Radionuclide Stochastic Distribution in the Irradiated Graphite - 15206

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ABSTRACT

In France, about 17,000 tons of irradiated graphite waste will be produced from the decommissioning of the six gas-cooled nuclear reactors operated by Electricité De France (EDF). Radionuclide measurements in irradiated graphite exhibit very high discrepancies especially when corresponding to precursors at trace level which is often the case for nuclear grade graphite. As evidenced by numerous studies nuclear graphite is a very pure material, however, it cannot be considered from an analytical viewpoint as a typical homogeneous material.

Geo-statistics do not give any notable results when the 3D neutron flux and power history are known. Without taking into account the very origin of the stochastic behavior of radionuclide, their properties may be assumed with the statistical properties which are used in the case of contaminated soils but which have no interest for irradiated graphite. At every scale, the huge discrepancy cannot be avoided but can be easily explained by Theory Of Sampling (TOS) of finely divided materials. The complexity of nuclear graphite is due to its high purity.

INTRODUCTION

EDF (Électricité De France) operated in France six gas-cooled reactors, all shutdown now for at least fifteen years. These reactors are of so-called in French, “UNGG” reactor type (Uranium Naturel Graphite Gaz – cf. Figure 1). They were graphite moderated, cooled by carbon dioxide and fuelled with natural metallic uranium. Due to the lack of uranium enrichment, graphite had a very high level of purity due to the necessity of the highest transparency to neutrons.

For the graphite waste producer (the reactors’ operator), assessing the radionuclide inventory of the graphite of UNGG reactors is needed because of final disposal. The inventory knowledge allows the computation of the repository sanitary impact during the future centuries and to decide if such a forecasted impact copes with the regulation. With the improvement of analysis techniques of analysis and the enhancement of data accessible through the computation of neutron flux, the radiological inventory of graphite can be calculated in order to be as close as possible to reality guaranteeing a conservative estimate.

This paper presents the stochastic phenomenon in measurements in order to give the best assessment of the radiological inventory based on large enough populations of samples.

