Innovative Extension of the EDF Inventory Methodology for Irradiated Graphite Sleeves – 16084

Bernard Poncet, EDF

ABSTRACT

About 17,000 t of EDF irradiated graphite wastes will be produced from the decommissioning of the six French gas-cooled nuclear reactors. Determining the radionuclide content of this graphite is of capital importance for safety purposes and in order to determine the best way to manage them.

A method using an identification calculation-measurement process has been proposed in order to assess a radiological inventory as precise as possible and has been validated by ANDRA, the French governmental radioactive waste management agency and by IRSN, the technical support of the French nuclear safety authority.

As presented in [1] at WM2013, this method is based on (a) radionuclide measurements from sufficient samples from a statistical point of view and (b) activation coefficients calculated from the neutron flux histories of the measured samples. For that purpose, it is important to know the precise location of each measured graphite sample, which is easy with pile sampling.

About 2,000 t of EDF irradiated graphite is composed of sleeves which contained fuel cartridges and they have been stored in silos of St-Laurent site. The general method needs to know the precise history including neutron flux of any sampled sleeve. As there is no possibility to individually identify sleeves, such a requirement on sleeves in a silo is impossible.

The paper presents both the general method for piles and an innovative way to replace that knowledge for long lived radionuclide of irradiated sleeves.

INTRODUCTION

Electricité De France (EDF) operated six gas-cooled reactors in France, all now shutdown. These reactors are called in French, ‘‘uranium naturel graphite gaz’’ reactor type (UNGG). They were graphite moderated, cooled by carbon dioxide and fuelled with metallic natural uranium. In the absence of uranium enrichment, graphite was used as a moderating material with a very high level of purity due to the necessity of the highest neutron transparency (nuclear purity). Graphite was also chosen as a mechanical support of the fuel cartridges (graphite sleeves) and as a biological shielding in some reactors. The irradiated graphite from the pile or from the biological

shielding still lies in the reactors. The graphite sleeves that are not already shipped to the final repository are stored in silos.

GENERAL METHOD APPLIED FOR THE PILES

Usual way used to assess the radionuclide inventory based on activation calculations of assumed impurity content in the graphite does not fit at all for the graphite from UNGG reactors. Most of the time, the impurity content of the graphite is not well known or actually unknown, remaining below the limit of chemical detection. A mathematical methodology, coupling computation tools and radiochemical analysis results, has been therefore investigated by EDF.
Nuclear graphite is a synthetic material manufactured from raw materials issuing from petroleum and coal that are natural products full of numerous impurities. Although subjected to thorough purification steps during the manufacturing process, some impurities remain at trace level in the nuclear graphite. The radioactivity of the graphite in UNGG reactors comes almost exclusively from the activation under neutron flux of these impurities and of the carbon constituting the graphite.

The easiest way to assess the radionuclide inventory of the irradiated graphite should proceed by interpreting radiochemical measurements on irradiated graphite sampled from the pile of the reactors. But in the particular case of irradiated graphite of nuclear purity, high discrepancies have been observed on radionuclide measurements, and particularly for radionuclide whose precursors are present at trace level (two or three orders of magnitude)

For EDF reactors, according to the dimensions and geometry of the piles, a set of 20–30 irradiated graphite samples were analysed mostly by radiochemistry on the 200 cores (samples) taken on average per reactor pile. This huge variability observed at the scale of a pile correlates (Bugey 1 data):

- neither with the variation in the neutron flux according the vertical distribution, which is less than one order of magnitude, as shown in fig.1 (x axis height of the pile in cm, y axis in arbitrary units),

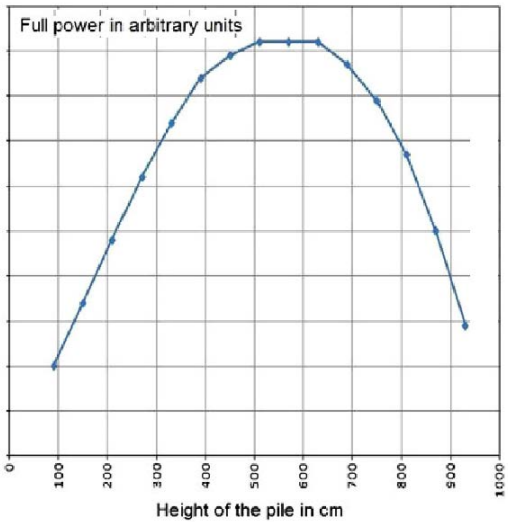

Figure 1: Shape of the thermal power correlated to the neutron flux into a fuel channel of the Bugey 1 reactor with x axis height of the pile in cm and y axis full power in arbitrary units

- nor with the other macroscopic operational parameters of the reactor such as the temperature which varies at full power approximately linearly from 280 to 530 °C between the top and the bottom of the pile.

