

EDF RADIOACTIVE GRAPHITE INVENTORIES BASED ON HUNDREDS OF MEASUREMENTS ARE CASTING DOUBT ON “A PRIORI” ACCEPTED IDEAS

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ABSTRACT

Data assimilation is a powerful methodology that makes efficient, accurate and realistic estimations possible, and that might not be feasible without it.

However, EDF has made impurity concentration estimates via data assimilation on 4 graphite piles with the minimum number of observations required by the Central Limit Theorem (CLT) and including the computation of confidence intervals in a stochastic context, because the analysed sample size is smaller than the Representative Elementary Volume (REV) of graphite.

Starting from hundreds of radiochemistry analyses, data assimilation applied to 4 dismantled graphite piles using Bateman equations only (i.e. activation by neutron flux only) has given 4 radioactive inventories concerning more than a hundred radionuclides.

The paper presents a comparison between the 4 sets of impurities that are intermediate results and illustrates significant matching of them with the origin of the graphite coke used to build each pile.

Although the origin of the coke is known, it has never been included in the input data, showing that the explanation by activation alone without any consideration of chemical behavior is sufficient for all radionuclides except for the C-14 explanation.

This coherence confirms the relevance of the data assimilation methodology that is based on computation of the activation and fitting impurity concentrations to obtain an explanation of the measurements.

If you don't take into account the concept of “Representative Elementary Volume” (REV), you may build any explanation about Cl-36 in graphite during operation. But, if you consider the measurement results and their very stochastic behavior, you will remember that no explanation can be given at a lower scale than the REV, which is the case in radiochemistry graphite samples. It is the reason why no R&D is needed to compute the effective quantity and localization of Cl-36, but only appropriate mathematical tools as “Central Limit Theorem” (CLT) and “Data Assimilation Method”.

In spite of an other a priori opinion about high content of nitrogen being activated to produce C-14 in graphite, if you take into account C-14 measurement results with the same tools, you will be aware that it is not the case.

Considering the computation results based on hundreds of radiochemistry analyses, the paper allows people to reconsider accepted ideas about “unknown Cl-36 behavior” and “almost 100% proportion of C-14 originating from nitrogen”.

Remember that the interest of these results is closely linked to the high sanitary impact of Cl-36 and C-14 on disposals for safety assessment purposes.

INTRODUCTION

Électricité de France (EDF), a radioactive waste producer, is responsible for managing waste from the dismantling of its permanently shut-down nuclear power plants. This dismantling process has been underway for a few years now and concerns 9 units. Of these, 6 units are gas-cooled graphite power plants. EDF is required to assess the radioactive inventory for each of its graphite radionuclides (RN) and communicate the information to French governmental disposal agency (ANDRA), for the purposes of their safety studies in connection with the identification of suitable storage sites.

We will start from the method of producing radioactive inventories and inventory calculations of BUG1, CHA3, SLA1 and SLA2, four French Gas Cooled Reactors decommissioned piles, and give a summary of intermediate results derived from this method and the comparison between them.

The method used is based on a mathematical demonstration using data assimilation. It consists of a physical measurement including uncertainty quantification processing. Although the final required result is radioactive inventories, the genuine measured magnitude is the adjusted impurities vector. The comparison must be made using values determined without any indeterminacy that make significant increases necessary.

