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A reverse method for the determination of the radiological inventory of irradiated graphite at reactor scale

Irradiated graphite waste will be produced from the decommissioning of the six gas-cooled nuclear reactors operated by Electricité De France (EDF). Determining the radionuclide content of this waste is an important legal commitment for both safety reasons and in order to determine the best suited management strategy. As evidenced by numerous studies nuclear graphite is a very pure material, however, it cannot be considered from an analytical viewpoint as a usual homogeneous material. Because of graphite high purity, radionuclide measurements in irradiated graphite exhibit very high discrepancies especially when corresponding to precursors at trace level. Therefore the assessment of a radionuclide inventory only based on few number of radiochemical measurements leads in most of cases to a gross over or under-estimation that can be detrimental to graphite waste management. A reverse method using an identification calculation–measurement process is proposed in order to assess the radionuclide inventory as precisely as possible.

Umkehrverfahren zur Bestimmung des radiologischen Inventars von bestrahltem Graphit im Reaktormaßstab. *Bei der Stilllegung der sechs von EDF betriebenen gasgekühlten Kernkraftwerke wird bestrahlter Graphit anfallen. Dabei ist die Bestimmung des Radionuklidgehalts dieser Abfälle eine wichtige auch rechtlich geforderte Pflicht sowohl aus Sicherheitsgründen als auch zur Festlegung der optimalen Strategie zum Umgang damit. Wie durch zahlreiche Studien belegt, ist Graphit, das in Kernkraftwerken verwendet wird, ein sehr reines Material, allerdings kann es aus analytischer Sicht nicht als ein gewöhnlich homogenes Material betrachtet werden. Aufgrund der hohen Reinheit des Graphits zeigen Radionuklidmessungen an bestrahltem Graphit insbesondere dann sehr hohe Diskrepanzen, wenn diese den Precursors im Spurenbereich entsprechen. Daher führt die Beurteilung des Radionuklidinventars allein auf wenigen radiochemischen Messungen zu großen Fehlern, die für das Abfallmanagement des Graphits schädlich sein kann. In diesem Beitrag wird ein Umkehrverfahren vorgeschlagen, das ein kombiniertes Verfahren aus Berechnungen und Messungen zur möglichst genauen Identifikation des Radionuklidinventars verwendet.*

1 Graphite waste management and radionuclide inventory

Electricité De France (EDF) operated six gas-cooled reactors in France, all now shutdown for at least 15 years. 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.

The design of UNGG reactors is, in principle, very close to that of the British Magnox reactors that were developed independently at the same period in United Kingdom. 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. The total amount of irradiated graphite for EDF UNGG reactors is about 17,000 tons. It consists of 13,000 tons of graphite blocks from the piles of the six UNGG reactors, 2,000 tons of graphite sleeves in silos and 2,000 tons of graphite used as biological shielding in ''integrated vessel'' type reactors.

The radionuclide inventory of nuclear waste is a quantitative description of – as precisely as possible – all containing radionuclides. It is a legal commitment that is needed and allows to anticipate an a priori classification of the waste and to develop the best suited management strategies.

For safety reasons, it is essential that the waste producer assesses the radionuclide inventory of the graphite. For dismantling purposes the radionuclide inventory is needed in order to choose the most suitable deconstruction tools and the radiological shielding to ensure the safety of the staff. As the behaviour of radionuclides may lead to significant discharges under dismantling conditions, radionuclide identification makes it possible to adapt the confinement and purification systems in order to limit the impact on the environment.

Concerning final disposal, the knowing the inventory allows computing the repository sanitary impact for millennia and coming to a decision if such a forecasted impact is in agreement with regulations. The radionuclide inventory declaration is the result of a rigorous procedure in accordance with the laws of physics, based on results of samples analysis and on calculations seeking to best reproduce the phenomena explaining the radionuclide presence. Sometimes in the absence of any quantifiable element, the result may be based on reasoned choices to ensure a reasonably achievable over-assessment of the radionuclide inventory to be declared by the graphite waste producer. With the improvement of analytical techniques and with the enhancement of accessible data through the implementation of new sampling and measurement campaigns, the radionuclide inventory of graphite can be made more precise in order to be as close as possible to

reality whilst guaranteeing an upper margin corresponding to a selected risk of under-assessment.

