CHARACTERIZATION OF GRAPHITE SLEEVES FROM BUGEY 1 EDF PLANT FOR PERMANENT DISPOSAL – MESUREMENT AND CALCULATION OF SCALING FACTORS FOR DIFFICULT-TO-MEASURE NUCLIDES

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ABSTRACT

Electricité De France's Bugey-1 reactor, with graphite moderator, was shutdown permanently in 1994. The natural uranium elements are encased in graphite sleeves to facilitate handling. 2,000 m^3 of concrete containers, containing non conditioned graphite sleeves, must be characterized and conditioned before shipment to the national repository site called "Centre de l'Aube".

The characterization work consists in quantifying Difficult-To-Measure nuclides (DTM) by the use of Scaling Factors (SF), which use Co-60 as tracer. Bugey developed an industrial method for the gamma counting of each package to perform easily and rapidly the measurement of the Co-60 content.

Depending upon the DTM radionuclide, Co-60 scaling factors are determined, or by measurement on graphite samples (case of C-14, Cl-36, Ni-63, H-3), either by using a calculation technique which is based upon the impurities present in the graphite sleeves.

This method is applied for the other pure beta emitters all DTM radionuclides :

Ag-108m, Be-10, Ca-41, Cd-109, Cd-113m, Co-57, Cs-135, Cs-137, Eu-155, Fe-55, Gd-153, Mo-93, Nb-93m, Nb-94, Ni-59, Pd-107, Pm-147, Sm-151, Sn-119m, Sn-121m, Sn-126, Sr-90, Tc-99, V-49 and Zr-93.

Calculations use six sleeve history cases : 1 year at 50% power, 2 years at 50 % power, 3 years at 50 % power, 4 years at 50 % power, 1 year at 100 % power and 2 years at 100 % power. The DTM nuclides have been calculated from impurity concentrations for each of these six cases, and the greatest scaling factor has been kept.

The calculation is based upon two impurity sets:

- First impurity set : a reverse activation calculation provides us with the best estimate value of impurities calculated from the measured mean gamma spectrum and from measured scaling factors. It consists in solving a system of simultaneous equations for the impurities as a function of the mean gamma radioactive spectrum and of the measured scaling factors. The concerned calculated impurities are Co, Cl, Li, Ag, Cs, Eu, Fe, Ni, Sb, Sc, Zn and Sn.

- Second impurity set: The other impurities which were not available by this reverse calculation are originated from the greatest value, which has been measured in the graphite and sometimes by using the detection limit.

This method allows us to avoid some detection limit problems and statistical weaknesses. It gets better, cheaper and faster characterization by mixing easy gamma spectrum measurement and simple linear calculation.

INTRODUCTION AND GENERAL PROBLEM

General problem of waste characterization

The general purpose of disposal management is to stay below acceptable exposure to the critical group. This critical group is a group of members of the public whose exposure to radiation is typical of individuals receiving the highest dose coming from the disposal in the future.

Such a calculation uses, for each nuclide, the most pessimistic way by dispersion through the system of engineered and natural barriers to its critical pathway. For a given disposal, such a safety study provides the facility with the global radioactivity limit for each nuclide having a sanitary impact.

It is the reason why nuclear waste producer needs to declare the activity of numerous nuclides shipped to the repository. If the producer systematically increases the declaration of several orders of magnitude for simplification purposes, it accelerates disposal permanent closure because of activity completion.

As confinement gives decreasing time to short lived nuclides, characterization of long lived nuclides is one of the main needs of waste producer. Knowledge of most of long lived nuclides is especially important for the repository safety, but still most of them are DTM nuclides.

Waste characterization is usually performed by two general types of method:

1. activity measurement
2. activation calculation from impurities measurement.

DTM nuclides are so called because the first type of method, activity measurement, is much more complicated than direct gamma spectrum measurement. It may give only a simple detection limit if the level remains below the used device accuracy.

Concerning the other type of method, impurities are often of a very low level, for example, they may be measured in part per billion (ppb) and still provide an high activity by activation because of their very large cross section. Then impurities are often too small even for chemistry best analysis methods.

Both radioactivity and impurity measurements are performed on small samples and may increase the measured value because of device detection limits. But, even when they exceed device detection limits, they both need a wide sampling to give a significant result from a statistical point of view, and that always increases their cost.

This paper describes a practical method which has been used to Bugey 1 plant graphite sleeves characterization in order to avoid some detection limit problems and statistical weaknesses.

Graphite sleeves waste

The graphite sleeves are essentially graphite tubes, which hold the GGR fuel elements in place and enable them to be handled. The sleeves are removed together with the spent fuel they contain, and the spent fuel is then removed from the sleeves for reprocessing.

As far as Bugey 1 EDF NPP was concerned, the irradiated sleeves were not stored in a silo on site (which is the case for St-Laurent A EDF GGR NPP). They were conditioned in concrete containers to be shipped to ANDRA (Government Agency for Nuclear Waste Disposal) shallow disposal. From the beginning of

Bugey 1 operation, this disposal was “Centre de la Manche”, near Cherbourg, which has been closed and replaced by “Centre de l'Aube”, near Troyes.

Difficult-To-Measure nuclides

Even if the nuclear grade graphite has been especially purified in order not to absorb neutrons, there were still enough impurities left to produce by activation numerous difficult-to-measure (DTM) nuclides.

The long-lasting nature and toxicity of some of these DTM nuclides have received in France particular attention relative to their disposal.