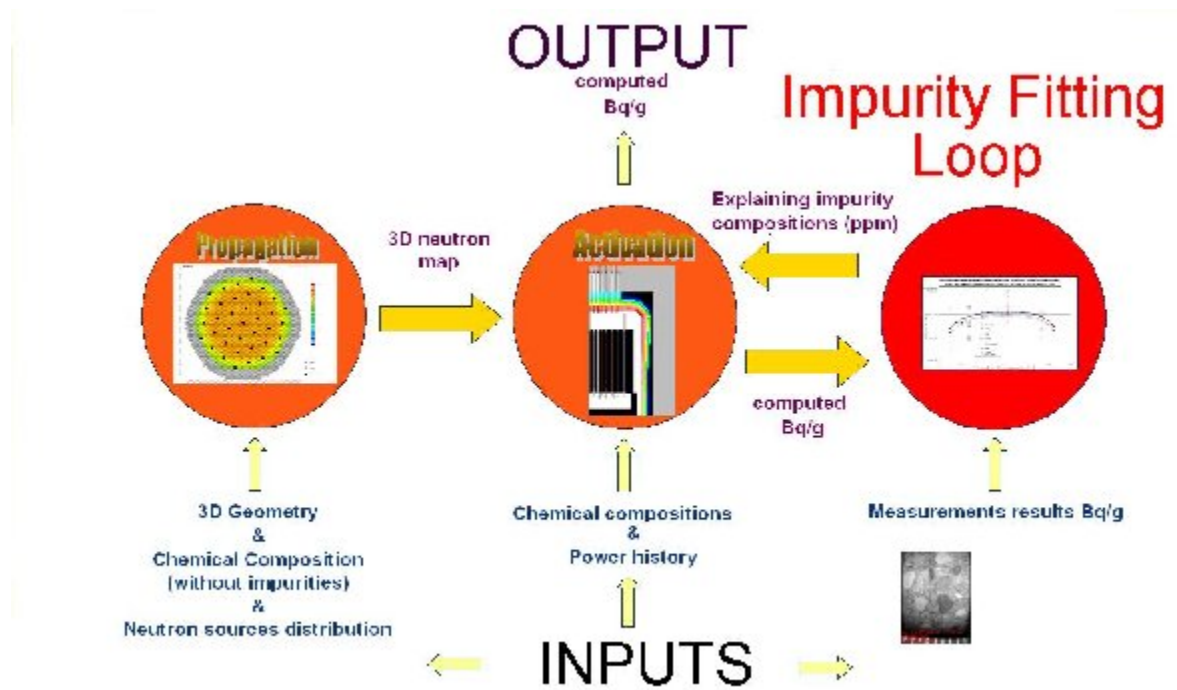


Fig. 1. Data Assimilation i.e. impurity adjustment by identification between measurements and computations of RN (Nothing here is specific to graphite)

Thus, as random variables, these impurities that explain inventories are the only means of making comparisons using the relative uncertainties calculation.

In the comparison of “explanatory compositions”, the term explanatory should be understood to mean the natural chemical precursor element that explains radioactivity measurements on samples as a function of the irradiation characteristics of these samples, and that also illustrates the classification of initial graphite in each reactor.

This classification is derived directly from

- RN (Radionuclides) measurements (several hundreds) made on taken core samples and
- the laws of physics that govern neutron fluxes and the resulting activity.

A comparison between "explanatory compositions" highlights a certain consistency that independently confirms the validity of the results, considering the sources of the graphite used. This consistency is highlighted, although it was not the initial objective of the inventory calculations.

Calculation of the radioactive inventory by data assimilation is now a routine process in the activated wastes domain. This type of calculation is nothing like a "numerical method" (as had been incorrectly considered or interpreted in the past) because it is based on **physical measurements** if there are enough of them, and the **laws of nature** (**Boltzmann's** equations for neutron fluxes and **Bateman's** equations for activation by neutron fluxes).

We should remain aware that the laws of interactions between neutron and nuclei are universal and that there is nothing specific to graphite.

REMINDER ABOUT FUNDAMENTALS

Mathematical language and credibility of the approach

The inescapable advantage of the method used is that, contrary to common belief, it provides a scientific demonstration that about thirty samples each weighing a few tens of grams (namely less than a kilogram of graphite) can be sufficient to give the value of the radioactive inventory of several thousand metric tons of graphite in a pile, an under estimation risk level being chosen.

The following describes how randomness is processed with certainty for a material such as graphite manufactured brick by brick (the bricks being replaceable) under the production conditions specific to nuclear graphite.

Processing of randomness through the average

For example, we can cite the mathematician Kolmogorov to help understand the probability approach used:

"The value [] of the probability theory is based on the fact that random phenomena generate a strict regularity at large scale, in which randomness has disappeared to a certain extent".

In physics, it is normal practice to define the concept of a "Representative Elementary Volume" (REV) for an heterogeneous material such as graphite. Very significant fluctuations in individual measurements, in other words large differences in the averages of these measurements, are observed when the size of the measured sample is very small compared with the size of the REV.

These fluctuations are described by regular probability laws with characteristics that are largely independent of the details of the physical phenomena from which they originate.

When the sample reaches the REV, by definition the concentration of the impurity is constant. In any case, the size of the graphite REV is very much larger than the size of a sample and is smaller than a brick, because the uniformity with which bricks were manufactured was also a manufacturing characteristic of nuclear graphite.