This paper shows that the 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, and the first results obtained have been analysed by IRSN, which has found out that results are promising, in the way that they exhibit physical robustness. These elements are the main topic of his paper.

2 Potential sources of radionuclides in irradiated graphite

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 [1]. As will be shown, 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 other radionuclides are naturally occurring radioactive elements such as ^{40}K and the various isotopes of natural uranium impurities.

In the context of preliminary safety studies, it has been chosen to arbitrarily focus this paper on three radionuclides considered to be representative of various phenomena that may occur in the reactor or considered to be significant for the management of graphite waste. They are the following:

- ^{3}H (tritium) is in terms of activity level a major radionuclide in irradiated graphite from UNGG reactors. It is a short-lived ($t_{1/2}$ = 12,33 years), low energy radionuclide whose potential impact concern mainly its management during the dismantling period and the operating period of a repository. Tritium can be produced through neutron activation of two impurities of the graphite, boron and lithium. The concentrations of lithium and boron impurities in the graphite are equivalent to around 0.1 mg/kg [2]. The tritium produced in UNGG graphite is almost exclusively due to the activation reaction of lithium ^{6}Li (n,α) ^{3}H, of which the neutron effective cross section is the highest [3].
- ^{14}C is also in term of activity level a major radionuclide in irradiated graphite. It is a long-lived ($t_{1/2}$ = 5730 years), pure β emitter which gives stable ^{14}N by radioactive decay. There are two main ways of forming ^{14}C by neutron activation in graphite: activation of the most abundant isotope of nitrogen (^{14}N) or activation of natural isotope 13 of carbon (^{13}C). The origin of ^{14}C in the graphite from the reactions ^{14}N (n,p) ^{14}C and ^{13}C (n,γ) ^{14}C highly depends on the nitrogen content in the graphite under operating conditions [2].
- ^{36}Cl is a long-lived radionuclide ($t_{1/2}$ = 301000 years). Although minor in terms of activity level in French irradiated graphite, ^{36}Cl is considered in impact studies as the first radionuclide to be released from the repository containment, due to its high mobility. It may be produced in graphite gas-cooled reactors in various ways by direct activation of stable ^{35}Cl and ^{39}K, and also by indirect pathway neutron activation of the stable ^{34}S. With the sulphur and potassium impurity contents in nuclear graphite, it appears that the reaction ^{35}Cl (n,γ) ^{36}Cl is by far the predominant reaction for the formation of ^{36}Cl in graphite of a UNGG reactor [4].

3 The issue of radionuclide measurement discrepancies

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 as noticeable in Fig. 4)

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 200 cores (samples) taken as average per reactor pile. For NPP Bugey 1, this huge variability observed at the scale of a pile correlates