Disposal by shallow burial “Centre de l'Aube” has been accepted by ANDRA only for the last 300 metric tons of Bugey 1 sleeves (i.e. 460 concrete containers) and scaling factors (SF) have been determined for them to compute all nuclides activities with only gamma counting of each package.

Main purpose was to cope with the DTM nuclides limitation by the regulatory body (DGSNR – French Nuclear Safety Governmental Authority) in the ANDRA shallow ground disposal. Within the frame of disposal safety analysis, Cl-36 is at the moment one of the most critical DTM nuclide.

ACTIVITY EVALUATION

Gamma spectrum

170 gamma spectrum coming from several hours of measurement with GeHP detector in front of each large side of 85 out of 460 containers, allow to compute a mean gamma spectrum of graphite sleeves at discharging time.

Co-60 (25,8%), Sb-125 (1,27%), Cs-134 (1,77%), Ce-144 (30,5%), Eu-154 (0,32%), Ru-106 (3,38%), Na-22 (0,23%), Eu-152 (0,039%), Zn-65 (3,97%), Mn-54 (1,09%), Ag-110m (0,61%), Ta-182 (0,18%), Sc-46 (11,2%), Co-58 (11,7%), Sb-124 (1,5%) and Fe-59 (6,44%).

There was no direct information about the mean burn-up corresponding to each container.

As Bugey 1 NPP shut down definitively in 1994, Co-60 remains the only gamma emitter which is greater than its detection limit after year 2000. Thus, Co-60 has been naturally the only possible gamma tracer and the DTM SF have been determined with respect to Co-60.

From sleeve spectrum to container counting

The several hour gamma spectrum HPGe detector measurement of concrete container was calibrated by individual measurement of each sleeve. Then, 170 gamma spectrum of containers allowed to calibrate a gamma counting “gate” with large detectors which are also used to control radioactivity of vehicles.

At last, it gave a efficient industrial method to measure the Co-60 activity of 20 concrete containers a day.

Cobalt impurity evaluation

The burn-up distribution of the 380 concrete containers corresponding to the last core fuel elements was globally described in the last fuel shipping sheets. By two ascending sorts, a one to one relation between burn-up and Co-60 activity has been established. Such a method gives the minimum standard deviation evaluation of cobalt concentration distribution.

Mean cobalt concentration computation for each container yields an average of 3.5 ppb and a standard deviation of 2.5 ppb is shown by Figure n°1. This standard deviation has been computed with 634 kg graphite samples (i. e. the containers).