Thus, the behaviors found can be qualified as universal and everyone will understand that this surprising universality of behaviors means that the results of the adjustments thus calculated have an excellent prediction capacity. They can be used to quantify the probability of the calculated values being exceeded. They lead to the conventional technical concept of a "confidence interval" taking account of a chosen risk in advance.

Central Limit Theorem (C.L.T.) and quantification

The simplest form of this phenomenon, clearly perceived by Laplace at the end of the XVIIIth century, is expressed by the fact that the sum of N random numbers with the same probability law is equal to N times the average value of one of these random numbers, when N is large enough.

The “Central Limit Theorem” (CLT) provides information not only about the average, but also about standard deviations from the average. More precisely, this theorem informs us about the probability that a certain fluctuation will occur as a function of its amplitude, in other words its deviation from the average.

As might be expected, large fluctuations that differ very much from the average, are less probable than small fluctuations. Deviations from the average are distributed according to a probability law illustrated by a bell curve, also called a Gauss curve.

The probability is maximum for a zero deviation (centered value). It is much lower for large values of the deviation. However, the width of the Gauss curve that measures the amplitude of probable fluctuations increases proportionally to \sqrt{N} , the square root of the number of measurements.

This Gaussian behavior is remarkable because it is particularly independent of the microscopic details due to the composite nature of the material (case of graphite that is very heterogeneous at low scale).

A physical measurement process is used

To use the conventional mathematical terminology, the method is a mathematical identification and it forms a global measurement process starting from all analyses on samples. These methods are also known nowadays under the term “data assimilation methods”.

The power of the method as applied in the practical context of activation also lies in the fact that it simultaneously processes the (relative instead of absolute) uncertainty that results from the same hundreds of RN measurements used as input data.

The method is verifiable

Physical laws form the sole basis for the approach. The term “numerical model” used to describe it is not suitable. The model is physical and its numerical aspect is simply due to the fact that mathematics provides the language and the basic tool used in physics. The fact that the numerical aspect of the solution of equations can be solved using different calculation methods should not suggest that the solution to the physical problem of adjustment on averages is not unique.

It is always possible to choose simplifying assumptions specific to approximation phases in which the error is quantified more briefly. This leads to an undue increase in the uncertainty. There is no hazardous simplification in the case of this implementation, and only a minimum number of input data are necessary.

Physical measurement characteristic

A physical measurement is always affected by an uncontrolled uncertainty. The term measurement error usually denotes a value that maximizes the absolute value of this uncertainty.

The effect of mathematical complexity lies in the uncertainty quantification process. This itself is dependent on the measurement means used and the expertise level necessary for its interpretation.

The evaluation of the radioactive inventory of graphite is also a measurement process. This process is undoubtedly advanced because

- it takes account of an activation model conforming with the laws of physics
- it includes an identification process relative to a set of analyses that supplies the uncertainty calculation while such information is not explicitly generated like a by-product for a simple standard measurement instrument.

Unlike a routine measurement and considering the context and very specific stakes, the evaluation of the radioactive inventory and its uncertainty must be accompanied by an explanation of the methods and means used (which are usually not done in many other domains).

Basic principles for processing uncertainties

In the approach, it was necessary to:

- develop the calculation starting from its basic principles that are:
 - (1) the REV concept for interpreting averages
 - (2) the CLT to quantify the uncertainty of these averages,
- explain this significant component corresponding to the uncertainty, although it is never mentioned in routine measurement processes because it is usually not very significant or neglected.

Remember that uncertainty is either related to random directly, or is related to phenomena that are so complex at very small scale that they cannot be used in anything other than probability laws.

- Made by random: In general terms, counting “disintegrations” is still the only way of obtaining a final result consisting of a radioactivity measurement after a more or less sophisticated physicochemical preparation. The radioactive measurement is inherently numeric, while counting is a perfect example of random because the physics itself explains how the occurrence of radioactive disintegrations is probabilistic. Remember that the measurement uncertainty corresponds to the uncertainty of the average of random variables derived from the same laws (the laws of physics). This gives a Gaussian quantification of a measurement uncertainty with 95% confidence through the application of the CLT and a width factor normally fixed at ≈ 2 (1.96 precisely). For example, this is the approach used in competent radiochemistry laboratories.
- Complexity of representation: The other aspect related to a complexity exceeding practical possibilities is the REV (Representative Elementary Volume) concept. This concept is used conventionally in geology and in all disciplines in which the studied medium is not a homogeneous liquid or gas solution due to Brownian stirring, but is rather due to mixing (frequently due to natural causes) that may be heterogeneous for very small volumes but is nevertheless homogeneous at the scale of the REV. This corresponds to the very definition of the REV: Global balances are respected at this scale, as what happens in a liquid or gaseous medium. Therefore, modeling of what happens at a scale smaller than the REV can only be probabilistic. The confidence interval can be quantified if a sufficient number of measurements are available for application of the CLT and “conventional” Student widening.