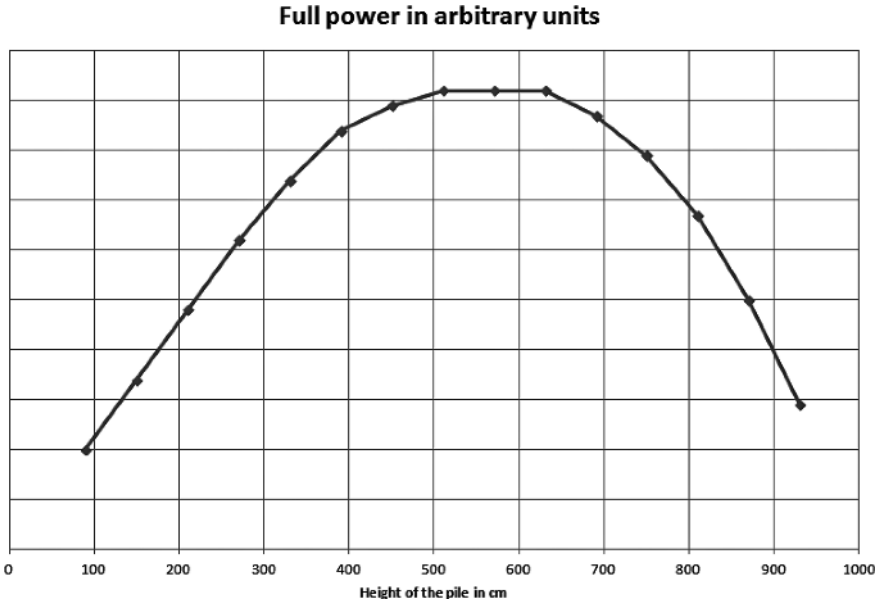

Fig. 1. Shape of the thermal power correlated to the neutron flux into a fuel channel of Bugey 1 reactor with x axis height of the pile in cm and y axis full power in arbitrary units

- 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),
- 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. Very pure nuclear graphite is a well-known polycrystalline material [1] that exhibits a huge heterogeneity from a structural point of view at micro and nanoscale. SIMS characterizations performed on some precursors of radionuclides such as ^{35}Cl also evidence a huge heterogeneity from a chemical point of view. For example, micrometric sized clusters of ^{35}Cl are observed [5]. Actually nuclear graphite cannot be considered from an analytical point of view like any other common homogeneous material (liquid solution, usual steel) 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 radionuclides and 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.

4 The identification calculation – measurement method

The easiest way to assess the radionuclide inventory of the irradiated graphite should proceed by interpreting radiochemical measurements on irradiated graphite sampled. The method of radionuclide inventory assessment by identification calculation-measurement described below is specifically designed for disposal sizing purpose. It consists in using the following process:

First step: ''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.

Second step: 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 Bateman equations. The adjustment process for the impurities explanatory values is iterative toward the minimization of the quadratic function Eq. (1):

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

where N_i is the number of available measurement activity of each i radionuclide (logarithm is used here to consider low measurement values participating at the adjustment process).

In a third step: to calculate a confidence interval taking into account that each measured radionuclide is characterized by a

