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Reference N°	Creation Date	
SCK CEN/54767932	2024-06-20	
Alternative Reference N°	Revision	Version
N/A	1.0	21
ISC	Revision Status	
Public	Approved	

ER-1250- Graphite model validation process.docx

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Review information since previous revision*

Name	Outcome	Date
Sébastien Caes	Reviewed	2024-06-20

Approval information for current revision*

Name	Outcome	Date
Elie Valcke	Approved	2024-06-20

Change log*

Revision	Version	Status	Date	Description of change
1.0	21	Approved	2024-06-20	

ISC motivation

ISC was automatically assigned.

ER-1250

Graphite model validation process

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Contract number: CO-90-02-1583
Contract information: Technical Liability
Publication date: 2024-06-20
Deadline: 31-03-2023

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SCK CEN – Publication date 2024-06-20
Stichting van Openbaar Nut - Fondation d'Utilité Publique - Foundation of Public Utility

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Glossary of abbreviations

BR1	Belgium Reactor 1
GDMS	Glow Discharge Mass Spectroscopy
EOL	End of Life
GRAPA	Graphite Processing Approaches
IAEA	International Atomic Energy Agency
IGA	Interstitial Gas analysis
i-graphite	Irradiated Graphite
LSC	Liquid Scintillation Counting
MCNP	Monte Carlo N-Particle code
NAA	Neutron Activation Analysis
NSO	Nuclear System Operation Unit
NSP	Nuclear System Physics Unit
ONDRAF/NIRAS	Belgian National Agency for Radioactive Waste and Enriched Fissile Material
PGA/B	Graphite Pile Grade Type A/B
RDW	R&D Waste Packages Research Unit
RN	RadioNuclide
R&D	Research and Development
W&D	Waste and Disposal Expert Group

Abstract

The BR1 reactor contains approximately 492 tons of graphite, which is converted to i-graphite during reactor operation and therefore classified as radioactive waste. The neutron irradiation converts stable elements, some of which being present as impurities in the virgin graphite, into radionuclides in the i-graphite. The type of radionuclides, their half-lives and their activity levels dictate the appropriate route for the waste disposal and therefore the importance of having an accurate inventory of the radionuclide activities in the BR1 i-graphite. The radionuclide inventory present in the i-graphite is determined by combining modelling (the activation calculations using Monte Carlo N-Particle code (MCNP)-based on the virgin graphite impurity levels and the reactor operation history) and the radionuclide activity measured in the i-graphite (for validation and model adjustment). SCK CEN has already developed the model for predicting the radionuclides activities of the BR1 graphite at the end of field life and during decommissioning. However, a complete validation of the model was not possible because some important information on the impurity and beta-emitting radionuclides concentration were not available. In this document, the process for the model validation is presented to support the assessment of graphite characterisation and inventory at the end-of-life of the BR1 for the correct waste category classification.

Keywords

Virgin graphite, impurities, NAA, GDMS, IGA, i-graphite, radionuclides, C-14, Cl-36, H-3 and Co-60, modelling and validation.