ADJUSTED OR NON-ADJUSTED IMPURITIES

This chapter explains the conditions under which the results of the four French graphite piles CHA3, SLA1, SLA2 and BUG1 inventories can be compared.

Adjustment without increase (injective)

The genuine result of the calculation/measurement identification is the average impurities vector of graphite without any irradiation. Corresponding to an average on a graphite volume larger than REV, this impurity volume is an "observable" variable.

This “observation” is made through processing of calculation/measurement deviations of the activities of the different RNs analyzed in different samples. Note that the samples are significantly smaller than the REV (this refers to the low volumes constraint required in radiochemistry).

This “observable” variable acts as a pivot to make the final activation calculation that uses this average vector that is the result of an optimum adjustment of calculations with measurements. The measurements made on samples very much smaller than the REV give the quantification of the uncertainty through the concept of the standard deviation, with the best global adjustment through the concept of the average.

If we exclude:

- adjustments by concurrent impurities that are impossible to discriminate with the result of RN measurements and that are nevertheless chosen so as to be conservative (the Carbon-Nitrogen pair through C-14 and the Sm-Eu pair)
- adjustments of a single impurity through two different RNs with the choice of the worst case adjustment (2 values of Ni through Ni-63 and Ni-59 and for Nb and U)
- impurities not accessible by adjustment due to the lack of information on RNs that they produce by activation

adjustments of the remaining impurities are injective.

This means that they fix a value of one and only one centered impurity through a reference RN without any systematic increase related to an over-abundance of unknowns in comparison with the equations.

Adjustment significantly increased by indetermination

Penalization of uranium, niobium and nickel

We will keep these elements in a first approach, because the penalty applicable to them can be judged to be low.

Indetermination of europium/samarium

The only available radioactive europium measurements are not sufficient to discriminate the impurity from which europium (Eu) and samarium (Sm) originated. After assuming that there is no Eu, the adjusted value of Sm is greater than the true value and conversely, after assuming that there is no Sm, the adjusted value of Eu is greater than the true value.

This increase so that the two opposing cases can cohabit is too large for it to be possible to use the selected values for Eu or Sm as being simultaneously significant (the proximities between Eu or Sm in different reactors limit the representativeness of the values obtained due this double increase required to remain conservative).

Impurity that cannot be accessed by measurement

Some impurities are not “observable”, in other words they are not accessible by RN measurements:

- either because the RNs resulting from them cannot be measured in the current state of radiochemistry,
- or the number of available analyses is too small to give an acceptable statistical validity.

Although it was evaluated for BUG1 and CHA3, the iron impurity (Fe) for SLA1 and SLA2 has been simply considered to be equal to the largest adjusted value for BUG1 on CHA3, in the absence of any available measurements of an explained RN. Consequently, it cannot be used for comparisons between reactors.

COMPARISON OF THE FIRST FOUR CALCULATED PILES

Use of significant impurities

The genuine measured magnitude is the adjusted impurities vector, but we restrict ourselves only to impurities obtained for which there is no indetermination leading to the systematic use of increased values (injective or almost injective impurities Ba, Cl, Co, Cs, Li, Nb, Ni and U, see above).

We will also consider thorium (Th) impurity being non-significant because the particularly low level of this impurity combined with the particularly high uncertainty associated with this very low value considerably weaken its significance.