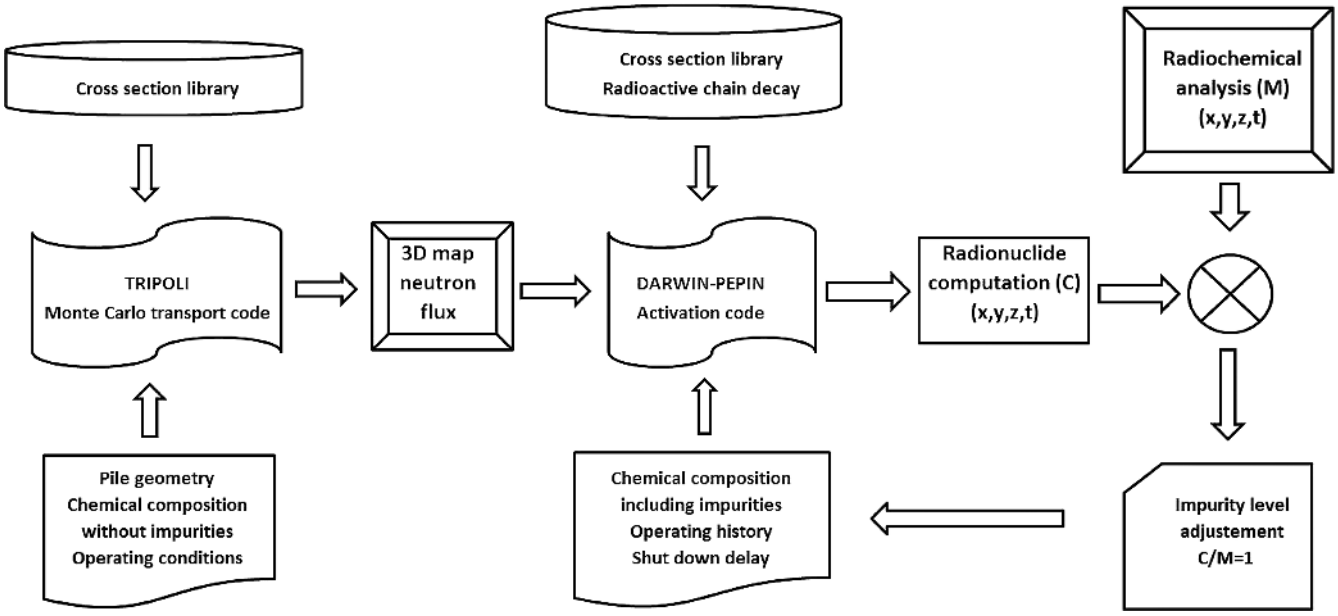

Fig. 2. Simplified description of step 1 and step 2 principles for calculating the radionuclide inventory of the pile in the EDF reactors by identification method

specific standard deviation that is dependent on the number of measurement points available; The standard deviation, which is individual to each measured radionuclide, is used to calculate a confidence interval for its mean calculated value. For an overall determination of the inventory, we consider the upper value of the confidence interval (which corresponds to a 2.5 % risk of under-evaluating the result) of the ratio with respect to the ^{60}Co γ-tracer, in order to remain consistent with the choices usually made in waste management practice because of the well-known ratio robustness.

In a fourth step, in order to determine the value of activity to be declared for each radionuclide, the upper value of the confidence interval of the ^{60}Co γ-tracer was considered. This value thus corresponds to a specific activity to which a 2.5 % risk of under evaluation margin is added. The upper value was multiplied by the upper value of the ratio previously calculated for the radionuclide concerned.

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. This data is processed using the Central Limit Theorem (CLT), employed in order to determine the maximum uncertainty in the activity (which follows a Gaussian probability distribution function by application of the CLT).

Since the variance of a mean is obtained by dividing the variance of the population of measurement points by the number of points, then it is guilty that the larger the number of measurement points, the smaller is the confidence interval on the mean value of the activity of a given radionuclide. This is used to directly obtain the uncertainty calculated from:

- statistical measurement data and the associated adjusted calculations
- number of points used
- under-assessment risk selected (2.5 % hereafter)

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. It is from an order of magnitude of about 30 points that the number of points can be seen as always being sufficient. The withdrawal of any of them does not significantly change either the estimate of the mean or its confidence interval. On the other hand, below 15 points, the calculation that employs the Student law introduces a penalty in terms of increasing the size of the confidence interval.

Step 1 and 2, which are the heart of the proposed method, are schematically described in Fig. 2. It is seen as *reverse* method in the sense that the radiological measurement results are some inputs.

5 Results and discussion

5.1 Radiological inventory, fluctuation regularisation, and cross section correlated spatial distribution

Figure 4 shows the axial distribution of activity weight concentration of ^{36}Cl, ^{14}C and ^{3}H in Bugey 1 reactor. These activities are the final ones, calculated by activation of the explanatory impurities whose concentration comes from the optimisation process. They are calculated in several radial channels of the pile, where measurements were done.

It is striking to see a first that the strong fluctuations in the measurements of these three radionuclides are regularised by the identification-calculation method, with a continuous profile that matches the parabolic neutron flux profile (clearly visible for ^{14}C).