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1 Introduction

Graphite is used in a number of nuclear reactors as a neutron moderator and reflector, and for structural components like fuel sleeves and air extraction channels. The Belgium reactor 1 (BR1), which is an air cooled type reactor, was built in 1956 and contains approximately 492 tons of graphite (used as moderator and as reflector), which is equivalent to approximately 300 m^3 of radioactive graphite waste [Nijst, 2014]. Before irradiation, the nuclear graphite is called virgin graphite and when irradiated by neutron particles the virgin graphite is being converted into irradiated graphite or i-graphite. The activation process converts stable elements, mainly those present as impurities in the virgin graphite, into radionuclides in the i-graphite. These impurities such as N-14, Li-6 and Cl-35, which are the precursors of C-14, H-3 and Cl-36, respectively, are always present due to the manufacturing process. The more important radionuclides identified in the i-graphite are C-14, Cl-36, H-3 and Co-60, with their respective half-lives being equal to 5730 years, 301000 years, 12.3 years and 5.3 years. Depending on the type of radionuclides (half-life) and activity, the i-graphite is classified in accordance with the waste categories A, B or C in an ascending order of severity as defined by ONDRAF/NIRAS [ONDRAF/NIRAS, 2013; ONDRAF/NIRAS, 2019-H6; ONDRAF/NIRAS, 2019-H14]. Category A deals with short-lived (half-life <30 years) low and intermediate-level waste. These wastes will be disposed of in a surface disposal site. Category B is for long-lived (half-life >30 years) low and intermediate level waste without heat emission. Category C is for long-lived high-level waste with heat emission. Both B and C category wastes require a very long isolation from the biosphere. They would be disposed of in a deep geological repository. The graphite waste should be preferably classified as category A for surface disposal due to its high volume and considering the higher costs involved for disposal of category B and C wastes. Therefore, the determination of the type of radionuclides and their activity levels (radionuclide inventory) in the i-graphite is crucial for the classification of the waste and consequently the route for disposal.

The graphite radionuclide inventory is determined by combining modelling (the activation calculations using Monte Carlo N-Particle code (MCNP)- based on the virgin graphite impurity levels and the reactor operation history) and the radionuclide activity measured in the i-graphite (for validation and model adjustment). Since 2000, SCK CEN has initiated the BR1 i-graphite characterisation by developing and performing modelling (activation calculations) through four Master Theses, as shown in Table 1.

Table 1. Previous work performed at SCK CEN on the modelling and characterisation of BR1 graphite.

Author and Year	Title
[Messaoudi, 2000]	Caractérisation radiologique des matériaux de structure du BR1 – Evaluation de l'activité des matériaux de structure de BR1 (Master Thesis)
[Bravo, 2010]	Characterisation of irradiated BR1 graphite (Master Thesis)
[Nijst, 2014]	Improving the radionuclide inventory determination of the irradiated graphite from BR1 in Mol (Master Thesis)
[Calder, 2018]	Classification of BR-1 irradiated graphite waste and the potential for thermal treatment (Master Thesis)

The work by Messaoudi [Messaoudi, 2000] used 2D geometry, cross section simplifications, and referenced old reaction data libraries. Bravo [Bravo, 2010] followed this study, using the same 2D neutron flux values as calculated by Messaoudi [Messaoudi, 2000], but applied more advanced depletion codes and a more complete methodology in order to study the radial effect of specific radioisotope production. Finally, both authors compared the resulting specific activities with the waste disposal criteria. Nijst [Nijst, 2014] then surpassed this study by implementing the more advanced Monte Carlo N-Particle Code to obtain a more accurate 3D representation of the BR1 geometry and the neutronic conditions following a detailed operational and fuel loading history. Nijst [Nijst, 2014] modelled and reported the activity of 20 radionuclides which could be generated in the i-graphite of the BR1 reactor. The relevant radionuclides were identified and the results of their activities were compared with the requirements of ONDRAF/NIRAS for surface disposal, which in 2010 defined a list of the 20 critical radionuclides for surface disposal [De Bock, 2010]. Nijst [Nijst, 2014] performed a preliminary model validation by using results of gamma spectroscopy of three PGA i-graphite samples taken in 2011 from channel C.2.3 in different locations along the axial position. The validation was performed using some selected gamma-emitting radionuclides (Ba-133, Co-60, Cs-134, Eu-152, Eu-154, Eu-155 and Zn-25) and did not include any beta-emitting radionuclides such as H-3, C-14 and Cl-36. Therefore, due to the lack of information on beta-emitting radionuclides and on some impurities (e.g. nitrogen), this work was only considered as a preliminary exercise.

