Updated Radiological Inventory of G1 Reactor Thanks to a Strengthened Data Processing – 17271

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ABSTRACT

The radiological inventory of G1 reactor is under refinement and consolidation. This reactor, located at the CEA Marcoule center in France, presents an issue for Chlorine-36 as graphite was used as moderator. The improvement of the 3D physical description and a better understanding of historical events enable the computation of activation levels thanks to a numerical model. These results are compared to old samples and more recent ones to quantify nuclide activity levels as well as their spatial distributions. In particular a spatial and statistical correction of sample bias leads to an update of the radiological inventory with an expected reduction of average activity levels.

Another part of the work consists in identifying missing pieces of information and still uncertain parts of the reactor (sample analysis of activated concrete for instance) to reinforce the radiological inventory of G1 reactor. This will be the basis of future investigation works.

INTRODUCTION

The G1 reactor (46MWt) was built in 1955 and operated between 1956 and 1968 in the Marcoule CEA Center [1]. It has been designed and used for military purposes (plutonium production), as well as an industrial prototype for reactors then developed by CEA (G2 and G3) and EDF (6 units). It was the first French reactor based on the use of natural uranium as the fuel and graphite as moderator. It initially had 30, then 46 megawatts of thermal power. It was air-cooled at atmospheric pressure by a central slit, dividing the reactor into two half-piles (loading and unloading sides).

Cleanup and dismantling began in 1969. All external circuits including the cooling circuits were removed and in 1996, the International Atomic Energy Agency's level 2 of dismantling classification was reached. The graphite stack is still inside the reactor. Nowadays the remaining structures/components are the concrete part of the reactor (31,000 t), several metallic pieces (thermal shield, ventilation…) and the graphite bloc (slightly more than 1,200 t).

Within the 2006 French regulation for the management of nuclear waste, the radiological inventory of long lived, intermediate and high activity have huge

consequences on the technical choices for final disposal by Andra (sub-surface repository / underground repository). Within this framework, CEA, as well as other French waste producers, put strong efforts in improving its knowledge on irradiated graphite [2], with a particular emphasis on specific nuclides such as Cl-36, as well as H-3 and C-14.

This paper then presents the recent characterization works conducted on G1 reactor:

- Improvement of the physical description (3D model) and understanding of historical events (paper records, interviews…);
- Computation of activation levels thanks to a numerical model (MCNPX/CINDER-based);
- Laboratory analysis in 2008 on historical graphite samples, collected in 1969 after the reactor shut-down, only in the loading side of the reactor;
- Collection of new graphite samples in the unloading part of the reactor, undergoing laboratory analyses.

One challenge is to gather and compare these different kinds of information to consolidate the radiological inventory thanks to statistical and geostatistical data analysis.

MATERIALS

Physical Modelling

The reactor pile constitutes the active element of G1 reactor. It comprises the graphite bloc and all metal elements ensuring the graphite stacking and its cooling air supply. The whole is placed inside a very thick structure of reinforced concrete, with large dimensions: length: 36 m, width: 28.75 m and height: 30 m.

The physical model is based on a SolidWorks project whose objects were exported in STL format and then imported as wireframes in Isatis. 788 objects allow representing the reactor (Fig. 1). Additional 35 objects by stopper lead to a total of more than 31,000 objects.

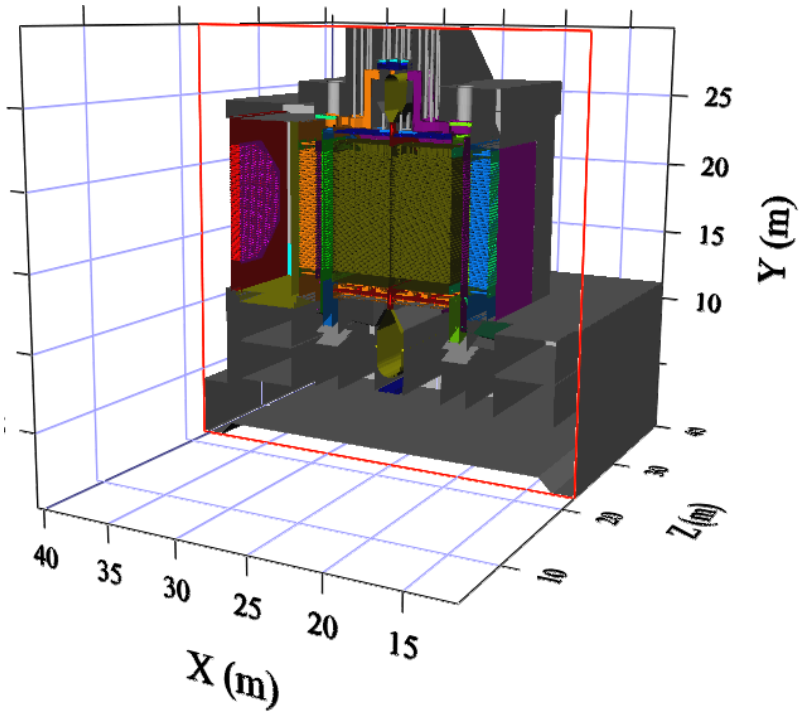

Fig. 1. Vertical clipping of the 3D physical model of G1 reactor.

