

# FIR 1 TRIGA activity inventories for decommissioning planning

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## ABSTRACT

*The objective of the study has been to estimate the residual activity in the decommissioning waste of TRIGA Mark II type research reactor FiR 1 in Finland. Neutron flux distributions were calculated with Monte Carlo code MCNP. These were used in ORIGEN-S point-depletion code to calculate the neutron induced activity of materials at different time points by modelling the irradiation history and radioactive decay. 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. Decommissioning waste consists mainly of ordinary concrete, aluminium, steel and graphite parts. Results include uncertainties due to assumptions on material compositions and possible diffusion of gaseous nuclides. Comparison to activity inventory estimates of two other decommissioned research reactors is also presented.*

## 1. Introduction

FiR 1 TRIGA Mark II type research reactor has been in operation in Finland since 1962. During its history FiR 1 has been used for training, scientific purposes, and isotope production. The reactor was permanently shut down in summer 2015 and will be decommissioned in following few years.

Decommissioning planning requires knowledge on the activation levels and amounts of radioactive decommissioning waste to design safe dismantling procedures. The activity inventories of different waste streams are also important for adequate waste packaging and planning both the interim storage and the final disposal of the decommissioning waste. Computational approach enables a non-destructive initial estimate, but systematical measurements are also needed later to verify these results.

In this paper the activity inventories of the FiR 1 decommissioning waste are estimated based on FiR 1 specific calculations. These are based on 3D Monte Carlo MCNP model estimating neutron flux in different components and using these fluxes in a point-depletion code ORIGEN-S to calculate actual activation of material taking into consideration the detailed irradiation history. Results are listed briefly for main materials, and some of the most important components are discussed in more detail. Latter ones are also compared to two other decommissioned TRIGA type reactors.

Material-wise total activities have been published in 2015 [1]. This work extends those results to more detailed component-basis and reviews the validation measurements performed so far.

## 2. FiR 1 TRIGA Type Research Reactor

FiR1 TRIGA Mark II reactor is entirely above ground and is surrounded by a concrete shield structure, as shown in Figure 1. The thermal reactor power was upgraded from 100 kW to 250 kW in 1967, and more concrete radiation shielding was added above the bottom octagon structure.

The reactor core and reflector assembly (see Figure 1) are located near the bottom of an aluminium tank 6.4 metres deep and 2.0 metres in diameter. Approximately 4.9 metres 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. They have been plugged in late 1980s'.

Originally, FiR 1 reactor has had a 1.2x1.2x1.7 metres graphite thermal column extending from the outer surface of the reflector assembly and penetrating the reactor tank and shield structure (see Figure 1). In 1995-1996, the thermal column was replaced by an epithermal boron neutron capture therapy (BNCT) beam. [2,3] BNCT irradiation room was also build of steel tube elements filled with density optimized heavy concrete and a heavy steel framed lead door. Based on the earlier shielding design calculations [4], it can be assumed, that these structures are not activated, and it is justified to exclude them from the activation inventory calculations. 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.