Moreover, if these profiles are regarded through the (thermal) neutron capture cross section (σ_{capt}) of the explanatory impurities, the results become very physical having in mind that:

- σ_{capt} of the ^{13}C (n,γ) ^{14}C reaction is 10^{-3} barn
- σ_{capt} of the ^{35}Cl (n,γ) ^{36}Cl reaction is 43 barns
- σ_{capt} of the ^{6}Li (n,α) ^{3}H reaction is 940 barns

The flat profile of tritium obtained by the proposed method is therefore not surprising: with such a high neutron capture cross section of the ^{6}Li impurity, the tritium can reach a saturation value, and it is therefore probable that all de ^{6}Li content has been activated, before the reactor final shutdown, leading to a loss of correlation between the axial distribution of the tritium concentration and the neutron flux. In the contrary to most shaped parabolic profile is found for ^{14}C axial distribution, whose ^{13}C explanatory impurity has a low neutron capture cross section, leading to a strong correlation with the neutron flux.

5.2 Explanatory impurities and activation scheme

Beside the results in terms of activity, an output of the method is the relation between activated impurities and radionuclides, and the global activation scheme of the pile. These results of the method are dependent of the method itself. If the nature of the impurities contributing to the formation of a radionuclide can be determined by individual activation calculations, the contributions of these impurities may be affected by the optimisation process or some user choices. For instance, for the present results (Bugey 1) the activation schemes of the three radionuclides are presented in Fig. 3.

More generally, 144 i-radionuclides are found by the activation of 52 k-impurities through the proposed method. Therefore an output result of the method is the activation matrix M whose terms m_{ki} represent the contributions of the k impurities to i radionuclides at the scale of the whole pile. This activation matrix M is a key result to compare from a pile to another to assess the physical robustness of the method, a comparison work that is planned as a next step.

6 Conclusion and further work

About 17,000 tons of irradiated graphite waste will be produced from the decommissioning of the six French gas-cooled nuclear reactors. Determining their radionuclide content is an important legal commitment for both safety reasons and in order to determine the best practice to manage them. It is

$$\left.\begin{array}{l} {}^{13}C\,(n,\gamma) \xrightarrow[73\%]{} \\ {}^{14}N\,(n,p) \xrightarrow[27\%]{} \end{array}\right\} \; {}^{14}C \text{ production } (100\%)$$

$$\left.\begin{array}{l} {}^{34}S\,(n,\gamma)\; {}^{35}S \xrightarrow[T_{1/2}=87d]{\beta} \; {}^{35}Cl\,(n,\gamma) \xrightarrow[96\%]{} \\ \qquad\qquad\qquad\qquad\quad {}^{35}Cl\,(n,\gamma) \xrightarrow[2\%]{} \\ \qquad\qquad\qquad\qquad\quad {}^{39}K\,(n,\alpha) \xrightarrow[2\%]{} \end{array}\right\} \; {}^{36}Cl \text{ production } (100\%)$$

$${}^{6}Li\,(n,\alpha) \quad {}^{3}H \text{ production } (\approx 100\%)$$

Fig. 3. Activation productions of ^{14}C, ^{36}Cl and ^{3}H

not possible to assess the radionuclide inventory by using a conventional activation calculation method alone as for a well-known simple activation of a metallic alloy.

But, another way to assess the radionuclide inventory of the irradiated graphite relies on statistically interpreting the radiochemical measurement population of irradiated graphite sampled from the pile of the reactors. This is particularly important with precursors at trace level.

As shown through sampling and measuring campaigns on EDF UNGG graphite piles, radionuclide measurements may exhibit a wide discrepancy. Starting from this very specific fact, EDF-DP2D has developed a method of 3D geographical reconstitution of the inventory of the nuclear piles based on an activation calculation adjusting the mean value of the several measurements available with the mean value of the calculation corresponding to the measurements (same position and same time). By using an averaging approach to the measurements, and not an approach based on the maximum measurement, the measured data is then made truly representative. The calculation of impurity contents from the measurements is performed on samples by reverse activation calculation. A particularly noteworthy result found by IRSN when expertizing this method is that the calculated activity concentration profiles are fully explainable, regarding the neutron capture cross section. The method seems therefore promising to assess the radiological inventories of all the radioactive nu-

