Scaling Factor Formation of FiR1 decommissioning waste

Finnish nuclear symposium, 1-2.2022 Antti Räty (Antti.Raty@vtt.fi)

Anumaija Leskinen (Anumaija.Leskinen@vtt.fi)

28/10/2022 VTT – beyond the obvious

Contents

- Introduction: FiR1 research reactor
- Characterisation roadmap
- Calculation and measurement methods in this project
- Challenges for nuclide vector determination
- How to divide the structures into nuclide vectors
- Example from activated concrete
- Conclusions

FiR1 TRIGA research reactor

- 250 kW TRIGA Mark II type research reactor
- Operational for years 1962-2015.
 - SNF was removed in 2020
- Training, Research and Isotope production reactor, General Atomics.
- LILW waste inventory less than 5 TBq or around 100 m³.
- Characterization approach
 - Scaling matrix method using pre-defined nuclide vector
 - ISO 21238-2007 standard
 - Loviisa waste acceptance criteria
 - Characterisation is a combination of calculations and measurements



Characterization roadmap





Basic idea

 Neutron activation is a linear process and radionuclides from a same source and behavior (speciation, charge, solubility, volatility, etc) maintain a constant relationship in their final activity concentrations



- Chemical and physical properties nedd to be similar enough
 - Special consideration on volatile nuclides
 - Nuclides with non-homogeneous distribution?
 - Samples must represent the studied material and plant areas well enough
- Special cases can occur
 - Li-enriched shielding plastics: not contain any homogeneously distribution gamma emitters
 characterization by sampling and measurements.

Definitions: scaling factor

- Nuclide vector determines the fraction of different nuclides forming the total activity in the item.
- A scaling factor, *SF*, is a ratio between the activity concentrations of two radionuclides in the item.

 $SF_{ij} = \frac{A_i}{A_i}$

- Activity concentration of the "denominator radionuclide" is called the key nuclide
- "numerator radionuclide" is the activity concentration of a difficult-to-measure radionuclide (DTM)
- After obtaining the SFs, activity concentrations of DTM's can be determined by multiplying the concentration of the key radionuclide with the SF.

-

Activity of a DTM nuclide (e.g. Ni-63)

NDE calculation is a two-step process



Repeat the process for individual components or reactor structural parts.

Measurements: Scaling matrix approach



Challenges regarding the methodology

- Complicated operating history, several structural modifications
 - Activation calculation had to be separated into several time periods.
- SNF still in the reactor tank until 2020
 - Some material could not be sampled, high underwater dose rates
 - Samples from outer parts of the reactor typically have very low activity
- Variety of materials and 1960's level of quality documentation
 - Simplify e.g. by handling all steel types with a conservative steel nuclide vector
 - Not only much information is missing, but also some of the original data is simply falce, e.g. composition impurities, but also dimensions.
- Some materials don't have a suitable key nuclide
- Non-nuclear requirements, e.g. chemical toxicity.
- How to separate contamination and activation?

VTT

Forming nuclide vectors



Different methods for different materials



VTT

Example: Concrete in FiR1 biological shield

Step 1: Collect the existing data Complete missing points (e.g. composition measurements)

Activation and dose rate calculations

Sampling plan and validate the results Dismantling plan (ALARA and optimizing the amount of active waste), waste packages and waste management

Dismantle the reactor. Measure the waste packages and classify them. Collect samples from waste packages for possible need for further validation

Possible pre-treatment and intermediate storage or final disposal Clearance measurement to free-release the building

VTT

Step 1: Collecting the existing data

- Construction drawings, dimensions, operating history (operating hours).
- If there significant changes in the reactor structures (power increase or such), the operating history need to be divided into different periods.
- Material composition may lack data on activating impurities
 - Effect on mechanical properties may be small.





Step 2: Complete the missing data

- Composition measurements from inactive parts.
- Prelimenary identification of the materials and structures to be dismantled
- Verifying the accuracy of original construction drawings
- Measuring dose rates and gamma spectra
- If some data is missing, use conservative assumptions:
 - Underestimate neutron shielding structures and material shielding properties
 - Overestimate activating impurities
 - Overestimate operating hours or underestimate cooling time.





