Nuclear Science and Technology Symposium - SYP2022
Helsinki, Finland, 1-2 November 2022

Scaling Factor Formation of FiR1 decommissioning waste

Antti Räty, Anumaija Leskinen
Technical Research Centre of Finland
Kivimiehentie 3, 02044 VTT, Finland
antti.raty@vtt.fi, anumaija.leskinen@vtt.fi

ABSTRACT

Characterisation of FiR1 decommissioning waste prior to dismantling has focused on formation of material specific scaling factors. In total, 15 scaling factors have been developed based on characterisation of original chemical compositions, activation calculations, and measurement of both easy-to-measure (ETM) and difficult-to-measure (DTM) radionuclides. Characterization work in the FiR1 project has improved VTT's capabilities e.g. activity calculation software, sampling activated materials and measuring long-lived DTM nuclides. Characterization will continue during the reactor dismantling together with Fortum by collecting samples from areas which have been earlier inaccessible.

1 INTRODUCTION

The first reactor in Finland, namely FiR1, is a 250 kW TRIGA Mark II type research reactor, which has been in operation in years 1962-2015. Spent nuclear fuel was sent for re-use to another TRIGA reactor in the USA and activated reactor structures will be dismantled in 2023.

Characterisation is a crucial part of a decommissioning project as it forms the basis for the waste management planning and cost estimation. As such, characterisation is needed prior to dismantling and one of the main efforts is in formation of material wise scaling factors. In the preliminary phase of FiR1 decommissioning project, VTT conducted activity calculations of the reactor components. The results were validated with various samples from the materials that were accessible at that stage of the project (i.e. before removal of the spent nuclear fuel). [1, 2, 3, 4]

This paper presents the formation of material specific scaling factors for FiR1 decommissioning project. The scaling factors will be utilised in calculation of Difficult-to-Measure (DTM) radionuclides in decommissioning waste after gamma spectrometric analysis of Easy-To-Measure (ETM) radionuclides.

2 SCALING FACTOR FORMATION

Formation of the scaling factors is based on ISO 21238:2007 standard [5]. Since the decommissioning waste will be disposed at Loviisa nuclear power plant site, their waste acceptance criteria was also followed.

The variety of materials (Figure 1) especially in research reactors poses a significant challenge in formation of nuclide vectors. VTT decided to form vectors for all activated assuming that e.g. different aluminium types are handled using the same vector. Altogether this still means that 15 scaling factors were formed [6].

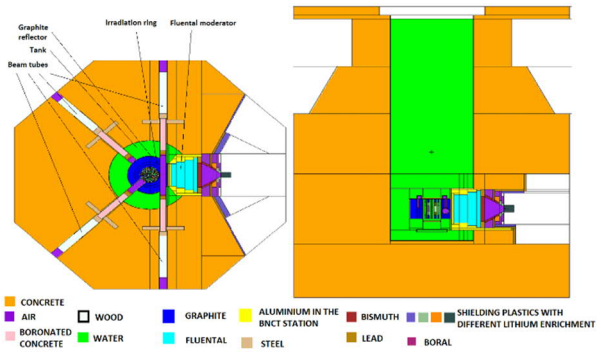

Figure 1: Materials in reactor structures. [3]

Formation of the scaling factors can be divided into four categories

1) scaling factors formed with analysis of chemical composition and activation calculations without validation (i.e. analysis of activated material)

2) scaling factors formed with combination of sampling and activation calculations with validation (i.e. analysis of activated material),

3) scaling factors formed with activation calculations using material standards or literature for chemical composition, and

4) scaling factor formed purely with sampling and analysis of active material.

The materials and scaling factor categories are listed in Table 1. Some of the scaling factors will be updated during the dismantling by Fortum and some may be used only for waste clearance.

Table 1 FiR1 scaling factor formation categories

Material	Scaling factor formation category
Steel	3
Aluminium	1
Reactor graphite	2
Concrete	2
Cadmium	3
Contamination	4
Ion exchange resin	4
Fluental	1
Bismuth	3
BNCT station lead	2
Lead in the beam port plugs	3
Boronated concrete	3
Li-rich plastic	4
Boral	1
Heavy concrete	2

Practically the process means updating the original calculated results using conservative assumption based on data from the sample measurements. The process in illustrated in Figure 2. This process requires enough redundant samples to avoid systematic errors from sampling or measurement technique. Statistical errors have been calculated following the approach in the ISO standard [5] with 95 percent confidence interval.

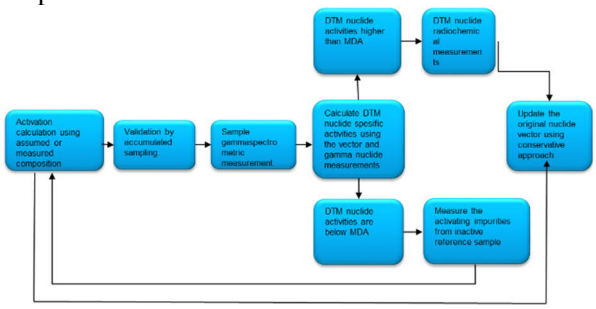

Figure 2: The process of updating calculated results using data from sample measurements.