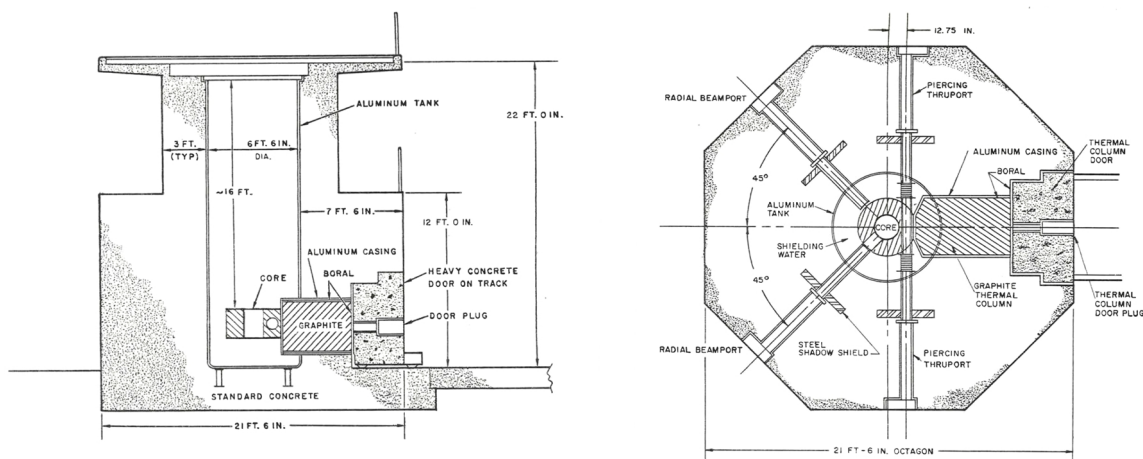


Figure 1 : Vertical and horizontal section views of FiR 1 reactor with thermal column.

## 3. Method of Calculation

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 [5] is presented in Figure 2.

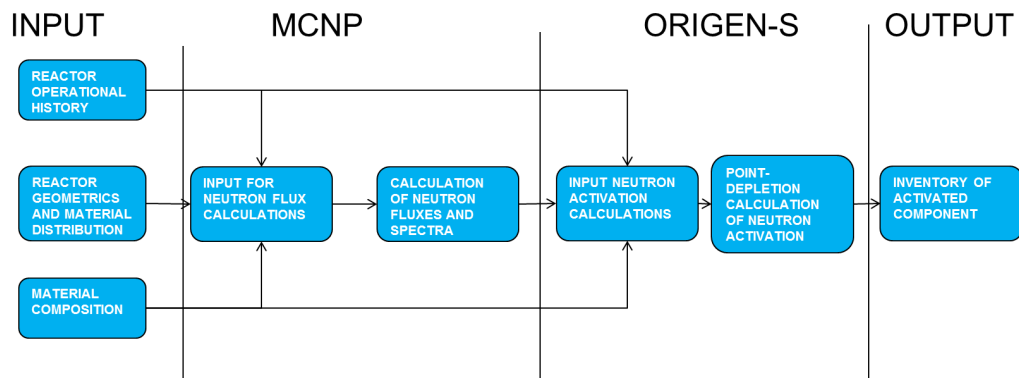


Figure 2 : Model scheme for activity inventory calculation [5].

In this study, neutron fluence rates in FiR 1 reactor were calculated with Monte Carlo code MCNP5 [6] 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. Considering all major modifications during reactor operational history, altogether three basic MCNP models were constructed:

1. MCNP model with thermal column and open beam ports
2. MCNP model with thermal columns and plugged beam ports
3. MCNP model with BNCT facility and plugged beam ports

Two-dimensional cross sections of the MCNP geometry are shown in Figure 3 for model with thermal and model with BNCT facility, respectively.

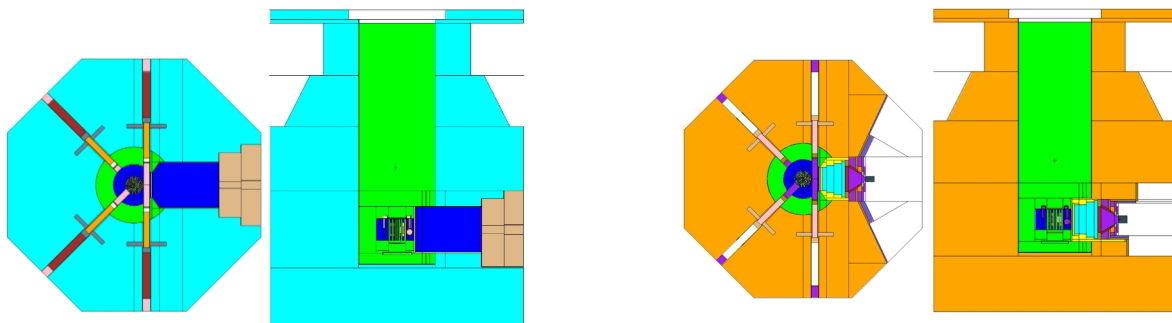


Figure 3: Horizontal and vertical cross sections of the MCNP geometry with the thermal column and BNCT facility. .