| näyte A | näyte K | näyte Y |
|---------|--|--|
| ppm | ppm | ppm |
| | | |
| 70300 | 67900 | 68050 |
| < 100 | < 100 | < 100 |
| 860 | 840 | 840 |
| 1730 | 1835 | 2165 |
| 91000 | 79000 | 95000 |
| 55 | 56 | 59 |
| 12 | 13 | 13 |
| 1,9 | 1,7 | 1,7 |
| 2,1 | 2,0 | 2,2 |
| 23000 | 21000 | 23000 |
| 4290 | 4150 | 4430 |
| 35300 | 36450 | 33650 |
| 36 | 27 | 39 |
| 9000 | 7700 | 8500 |
| 350 | 310 | 370 |
| < 200 | < 200 | < 200 |
| 19000 | 20000 | 19000 |
| < 50 | < 50 | < 50 |
| < 500 | < 500 | < 500 |
| 3200 | 2400 | 2900 |
| 258000 | 258500 | 258500 |
| 8,3 | 8,0 | 13 |
| 1380 | 1210 | 1370 |
| 3,0 | 3,4 | 4,0 |
| | nityte A ppm 70300 < 100 860 1730 91000 55 12 1,9 2,1 23000 4290 35300 36 9000 350 < 200 19000 < 50 < 500 < 500 258000 8,3 1380 3,0 | $\begin{array}{c c c c c c c c c c c c c c c c c c c $ |



Step 3: Activation and dose rate calculatios

- Two step process: neutron transport calculations and activation calculations.
- In case of power increase (or other modifications), build separate models for each operating period.
- Utilize the results for prelimenary dismantling and waste management plans



Step 4: Sampling and validation

- Collect samples of the activated parts
- Measure the nuclide-wise activities and form the SFs
 - Validate and/or update the calculated data.
- Refine the data for dismantling and waste managements plans
 - If conservative assumption had been used, the amount of activated waste may be smaller.
 - Some part of volatile nuclides may have diffused, but use conservative assumptions.



Step 4: Sampling and validation – closer look at the experimental work

- General concept
 - Elemental and DTM analyses with destructive methods
 - ETM analysis with non destructive methods
- DTM analyses
 - Destruction of the matrix
 - Separation of the radionuclide of interest
 - Measurement
- Validation of the DTM analysis
 - No commercial reference materials
 - Intercomparison exercises organised
 - Activated steel
 - Activated concrete
 - Spent ion exchange resin











Step 4: Sampling and validation – closer look at the experimental work



- First sampling campaign for determination of chemical composition for activation calculations
- Second sampling campaign for ETM and DTM analyses
 - Coring of activated concrete cores
 - Subsampling of cores to produce fine power subsamples
 - Determination of volatile H-3 and C-14 during sampling
 - Same method utilised also for subsampling of activated graphite







Step 4: Sampling and validation – closer look at the experimental work



- Radiochemical method development
 - FiR1 decommissioning project
 - KYT-DEMONI project with Helsinki University
 - Various MSc studies
- Method validation via an intercomparison exercise
 - Participants from several Nordic laboratories, partially NKS funding
 - Difficulties with solubility
 - Difficulties with low activity



Step 5: Dismantling and waste management plan

- Choose the dismantling method
 - Include mechanical requirements (e.g. drilling vs. wire saw)
 - Logistics (both inside the reactor hall and highway transportation)
 - Dose estimate to personnel and contamination control
- Plan the packages
 - Logistics (both inside the reactor hall and highway transportation)
 - · Compatibility at the waste final disposal site









Lessons learned

- Scaling factor approach enables fluent dismantling by performing most of the characterization work beforehand.
- Reseach reactor contain special characteristics, which require assumptions and simplification in basic methodology.
- Having real samples is crusial for characterisation
 - Real compositions and operating history are crusial for the calculations.
 - Radiochemical method development cannot be performed with only pure reference materials.
 - Samples or redundant construction material should be available in early phase of a decom project.
- The decommissioning project and related KYT and NKS projects have contributed to method development, sampling, activity validation and education of experts.
 - Fit-for-purpose sampling techiniques (e.g cross contamination, volatility, form)
 - Material specific radiochemistry methods, method validation via intercomparison exercise
 - 1 PhD thesis, 5 MSc theses, ~10 peer-reviewed journal articles on characterisation topics.