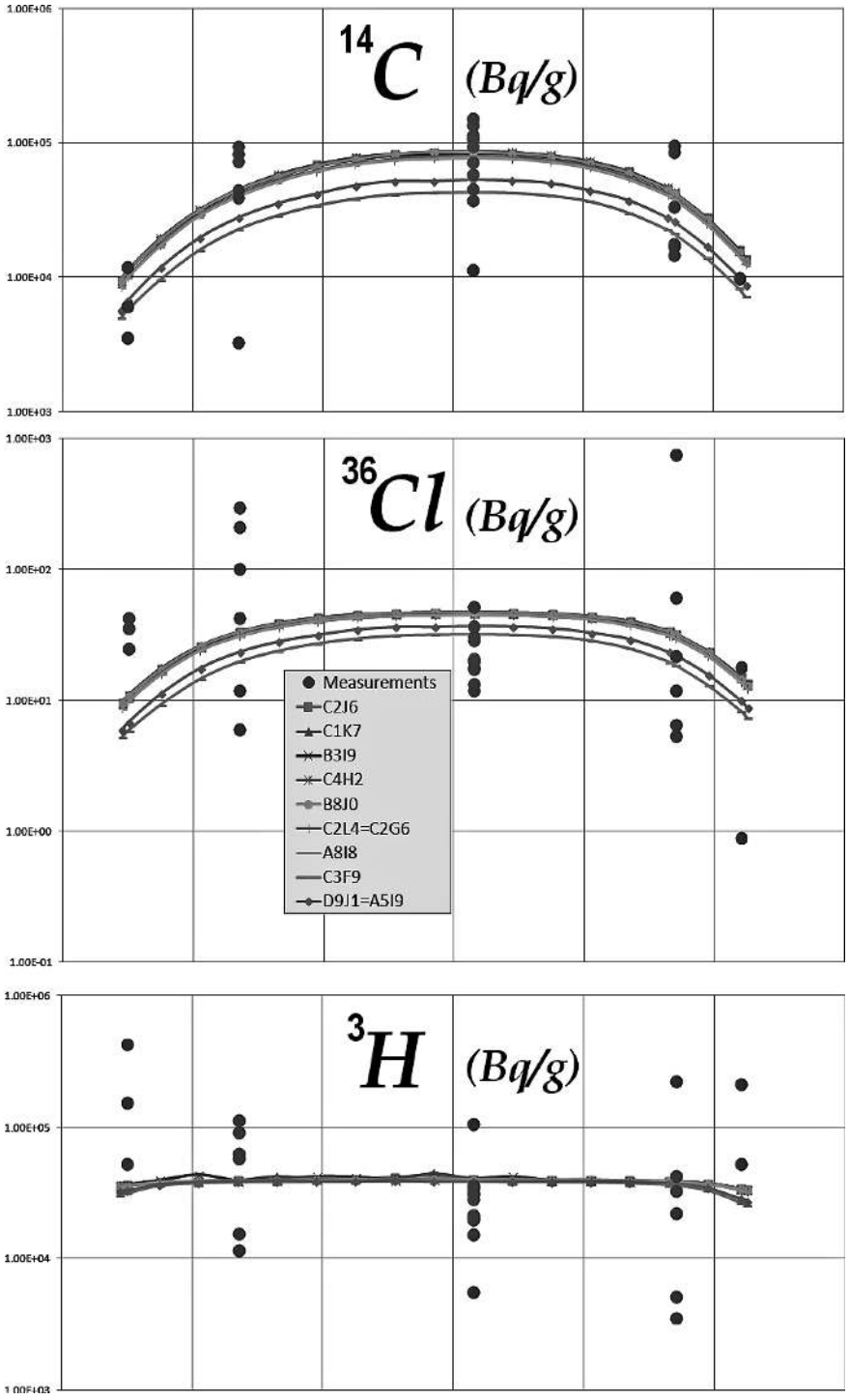

Fig. 4. Activity concentration profiles calculated by the proposed method: y axis are Bq/g of ^{14}C, ^{36}Cl and ^{3}H versus x axis which is vertical coordinate of Bugey 1 graphite pile with black balls representing sample measurements, compared to average calculated curves for the 11 corresponding sampled channels. The more or less parabolic shapes of the calculated channel profiles are fully explained by the neutron capture cross section of precursor impurities