Historical Data

In 2008-2009, the CEA carried graphite analyzes on several samples taken in 1969, after the shutdown of the reactor. Other data during operation or after shutdown of the reactor are also available. The focus is given to the most recent values as previous ones were not intended in a dismantling and decommissioning purposes. Their traceability and exhaustively (mostly for measured nuclides) may present weakness to some extent.

The 2009 results make possible a relevant exploratory analysis in order to characterize the homogeneity (or heterogeneity) of the radionuclides among themselves and their spatial distribution. However, they only investigate the loading part of the reactor, for accessibility reasons in 1969.

All samples were analyzed for Chlorine-36 quantification but only 9 out of 14 gave results for other nuclides such as H-3, C-14, Co-60, Ni-63, Sr-90, Nb-94, Ba-133, Cs-137, Eu-152, Eu-154, Eu-155, 241-Am and uranium and plutonium compounds.

Correlations between nuclides are analyzed through scatter plots as well as principal component analysis.

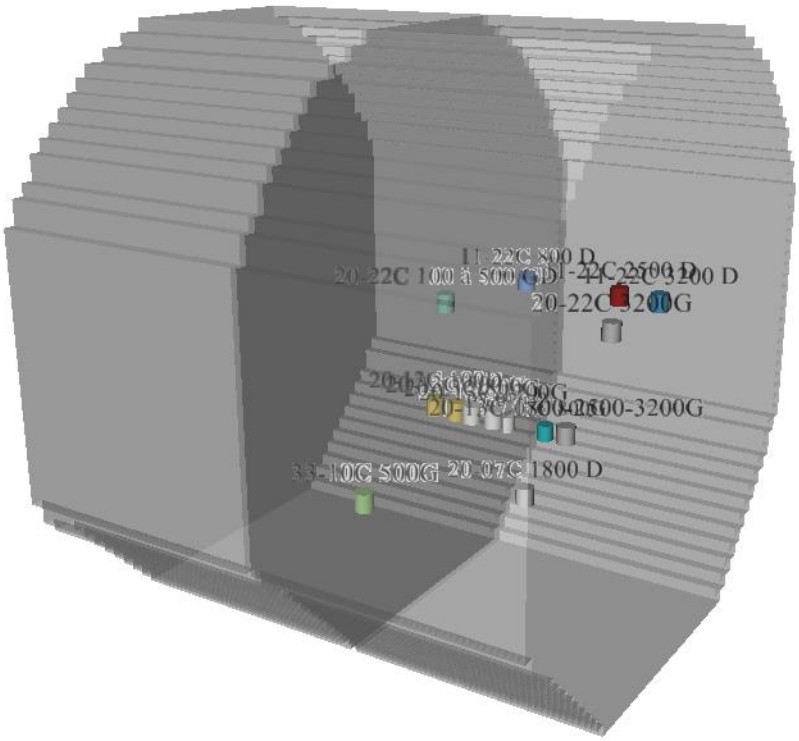

Fig. 2. Location of the samples in the graphite block.

The findings from this analysis can be compared with the different parameters of the reactor (geometric / operational) to identify similar or atypical points. Additional pieces of information such as the control rod network (neutron-absorbing) and sheath ruptures are integrated.

Activation Model

Activation calculations on graphite, steel materials and concrete structures were carried out by DEN/DANS/DM2S/SERMA/LPEC. They allow having a 3D model of the distribution of the radionuclides (Fig. 3) resulting from the neutron effect on the chemical elements within the different materials.

Neutron calculations were carried out using the probabilistic MCNP-X (Monte Carlo N-Particle) code developed at the Los Alamos laboratory (USA) [3]. Then material activation is calculated with the deterministic code CINDER '90, also developed in the laboratory of Los Alamos [4].

Some hypotheses have to be taken as regards the variations of the power of the reactor as well as the impurity levels in the different materials, the control rod and the absorbent network positioning.