After calculating the fluxes in three-group form, they were used to model the decay chains of different nuclides in all the structures with point-depletion code ORIGEN-S [7]. Irradiation history was modelled according to reactor's actual operational history for years 1962-2015 taking into account thermal power increase, beam port plugging and building of the BNCT station. After irradiation, long decay period was modelled to study the radionuclide inventory both during dismantling and final

disposal. Total neutron flux was considered constant during all irradiations and zero in between them. Altogether, FiR 1 has been used for around 11 500 MWh.

ORIGEN-S calculations were performed on component or structure base resulting in around 200 output files. Detailed results will be used during practical dismantling work, but only most important components and material-wise total values are listed here.

## 4. Results

Material-wise total activities are listed in Table I. [1] Thermal column structures were modelled only for years 1962-1995, and BNCT structures for years 1997-2015, respectively. Material definitions were mostly based on original construction specifications. Since activity is mostly due to small impurities, their fractions were overestimated conservatively. Some of the most important materials and components are studied in following subchapters.

Table I: All the parts to be decommissioned. [1]

Material	Volume (m <sup>3</sup> )	Mass (kg)	Main isotopes	Total activity (TBq)
Concrete	25	61 000	H-3, Fe-55, Co-60, Eu-152, K-40	0.105
Graphite	2.61	4447	H-3, C-14, Co-60, Ba-133	0.859
Steel	0.449	3 540	Ni-63, Fe-55, Co-60, Ni-59, C-14	1.577
Aluminium	0.836	2220	Fe-55, Zn-65, Ni-63, Co-60, Mn-54, Fe-59	0.385
Fluental	0.45	1330	H-3, C-14	36.50
Other	10	2700	H-3, C-14, Ag-108M, Ni-63	0.416
<b>Sum</b>	<b>39.5</b>	<b>75100</b>		<b>39.821</b>

### 4.1 Graphite reflector

FiR 1 contains a 550 kg graphite reflector around the core. According to General Atomics specifications and Paul Scherrer Institute measurements [8] it is AGOT graphite type with porosity of 24 percent. Porosity is especially important because the nitrogen in air of the porous holes produces large amount of radioactive carbon-14 via N-14(n,p)C-14 reaction.

The reflector has been in  $1.8 \times 10^{12}$  n/s flux during whole reactor power history and total activity is around 290 MBq. This is mostly due to tritium. Other important nuclides are e.g. C-14, Be-10, Co-60 and Ba-133. Measurements on reflector graphite have not been conducted so far, but preliminary combustion measurements on thermal column graphite have been performed with the University of Helsinki on thermal column graphite, and the results for C-14 were on the same scale.

Because all TRIGA reactors have somewhat similar graphite reflector, this paper compares FiR 1 inventory calculations results to two decommissioned TRIGA reactors: MHH at Medical University of Hannover [9] and KRR-1 in Korea Atomic Energy Research Institute [10]. MHH started its operation in 1973 and KRR-1 in 1962. MHH and KRR-1 reactors were shut down in 1997 and 1995, respectively, The main difference between FiR 1, MHH and KRR-1 is that the thermal column in

FiR 1 was disassembled in 1995 and TRIGA Mark I type MHH has only one radial beam tube. KRR-1 has been used altogether for around 3 700 MWh, MHH for around 2 188 MWh and FiR 1 around 11 500 MWh.

Table II list the specific activities of the reflector graphite in these three reactors. Specific activity in FiR 1 is the highest, as expected due to longer power history. However, all reflectors have the same nuclides with similar fractions of specific activities, although porosity in KRR-1 and MHH was not known.

Table II: Specific activities of graphite reflectors in FiR 1, KRR-1 and MHH.