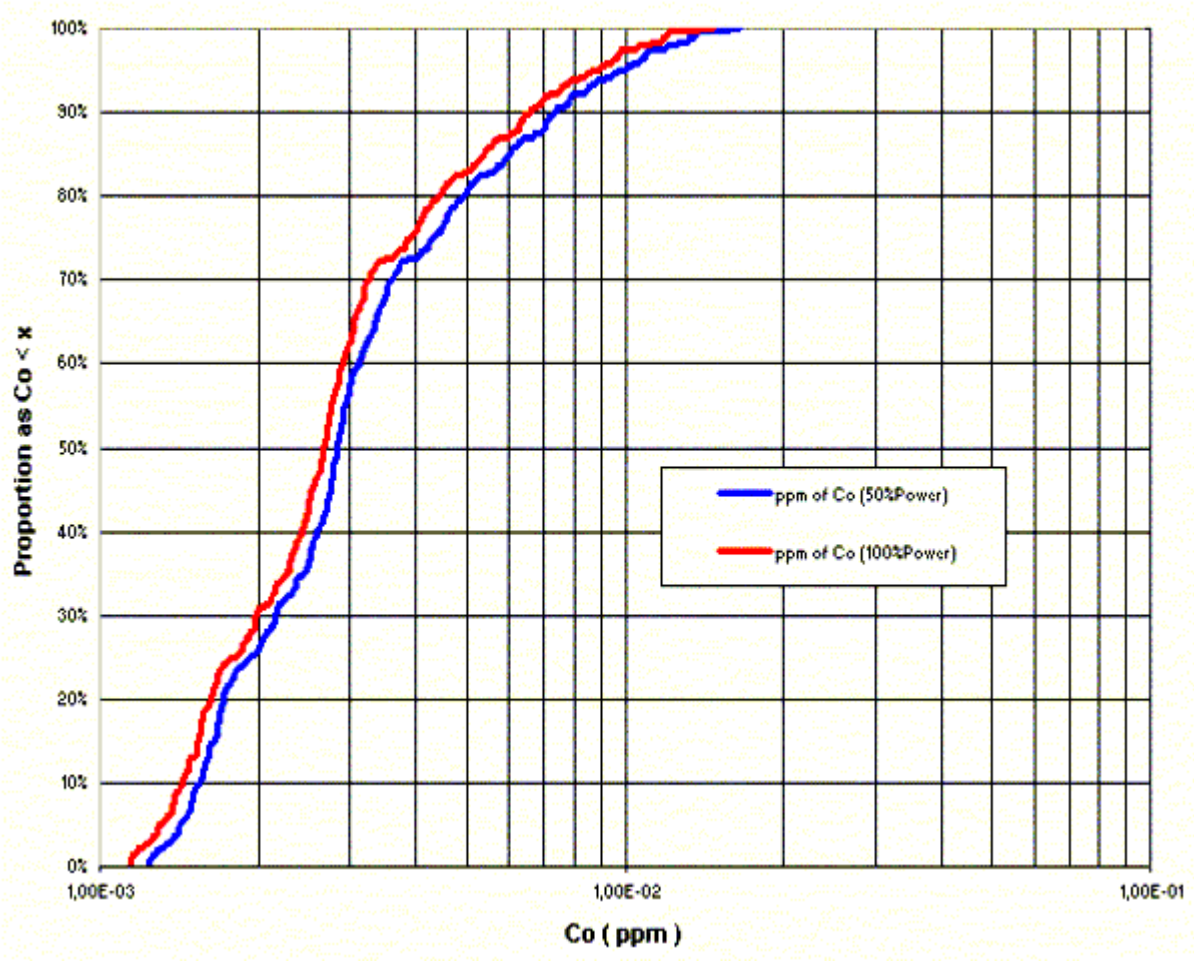

Figure n°1:
Statistical repartition of ppm values for the last core discharged. Average of 3.5 ppb and a standard deviation of 2.5 ppb for a population of 380 samples of 634 kg graphite.
N.B. : 100%Power curve is slightly above 50%Power and a mean value has been chosen

From container to radiochemistry sample

Standard deviation corresponding to 26 g radio-chemistry samples is 156 times greater. It is naturally the square root of the ratio of the sample masses.

As far as graphite cobalt concentration of about 26 g samples is concerned, standard deviation upon mean value, i.e. “relative error at one standard deviation” is then greater than 100.

Measured scaling factors

We completed information from the mean gamma spectrum with several radio-chemical analysis of graphite samples to estimate the following SF at discharging time :

H-3/Co-60 = 13, C-14/Co-60 = 0.8, Cl-36/Co-60 = 0.1, Ni-63/Co-60 = 0.5,

With ANDRA agreement, these SF have been estimated at a bigger than average level which is reasonably high for storage safety purposes.

Scaling factors uncertainty

Let B, the graphite burn-up. B is between 0 and $B_{max.}$
Let C_j the number of ppm of impurity j. Let C_o those of cobalt.
Let R_i the nuclide i scaling factor relative to Co-60.
Let n_i be the number of impurity giving nuclide i radioactivity.

We can compute an estimation of SF relative uncertainty (see appendix 1).

$|\Delta C_0/C_0| > 100$ being a result from chapter above, $|\Delta B/B_{max}| < 1$ and (see appendix 1) $n_i < 4$ being known, if we use equation 17 from appendix 1, we can see that maximum contribution of burn-up to the scaling factor uncertainty is less than 1%.

Radiochemistry samples mass make the SF more than 100 times more dependent on cobalt concentration variability than on burn-up variability. Thus, measured SF have been considered valuable enough for all neutron flux histories.

SCALING FACTOR COMPUTATION

Linear activation computation

We used measured data to compute the maximum value of the others DTM SF for different neutron flux histories as 1, 2, 3 or 4 years at 50% power and 1 or 2 years at 100% power (cases 1 to 6).

These 6 cases approximately cover the possible history because of the limitation of time in core and because of maximum burn-up limitation of GCR fuel. This even includes the low burn-up fuel case actually occurring at the last discharging of the core.

If we perform an activation computation with a given impurity vector V_1 (respectively V_2), we get a radioactive nuclides vector W_1 (respectively W_2). We intuitively understand that the computation performed with impurity vector $V_1 + V_2$ will give radioactive nuclides vector $W_1 + W_2$. We also intuitively understand that the computation performed with impurity vector $\lambda.V_1$ will give radioactive nuclides vector $\lambda.W_1$.

The function which computes activated nuclides vector from impurity vector is then linear. That explains the linear equation presentation of appendix 2 which sums up activation computations for 2 cases.

Activation reverse calculation

To compute the maximum reached by the SF, we need to take the minimum of activity for Co-60 which is at the denominator.

The high variability of C_o (cobalt concentration in ppm) would lead us to zero to reach the absolute minimum but it makes the SF difficult to reasonably quantify ! It is the reason why we used a global evaluation of minimum mean cobalt concentration from mean radioactive measurements.

Assuming that the scaling factor C-14/Co-60 is known (because its comes from a measurement), we should thus take the minimum of activity for C-14, which implies for oxygen and nitrogen to be zero (O = N = 0) because the quantity of carbon in graphite is given ($C = 10^6$ ppm).

We can verify that Be-10/Co-60 remains maximum even if oxygen and nitrogen give Be-10 by activation (this nuclide is the only one in this case).

Let N = O = 0 to minimize Co-60. Let in graphite $C = 10^6$ ppm.
From chemical measurement, we have the values of other impurities: Cd = 0.1 ppm, K = 1 ppm, Ba = 0.9 ppm, Gd = 0.1 ppm, B = 1 ppm, Be = 0.1 ppm and Ca = 5 ppm.

C-14 (function of O, N and C) is computed from N = O = 0 and $C = 10^6$ ppm.
Co-60 is given by the measured SF : C-14/Co-60.

Co-60 being known, note that 12 impurities can bee computed for each case of neutron flux history. It is simply a system of 12 linear equations in 12 unknown which allows us to determine Co, Li, Sb, Sn, Ni, Fe, Zn, Ag, Cl, Eu, Sc and Cs.