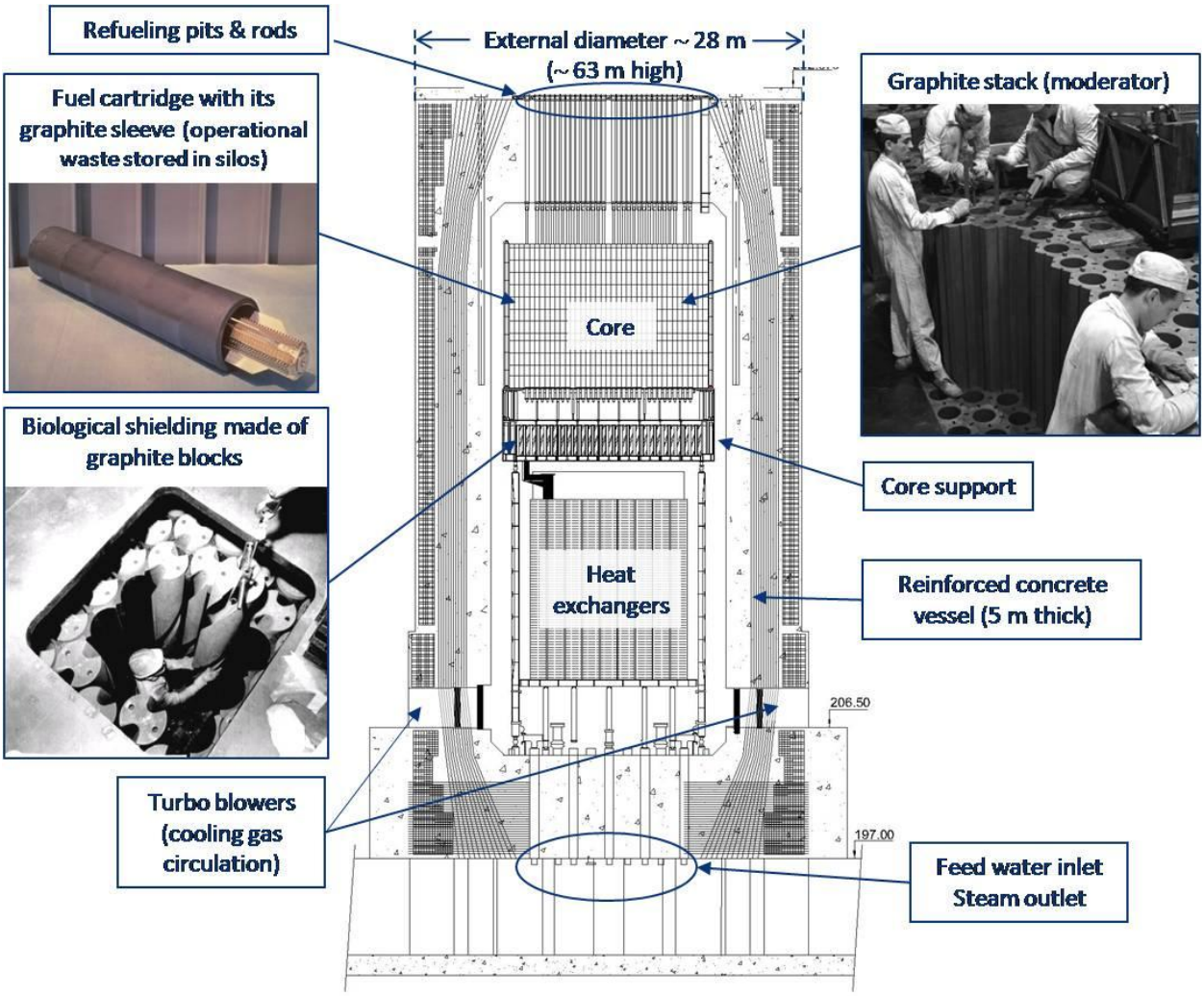

Fig. 1. Graphite in EDF UNGG reactors and transverse section diagram of an integrated vessel type reactor

EDF follows the methodology coupling modeling tools and radiochemical analysis results.

It was fully consistent with IAEA recommendation [1] for the assessment of a radiological inventory that “the result of any theoretical calculations should be compared with the data obtained by experiments to obtain a validation of the calculations”.

TRACES AND LACK OF SPATIAL CORRELATION

Computing Inverse Activation From Activity to Impurity

The particular sensitivity of radio-chemical measurement provides access to elements that would only yield only detection limits in chemical context. When in the presence of nuclear grade graphite, the level of impurities is not chemically measurable and only radioactivity measurement is likely to exceed the detection limit. To convert radio-chemical measurements in

chemical concentrations, it is necessary and sufficient to know the neutron flux at the location of the graphite sample and its history, thus allowing, by reverse activation calculation, the computation the values of explanatory chemical impurities, i.e. the values of chemical concentration which would explain the measured RN. The average chemical composition is precisely the input data used to compute radionuclide inventories of EDF graphite.

Impurities Spatial Correlation may only Happen with Deposits

Radioactivity is only coming from activation by the neutron flux and the spatial correlations of each radionuclide provide no information because the spatial distribution of neutron flux history is known and the link between impurities and radioactivity is determined at any sampling point, which is why the study of the spatial distribution can only focus on the explanatory concentrations inducing measured radionuclide.

To consider spatial correlations of these concentration levels, we must be aware that the pile was built from numerous thousands of bricks. Their position is coming from a draw between the fabrication and the pile. Obviously, it is impossible to know where exactly the brick is going to be placed in the stack. It is not rational to consider a spatial correlation of initial concentrations between two points that do not belong to the same brick. It shows that a variogram, a type of graph which illustrates the spatial correlation as a function of distance, can only make sense while explaining events of radionuclide migration during the operating period. We have just to take into account the shuffling of bricks which initial positions are naturally "randomized".

On chlorine for example, it can be shown that in the physical conditions of operation (temperature and pressure), the only physically possible migration is not inside the graphite but only escaping from chlorine to the outside coolant gas. In these circumstances, the only possible spatial correlation would correspond to a process of release/deposit via carbon dioxide gas.

As a coolant undergoing continuous mixing during at each cycle, it can be considered that all the channels are equivalent. The variogram must then use the vertical distance between all pairs of measurements of a given RN considered from a single channel (same CO_2). But, even with Gaussian anamorphosis to get a better variogram, a strong “nugget effect" is obtained which is incompatible with the vertical temperature gradient which is supposed to drive such a deposit according to the height in the channel. Remember that a 100% “nugget effect” represents the lack of any spatial correlation.