Component	Specific activity in FiR 1 (Bq/g)	Specific activity activity in KRR-1 (Bq/g)	Specific activity in MHH (Bq/g)
Core reflector	H-3: $4.95 \times 10^5$ C-14: $8.79 \times 10^3$ Co-60: $2.82 \times 10^4$	H-3: $4.13 \times 10^3$ C-14: $1.92 \times 10^2$ Co-60: $1.39 \times 10^2$	Co: $2.0 \times 10^3$

## 4.2 Irradiation ring

Rotary specimen rack or irradiation ring is a structure in the graphite reflector that contains several smaller steel and aluminium parts. A detailed CAD model was also build, but results are presented for the whole component only, since it is plausibly going to be dismantled in one piece. The overall mass is around 57 kg, of which 6.7 kg is steel. Irradiation rings has been in the reactor under very heavy flux for the whole operating history, and is likely to be highly active especially due to impurities of cobalt and nickel.

Total calculated activity of the FiR 1 irradiation ring is about 1.7 TBq, of which large part is due to beta active iron and nickel isotopes. More important cobalt activities are compared with irradiation rings in MHH and KRR-1 in Table III. Specific activity in FiR 1 is higher, as expected considering longer operating history of FiR 1. However, fractions of specific activities of are also on the same scale.

Table III : Specific activities in the irradiation ring.

Component	Specific activity in FiR 1 (Bq/g)	Specific activity activity in KRR-1 (Bq/g)	Specific activity in MHH (Bq/g)
Rotary specimen rack (steel parts)	Fe-55: $1.36 \times 10^7$ Co-60: $2.77 \times 10^7$ Ni-59: $4.53 \times 10^5$	Fe-55: $8.93 \times 10^5$ Co-60: $7.01 \times 10^6$ Ni-59: $3.00 \times 10^4$	Co-60: $5.0 \times 10^6$

## 4.3 Biological shield concrete

The largest mass of the decommissioning waste comes from biological shield concrete. Used composition is based on in-house measurements. [11] These show e.g. 2.2 ppm of europium, 13 ppm of cobalt, 50 ppm of nickel and 4.0 ppm of uranium, for instance.

Inventory calculations of concrete are based on analysing in more detail the specific activity per 1 kg of concrete at 30 different points. Following the clearance regulations of the Finnish Radiation and Nuclear Safety Authority [12], calculations estimated that concrete is activated around the core in about 1.5 m radius and around the beam tubes in about 0.5 m radius. Fluxes in activated areas are demonstrated in Figure 4. Final estimated average specific activity of concrete is 1.7 kBq/g. For instance, MHH has 1.1 kBq/g.[9].

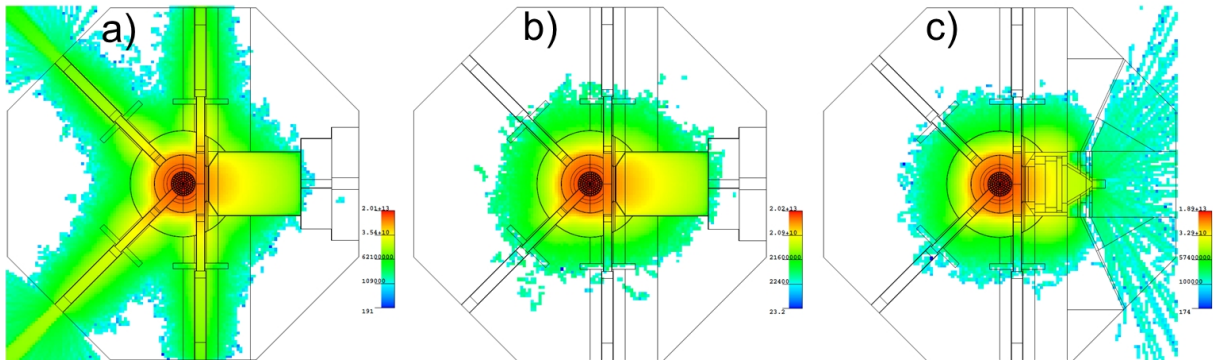


Figure 4 : MCNP simulated total neutron fluence rates a) with thermal column and open beam ports, b) with thermal column and plugged beam ports, and c) with BNCT beam.

