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Activity Characterisation Methods in FiR1 Decommissioning Project

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ABSTRACT

FiR1 is a 250 kW TRIGA Mark II type open pool reactor from General Atomics. It has been in operation for education, research, BNCT treatments and isotope production in Finland for years 1962-2015 and it is to be decommissioned in following few years. Overall the reactor has been in use for around 11 500 MWh. The presentation introduces the scaling factor method that is used to characterise the active decommissioning waste. The method is based on a combined MCNP-ORIGEN-S calculation model, which is verified with gammaspectrometric and radiochemical activity measurements.

1 INTRODUCTION

FiR1 is a 250 kW TRIGA Mark II type open pool reactor from General Atomics. The reactor is entirely above ground and surrounded by a concrete shield structure, as shown in Figure 1. [1]

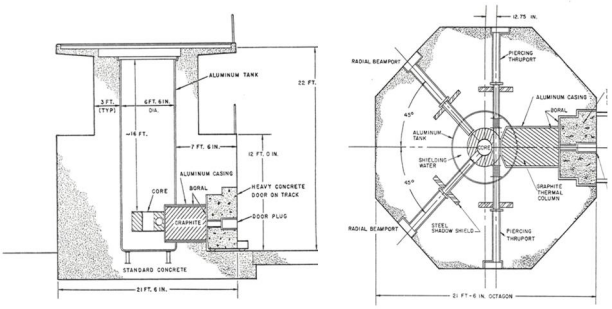

Figure 1: Vertical and horizontal view of the FiR1 research reactor. [1]

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.

Originally, FiR 1 reactor 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]

Radioactive characterisation means studying the radioactive properties in the facility materials by giving estimations of component level activations and radionuclide inventories. Aim is to ensure safe dismantling (minimizing the radiation doses to the workers) and optimised waste management of the decommissioning waste (minimizing the amount of active final disposal waste).

In 2019 the FiR1 reactor is at extended shutdown state. Official decommissioning licence from the state council is expected in 2019. Actual dismantling will take place after returning the spent nuclear fuel to US, plausibly in 2021-2022. Currently, FiR1 reactor components have been modelled and verification of the results and formation of scaling factors are carried out using non-destructive and destructive analytical methods for active samples of various materials.

2 SCALING FACTOR METHOD

The nuclide vectors of the decommissioning waste are divided into easy-to-measure gamma active key nuclides and difficult-to-measure beta active nuclides.

During the dismantling of a nuclear reactor, dozens or even hundreds of decommissioning waste packages need to be characterised without disrupting the dismantling process. This requires fast and reliable sampling and characterization methods to determine the inventory and activity concentration of the radionuclides. Because measuring beta active nuclides can take several days, an optimal approach needs to be found in sampling and activity analyses. According to Finnish regulatory guides for nuclear and radiation safety [4], 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 difficult-to-measure beta activities assuming the same ratio between nuclide-wise activities as in the samples. The procedure is illustrated in Figure 2.

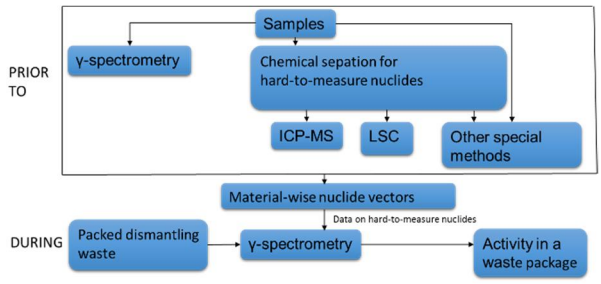

Figure 2: Scaling matrix approach in activity characterisation.

3 CALCULATION MODEL

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 3.

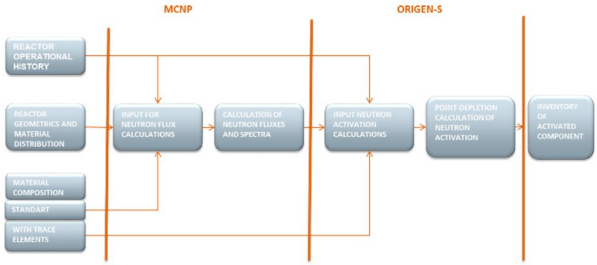

Figure 3: Calculation scheme.