Calder [Calder, 2018] used the SCK CEN proprietary code ALEPH, combining the computational Monte Carlo N-Particle transport code (MCNP) with a deterministic activation and depletion algorithm. In this study, the author concentrated on the activity predictions of four main radioisotopes (C-14, Cl-36, Co-60 and H-3) to determine the waste category and the disposal route of the BR1 i-graphite. The sources and production route of these radionuclides originated mainly from the impurities in the virgin graphite. For C-14 generation, the two main sources were identified as C-13 and N-14. N-14 was identified as a relevant impurity but the concentration of this element in the virgin graphite of the BR1 was not reported or measured. Therefore, a sensitivity analysis using initial N-14 concentrations of 10, 70 and 300 mg kg^{-1} in the virgin graphite was performed. Calder [Calder, 2018] did not perform any model validation but the simulations indicated the importance of assessing both graphite types, e.g. Pile Grade A (PGA) and Pile Grade B (PGB), since BR1 contains approximately 172 tons of PGA and 320 tons of PGB graphite. PGA possesses a high purity and it is used in the core of the reactor as neutron moderator. PGB possesses slightly more impurities than PGA and it is used in the outer region of the reactor as neutron reflector [Nijst, 2014; Calder, 2018].

Because measured data of the main beta-emitting radionuclides (H-3, C-14 and Cl-36) and results of some impurities were not available (e.g. N-14) at the time of the activation calculations, a report on the BR1 graphite status was issued [De Souza, 2020 (a)] where the gaps to perform characterisation of the graphite were identified and a research plan was initiated [De Souza, 2020 (b)]. This research plan focuses on the radionuclide inventory and the leaching behaviour of the i-graphite from the BR1, in which the status and progress have been reported thought different annual and interim reports issued from December 2020 to January 2023 [De Souza, 2021; De Souza, 2022; De Souza, 2023].

In this document, a high-level process for the model validation is presented, which indicates the steps and phases required for validating the activation calculations already developed at SCK CEN.

2 Objectives and scope of the document

The objective of this document is to describe a high-level process for the model validation of the graphite activation calculation and radionuclide activity predictions.

3 Validation process and process flow chart

GRAPA (Graphite Processing Approaches) [GRAPA, 2021] indicated and provided some examples of organisations that performed graphite radiological inventory by combining modelling (the activation calculations using Monte Carlo N-Particle code (MCNP)-based on the virgin graphite impurity levels and the reactor operation history) and the radionuclide activity measured in the i-graphite for validation and model adjustments. Figure 1 shows an example of the modelling approach used for L-54M nuclear research reactor of Politecnico di Milano [GRAPA, 2021]. The approach was divided in three steps as follows: i) simulation of the nuclear reactor and model verification with experimental criticality and fluences data; ii) simulation of the main nuclear activation reactions, accordingly with the radionuclides of interest; iii) simulation of graphite activation and model validation with experimental data from radiological characterisation. The approach shows that Monte Carlo N-Particle code (MCNP) was used in this example. It is noticed that step ii) utilises the data from the selected relevant impurities of the virgin graphite and step iii) the validation with the radiological characterisation (experimental values), which has similarities on the approach adopted at SCK CEN as it will be shown in the following paragraphs.