#### 4.4 Fluental Neutron Moderator

Fluental™ [13] is a special neutron moderator material used in the BNCT facility to shape the neutron energy spectrum to an epithermal neutron beam. It is a special mixture of 69 w%  $\text{AlF}_3$ , 30 w% Al and 1 w% LiF with density of  $3 \text{ g/cm}^3$ . [14]. The BNCT beam structure is illustrated in Figure 5.

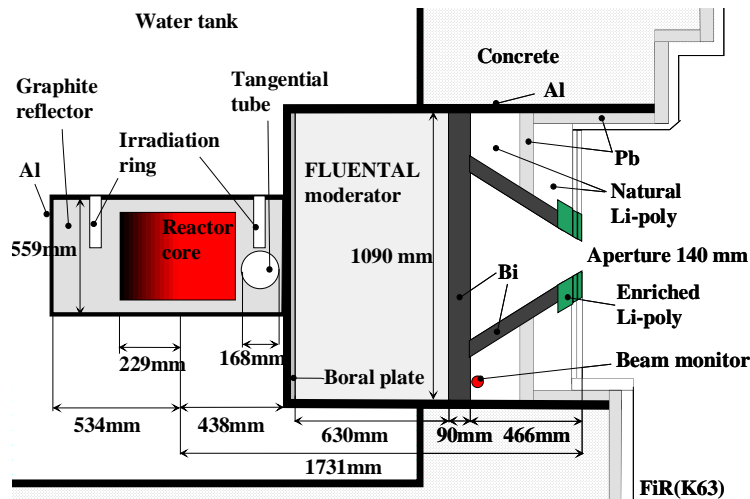


Figure 5 : Vertical section view of epithermal BNCT beam.

In the calculated activity inventories, tritium ( $\text{H-3}$ ) is one of the major contributors to the total activity. The most important nuclear reaction, which yields the beta active tritium ( $\text{H-3}$ , half-life 12.323a) is  $\text{Li-6}(n,\alpha)\text{H-3}$ . With fast neutrons,  $\text{Li-7}(n,\alpha)\text{H-3}$  reaction also occurs. Cross section for these reactions and fluxes in Fluental are illustrated in Figure 6.

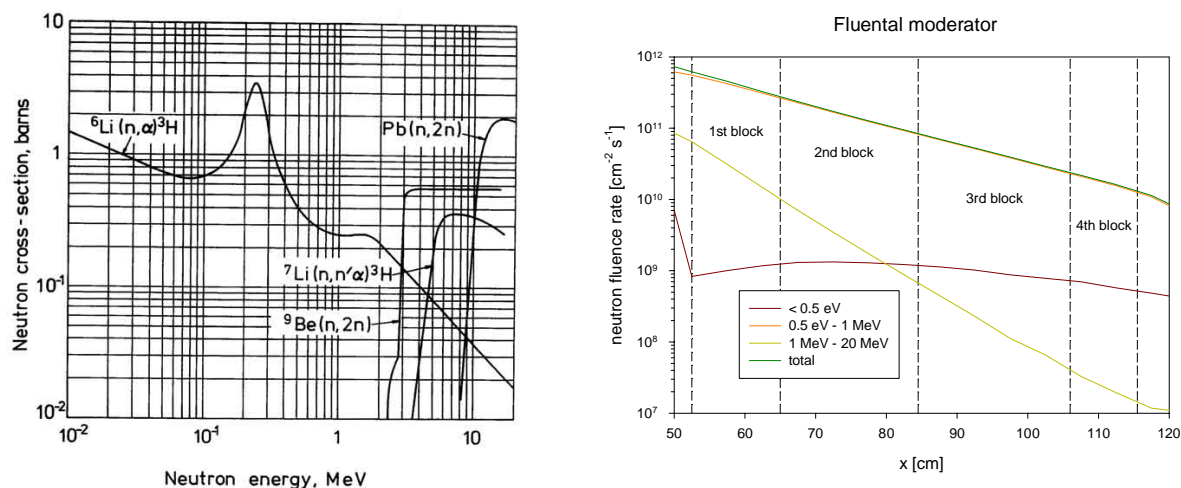


Figure 6 : Li-6 and Li-7 neutron cross sections [15] and neutron fluxes in Fluental.