2.1 Activation calculations

VTT's calculation model combined several MCNP neutron flux models representing different reactor operation phases to ORIGEN-S point-kinetic calculations taking into account the operation hours of each configuration (Figure 3) [1].

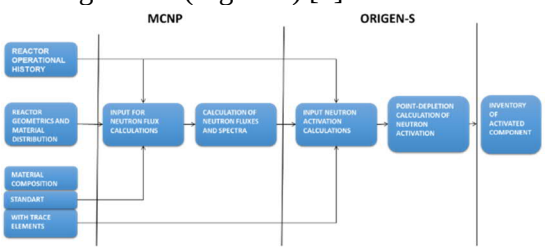

Figure 3: Illustration of the calculation system.

Material chemical composition, irradiation history and neutron fluxes are required as input data in the calculation model. Because the model assumes everything to be homogeneous, the calculations were repeated for around 200 components dividing the structures similarly as they would be actually dismantled.

2.2 Characterisation measurements

Characterisation measurements have included chemical analyses of original chemical compositions, radiochemical analyses of DTMs and gamma spectrometric analyses of ETMs. The main tools for the chemical analyses have been inductively coupled plasma optical emission spectrometry (ICP-OES) and mass spectrometry (ICP-MS). The ICP-OES and ICP-MS analysis techniques require dissolving of the material into a liquid form. Most often mixtures of acids, such as HNO_3 and HCl, are used with an analytical microwave oven, which utilise temperature and pressure for complete destruction of the material matrix. In some cases, such as with concrete, especially dangerous acid HF is requires for the complete material destruction. The ICP-OES is utilised for elemental analysis at macro level (ppm) whereas ICP-MS enables both elemental and isotopic measurements at micro level (ppb-ppt).

Dissolution of the material is also required in DTM analysis, in which the analyte of interest is separated from interfering elements and radionuclides using radioanalytical techniques such as ion exchange or chromatographic resin treatments and precipitations. Dissolution difficulties have been encountered especially for concrete and graphite, which required usage of HF and $HClO_4$ [7,8,9]. Another technique for the total destruction of the material is alkali fusion, for which method development is currently carried out in a master's thesis [10]. DTM analyses are further discussed in section 2.4.

The gamma spectrometric analyses of ETMs is a non-destructive analysis technique, which can be carried out in solid or liquid form and therefore it requires only preparation of a measurement sample. However, sampling, which can be a significant source of uncertainty, is an essential step in characterisation for both ETMs and DTMs. Sampling is further discussed in section 2.3.

2.3 Sampling

Perquisite for representative sampling is in understanding of the material

homogeneity/heterogeneity and radionuclide distribution in the material and their behaviour during sampling and storage. Practical challenges in the FiR1 project are typically related to the variety of material, relatively low specific activities and handling volatile nuclides. Some examples can be listed [11]

- If the activity of DTM nuclides is below MDA, the vectors can only be validated using measured impurities from inactive reference samples.
- In case of small quantities of several steel types the best approach has been to form only one vector using conservative assumptions and checking with the waste acceptor that the list contains all the relevant nuclides.
- A relevant gamma active key nuclide cannot be identified for some of the special materials (lithiated shielding plastics, Fluental neutron moderator, bismuth).
- How to measure the nuclides as a result of contamination from reactor operation (isotope production and activation analysis)? Current approach is to measure only gamma active nuclides and supplement the data with historical records of reactor operation.
- Since some of the material are inaccessible before dismantling commences, These vectors will be further refined and validated by Fortum during dismantling
- When sampling biological shield concrete, a homogenous sample was taken using an in-house developed drilling set-up, which enabled studying of release of H-3 and C-14 during drilling [12]. The results showed that significant amount of H-3 was released during drilling [13]. The drilling set-up was upgraded to include temperature control and successfully utilised in drilling of thermal column graphite (results to be published).

2.4 DTM measurements

As with any analytical measurement, validation of the results is required. Analysis of reference materials is a wide used way for the result validation. However, the lack of commercially available reference materials of DTMs in decommissioning waste prompted intercomparison exercise projects within the Nordic nuclear safety research (NKS) community. Radiochemical analysis of H-3, C-14, Fe-55, Ni-63, Sr-90 and gamma emitters have been studied in steel, concrete and spent ions exchange resin [14-18]. The exercise and statistical analysis of the results were based on ISO 13528 standard [19]. The main conclusions for the steel analysis were related to the Fe-55 quenching in liquid scintillation measurement (i.e., quenching of Auger electrons and low energy X-rays from electron capture decay) and removal of interfering Co-60 from Ni-63 fractions (i.e., chemical similarity of Co and Ni) [20,21]. The main conclusions for the concrete analyses were related to the problems in dissolution of the material, low activity of the material and original high Fe amount [22,23]. The main conclusions in the spent resin analysis were related to the ETM analyses have also their difficulties (i.e., complex matrix of radionuclides) whereas the DTM analyses were in general well aligned [24].

3 CONCLUSIONS

FiR1 characterisation has been a multi-year effort in the decommissioning project. Characterisation will continue also during the dismantling. In total, 15 scaling factors have been produced and some of them will be updated. Wide range of analytical capabilities has been developed and validated.