clear graphite coming from UNGG reactors in France. A next step of validation is to apply this method to every pile, and compare the activation matrix from a pile to another. They are expected to be very similar, ensuring that the results given by the method are not dependant on the considered reactor.

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The Safety Culture of an Effective Nuclear Regulatory Body. Published by the OECD/NEA, NEA Report No. 7247, 35 pp., 2016, available online at: www.oecd-nea.org/rp/pubs/2016/

Although the national regulator plays an essential role in each country, operating experience has shown that accidents may impact other countries and may involve other national regulators. Safety is therefore not bounded by national borders. The implications of this global nuclear safety approach should be taken into account when addressing the safety culture of the national regulator.

It is important to emphasise that, although the mission of the regulatory body is to provide oversight on nuclear safety, the prime responsibility for the safety of a nuclear installation remains with the licensee or plant operator. The regulatory body nevertheless has an important responsibility in assuring that the licensee meets its primary goal of ensuring the safety of nuclear installations. With its regulatory strategy, the way it carries out its daily oversight work, the type of relationship it cultivates with licensees, the values it conveys and the importance it gives to safety – in short, with its own safety culture – the regulatory body profoundly impacts the licensee's safety culture and its sense of responsibility for safety. Hence, the regulatory body needs to be conscious of its own safety culture's impact on the safety culture of the organisations it regulates and oversees in order not to hamper those organisations' willingness and efforts to take on their primary responsibility for safety. For this reason, it is paramount that the regulatory body not only consider safety culture as a matter of oversight, but also as a matter of self-reflection. It should actively scrutinise how its own safety culture impacts the licensees' safety culture. It should also reflect on its role within the wider system and on how its own culture is the result of its interactions with the licensees and all other stakeholders.

A regulatory body should have public safety as its primary focus and a healthy safety culture is essential in this regard. Such a safety culture should encompass individual staff members, leaders and the organisation as a whole.

This Nuclear Energy Agency (NEA) regulatory guidance report identifies and describes five principles and their associated attributes that underpin and support the safety culture of an effective nuclear regulatory body. Each of the characteristics – the principles and the attributes – discussed in this report is a necessary feature of the safety culture of an effective nuclear regulatory body, but no one characteristic is sufficient on its own. It is the combination of these characteristics – the principles and the attributes – that leads to a healthy safety culture within the nuclear regulatory body.

The report concludes that the following elements support a healthy safety culture within the regulatory body:

- Excellence in leadership for safety at all levels of the organisation to demonstrate the importance of prioritising safety above all else.
- Strong sense of personal accountability so that everyone takes personal ownership of their actions and decisions with respect to safety.
- Formal direction on safety culture (i.e. a clear corporate policy on safety culture in the form of statements, guidance or a code of conduct).
- Staff who are aligned and engaged: a healthy safety culture is supported by staff who know what they are doing.
- Open and transparent communication, internally and externally.
- Informed, balanced accountability that encourages open and honest reporting and respects safety information.
- A comprehensive and systemic approach to the regulatory environment which is a complex and interdependent system that requires a holistic approach to its management.
- A clear and appropriate regulatory framework.
- Continuous improvement and learning: an open, adaptable and learning attitude in technical, regulatory and organisational areas helps avoid complacency by continuously challenging existing conditions and activities.
- Self-assessment: assessment of the safety culture of the regulatory body supports continuous improvement. At the same time, more work is needed in the development of assessment methodologies and appropriate performance indicators.
- Benchmarking to ensure consistency with peers, share experiences and support a global safety approach.