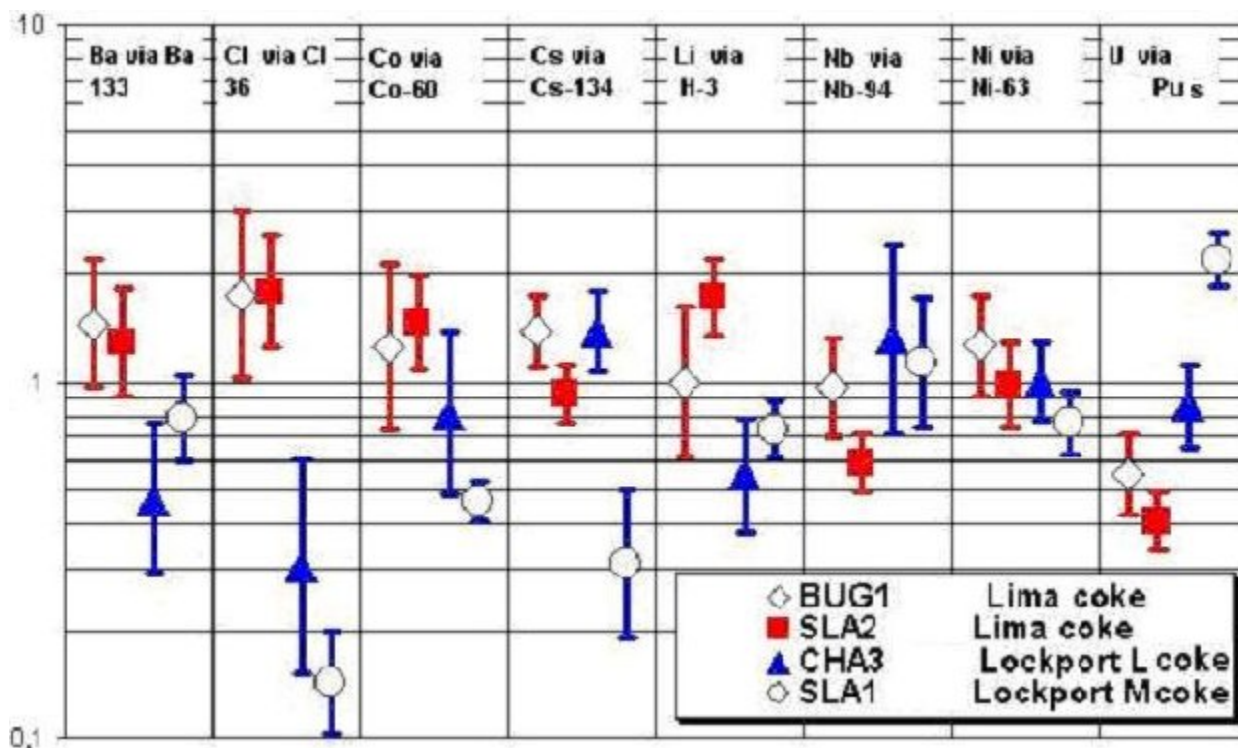


Fig. 2. Eight impurities adjusted from RN measurements and neutron fluxes for 4 piles – Concentrations divided by the 4 piles average given with 95% confidence intervals.

Analyses of results and comments

The coke from which the graphite originated seems explanatory

Similarly to what is predicted by the standardization of chemical analyses of solids in which two test sample measurements are considered to be coherent if the intersection of their confidence intervals is not null, the consistency of the “measured impurities” corresponds to the intersection of intervals shown as uncertainty bars in the graph.

The proximity of BUG1 and SLA2 (LIMA coke) is confirmed in all cases, although the cross check with LOCKPORT coke is not systematic due to the fact that SLA1 and CHA3 use two different LOCKPORT variants.

Thus, the LIMA/LOCKPORT difference appears consistent with the results derived from application of the data assimilation on four UNGG reactors. It thus appears as a factor justifying differences of “explanatory” compositions and consequently providing an additional candidate explanation for radioactive inventories that were not included in working assumptions.

The reactor operating mode does not appear to have any influence

It has been envisaged that the difference between operating conditions of the reactor (for example renewal of the heat transporting CO₂ or the pollution of fuel cladding) could be the cause of an effect on an “explanatory composition” by activation.

The explanatory nature of the origin of the coke would appear to be sufficient without any need for an explanation dependent on operation of the reactor.

“A PRIORI” IDEAS ON CHLORE-36 APPEAR TO BE CONTRADICTED

No need for R&D for consolidation

“A priori” ideas about Chlorine 36 R&D needs are apparently still present in national and international publications but no particular R&D action was or is necessary to determine the reasonable envelope radiological inventory derived from available measurements on EDF piles and obtained using data assimilation on an activation model.

Consolidation is achieved through a simple objective consideration of the laws of nature and their use by mathematical data assimilation tools. This is perfectly sufficient for inventory needs for waste elimination purposes. We should remain aware that the laws of interactions between neutron and nuclei are universal and that there is nothing specific to graphite.