This observation is also seen for other radionuclides and in other UNGG reactors. It has nothing to do with the quality of sampling, nor with the quality of the analysis method.

The fundamental physical phenomenon that explains the issue of the discrepancy of the analyses is directly linked to the high purity inducing an almost paradoxical and very specific heterogeneity of the nuclear graphite. Actually nuclear graphite cannot be considered from an analytical point of view like any other common homogeneous material where impurities may exhibit a continuous spatial concentration, but as a finely divided material to which the geostatistical concept of the ‘‘nugget effect’’ can be applied. It concerns a regionalized random function for which the arbitrary closeness of points corresponds nevertheless to a high variance.

These observations demonstrate that it is impossible to get representative and reproducible measurements to the scale of an entire pile for impurities at trace level. Therefore, for establishing the radionuclide inventory of the irradiated graphite, an approach based on a statistical tool through the calculation of a mean of several measurements is a necessity.

The easiest way to assess the radionuclide inventory of the irradiated graphite should proceed by interpreting radiochemical measurements on irradiated graphite sampled.

Identification calculation-measurement method

The method of radionuclide inventory assessment by identification calculation-measurement is described below. It consists in using the following process:

- ‘‘3D’’ map computation of the flux density of neutrons (with 315 intervals for neutron energy) based on the geometry of each pile. It is computed with the TRIPOLI calculation code devoted to the transmission of particles (neutrons) by solving the Boltzmann equations and coupled with international nuclear databases.

- from this flux map and incorporating the history of reactor operation to reconstruct a global inventory of the radioactivity produced by this flux throughout the geometry of the reactor pile; the inventory is then created by incorporating the impurity levels, which are adjusted to their explanatory values (Fig. 2), from the result of the activation calculations (C) with the available measurements (M) of the corresponding radionuclides. These activation calculations are performed by using the DARWIN-PEPIN code which integrates all the phenomena leading to radionuclide production (activation, fission, radioactive decay, etc.) by solving the generalized Bateman equations ([2] and [3]). The adjustment process for the impurities explanatory values is iterative toward the minimization of the quadratic function:

$$\sum_i \mu_i^2 = \sum_i \left(\frac{1}{N_i} \sum_{j=1}^{j=Ni} \ln \frac{C_{i,j}}{M_{i,j}} \right)^2$$

where N_i is the number available measurement activity of each *i* radionuclide (logarithm is used here to let low measurement values participate to the adjustment process).

The relevance of the method for inventory calculation by adjustment calculation-measurement is based upon the availability of a sufficient number of measurements of the wanted radionuclides.

In practice, for each reactor, EDF has arranged for the analysis of some 30 radionuclides on some 30 samples of graphite taken from the reactor. The heart of the proposed method is schematically described in the following fig. 2. It is seen as *reverse* method in the sense that the radiological measurement results are some inputs.