The largest tritium content is calculated to be in the Fluental moderator blocks of the BNCT beam structure. In addition, some tritium is also calculated to exist in graphite and even in small extent in the concrete. Concerning the Fluental and thermal column graphite, the actual tritium inventory could be less than the calculated inventory. This is due to the fact, that in the thermal column and current BNCT beam structure, constant argon ventilation exists. Argon ventilation is used to ventilate the radioactive Ar-41 from the reactor building through specific piping leading to the exhaust tower. Ar-41 is created during reactor operation in all structures containing air and being exposed to neutron radiation. The argon ventilation system is original TRIGA reactor design. It is somewhat unclear how much tritium from of the thermal column graphite and Fluental has been ventilated into air outside of the reactor building, together with Ar-41. Specific argon release monitoring system is installed in the ventilation system and the annual release of Ar-41 from FiR 1 reactor is estimated based on the measured values (with a Geiger tube measuring gamma radiation through the ventilation piping). However, tritium emits only low energy beta particles with average energy of 5.7 keV, which cannot be easily measured. Some preliminary tritium concentration measurements in argon ventilation have been performed in 2015 using air bubbling through non-tritiated water and measuring the water samples with liquid scintillation counting (LSC) technique. Extrapolating these preliminary results shows that total release for years 1997-2015 could be around 6 GBq. This is less than a percent of calculated total production via neutron activation. Plausibly tritium is released only from the outer parts of Fluental and diffusion from the middle parts is much slower than the amount of produced new tritium. Moreover, tritium is also formed in lithiated plastic parts in the BNCT facility, and these measurements cannot distinguish tritium release from different sources.

In June 2015 three non-active samples of Fluental were irradiated in the reactor irradiation ring for 14 hours. These have been studied by dissolving them in water for few weeks. Measuring the tritium activity in water with LSC technique estimated that specific activity is gradually increasing, but still stays under one percent of calculated estimate. Plausibly tritium does not dissolve into water easily at room temperature, as have been noticed with other aluminium-lithium alloys [16, 17, 18]. Future work will include chemical separation with acid dissolution and higher temperatures, but the mechanisms of diffusion of tritium from Fluental are still not known well enough.

For the moment it is assumed that almost all of the produced tritium still remains in Fluental material. Further studies are needed before and during dismantling. Since beta-activity measurements with LSC technique (including tritium measurements) are time taking, requiring typically first the chemical separation of elements, suitable key nuclides and phenomena related to tritium diffusion should be studied further.

#### 4.5 Aluminium and steel parts

Neutron fluxes in the reactor core area have been around  $2 \times 10^{13}$  n/s (Figure 7), but vary a lot outside the reactor core, depending on distance from the core and attenuating materials between the core and the region of interest. Nevertheless, neutron flux close to the reactor is such that consequently all metal structures near the core are highly active. Most important nuclides are cobalt and nickel, which are impurities in both steel and aluminium.

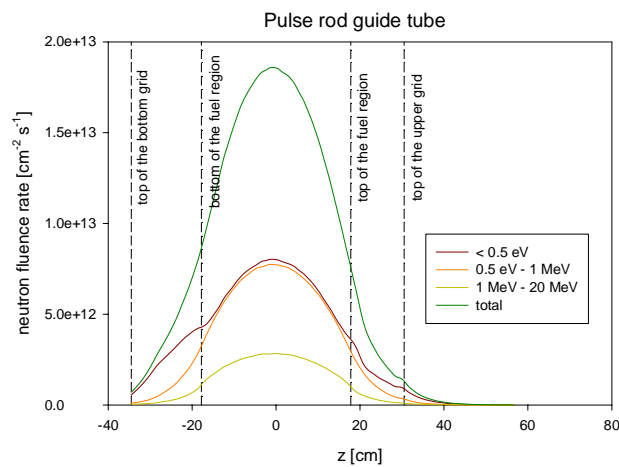


Figure 7 : Vertical neutron flux distribution through the reactor core along the pulse rod guide tube.