Table 1 : Calculated intermediary impurities

Impurities	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6
Ag (ppm)	1.85E-04	3.00E-04	4.54E-04	6.50E-04	2.04E-04	3.65E-04
Cl (ppm)	9.64E+00	9.77E+00	9.90E+00	1.00E+01	9.77E+00	1.00E+01
Cs (ppm)	1.02E-04	1.25E-04	1.51E-04	1.81E-04	1.07E-04	1.38E-04
Eu (ppm)	3.74E-05	5.96E-05	9.21E-05	1.38E-04	5.77E-05	1.32E-04
Fe (ppm)	8.04E-01	1.12E+00	1.47E+00	1.87E+00	8.06E-01	1.12E+00
Li (ppm)	5.88E-03	7.48E-03	9.37E-03	1.16E-02	7.24E-03	1.08E-02
Ni (ppm)	4.47E-01	8.94E-01	1.34E+00	1.80E+00	6.42E-01	1.29E+00
Sb (ppm)	2.23E-04	4.42E-04	6.67E-04	8.95E-04	2.23E-04	4.45E-04
Sc (ppm)	2.63E-04	4.82E-04	7.11E-04	9.45E-04	2.51E-04	4.38E-04
Zn (ppm)	7.65E-03	1.14E-02	1.57E-02	2.04E-02	7.76E-03	1.17E-02
Co (ppm)	2.32E-03	2.50E-03	2.68E-03	2.88E-03	2.34E-03	2.54E-03
Sn (ppm)	1.22E-01	1.37E-01	1.54E-01	1.72E-01	1.22E-01	1.38E-01

These estimated impurities are different because the neutron flux history are different.

Cobalt impurity global estimations of 3.5 ppb is greater than these 6 estimations, which give an idea of the effect of N = O = 0 hypothesis.

Direct activation calculation

Together with these 12 impurities, let us take the maximum values measured for Mo, Nb, Ru, Sm et Zr, even sometimes their chemical detection limit if no better information is available.

Let Mo = 0.2 ppm, Nb = 0.1 ppm , Ru = 0.1 ppm, Sm = 0.003 ppm and Zr = 0.1 ppm.

N.B. : Sm has been measured 20 years ago with activation techniques, which allow to reach the ppb.

This information allows us to compute, again for each of the 6 cases, now in a direct way, the activation and the other scaling factors. See table 2:

Table 2 : Computed scaling factors

Scaling factors	Maximum	Case 1	Case 2	Case 3	Case 4	Case 5	Case 6
Ag-108m/Co-60	1.00E-05	3.30E-06	5.10E-06	7.35E-06	1.00E-05	3.48E-06	5.66E-06
Be-10/Co-60	8.26E-04	8.25E-04	8.26E-04	8.26E-04	8.26E-04	8.25E-04	8.26E-04
Ca-41/Co-60	1.64E-03	1.64E-03	1.64E-03	1.64E-03	1.64E-03	1.64E-03	1.64E-03
Cd-109/Co-60	5.44E-02	5.44E-02	4.15E-02	3.28E-02	2.69E-02	5.21E-02	3.86E-02
Cd-113m/Co-60	1.75E-03	1.75E-03	8.77E-04	5.88E-04	4.43E-04	8.79E-04	4.44E-04
Co-57/Co-60	1.49E-03	7.50E-04	1.04E-03	1.16E-03	1.20E-03	1.07E-03	1.49E-03
Cs-135/Co-60	1.09E-08	2.28E-09	4.85E-09	7.74E-09	1.09E-08	4.61E-09	9.99E-09
Cs-137/Co-60	2.00E-07	2.00E-07	1.98E-07	1.96E-07	1.94E-07	2.00E-07	1.99E-07
Eu-155/Co-60	7.62E-03	4.90E-03	6.51E-03	7.13E-03	7.40E-03	6.65E-03	7.62E-03
Fe-55/Co-60	3.52E+00	2.13E+00	2.63E+00	3.10E+00	3.52E+00	2.14E+00	2.64E+00
Gd-153/Co-60	4.37E-01	4.37E-01	1.27E-01	4.95E-02	2.17E-02	1.35E-01	2.29E-02
Mo-93/Co-60	3.10E-05	3.10E-05	3.09E-05	3.08E-05	3.07E-05	3.09E-05	3.07E-05
Nb-93m/Co-60	2.32E-06	6.24E-07	1.21E-06	1.77E-06	2.32E-06	6.86E-07	1.31E-06
Nb-94/Co-60	3.63E-04	3.63E-04	3.61E-04	3.59E-04	3.56E-04	3.61E-04	3.56E-04
Ni-59/Co-60	3.91E-03	1.05E-03	2.05E-03	3.00E-03	3.91E-03	1.47E-03	2.81E-03
Pd-107/Co-60	3.51E-13	1.72E-13	2.17E-13	2.77E-13	3.51E-13	1.78E-13	2.39E-13
Pm-147/Co-60	3.14E-08	3.14E-08	2.49E-08	2.00E-08	1.62E-08	2.80E-08	2.00E-08
Sm-151/Co-60	2.89E-03	2.89E-03	1.36E-03	8.52E-04	6.01E-04	1.36E-03	6.01E-04
Sn-119m/Co-60	3.61E-02	3.60E-02	2.89E-02	2.43E-02	2.13E-02	3.61E-02	2.90E-02
Sn-121m/Co-60	5.97E-05	4.37E-05	4.87E-05	5.40E-05	5.97E-05	4.36E-05	4.85E-05
Sn-126/Co-60	4.38E-12	1.91E-12	2.19E-12	2.47E-12	2.76E-12	3.82E-12	4.38E-12
Sr-90/Co-60	1.88E-11	4.75E-12	9.42E-12	1.40E-11	1.85E-11	9.49E-12	1.88E-11
Tc-99/Co-60	5.94E-06	5.94E-06	5.91E-06	5.85E-06	5.80E-06	5.87E-06	5.78E-06
V-49/Co-60	4.43E-15	2.05E-15	2.21E-15	2.27E-15	2.31E-15	4.10E-15	4.43E-15
Zr-93/Co-60	1.15E-07	1.15E-07	1.15E-07	1.15E-07	1.15E-07	1.15E-07	1.15E-07

CONCLUSION AND LESSONS LEARNED

Better and cheaper characterization method

Such a method which consists in coupling radionuclide measurements with “reverse” and “direct” activation computation gives a powerful tool to avoid too pessimistic overestimations of DTM nuclides.