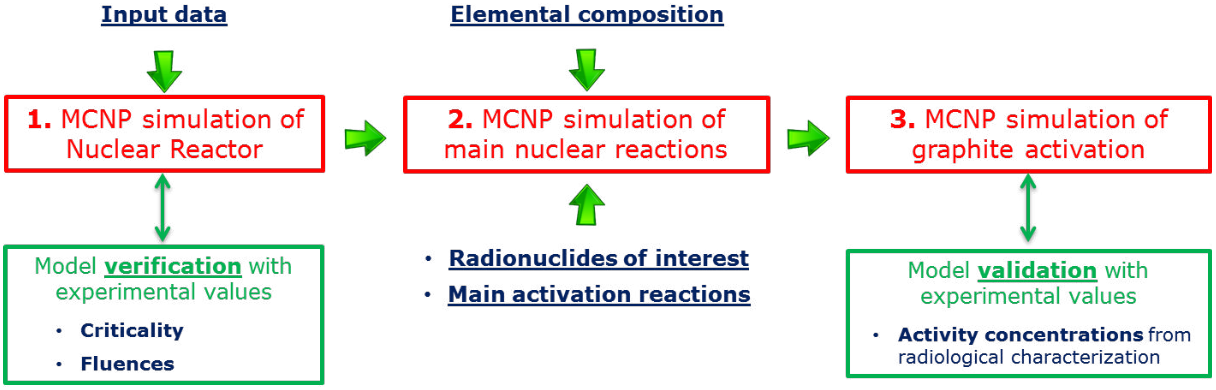

Figure 1. Monte Carlo N-Particle computational approach used for the L-54M model activation analysis [GRAPA, 2021].

The model for the activation calculation code has already been developed by SCK CEN as indicated in Table 1. The validation process is described below in five phases and summarised on the flow chart in Figure 2.

- *Phase 1*:
    - Collecting data from the impurities content and range in the PGA and PGB virgin graphite. This is equivalent to step two in Figure 1 with the selection of the relevant impurities and activation reactions. This has been already prepared in DeSouza *et al.* [DeSouza, 2022], where the impurities were measured in five PGA and five PGB blocks using glow discharge mass spectroscopy (GDMS) and neutron activation analysis (NAA). The average and maximum values are already presented for the main impurities, such as Cl-35 and Li-6.
    - N-14 was measured by interstitial gas analysis (IGA) and irradiation of virgin graphite in a controlled manner (PGA graphite) as reported by De Souza *et al.* [De Souza, 2022] and Borms [Borms, 2023] respectively. The results provide a range of N-14 contents expected in the BR1 graphite.
    - In this phase, the criteria for near surface disposal should be established. The main radionuclides will be listed and these radionuclides will dictate the main impurities to be addressed in more detail, as for instance Cl-35, Li-6, N-14, Nb-93. The analysis of Phase 1 will be used to support the analysis in Phase 5.
- *Phase 2:*
    - Collecting PGA and PGB i-graphite samples from BR1 and measure their radionuclide concentrations by gamma spectroscopy for gamma-emitting radionuclides (e.g. Co-60, Nb-94, Cs-134, Cs-137) and by liquid scintillation counting (LSC) for measuring H-3, C-14 and Cl-36, which are first extracted and separated using a pyrolysis method [De Souza, 2022; De Souza, 2023]. The measured radionuclide concentrations, especially C-14 and Cl-36, are compared with the acceptance criteria for near surface disposal as established by ONDRAF/NIRAS.

- *Phase 3*[1]*:*
    - Verifying the reactor operation history data, and estimating or defining the BR1 end of field life and decommissioning period. The reactor irradiation history, the fuel configuration and the neutron flux are relevant inputs for modelling.
- *Phase 4*[1]*:*
    - Verifying and updating the models and activation calculations prepared in previous works [Calder, 2018; Nijst, 2014]. In this phase, the models need to be revisited and updated with the irradiation history and field life information of the BR1 reactor.
- *Phase 5:*
    - Performing sensitivity analysis by applying, for the precursors of the main radionuclides of interest (e.g. N-14 and Cl-35), different ranges of concentrations in the activation calculation model, from the very conservative to the more realistic concentrations. For instance, Calder [Calder, 2018] has already performed a sensitivity analysis for N-14 by but the new data acquired later than that work (measured data – impurities and beta-emitting radionuclides) need to be taken into consideration when re-running the model (activation calculations).
    - Comparing the predicted activities of H-3, C-14 and Cl-36 with the measured values in different regions of the reactor for PGA and PGB, including an assessment of the data from the sensitivity range provided for the impurities in Phase 1.
    - Using measured radionuclides concentration from short-term irradiation test: Virgin graphite samples from PGA type were irradiated (under a controlled manner) for 21 days in channel Y4 of BR1 [Borms, 2023] and C-14 and Co-60 were measured after irradiation. The results from this task may be useful to support the validation process. The impurities of the virgin graphite sample were also measured before irradiation.
    - Performing adjustment to the model based on the measured H-3, C-14 and Cl-36 data (this step is similar to Step 3 in Figure 1).