This document has shown that a comparison between explanatory impurities through their neutron activation of four EDF piles can actually determine the proximity of cokes from which the graphite concerned originated, which shows a significant match between the activation model and the measured reality.

There is no need for any research action on the precise subject of the EDF graphite Cl-36 inventory, it would not be useful.

Elementary explanation by the R.E.V.

An R&D approach is not necessary for the central concept of the understanding of phenomena because it is simply that the analyzed samples are necessarily small and therefore significantly less than the REV (Representative Elementary Volume) of the graphite material.

As we have seen above, below the REV, the average value on the sample is not the same as the real average value for the material, and that the real value is obtained either from a REV or a larger volume (large samples cannot be taken from an active pile under the existing industrial and regulatory conditions), or by taking the average of statistically sufficient values for small samples so that mathematical standards associated with the CLT can be applied (which was done with a sufficient number of samples).

We have seen above that, in the mathematical method used, this consubstantial variability of a locally heterogeneous material like graphite is quantified by the size of the confidence interval surrounding the centered calculation of RNs.

The figure shows that the sizes of these confidence intervals are comparable for Cobalt-60 and Chlorine-36. Since these sizes are simply the image of the variability of the original impurity, it can be seen that the Chlorine-36 present in the graphite is not due to any particular mobility. We would have to conclude that Cobalt-60 has at least an equivalent mobility, which seems highly unlikely ...

Obviously, the distribution of chlorine in graphite is not homogeneous, but unlike some “a priori” opinions, the distribution of chlorine is not more dependent on the location of graphite in the reactor or the temperature reached, than the cobalt distribution.

No matter how attractive it may be, such a dependence would be incompatible with the particularly discontinuous nature (therefore without correlation in space and without correlation in operating temperature) of the measurements made. The lack of spatial continuity is the first result that is immediately obvious when looking at the measurements. Therefore any explanation other than activation of a small scale stochastic impurity, for example such as an explanation by a position or by a temperature (continuous field) is quite impossible.

Confirmation by S.I.M.S. analyses

This local variability that is large at the scale of the sample was also recently confirmed by CEA analyses on activated graphite using the SIMS (Secondary Ion Mass Spectrometry) tool. The operator explains in his report that the observation of the Chlorine 36 (very recognizable mass 36) only lasts for a few fractions of a second because the ion beam very quickly volatilizes the very small clump of Chlorine 36, despite the fact that it is extremely thin. This is the “visual” confirmation of the very strong local heterogeneity that gives its full meaning to the stochastic nature of the impurity below the REV (consequently to the activity per unit mass).

Everyone realizes that the measurement of the activity per unit mass of Chlorine 36 in a sample much smaller than the REV, is not at all representative, and this was the reason why the inventory was adjusted over a statistical sample of about 30 samples to obtain the required representativeness and the associated confidence interval.

Similarly, the leached fraction of the Chlorine 36 activity in a sample much smaller than the REV is not representative either, which perfectly explains the observed experimental variability of this leached fraction (between 20% and 80%).

The stochastic nature takes precedence

Without any other evidence of it, there is no need to introduce an explanation with several chlorines or several diffusion regimes because diffusion is inherently a stochastic regime that accumulates many elementary random behaviors of materials at a sufficient scale.

This sufficient scale only appears to start when the sample is representative and this is never the case even for grouped samples because the quantity involved is always smaller than the REV.

Furthermore, the leached fraction closely depends on renewal conditions of the leaching water and the fact that the measurement in the leachate is less than the detection limit (<DL) so that it is impossible to escape from the total renewal required to represent null Dirichlet conditions (null conditions at boundaries).

In order to be able to interpret the stochastic results obtained, their random nature needs to be eliminated by an averaging process on a sample statistically comparable to that used for building up the inventory. Another application of the CLT has to be made.

It is only after such an approach that a possible correlation with the position and/or the temperature reached by the graphite could be envisaged, with a quantifiable confidence level. However, remember that such a correlation is already definitively contradicted by the particularly discontinuous (i.e. highly uncorrelated) measurements of the activity per unit mass of Chlorine 36.

CARBON 14 AND RADIOLYTIC CORROSION

This chapter also contains key information to help understand position differences between British calculations and EDF measurements particularly about the activation of nitrogen, that became clear at the most recent conferences.