At VTT, 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 energy group structure as specified in ORIGEN-S manual. All major modifications during reactor operational history were taken into account by creating separate models for different reactor configurations.

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. ORIGEN-S calculations were performed on component or structure base resulting in around 200 output files. In some cases data was not available and then conservative assumptions were made for missing data. Elemental analysis tools were used for the chemical composition analysis in cases when original materials were available. An overview of waste quantities is presented in Table 1.

Table I: Calculated total activities

Material	Volume (m3)	Mass (tons)	Main isotopes	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

4 MEASUREMENT TOOLS

Characterisation is a combination of modelling and experimental analysis. Calculation model provides an non-destructive approach to early phases of a decommissioning project, but it often contains approximations and the results need to be validated with sample measurements. Moreover, composition measurements provide initial data on activating impurities for activity calculations.

Experimental analysis is divided to non-destructive analysis (NDA) e.g gamma spectrometry and destructive analysis (DA) e.g elemental analysis of stable isotopes and radiochemical analysis of difficult to measure (DTM) radionuclides. Analytical techniques used in the characterisation of FiR1 materials are mainly ISOCS gamma spectrometry, HR-ICP-MS, ICP-OES and Hidex liquid scintillation counter.

Activating nuclides are typically small impurities on construction materials, e.g. Co, Eu, Ni etc. For example, trace levels of impurities in concrete do not have any mechanical effect and therefore they are not necessarily included in the chemical composition listed in original construction spesifications. Therefore, the initial composition of the materials used in the calculations required

measurements. VTT has measured the compositions of FiR1 materials using Thermo Scientific ELEMENT 2TM HR-ICP-MS, which is able to analyse trace level concentrations as low as ppt level (Figure 4). Prior to the elemental analysis, the solid material needs to be dissolved and most often microwave digestion is the used technique. Agilent SVDV 5100 ICP-OES is another analytical tool used for the analysis of stable elements in ppm level. In the case of FiR1 characterisation, ICP-OES has been especially used in determination of yields after radiochemical analysis of DTM radionuclides and measuring impurities on concrete composition. [8-10].

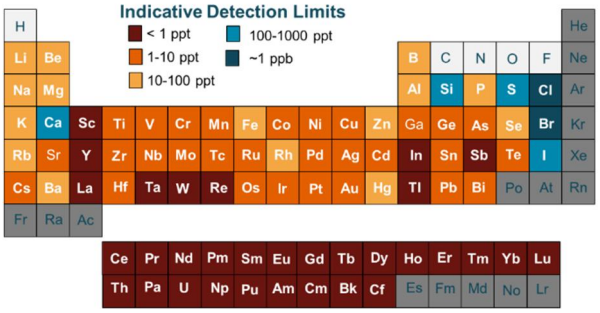

Figure 4: Indicative detection limits for HR-ICP-MS at VTT the Centre of Nuclear Safety. [11]

Gammaspectrometric analyses are used to measure the key nuclides in sample nuclide vectors and to classify the final waste packages using scaling matrix approach. VTT uses Canberra ISOCS (In-Situ Object counting system) with an HPGe detector (5-2700 keV). It enables performing the efficiency calibration also for objects of complicated geometries. A rotating barrel measurement setup is presented in Figure 5.

Figure 5: Rotating gammaspectrometric measurement setup.

Radiochemical methods are used to measure the alpha and beta emitters in decommissioning waste. These require dissolving and chemical separation to purify the radionuclide of interest from each other and possible interfering radionuclides. VTT has also used oxidizers to separate volatile nuclides from e.g. graphite and lithium-enriched shielding materials. [8-10] The activity measurements are carried out using Hidex 300 SL liquid scintillation counter with a TDCR technology and yield measurements using ICP-OES.

5 CONCLUSIONS

The characterisation results in FiR1 decommissioning project provide data for many parts of the project, e.g. radiation safety, waste management and dismantling planning.
Current work is focused on sample measurements and VTT is gradually shifting towards applying the methods in actual dismantling process. This development work provides valuable experience also for characterisation work of other future decommissioning projects.

ACKNOWLEDGEMENTS

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