It is important to mention that Phases 1 and 2 can be performed in parallel.

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[1] This phase is performed by the Nuclear System Operation (NSO) and the Nuclear System Physics (NSP) groups at SCK CEN. For more information please refer to these groups.

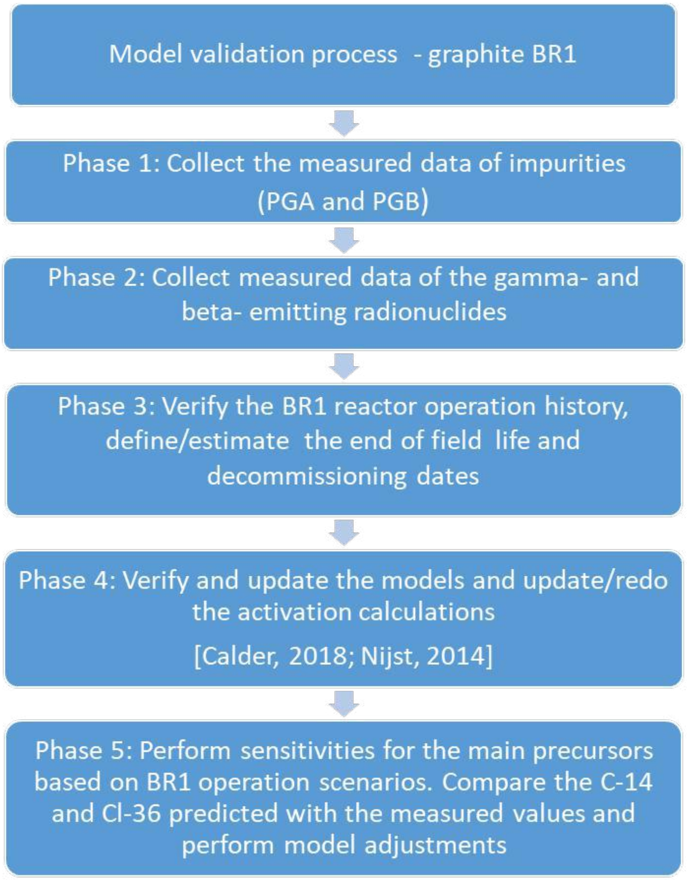

Figure 2. Flow chart showing the steps or phases of the modelling validation process.

4 Conclusion

The BR1 reactor contains approximately 492 tons of graphite, which is converted to i-graphite during reactor operation and therefore classified as radioactive waste. The neutron irradiation converts stable elements, some of which being present as impurities in the virgin graphite, into radionuclides in the i-graphite. The type of radionuclides, their half-lives and their activity levels dictate the appropriate route for the waste disposal and therefore the importance of having an accurate inventory of the radionuclide activities in the BR1 i-graphite. The graphite radionuclide inventory is determined by combining modelling (the activation calculations using Monte Carlo N-Particle code (MCNP)-based on the virgin graphite impurity levels and the reactor operation history) and the radionuclide activity measured in the i-graphite (for validation and model adjustment). SCK CEN has already developed the model for predicting the radionuclides activities of the BR1 graphite at the end of field life and during decommissioning. However, a complete validation of the model was not possible because some important information on the impurity concentration in the PGA and PGB virgin graphite types (e.g. N-14) and the concentrations of beta-emitting radionuclides in the i-graphite (H-3, C-14 and Cl-36) was still missing. After collection/determination of data from phases 1 and 2, the validation process can be done by performing the following phases: (3) by updating the irradiation history of the BR1 and establish the end of field life, (4) by updating the models previously developed with the information from steps (1,3), and (5) by calculating the radionuclide concentration at the end-of-life of the BR1 (also in the zones of the reactor where graphite sampling is difficult). In phase 5, an adjustment of the model with the measured radionuclides values and classification of the graphite in the correct waste category are also performed.

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