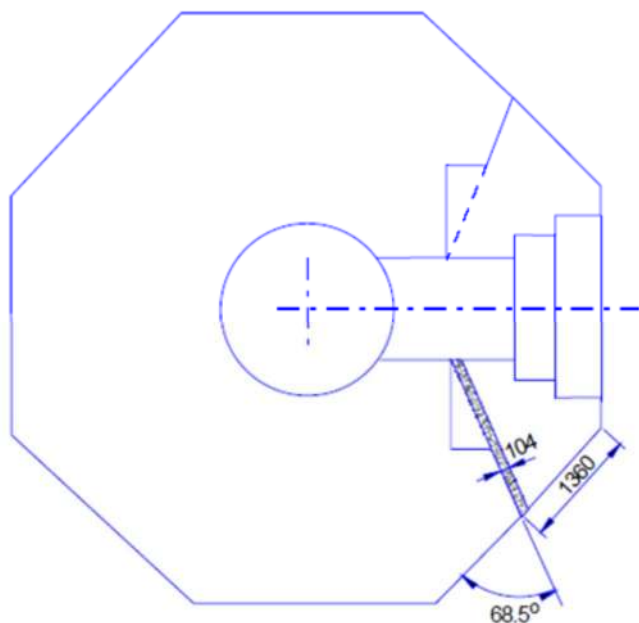
Figure 16: Original location of the drill core.

Gammaspectrometric measurements were performed using ISOCS system [15]. Used detector was BE2020 and the calibrations were done using ISOCS version 4.3. Distance from source to detector was 540 mm. The measured spectrum is illustrated in Figure 17. Results and comparison with calculation are listed in Table X.
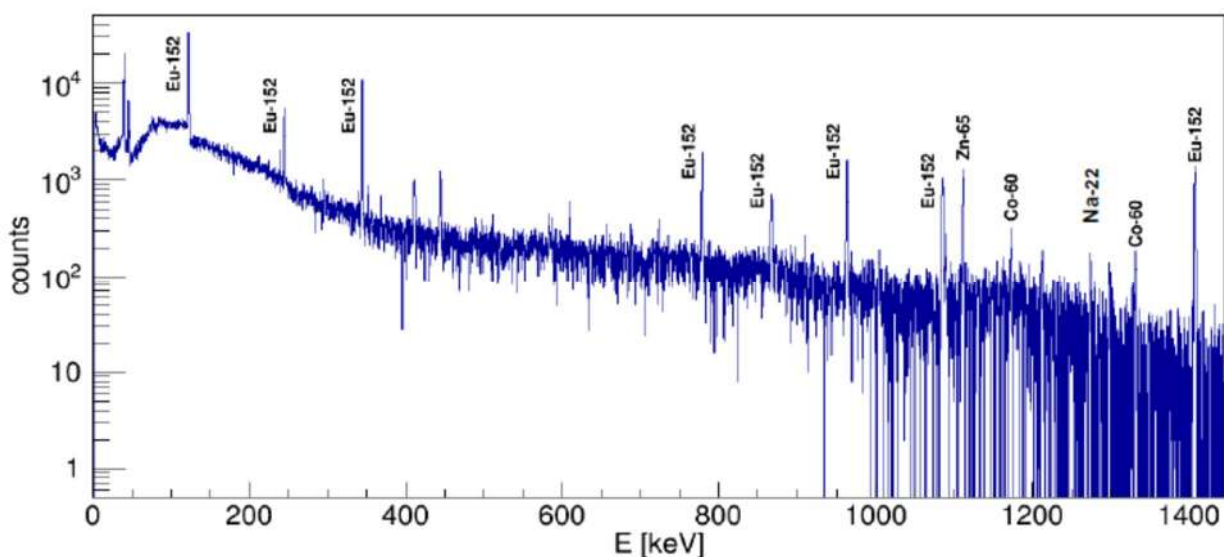

Figure 17: The gamma spectrum of the concrete sample.

Table X: Comparison of measured and calculated gamma activities in concrete.

Nuclide	Measured (Bq/g)	Measurement uncertainty (%)	Calculated (Bq/g)	Calculation uncertainty (%)
Co-60	2.5E-02	6.93	2.6E-02	10
Eu-152	9.6E-01	2.29	8.1E-01	10

Uncertainties in the calculation arise from the facts input neutron spectrum cannot be modelled exactly. Moreover, the material composition of the samples has been assumed to be homogenous, but in real life some variations always occur. However, a separate calculation was performed for this exact location and measured concrete composition was used as input quantity. An uncertainty of ten percent was estimated. Measurement uncertainties arise mainly from separating the background from the measurement data and converting the cpm to Bq/g. Bearing these in mind, specific activities correspond to the values reasonably well, although the measured sample was quite old and had a very low activity. Co-60 and Eu-152 will be chosen as key nuclides for future concrete waste measurements.

IV. CONCLUSION AND FUTURE WORK

This paper describes preliminary measurements that will be used in decommissioning project of FiR1 TRIGA research reactor. Computational estimates of total activity inventories have been published earlier [7], but measured values are required to verify the results and to plan safety of workers during dismantling and all legal requirements for waste packaging and final disposal.

The presently completed measurements support earlier calculations, although only low activity samples have been studied so far. To determine the radiological condition of the facility in detail, samples need to be taken also from more active parts with a systematic sampling plan. Work will continue with collecting more samples from different parts of the facility (especially from biological shield concrete) and from material that have not been drilled yet, such as steel inside the tank, Fluental moderator and beam tube plugs. This requires a detailed sampling plan and well-established measurement methods. Moreover, beta active nuclides such as Ni-63, Fe-55 and Ca-41 need to be measured via chemical dissolving, radiochemical separation and liquid scintillation counting. Only after that, full nuclide vectors and scaling matrices can be reported.

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