Even if such a deposit phenomenon seems to appear because it is possible to see a black deposit during TV inspections, its impact on measures of radioactivity is not significant. Even assuming the hypothesis of deposits, their impact remains undetectable in explaining spatial (geo-) statistic distributions.

GEO-STATISTICS BRING LITTLE FOR COMPUTING THE INVENTORY

Such contamination problems could be solved by taking into account methods based on correlation with respect to space called “geo-statistic”. Such methods which represent a kind of generalization of stochastic continuity with respect to space are not suitable with graphite. The very nature of the material and the known neutron flux history show that such methods are definitively useless for activated graphite when its neutron flux history is known.

Such a hypothesis is unfit because it would be the exact opposite of the “nugget effect” (Figure 2 which gives the proof of it) clearly observed in graphite measurement.

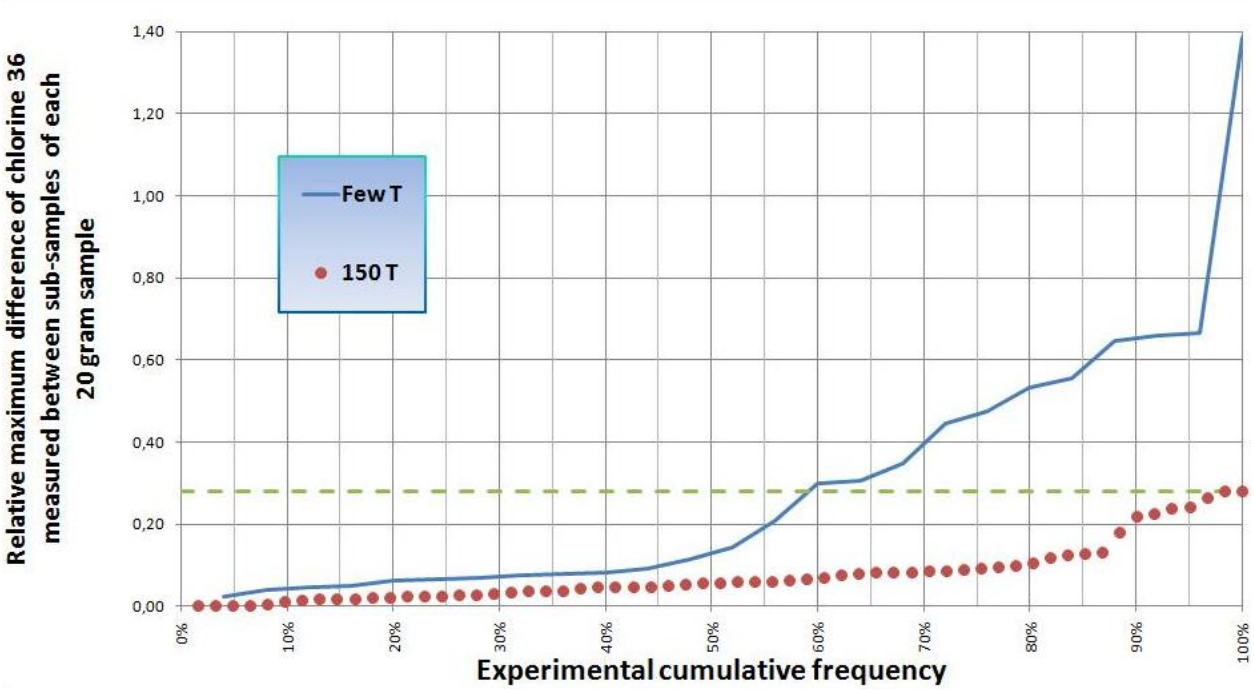

Fig. 2. Experimental cumulative density functions of relative maximum difference of Cl-36 measured between sub-samples for two laboratories with very different grindings

By definition, this effect is the very opposite of a meaningful deposit because it demonstrates the total lack of any correlation between the same radionuclide measured at two points even when very close together. We should remember that the global inventory of each RN is the only method to quantify the ratios between RN and that these ratios have nothing to do with correlation coefficients as this is possible only when in the presence of particularly simple contamination phenomena.

The links between RN and gamma measurement aren’t effective and it is non-sensical to hope that using the results of spatial distribution of activity like those of geo-statistics for the treatment of contaminated soils by restricting the remediation to specific areas. In the case of graphite pile waste, making such discrimination would be questionable, but even if such a selection was possible, it first would induce only additional costs compared to a systematic packing and measuring the gamma of the whole waste basket.