ACKNOWLEDGEMENTS

The authors would like to thank the FiR 1 research reactor personnel participating in the decommissioning project and national nuclear waste management fund for supporting the KYT-VAMMA and KYT-DEMONI projects developing measurement methods and their validation intercomparison exercises, which were partly funded by the Nordic Nuclear Research NKS-B programme.

REFERENCES

[1] A. Räty, P. Kotiluoto, “FiR 1 TRIGA Activity Inventories for Decommissioning Planning”, Nuclear Technology 194:1 (2016)

[2] A. Räty, “Activity Characterization Studies in FiR 1 TRIGA Decommissioning Project”, Doctoral thesis, Helsinki University, 2020

[3] A. Räty et al., “Preliminary Waste Characterisation Measurements in FiR 1 TRIGA Research Reactor Decommissioning Project”, Nucl. Tech., 203 (2018)

[4] A. Räty et al., “Characterization measurements of fluental and graphite in FiR1 TRIGA research reactor decommissioning waste”, Nuclear Engineering and Design 353 (2019)

[5] International Organization for Standardization, Scaling factor method to determine the radioactivity of low- and intermediate-level radioactive waste packages generated at nuclear power plants, ISO 21238:2007, 2007

[6] A. Räty & A. Leskinen, ” FiR1-tutkimusreaktorin aktivoituneiden rakenteiden esivaiheen nuklidivektorit”, VTT tutkimusraportti VTT-R-00114-22, 2022

[7] A. Räty et al., “Characterization measurements of fluental and graphite in FiR1 TRIGA research reactor decommissioning waste”, Nuclear Engineering and Design 353 (2019)

[8] A. Leskinen et al, ”DTM-Decom II - Intercomparison exercise in analysis of DTM in decommissioning waste” NKS-441, Roskilde, Denmark, 2021

[9] A. Leskinen et al., “Intercomparison exercise on difficult to measure radionuclides in activated concrete - statistical analysis and comparison with activation calculations”, Journal of Radioanalytical and Nuclear Chemistry 329 (2021)

[10] V. Meriläinen “Analysis of reactor graphite DTM and comparison of acid digestion and alkaline fusion as sample digestion methods for different matrices”, MSc thesis, Helsinki University (to be submitted 2022)

[11] M. Airila et al., “Case Study on Decommissioning of the FiR 1 TRIGA reactor”, VTT research report VTT-R-00090-22, 2022

[12] A. Leskinen, J. Hokkinen, T. Kärkelä, T. Kekki “Release of H-3 and C-14 during sampling and speciation in activated concrete”, Journal of Radioanalytical and Nuclear Chemistry 331 (2022)

[13] A. Leskinen, J. Hokkinen, T. Kärkelä, T. Kekki “Release of H-3 and C-14 during sampling and speciation in activated concrete”, Journal of Radioanalytical and Nuclear Chemistry 331 (2022)

[14] A. Leskinen et al., ”Intercomparison exercise in analysis of DTM in decommissioning waste”, NKS-429, Roskilde, Denmark, 2020

[15] A. Leskinen et al., ”DTM-Decom II - Intercomparison exercise in analysis of DTM in decommissioning waste” NKS-441, Roskilde, Denmark, 2021

[16] A. Leskinen et al., ”DTM-Decom III - Intercomparison exercise in analysis of DTM beta and gamma emitters in spent ion exchange resin”, NKS-457, Roskilde, Denmark, 2022

[17] A. Leskinen et al., “Intercomparison exercise on difficult to measure radionuclides in activated steel: statistical analysis of radioanalytical results and activation calculations”, Journal of Radioanalytical and Nuclear Chemistry 324 (2020)

[18] A. Leskinen et al., “Intercomparison exercise on difficult to measure radionuclides in activated concrete - statistical analysis and comparison with activation calculations”, Journal of Radioanalytical and Nuclear Chemistry 329 (2021)

[19] International Standard, “ISO 13528:2015(E) Statistical methods for use in proficiency testing by interlaboratory comparison”, Geneva, 2015

[20] A. Leskinen et al.,”Intercomparison exercise in analysis of DTM in decommissioning waste”, NKS-429, Roskilde, Denmark, 2020

[21] A. Leskinen et al., “Intercomparison exercise on difficult to measure radionuclides in activated steel: statistical analysis of radioanalytical results and activation calculations”, Journal of Radioanalytical and Nuclear Chemistry 324 (2020)

[22] A. Leskinen et al., ”DTM-Decom II - Intercomparison exercise in analysis of DTM in decommissioning waste” NKS-441, Roskilde, Denmark, 2021

[23] A. Leskinen et al., “Intercomparison exercise on difficult to measure radionuclides in activated concrete - statistical analysis and comparison with activation calculations”, Journal of Radioanalytical and Nuclear Chemistry 329 (2021)

[24] A. Leskinen et al., ”DTM-Decom III - Intercomparison exercise in analysis of DTM beta and gamma emitters in spent ion exchange resin”, NKS-457, Roskilde, Denmark, 2022