Current situation

Concerning the interpretation of C-14 measurements, we will see that it appears necessary to take account of the radiolytic corrosion effect.

In particular, this phenomenon provides explanations about the general and comparable existence of samples from the 4 piles, such that the measured C-14 is less than the C-14 derived from the carbon matrix of the graphite. At a scale less than REV, the porosity distribution is very variable, in the same way as the impurities. Therefore, the chemical corrosion reaction does not respect the isotopic ratio at this scale.

The radiolytic corrosion phenomenon originates from oxidizing species in the gaseous heat transporter that transit through open pores. Even in the case of a chemical oxidation, these measurements show that the isotopic ratio is effectively not always conserved. It can be observed that some cores contain less C-14 activity than should have produced by carbon activation alone. Therefore, apparently less stable carbon was oxidized concomitantly perhaps because of higher link with the matrix.

In the activation producing C-14, the influence of oxygen remains negligible compared with the influence of nitrogen and it was decided to adopt conservation of the oxygen/nitrogen ratio in the atmosphere.

Remember the principles adopted to remain conservative maximize the increase in the estimate for nitrogen. However, even this increased value is lower than British estimates that are not based on C-14 measurements but rather on nitrogen measurements made under conditions that appear to be significantly different from conditions in the reactor.

We suggest the following explanation:

- the surplus C-14 above the quantity derived from activation of the graphite matrix is interpreted as being the effect of activated occluded air (corresponding to simply adsorbed C-14).
- the deficit of C-14 below the quantity derived from activation of the graphite matrix is interpreted as being the consequence of radiolytic corrosion of the carbon matrix (which seems to match with the energy magnitude given to C-13 by the impact of the activation neutron) once the occluded air activated in C-14 is depleted. This latter assumption is adopted because it is the assumption that increases the average proportion of nitrogen. However, this is not a certainty but it is simply an assumption used to create an upper limit for nitrogen.
- unless other information is available and to remain conservative, the corrosion phenomenon is not considered and the resulting C-14 deficit is neither used nor generalized during the global calculation of the C-14 inventory.

Therefore the assumption that was chosen as being the most probable consists of postulating that radiolytic corrosion consumes C-14 originating from activation of nitrogen in priority, and then C-14 originating from activation of C-13.

Difference with the British

The “envelope estimate” nitrogen level on the four compared reactors (BUG1, CHA3, SLA1 and SLA2) does not significantly exceed 10 ppm despite the drastic methodological overestimate that has just been described and this value remains significantly lower than values announced by the British at the EPRI conference in LYON in October 2008. It was a lively debate at the EPRI Lyon meeting and further discussions occurred at the following EPRI conference in Hamburg in October 2009, but there is always no British publication of activated graphite C-14 measurements.

A priori, there would seem to be only two possible explanations for such a difference:

- (1) The physicochemical operating conditions in British and French graphite piles are sufficiently different (comparative compositions of real heat transporting gases).
- (2) The fact that the British made a direct measurement of nitrogen in the graphite and that the high level of this measurement is related to the ambient atmosphere more closely than to the genuine nitrogen in the graphite that is actually occluded, that had actually been activated and that remained in place in the graphite waste after activation despite its simple adsorbed form.

Without saying that assumption number 1 is impossible, explanation number 2 seems plausible because it can be demonstrated how the direct measurement of the impurity must be reserved for cases in which there are no RN measurements that can be used to obtain the explanatory impurity, for reasons related to the measurement precision.

An indirect measure is more judicious because it is based on real irradiation and can benefit from the precision possibilities using radiochemistry that is better than what is possible with chemistry.

A chemical measurement can be particularly penalizing, for example as happens when a direct chemical measurement of 1 ppm of cobalt in LD gives 40 times more Co-60 on Bugey 1 than an indirect evaluation through the identification of measured and calculated Co-60 (data assimilation method).

Nevertheless, considering the conditions of a direct measurement of nitrogen in graphite, the British approach probably quantifies a C-14 inventory close to what is produced in the graphite and that effectively originates almost exclusively from nitrogen. Nevertheless, there is nothing to confirm that this quantification corresponds to the final waste and measurements on the first four EDF reactors suggest the contrary. If the C-14 derived from nitrogen was produced in larger quantities (a plausible British assumption), this C-14 did not remain in place in the graphite in its final waste stage (demonstration provided by EDF measurements).