MEASUREMENT STOCHASTIC MODELING WITH POISSON LAW

Graphite without Taking Anything Else into Account

Considering only graphite, that is to say, without taking into account the stochastic effects from the process used to perform measurements on samples, we can assume it is characterized by a stochastic distribution of impurities, a random variable. To cope with continuous modeling, this

random variable will be an affine transformation of a spatially distributed “Poisson” random variable.

In probability theory, a Poisson point process is a particular kind of random process by which a set of isolated points are scattered about a three-dimensional space. The concept is named after the French mathematician Siméon Denis Poisson. The Poisson point process is characterized by the following properties:

- The numbers of isolated points falling within two regions A and B are independent random variables if A and B do not intersect each other;
- The expected number of isolated points falling within a region A is the measure of the region A. This "measure" is proportional to the volume of A. But the measure must be defined in such a way that the measure of the union of regions that do not intersect each other is simply the sum of their measures.

The explanation is given by representing graphite, a finely divided material, in the form of sufficiently small balls without impurity of interest on the one hand and made of impurity of interest on the other hand, wherein a sample of balls is drawn at random without replacement.

Given the number of balls, an "hypergeometric" drawing (without replacement) substantially coincides with a random "binomial" drawing (with replacement) which, by passing to the well-known limit, leads to the Poisson drawing of number of balls corresponding to the impurity of interest from the sample.

Taking into Account the Global Process of Measurement

The affine transformation of a spatially distributed “Poisson” RV seems to be a natural choice because Poisson random variable links between variance and average are fitting with Pierre Gy sampling law. Anyway, these elements are sufficient to compute the average value of each explanatory impurity, but we should admit that, if we want to have a fitting both for skewness and kurtosis, we need to use a more complex modeling to introduce the method of grinding, mixing and sampling and the way of measurement it-self. It is a matter of fact that the stochastic distribution of impurities in the graphite does not give the true explanation of the stochastic distribution of measures.

CONSEQUENCES ON “A PRIORI” IDEAS

The False Issue of RN Measurement Reproducibility

In order to assess the radiological inventory of the irradiated graphite, EDF uses radiochemical measurement on irradiated graphite sampled from the core of the reactors. For EDF reactors, according to the dimensions and geometry of the piles, a set of 20 to 30 irradiated graphite samples were in general analyzed by radiochemistry on the 200 cores taken on average per reactor pile. The radiochemical analyses were performed by CEA according to state of the art methods whose main principles are presented in an IAEA technical report on radiological characterization of nuclear reactors [1]. In the case of the RNs present in very small quantities such as chlorine 36, the reproducibility of the results has no interest.

The fundamental physical phenomenon that explains the clear impossibility of the reproducibility of the analyses for the elements present at trace level has nothing to do with the quality of the sampling and neither with the quality of analyses. The variability of the results is well known and completely impossible to avoid because it is inherent to sampling process of finely divided material. The performance of the measuring laboratory is also not at all in question. Sharing some samples of graphite for comparison purposes between different laboratories is very questionable. It is not possible to do it with that material even if it is completely acceptable for example with samples coming from the same casting phase of a metallic alloy. Concerning graphite, the differences are coming from the sampling.

If you send different graphite samples to the laboratories, first of all, you will compare the different samples and you will discover again the “Theory of Sampling”. Any “Round Robin test” would only be a kind of “coin toss”. It is a case of the variance that affects the measurement of a sample for representing a concentration (chemical content or specific activity) of a given batch, according, among other parameters, to the concentration, the mass of the sample or the mass of the batch. This phenomenon, which it will be attempted to explain briefly below, was in particular theorized by Pierre Gy. We should admit that such demonstration usually begins to upset the chemist, even the best, who is used to aqueous solutions, which obviously never present any sampling variability because of the liquid phase, exactly as what occurs with an alloy which was also a liquid phase during the casting.

The characteristic of the impurities of nuclear graphite is their trace proportion, for reasons that are fully understandable related to the neutron performances necessary for obtaining criticality despite the use of “non-enriched” uranium (UNGG).