Excluding the irradiation ring, most of the steel activity comes from the bellows and clamps around the beam tubes (1.5-8.5 MBq/g) and small bolts and dowel pins inside the tank (0.25-750 MBq/g). Steel shadows around the beam tubes are heavy, but their activity is relatively low (1.5-12 kBq/g).

Aluminium is used mostly for the reactor tank, beam tubes and the tubes and grids inside the tank. All the parts inside the tank were assumed to be of the AlMg3 type. Calculated aluminium activity differs significantly from less than 1 Bq/g to around 1 MBq/g, but stays much lower than for the steel parts. Activity relevant from the dismantling point of view is due to activation of impurities in the material, such as Fe, Co and Zn.

Specific activities of some metal parts are compared in

Table IV between FiR 1 and MHH reactors. Values in FiR 1 are higher, but still on the same scale. Same applies to screws and small steel components inside the tank. These may have variation depending on initial compositions, but higher values were also expected, since FiR 1 has longer power history.



Table IV: Activities of certain components in MHH TRIGA reactor [9] and corresponding FiR 1 values.

Component	Material	Specific activity in MHH (kBq/g)	Specific activity in FiR 1 (kBq/g)
Upper grid plate	AlMg3	39.0	68.9
Steel components (screws etc.)	Stainless steel	17 370	50 200
Central irradiation tube	AlMg3	182.8	202.4

## 5. Discussion

Main uncertainty in the activity inventory of FiR 1 is related to tritium gas in Fluenta moderator. Tritium emits only low energy beta particles which makes the measurements challenging. Tritium is also highly mobile. Consequently some part of the tritium inventory may have evaporated from the materials. The same problem occurs with all other gaseous nuclides as well. However, preliminary measurements show that this diffusion mechanism is still unclear.

Other main sources of uncertainty are initial assumptions on material compositions. Because activity is usually due to small impurities in materials, even low concentrations of e.g. cobalt, nickel, europium or zinc may have notable effect on total activities. This was taken into account by assuming conservatively high impurities, but results should be verified later with systematic measurements. Even smaller quantities especially in gamma active nuclides are important, because during dismantling process, waste is usually classified according to keynuclides and activity of other nuclides is determined by multiplying with their fractions in total activity. As gamma active nuclides are easy to measure, they are a natural choice for keynuclides.

This paper does not list nuclides according to their decay modes, biological hazardousness or chemical properties. E.g. iron-55 and tritium are not highly important from the point of dismantling, but constitute a major part in the total activity. In addition, the importance of the nuclides is different from the occupational health and safety point of view during dismantling operations, and from the long-term safety point of view concerning the final disposal of the decommissioning waste.

## 6. Conclusions and Future Work

This paper describes the results and methodology used in activity inventory calculations for FiR 1 research reactor decommissioning planning. Total inventory was around 39.5 TBq with main contributing nuclides being H-3, C-14, Fe-55, Co-60, Ni-59, Ni-63, Eu-152 and Zn-65.

According to the inventory calculations, Fluenta moderator of the BNCT facility has accumulated a huge amount of tritium, which constitutes the major amount of overall activity in the FiR 1 decommissioning waste. However, some part of the tritium inventory may have evaporated. Consequently, current computational activity inventory results still contains large uncertainties.

FiR 1 reactor was permanently shut down in summer 2015. Current plan is to return the fuel to U.S. under U.S.DOE Spent Nuclear Fuel Acceptance Program [19]. All other waste will be eventually disposed to Finnish NPP's final disposal sites in 2030's.

Main work in near future is to verify computational activity inventory results with systematic radioactivity measurements and to study the requirements for both interim storage and final disposal of the FIR 1 decommissioning waste.

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