References on methodology

- IAEA Safety report series No. 95: "Methodologies for Assessing the Induced Activation Source Term for Use in Decommissioning Applications"
- IAEA: Technical Report Series 389: "Radiological Characterization of Shut Down Nuclear Reactors for Decommissioning Purposes"
- ISO 21238:2007 (Scaling Factor Method to Determine the Radioactivity of Low-and Intermediate-Level Radioactive Waste Packages Generated at Nuclear Power Plants).
- IAEA Technical Report "International Experience in the Determination and Use of Scaling Factors in Waste Characterization".
- NUREG/CR-4101 "Assay of Long-Lived Radionuclides in Low-Level Wastes From Power Reactors".
- EPRI (USA): "Updated Scaling Factors in Low-Level Radwaste" (1987)
- EPRI (USA): "Utility Use of Constant Scaling Factors" (1999)

VTT references

- A. Räty, "Activity Characterization Studies in FiR 1 TRIGA Decommissioning Project", Doctoral thesis, Helsinki University, 2020
- A. Räty and P. Kotiluoto, FiR 1 TRIGA Activity Inventories for Decommissioning Planning, Nuclear Technology, Volume 194, Issue 1, April 2016
- A. Räty et al., "Preliminary Waste Characterisation Measurements in FiR 1 TRIGA Research Reactor Decommissioning Project", Nucl. Tech., 203 (2018)
- A. Räty et al., "Characterization measurements of fluental and graphite in FiR1 TRIGA research reactor decommissioning waste", Nuclear Engineering and Design 353 (2019)
- A. Räty, M. Tanhua-Tyrkkö, P. Kotiluoto, T. Kekki, "Validation and Optimization of Activity Estimates of the FiR 1 TRIGA Research Reactor Biological Shield Concrete", Nuclear Science and Engineering, Vol. 196, Issue 6, 2022

More VTT references

- Iso-Markku (2019) Difficult-to-measure beta active radionuclides in nuclear decommissioning waste, MSc thesis, Helsinki University
- Laurila (2021) Vaikeasti mitattavien radionuklidien määrittäminen aktivoidusta betonista ja ioninvaihtomateriaalista, AMK insinöörityö, Metropolia Ammattikorkeakoulu
- Leskinen A, Salminen-Paatero S (2021) Development of ³H, ¹⁴C, ⁴¹Ca, ⁵⁵Fe, ⁶³Ni radiochemical analysis methods in activated concrete samples. J Radioanal Nucl Chem https://doi.org/10.1007/s10967-021-08073-4
- Leskinen A, Gautier C, Räty A, Fichet P, Kekki T, Laporte E, Giuliani M, Bubendorff J, Laurila J, Kurhela K, Fichet P, Salminen-Paatero S (2021) Intercomparison exercise on difficult to measure radionuclides in activated concrete statistical analysis and comparison with activation calculations. J Radioanal Nucl Chem 329:945-958
- Leskinen A, Tanhua-Tyrkkö M, Kekki T, Salminen Paatero S, Laurila J, Kurhela K, Hou X, Stenberg Bruzell F, Suutari T, Kangas S, Rautio S, Wendel C, Bourgeaux-Goget M, Moussa J, Stordal S, Isdahl I, Fichet P, Gautier C, Laporte E, Giuliani M, Bubendorfd J, Fichet P (2021). DTM-Decom II Intercomparison exercise in analysis of DTM in decommissioning waste. NKS-441, NKS-B, Roskilde, Denmark

More VTT references

- Leskinen A, Salminen-Paatero S, Gautier C, Räty A, Tanhua-Tyrkkö M, Fichet P, Kekki T, Zhang W, Bubendorff J, Laporte E, Lambrot G, Brennetot R (2020) Intercomparison exercise on difficult to measure radionuclides in activated steel - statistical analysis of radioanalytical results and activation calculations. J Radioanal Nucl Chem 324:1303-1316
- Leskinen A, Salminen-Paatero S, Räty A, Tanhua-Tyrkkö M, Iso-Markku T, Puukko E (2020) Determination of ¹⁴C, ⁵⁵Fe, ⁶³Ni and gamma emitters in activated RPV steel samples - a comparison between calculations and experimental analysis. J Radioanal Nucl Chem 323:399-413
- Leskinen A, Tanhua-Tyrkkö M, Kekki T, Salminen Paatero S, Zhang W, Hou X, Stenberg Bruzell F, Suutari T, Kangas S, Rautio S, Wendel C, Bourgeaux-Goget M, Stordal S, Isdahl I, Fichet P, Gautier C, Brennetot R, Lambrot G, Laporte E (2020). Intercomparison exercise in analysis of DTM in decommissioning waste. NKS-429, NKS-B, Roskilde, Denmark
- Leskinen A, Hokkinen J, Kärkelä T, Kekki T (2022) Release of H-3 and C-14 during sampling and speciation in activated concrete. J Radioanal Nucl Chem 331:859-865
- Meriläinen (2022) Analysis of difficult to measure radionuclides in reactor graphite and comparison of acid digestion and alkaline fusion as sample digestion methods for different matrices, MSc thesis, Helsinki University