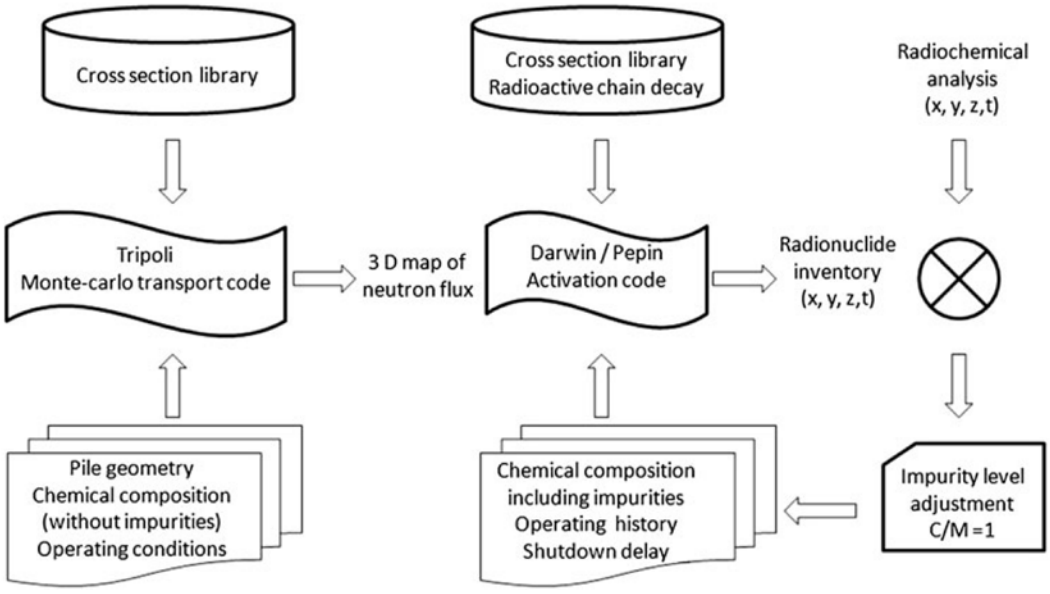

Figure 2: Simplified description of principles for calculating the radionuclide inventory of the piles in the EDF reactors by identification method

Particular case of graphite sleeves

About 1,900 tons of graphite sleeves from fuel cartridges used in both reactors SLA1 and SLA2 were stored in two silos on the EDF site of “Saint-Laurent des Eaux”. Individual precise neutron flux and operating history are unknown for these sleeves in bulk storage. Global activation inventory depends on the whole cartridge renewal during reactor operation.

Sleeves have different histories and spectra. This is why activity computation process cannot be carried out as in the case of a pile. A pile has its own mapping of the neutron flux and a unique history.

A typology of sleeve histories in 32 types has been defined using the unloading chronicle of the reactor. Channels have been unloaded from 3 to 6 times depending on their irradiation level. Based on this information, a total unloading chronicle for 29,712 fuel cartridges was established. For each pile, 25 neutron flux classes were determined using an iterating classification method known as “Diday Dynamic Clusters”, a nonhierarchical clustering.

Let’s give a short explanation of such a clustering: Having initialized k = 25 neutron flux vector Φ as centers, all neutron flux vectors, corresponding to a geometric position, are assigned to the class whose center is closest (Euclidean distance). Centers of gravity of neutron flux vectors of these classes are now the new centers. The process (assignment of each neutron flux vector to the closest center, determining centers of gravity) is iterated until full convergence is achieved.

The piece of steel which was included in the sleeve in order to support the fuel cartridge will be used. In that piece of stainless steel, which received the same neutron flux as the graphite, stable and radioactive isotopes of stainless steel components can be measured. This allows the computation of an equivalent of the flux history. By including the building of families of neutron flux seen by the unloaded sleeves by a nonhierarchical clustering, their fluencies can be computed for each flux family Φ. For any long lived radionuclide, the fluency is a way to replace the detailed history to compute activation by neutron flux.

Bateman equations in steel and graphite with the same neutron flux Φ

Radionuclide decay is neglected because of high half-lives. This choice is consistent with a simplified calculation to total nominal flow where t represents an equivalent period of time to full power. For a general power history H(t), Φt is replaced by $\Phi\int_0^{\Delta t} H(\tau)d\tau$ where Δt is the real time of irradiation. Then $t=\int_0^{\Delta t} H(\tau)\,d\tau$.

Let S_{XX-nn} the specific activity of ^{nn}XX in Bq per gram of activated material. Cross sections σ_{XX-nn} is a function of neutron spectrum Φ.