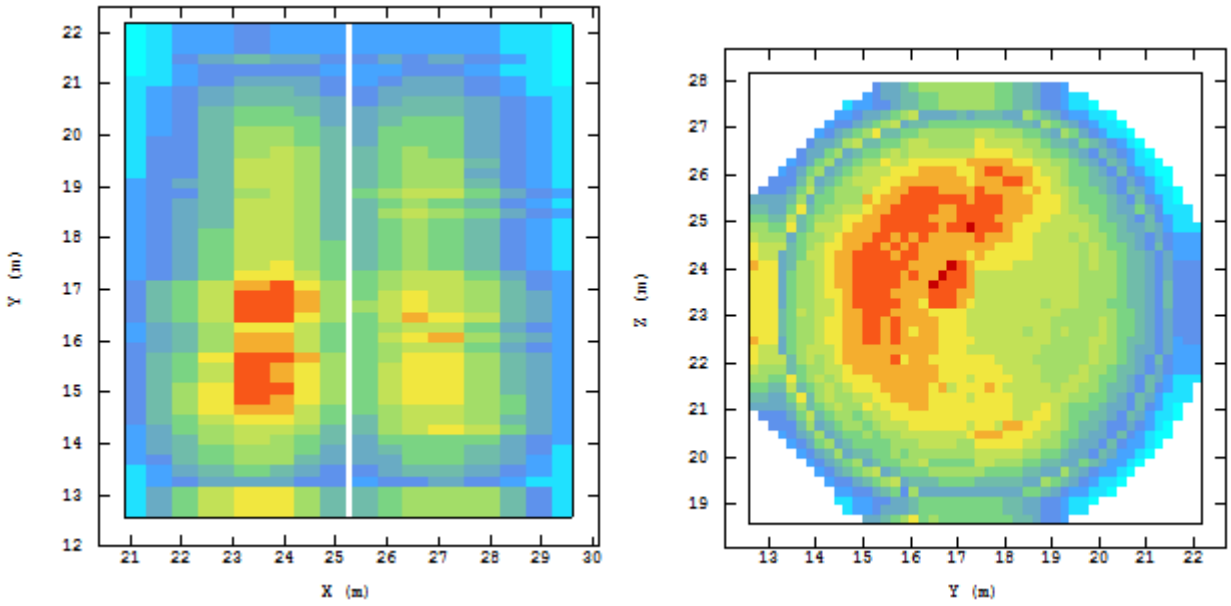

Fig. 3. Horizontal section (on the left) and radial section (on the right) of total activity estimates from neutron activation computation code.

That way, results from this numerical model have to be considered as relative quantification rather than an absolute one. It gives activity variations in the whole graphite block.

RESULTS AND DISCUSSION

Methodology and Radiological Inventory Update

Several years ago, the radiological inventory initially derived only from a statistical average of several samples from 1969 that were analyzed in laboratory in 2008-2009 (DEN/CAD/DEC/SA3C/LARC and AREVAMARCOULE/DTN/ STPA/LPC). Theses samples were considered as representative of mixed activation and contamination phenomena within the reactor. Due to the observed sampling bias in the spatial distribution of the data, the total activity was probably overestimated assuming that the highest activity levels are expected in the central part of the graphite block.

For that reason, the combination of both analyses on graphite samples and neutron activation computation has been used to refine and update the radiological inventory of G1 reactor. For each nuclide, a correlation between activity levels on samples (absolute quantity) and results from activation model (relative quantity) is adjusted.

These updated results show a significant decrease of the radiological inventory in comparison to values obtained solely on the basis of the historical data (2008 measures). Thus, the sampling bias (non-homogeneous positioning of the sampling data) is adequately corrected by the information provided by the activation calculation results, after recalibration in mean value by the linear regression without constant. Only Eu-152 appears to be significantly underestimated (by a factor of 5) on the basis

of historical samples due to the fact the strongest values by activation calculation are located in the reflector part (not investigated by historical sampling and presenting different burning rates in the two zones).

As expected, most of the radiological inventory is composed of C-14 and H-3. Europiums and Ni-63 follow. Cl-36 is quite at a low level but is one of the most impacting nuclides form long-term storage issues.

Discussion and Ongoing works

The linear regression without constant between samples and model can be seen as an *a posteriori* correction of impurity contents as input data for the activation model. However, the numerical model is not considered as the reality, first because it only intends to reproduction activation phenomenon and second because some strong assumptions may significantly influence local variations of nuclide activity levels (average control rod positioning, absorbent network…). That way, radiological inventory results are based on real samples that are statistically improved by auxiliary information.

This analysis also points out some "shadow" areas. For instance the symmetry of the reactor block has to be considered as it is a physical fact but not from an operational point of view. The neutron flux tries to model a possible asymmetry due to the burning rate of the uranium fuel assemblies. But it needs to be calibrated experimentally due to the limited amount of information. Similarly the moderator part of the graphite block is only quantified through the results form activation computation. Even if Eu-152 is not the biggest challenge (as for total inventory or dose rate consideration), the specific phenomenon that occurred in the moderator part has to be confirmed by graphite samples.

CONCLUSIONS

The main issue is to combine activity levels measured on destructives samples on the one hand and numerical results from activation computation on the other hand. This strengthened data processing enables improving the understanding and the quantification of nuclide activity levels as well as their spatial distributions. For instance, the spatial bias correction from graphite samples (due to their preferential localization in the expected highest activated part of the reactor) leads to a possible reduction of the radiological inventory, compared to first estimations. These updated results also play a crucial part for the preparation of future decommissioning works (planning, funding, waste package decision, waste volumes, waste categories…).

New graphite samples are under collection and laboratory analysis (DEN/CAD/DEC/SA3C/LARC and DEN/DANS/DPC/SEARS/LASE) to reinforce this approach and to reduce the main limitations described hereinbefore. It still remains concrete and steel materials that are characterized only on the basis of the numerical activation model.

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