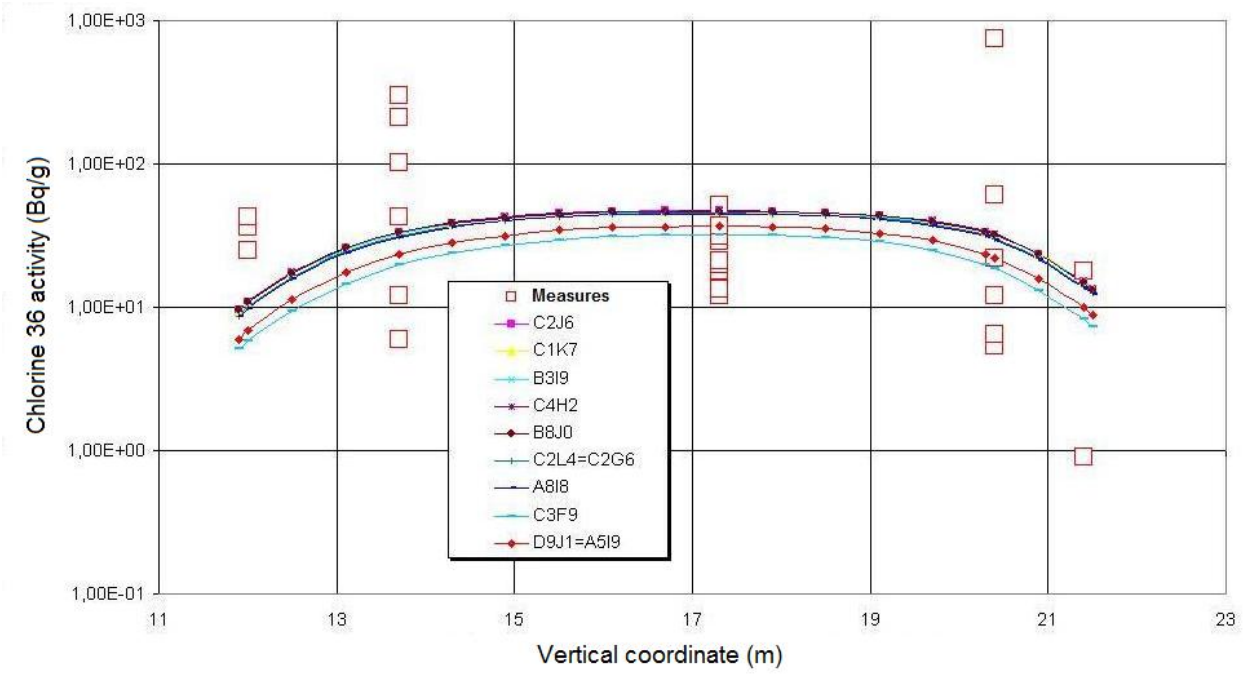

Fig. 3. Average calculated values (in Bq/g) of chlorine 36 for the graphite of some fuel channels in the Bugey 1 graphite pile (squares represent measurements - x-axis: height of the pile). The average value is 20 ppb of chlorine 36.

The False Issue of RN Measurement Spatial Variability

This very great variability observed to the scale of a pile cannot be explained by the variability of the neutron flux, giving rise to the production of chlorine 36. Logarithmic Figure 1 squares reproduce the dispersion of the chlorine 36 measurements in the samples from the Bugey 1 reactor according to their sampling point in the graphite pile. The link with the purity and the sampling size is coming from "Theory of Sampling" by P. Gy [2] as explained above.

Without taking into account the fact that a difference of level of activation between the sampled bricks which explains no more than 1 order of magnitude, it appears impossible to get representative and reproducible measurement to the scale of an entire pile. This finding shows that for establishing the radionuclide inventory of the irradiated graphite in the piles, an approach based on a statistical tool through the calculation of a mean of several measurements is a necessity.