We will now consider the C-14 inventory in ultimate disposal, in other words what effectively remains in the graphite waste, namely the value estimated with the maximum conservatism due to our approach.

This disposal inventory no longer contains more C-14 than would correspond to:

- 100% of the carbon matrix (ignoring wearing-out) and
- an envelope (maximum) average nitrogen "explanatory impurity" equal to less than 10 ppm \pm 10%.

Remember that the term "explanatory impurity" (by activation) was defined above.

Assumptions adopted by EDF at the present time

The envisaged explanation for the 4 inventories is based on preferred elimination by radiolytic corrosion of C-14 derived from the following, in order,

- (1) firstly the transformation of adsorbed Nitrogen and Oxygen (weaker link),
- (2) then C-14 derived from C-13 (because of the delivered energy).

The latter, at the instant at which they are formed, could plausibly be in a chemically more activated state than the available C-12, that would increase the probability of occurrence of their radiolytic oxidation when adsorbed C-14 originating from N and O has been completely consumed.

In the 4 inventories (BUG1, SLA1, SLA2, CHA3) transmitted to French governmental disposal agency (ANDRA), the reasonable envelope choice consisted of "artificially" increasing "apparently aberrant" measurements so that they respected the minimum calculated from Carbon alone, so as to never show any "explanatory" Nitrogen with a negative fraction per unit mass and to ignore this no quantified wearing-out effect to avoid the risk of underestimating the inventory.

Unlike the flux and activation calculations based on known and universally shared laws of nuclear physics, the current model of radiolytic corrosion of graphite (even though it is recognized because it was used as a justification for Bugey 1 final shutdown) cannot provide an efficient and scientifically justified quantification.

Therefore, EDF deliberately overestimated the C-14 inventory to get around the problem of "apparently aberrant" C-14 measurements probably related to radiolytic corrosion of graphite under flux.

C-14 specifications and sanitary impact during disposal

The British did not hesitate to talk about the occurrence of an organic form of C-14 which appears to be important in quantification of the sanitary impact of C-14, although apparently without any rigorous and proven justification because they are apparently values measured on mixes in which organic waste competes with irradiated graphite.

In particular, a more detailed study of the link between C-14 and preferred radiolytic corrosion of C-14 could confirm that the best opportunity for C-14 to become organic (for example the formation of ^{14}CO or $^{14}\text{CH}_4$) occurs when it has definitively left the graphite during operation through the gaseous path due this corrosion.

The strong spatial uncorrelation of measurements of the different universally observed RN on EDF piles show that any organic depositions (some even envisage "organic-hydrogen-chlorine" deposits) do not contain any measurable RN. Therefore, there is no measurable re-deposition of C-14 removed by radiolytic corrosion that could become organic just at this time.

On the other hand in the extreme case, in other words beyond C-14 originating from adsorbed nitrogen and corresponding to "apparently aberrant" EDF measurements (measurements less than the C-14 originating from carbon alone), radiolytic corrosion could be the argument demonstrating that only the C-14 remaining in the waste is likely to be inorganic.

In this case, organic C-14 would be more specific to operation of the reactor.

CONCLUSION

This document contains a general summary of the fundamental mathematics on which the calculation of the radioactive inventory of graphite piles is based.

The very general nature of scientific and technical justifications of the approach related to neutron activation now make it possible to broaden it to include a general application to cases of metals and concretes.

Indeed, the results of data assimilation for the BUG1, CHA3, SLA1 and SLA2 reactors have confirmed that a typology of radioactive inventories of pile graphite related to their original coke is acceptable.

This data is in no way an input data for the method and this is why it could have an explanatory usefulness independently of this method.

In other words, although obtained by completely independent methods, this observation offers a coherent check of results drawn scientifically from radioactivity measurements of samples and the neutron flux map.

Finally, it provides a basis for a justified reconsideration of all a priori ideas on Chlorine 36 and Carbon 14 that are currently in the community of the different graphite waste stakeholders, both nationally and internationally.

Especially, the only need is RN measurement in a number large enough to apply the Central Limit Theorem and we can now consider in the four EDF graphite piles (BUG1, CHA3, SLA1 and SLA2):

- the large enough number of RN measurements has been reached by EDF,
- there is no R&D need for Cl-36 quantification and localization purposes,
- there is no more than 10 ppm Nitrogen explaining the C-14 EDF measurements.

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