It is a way to replace the maximum of radio-chemistry measurement on small samples. Their poor measurement accuracy and statistic reliability lead to increase the radioactivity values for safety reasons.

The method replaces them by mean gamma spectrum (with as many short lived as possible) which measurement accuracy and statistic reliability are much better. Present quality of neutron flux and activation computer codes allows us to compute the maximum of impurities from the mean gamma spectrum.

Obviously, gamma spectrum measurement and activation computation are always cheaper than all the operations connected to the radio-chemistry analysis of a selection of samples.

Especially for activated dismantling waste management, all you have seen so far shows that you should, to put it simply and shortly:

1. First of all, measure the gamma spectrum of a rather large selection of each type of waste within the first year after plant operation is over in order to get a spectrum of good statistic value and including short lived.
2. Then calculate, for the plant neutron flux history, each nuclide activation equation as a function of impurities.
3. Then compute the impurity set which satisfies these equations, the gamma spectrum giving the second hand of the system of simultaneous equations of gamma emitters.
4. And finally, compute with the impurities, all the others DMT-nuclides with the rest of activation equations.

This method mixes measurement and calculation to get better, cheaper and faster characterization

Large variability of cobalt concentration

The very large variability of cobalt concentration in graphite has been proved. DTM SF measurement is more than 100 times more dependent on cobalt concentration variability than on burn-up variability. As far as graphite is concerned, measuring different burn-up samples to estimate DTM SF is of low interest.

For example, in case of Bugey 1, we could reasonably spare money, time and dose by using several samples from the same sleeve to measure C-14, Cl-36, Ni-63, H-3 and tracer Co-60 to get SF.

Practical consequences

French Ministry (DGSNR) has given allowance for storing maximum 0.4 TBq of Cl-36 on “Centre de l'Aube” repository, which exactly corresponded to the Bugey 1 graphite sleeves need. Then, ANDRA accepted the presented DTM SF calculation with tracer Co-60 and gave a shallow disposal agreement.

A few months later, ANDRA accepted the new measurement method by gamma counting of Co-60 which has been authorized later on by DGSNR before any mortar injection. The mortar injection on Bugey site has been authorized in January 2002 after a safety study and the beginning of shipment to ANDRA disposal began at the end of 2002.

APPENDIX 1 – SCALING FACTOR UNCERTAINTY CALCULATION

- Let B the sleeve burn-up. B is between 0 and B_{max} and let $X = B/B_{max}$.
- Let C_j (sub j) the number of ppm of initial impurity j activated by neutron flux.
- Let A_i (sub i), disregarding the third order and following, nuclide i mass activity at the last shut down.

We obtain :

$$A_i = \sum_j P_{ij} \qquad \text{(Eq. 1)}$$

$$P_{ij} = K_{ij}\left(X - \alpha_{ij} X^2\right) C_j \qquad \text{(Eq. 2)}$$

- K_{ij} is a positive constant.

- Parameter α_{ij} is positive and it is decreasing with nuclide i half-live because of it radioactive decay which occurs at the same time as creation by activation.

As our nuclides of interest are Co-60, ^{3}H, ^{36}Cl, ^{14}C and ^{63}Ni, maximum parameter $\alpha_{ij} = \alpha_{00}$ is those of Co-60 (i = 0) (it is the shorter half-life) and from impurity Co activation (j = 0) (other impurities giving Co-60 are negligible).

- $\alpha_{00} < 1/3$ as we can see it by plotting Co-60 GBq/kg per cobalt ppm against burn-up for 50% and 100% core power as shown by Figure n°2.