FIRST RESULTS FOR FOUR PILES

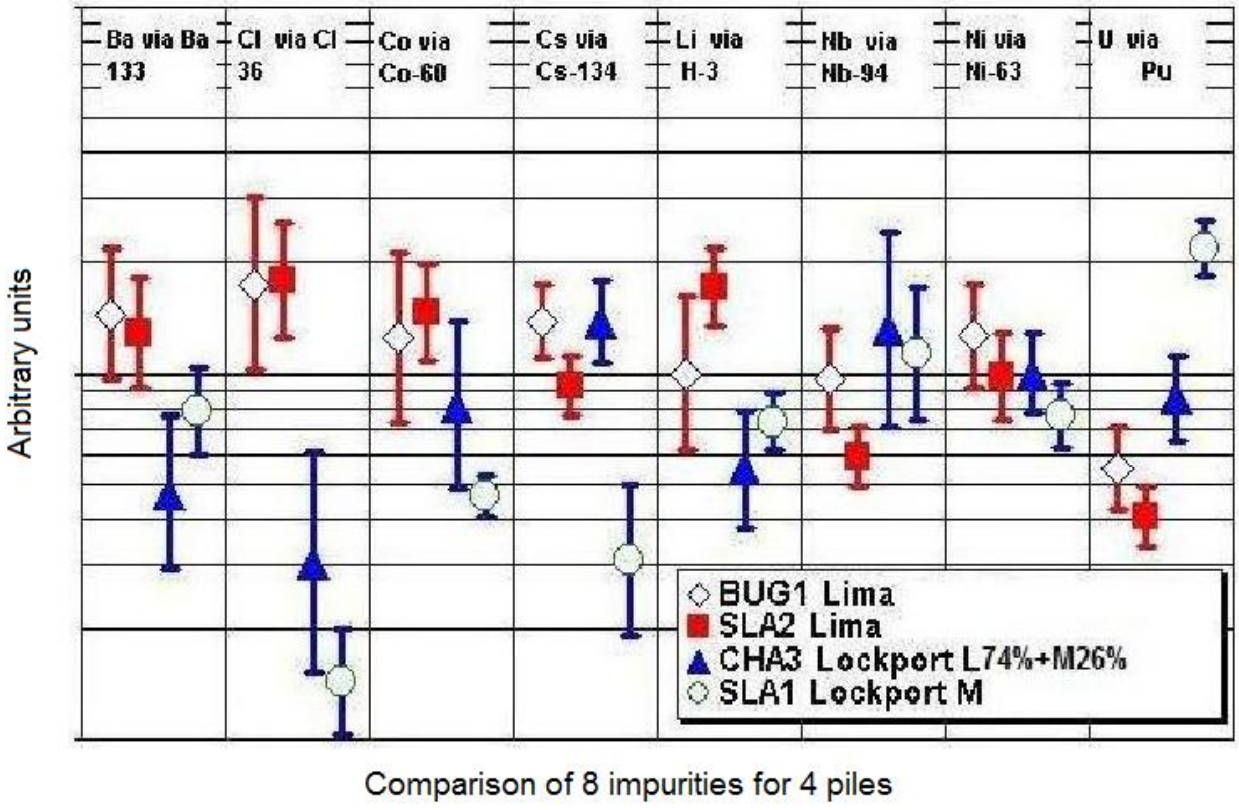

Fig. 4. Arbitrary unit 95% confidence interval of Ba, Cl, Co, Cs, Li, Nb, Ni and U impurities adjusted for Bugey 1, St-Laurent A2, Chinon A3 and St-Lauren A1 piles

The use of a mean value is in fact the only method that makes it possible to divide the variance observed by the number of measurements and is reliable, whereas the extrema (minima and maxima) represent by definition singular measurements very far from average reality because we know the pure random origin of the observed variability.

Figure 4 shows the results of EDF method for 4 piles with “confidence intervals” that illustrates the very stochastic nature of the inventory assessment. The other interest of Figure 4 is also the re-discovery of cokes at the origin of the graphite of the 4 piles because the confidence intervals corresponding to the same coke are very close. Figure 4 is also a notable demonstration of the quality of results and that stochastic modeling is the efficient way to study graphite.

CONCLUSIONS

Anyway, with only one sample and only one measurement, you never have any standard deviation of radionuclide measurement. It is the reason why, when you don’t accept to take into account any discrepancy, you only use a single measurement. Anyhow, the quality of the corresponding results remains questionable because a single other measurement appears to be very different, even sampled very closely from the first one. This lack of correlation with respect to space is always the “nugget effect”. Stochastic behavior modeling of graphite and measurement improves the understanding of populations of analysis in order to compute better radionuclide inventories. Assessing now a more precise and lower level of ^{36}Cl may open a type of a repository which could be less expensive than deep geological disposal for EDF graphite.

REFERENCES

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[2] Gy P, (1992) Sampling of heterogeneous and dynamic material systems, theories of heterogeneity, sampling and homogeneizing. Elsevier, Amsterdam, 653p