Let $^{58}Ni\,(n,\gamma)\,^{59}Ni$ **in the steel:**

T being the half-life, χ being the mass fraction of a chemical element, θ being the natural abundance of the isotope of a chemical element and N_A being the Avogadro number, with $\left[^{59}Ni\right]=\frac{T_{Ni-59}}{\ln 2}S_{Ni-59}$ and $\left[^{58}Ni\right]_0=\theta_{Ni-58}\frac{N_A}{58}\chi_{Ni}$, it follows that :

$$\begin{cases} \dfrac{d}{dt}\left[^{58}Ni\right]=-\sigma_{Ni-58}\Phi\left[^{58}Ni\right]\Rightarrow\left[^{58}Ni\right]=\left[^{58}Ni\right]_0 e^{-\sigma_{Ni-58}\Phi t} \\ \dfrac{d}{dt}\left[^{59}Ni\right]=\sigma_{Ni-58}\Phi\left[^{58}Ni\right]\Rightarrow\left[^{59}Ni\right](t)=\left[^{58}Ni\right]_0\left(1-e^{-\sigma_{Ni-58}\Phi t}\right) \end{cases}$$

Then : $\frac{\theta_{Ni-58}}{58}\chi_{Ni}\left(1-e^{-\sigma_{Ni-58}\Phi t}\right)-\frac{T_{Ni-59}}{N_A\ln 2}S_{Ni-59}=0$

Let $^{35}Cl(n,\gamma)^{36}Cl$ **in the graphite:** $\frac{\theta_{Cl-35}}{35}\chi_{Cl}\left(1-e^{-\sigma_{Cl-35}\Phi t}\right)-\frac{T_{Cl-36}}{N_A\ln 2}S_{Cl-36}=0$

Using fluency, let: $\chi_{Cl}\left(\Phi t,S_{Cl-36}\right)=S_{Cl-36}\left(\frac{T_{Cl-36}}{N_A\ln 2}\frac{35}{\theta_{Cl-35}}\right)\cdot\frac{1}{1-e^{-\sigma_{Cl-35}\Phi t}}$.

CONCLUSION

The steel has received the same flux history than the graphite because the maximum size of the sleeve is equal to the migration length of UNGG reactor.

Only using information coming from chemical Ni and ^{59}Ni measurements in the steel, a first way using activated Ni gives even an explicit formula knowing neutron flux Φ.

Let $a_{Cl,j}=\dfrac{1-\left(1-\frac{58}{\theta_{Ni-58}}\frac{T_{Ni-59}}{N_A\ln 2}\cdot\frac{S_{Ni-59}}{\chi_{Ni}}\right)^{\frac{\sigma_{Cl-35}}{\sigma_{Ni-58}}}}{\frac{35}{\theta_{Cl-35}}\frac{T_{Cl-36}}{N_A\ln 2}}$, for graphite sample j.

With $a_{Cl,j}$ and criteria $\sum_i \mu_i^2 = \sum_i \left(\frac{1}{N_i} \sum_{j=1}^{j=Ni} \ln \frac{C_{i,j}}{M_{i,j}} \right)^2$ minimization, for i corresponding to Cl-36, we have $M_{Cl-36,j} = a_{Cl,j} \cdot \chi_{Cl,j} = S_{Cl-36,j}$ and $C_{Cl-36,j} = a_{Cl,j} \cdot \chi_{Cl}^{computed}$.

Seeking the minimum of the quadratic criteria implies the following result:

$$\mu_{Cl-36} = 0 \Leftrightarrow \frac{1}{N_{Cl-36}} \sum_{j=1}^{j=N_{Cl-36}} \ln \frac{C_{Cl-36,j}}{M_{Cl-36,j}} = 0$$

$$\Leftrightarrow$$

$$\chi_{Cl}^{computed} = \exp\left[\frac{1}{N_{Cl-36}} \sum_{j=1}^{j=N_{Cl-36}} \ln \frac{\frac{35}{\theta_{Cl-35}} \frac{T_{Cl-36}}{N_A \ln 2} M_{Cl-36,j}}{1 - \left(1 - \frac{58}{\theta_{Ni-58}} \frac{T_{Ni-59}}{N_A \ln 2} \cdot \frac{S_{Ni-59}}{\chi_{Ni}} \right)^{\frac{\sigma_{Cl-35}}{\sigma_{Ni-58}}}} \right]$$

Inverse method knowing the irradiation history has been developed for the piles and has been presented as a basis. Then, it has been demonstrated that a way does exist generalizing it to the cases without history as sleeves owning an activated component made of material which contains measurable non radioactive nickel, which is not the case of too pure graphite.

REFERENCES

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[3] CETNAR, J., 2006. General solution of Bateman equations for nuclear transmutations. Annals of Nuclear Energy 33, 640–645.