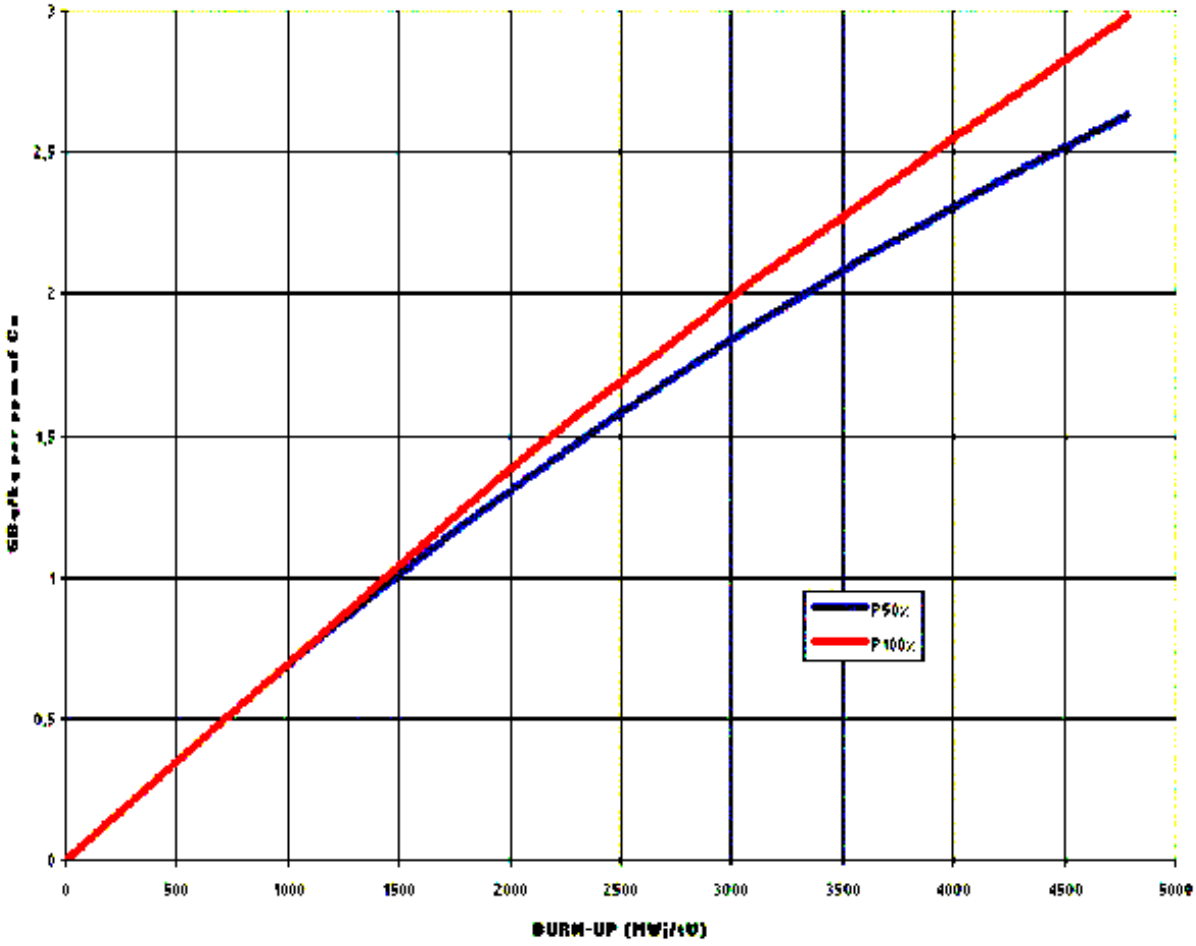

Figure n°2:
Co-60 activity in GBq/kg/ppm of cobalt versus burn-up (MWd/t_U) for 50% and 100% power
100% curve is above 50% because, for the same burn-up, the time of decreasing is smaller

For Co-60 specific case, we obtain :

$$A_0 = P_{00} = K_{00}\left(X - \alpha_{00}X^2\right)C_0 \qquad \text{(Eq. 3)}$$

because only cobalt has a significant influence. Fe and Ni are negligible (see appendix 2).
N.B. : When half-live tends to infinity, α_{ij} parameter tends to zero.

- Let nuclide i scaling factor with Co-60, $R_i = A_i/A_0$.
- With $R_{ij} = Pij/A_0$, we obtain :

$$R_i = \sum_j R_{ij} \qquad \text{(Eq. 4)}$$

Partial derivative of $\ln(R_{ij})$ with respect to X is between 0 and ½.

As

$$R_{ij} = \frac{K_{ij}}{K_{00}} \cdot \frac{1 - \alpha_{ij} X}{1 - \alpha_{00} X} \cdot \frac{C_j}{C_0} \qquad \text{(Eq. 5)}$$

Because $\alpha_{ij} \le \alpha_{00}$

$$\frac{\partial}{\partial X}\left(\ln\ R_{ij}\right) = \frac{\alpha_{00}}{1 - \alpha_{00} X} - \frac{\alpha_{ij}}{1 - \alpha_{ij} X} \ge 0 \qquad \text{(Eq. 6)}$$

Because $\alpha_{00} \le 1/3$ (see above)

$$\frac{\partial}{\partial X}\left(\ln\ R_{ij}\right) \le \frac{\alpha_{00}}{1 - \alpha_{00} X} \le \frac{1}{2} \qquad \text{(Eq. 7)}$$

We finally obtain:

$$0 \le \frac{\partial}{\partial X}\left(\ln\ R_{ij}\right) \le \frac{1}{2} \qquad \text{(Eq. 8)}$$

X and Cj being independent, we obtain classically R_{ij} relative error with $0 \le \varepsilon_{ij} \le ½$:

$$\left|\frac{\Delta R_{ij}}{R_{ij}}\right|^2 = \varepsilon_{ij}^2 \left|\Delta X\right|^2 + \left|\frac{\Delta C_0}{C_0}\right|^2 + \left|\frac{\Delta C_j}{C_j}\right|^2 \qquad \text{(Eq. 9)}$$

Thus

$$\left|\Delta R_{ij}\right|^2 = \left|\frac{P_{ij}}{A_0}\right|^2 \cdot \left[\varepsilon_{ij}^2 \left|\Delta X\right|^2 + \left|\frac{\Delta C_0}{C_0}\right|^2 + \left|\frac{\Delta C_j}{C_j}\right|^2\right] \qquad \text{(Eq. 10)}$$

We then get :

$$\left|A_0 \times \Delta R_i\right|^2 = \left|A_0\right|^2 \sum_j \left|\Delta R_{ij}\right|^2 = \sum_j \left\{P_{ij}^2 \left[\varepsilon_{ij}^2 \left|\Delta X\right|^2 + \left|\frac{\Delta C_0}{C_0}\right|^2 + \left|\frac{\Delta C_j}{C_j}\right|^2\right]\right\} \qquad \text{(Eq. 11)}$$

Which may bee written :

$$\left|A_0 \times \Delta R_i\right|^2 = \left|\Delta X\right|^2 \left[\sum_j P_{ij}^2\ \varepsilon_{ij}^2\right] + \left|\frac{\Delta C_0}{C_0}\right|^2 \left[\sum_j P_{ij}^2\right] + \sum_j P_{ij}^2 \left|\frac{\Delta C_j}{C_j}\right|^2 \qquad \text{(Eq. 12)}$$

Hence :

$$\left|\frac{\Delta R_i}{R_i}\right|^2 = \frac{\sum_j P_{ij}^2\ \varepsilon_{ij}^2}{\left[\sum_j P_{ij}\right]^2} \cdot \left|\Delta X\right|^2 + \frac{\sum_j P_{ij}^2}{\left[\sum_j P_{ij}\right]^2} \cdot \left|\frac{\Delta C_0}{C_0}\right|^2 + \frac{\sum_j P_{ij}^2 \left|\frac{\Delta C_j}{C_j}\right|^2}{\left[\sum_j P_{ij}\right]^2} \qquad \text{(Eq. 13)}$$

$$\frac{\sum_j P_{ij}^2\ \varepsilon_{ij}^2}{\left[\sum_j P_{ij}\right]^2} \le \frac{\sum_j P_{ij}^2\ \varepsilon_{ij}^2}{\sum_j P_{ij}^2} \le \frac{1}{4} \qquad \text{(Eq. 14)}$$

$$\frac{1}{n_i} \le \frac{\sum_j P_{ij}^2}{\left[\sum_j P_{ij}\right]^2} \le 1 \qquad \text{(Eq. 15)}$$

with n_i being the number of impurity giving A_i radioactivity.

$$\frac{1}{n_i} \cdot \frac{\sum_j P_{ij}^2 \left|\frac{\Delta C_j}{C_j}\right|^2}{\sum_j P_{ij}^2} \le \frac{\sum_j P_{ij}^2 \left|\frac{\Delta C_j}{C_j}\right|^2}{\left[\sum_j P_{ij}\right]^2} \le \frac{\sum_j P_{ij}^2 \left|\frac{\Delta C_j}{C_j}\right|^2}{\sum_j P_{ij}^2} \qquad \text{(Eq. 16)}$$

The right hand member of that equation is between $\text{Min}_j \left|\Delta C_j/C_j\right|^2$ and $\text{Max}_j \left|\Delta C_j/C_j\right|^2$ and we obtain :

$$\left|\frac{\Delta C_0}{C_0}\right|^2 + \frac{1}{n_i} \cdot \underset{j}{Min} \left|\frac{\Delta C_j}{C_j}\right|^2 \le \left|\frac{\Delta R_i}{R_i}\right|^2 \le \left|\frac{\Delta C_0}{C_0}\right|^2 + \underset{j}{Max} \left|\frac{\Delta C_j}{C_j}\right|^2 + \frac{1}{4} \cdot \left|\Delta X\right|^2 \qquad \text{(Eq. 17)}$$

APPENDIX 2 – ACTIVATION EQUATION SETS (ONLY TWO EXAMPLES)

From GUEPARD PC v1.0.5 (an EDF activation code), let's only show the example of activity equations for 2 of the 6 neutron flux histories (case 1 and case 6), in GBq/kg of graphite as linear function of chemical elements given in ppm.

The text format is the internal one from EXCEL formula and it gives an idea of the equations used for 2 cases (one year at 50% power and two years at 100% power).

Case 1

C14 = 0,000000007343*O + 0,00000000152708*C + 0,000148069*N
AG110M = 0,000000803941*CD + 0,243576*AG
CL36 = 0,00000000393787*K + 0,0000000000631609*AR + 0,000019792*CL
CS134 = 0,0000000021184*BA + 1,28384*CS
EU154 = 0,000000000206339*GD + 0,631947*EU
MN54 = 0,000100061*FE
H3 = 0,0000000223669*O + 0,0000000000859762*C + 0,00019147*B + 0,0000977924*BE
+ 4,17144*LI + 0,00033051*HE
CO58 = 0,00193127*NI
SB124 = 0,49374*SB + 0,00000586089*SN
SC46 = 0,00000918744*CA + 2,9664*SC
ZN65 = 0,0382914*ZN
CO60 = 0,00000212166*NI + 0,000000542354*FE + 0,822074*CO
SB125 = 0,000197773*SB + 0,000769091*SN + 0,0000000125357*TE
EU152 = 0,00000000000274346*GD + 0,0741844*EU
FE59 = 0,00000102399*NI + 0,000584227*FE
NI63 = 0,000563977*NI + 0,000000000016665*CO + 0,0000000299043*ZN
+ 0,00000380538*CU
AG108M = 0,000000000307734*CD + 0,0000338949*AG
BE10 = 7,29229E-17*O + 0,00000000000157511*C + 0,0000000000842063*B

+ 0,00000000391103*BE + 4,29907E-18*N
CA41 = 0,000000626425*CA
CD109 = 0,00103706*CD + 0,000742813*AG
CD113M = 0,0000333133*CD + 0,0000000000183514*AG + 0,0000000000137913*SN
+ 0,0000000000719155*IN
CO57 = 0,00000319808*NI
CS135 = 8,24433E-15*BA + 0,0000000424905*CS + 0,000000000206856*XE
CS137 = 0,000000000424893*BA + 0,00000000078108*CS + 0,0000381424*XE
EU155 = 0,000000000104144*GD + 0,249726*EU
FE55 = 0,0000456989*NI + 0,00503696*FE
GD153 = 0,00833752*GD
I129 = 0,000000000365645*TE
MO93 = 0,00000029576*MO + 0,0000000000903345*RU
NB93M = 0,00000000595112*MO + 0,00000000000163049*RU
NB94 = 0,0000000000218218*MO + 5,09936E-19*RU + 0,00000692923*NB
NI59 = 0,00000448963*NI
PB210 = 3,99432E-15*BI
PD107 = 1,83352E-15*CD + 0,000000000000783925*AG + 0,00000000215843*PD
PM147 = 0,0000000199621*SM
SE79 = 0,00000019301*SE + 0,0000000005174*BR
SM151 = 0,00000000000194251*GD + 0,00000000000775314*EU + 0,00183649*SM
SN119M = 0,000564025*SN
SN121M = 0,00000000215815*SB + 0,000000684511*SN
SN126 = 0,000000000000298965*SN + 1,96324E-18*TE
SR90 = 0,000000000000905294*ZR + 6,17595E-18*MO + 9,16613E-30*RU
+ 1,13839E-16*NB + 0,00000000000675605*Y
TC99 = 0,0000000566901*MO + 0,00000000000898812*RU
TL204 = 0,000000000965852*PB + 0,0332759*TL
V49 = 4,85692E-18*FE + 0,00000016165*CR + 0,0000000114179*V
ZR93 = 0,0000000021961*ZR + 0,000000000000149836*MO + 4,42854E-25*RU
+ 0,00000000000276191*NB

Case 6 :

C14 = 0,0000000293783*O + 0,00000000610796*C + 0,000591398*N
AG110M = 0,0000140964*CD + 0,4914*AG
CL36 = 0,00000001561*K + 0,000000000249824*AR + 0,0000761168*CL
CS134 = 0,00000000662786*BA + 3,80794*CS
EU154 = 0,000000000322198*GD + 0,719207*EU
MN54 = 0,000286905*FE
H3 = 0,0000000870163*O + 0,000000000491031*C + 0,000349614*B + 0,00104482*BE
+ 9,11131*LI + 0,00033505*HE
CO58 = 0,00268078*NI
SB124 = 0,979162*SB + 0,0000532157*SN
SC46 = 0,000135321*CA + 5,99853*SC
ZN65 = 0,100489*ZN
CO60 = 0,0000277466*NI + 0,00000972024*FE + 2,98239*CO
SB125 = 0,00159932*SB + 0,00271171*SN + 0,0000000445261*TE
EU152 = 0,000000000000573453*GD + 0,000000403323*EU
FE59 = 0,00000917439*NI + 0,00120276*FE
NI63 = 0,00218624*NI + 3,04019E-15*FE + 0,00000000407039*CO + 0,000000117015*ZN

+ 0,000014847*CU
AG108M = 0,00000000442183*CD + 0,000117328*AG
BE10 = 1,17037E-15*O + 0,00000000000630292*C + 0,000000000101923*B
+ 0,0000000156436*BE + 6,87197E-17*N
CA41 = 0,00000249727*CA
CD109 = 0,00291382*CD + 0,00913248*AG
CD113M = 0,0000338995*CD + 0,00000000102439*AG + 0,000000000013794*SN
+ 0,0000000000650273*IN
CO57 = 0,00000884911*NI
CS135 = 0,0000000000000330031*BA + 0,00000055409*CS + 0,000000000486819*XE
CS137 = 0,00000000167456*BA + 0,0000000918969*CS + 0,000150792*XE
EU155 = 0,000000000188534*GD + 0,442217*EU
FE55 = 0,000160978*NI + 0,0177772*FE
GD153 = 0,00174998*GD
I129 = 0,00000000143243*TE
MO93 = 0,00000117358*MO + 0,000000000358129*RU
NB93M = 0,0000000499338*MO + 0,00000000001277*RU
NB94 = 0,0000000000858496*MO + 1,60932E-17*RU + 0,0000272019*NB
NI59 = 0,0000166204*NI
PB210 = 0,0000000000000604445*BI
PD107 = 7,27733E-15*CD + 0,0000000000030074*AG + 0,00000000855729*PD
PM147 = 0,0000000509302*SM
SE79 = 0,000000764134*SE + 0,00000000201536*BR
SM151 = 0,0000000000167054*GD + 0,00000000000013264*EU + 0,00152962*SM
SN119M = 0,00160094*SN
SN121M = 0,00000000829684*SB + 0,00000267867*SN
SN126 = 0,0000000000000242223*SN + 1,60083E-17*TE
SR90 = 0,0000000000143343*ZR + 9,76695E-17*MO + 1,15555E-7*RU
+ 0,0000000000000018004*NB + 0,000000000058387*Y
TC99 = 0,000000220528*MO + 0,000000000112013*RU
TL204 = 0,000000000348487*PB + 0,118733*TL
V49 = 3,01409E-17*FE + 0,000000467721*CR + 0,0000000322123*V
ZR93 = 0,00000000875911*ZR + 0,0000000000000596375*MO + 1,39921E-23*RU
+ 0,0000000000109937*NB