

## **ALGEBRAIC METHOD APPLIED TO PWR NPP ACTIVITY INVENTORIES FOR DISMANTLING PREVISIONS**

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### **ABSTRACT**

Low-level and intermediate-level dismantling waste from 58 P.W.R. nuclear plants of Électricité de France (E.D.F.) will be shipped to surface disposal.

The only E.D.F.-P.W.R. comprehensive activation computation is now available on a 300 MWe plant that was shutdown a few years ago. Anyway, what best can be done without further long and detailed computation is simply considering this plant as a 1/3-scale model of a 900 MWe PWR plant but the scale effect is not enough.

Indeed, for each radio nuclide, in order to take into account the effect of changing the operating cycle history, some algebraic properties of activation calculations should be introduced. Such elementary properties are almost never enhanced.

Nevertheless, these properties prove very useful for either “precise” or “approximated” computations. What is here called “precise” computation considers the fact that target parent isotopes are used whereas “approximated” computation neglects it.

### **INTRODUCTION AND GENERAL PROBLEM**

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 global activity of numerous nuclides which will be shipped to the repository. EDF needs to know the order of magnitude of global activity for the main radionuclides from dismantling.

The main purpose of this paper is to present some useful algebraic properties of activation under neutron flux.

Such properties allow us to compute sensibility calculation with respect to initial chemical compositions and operating cycle history of the plant.

In order to globally evaluate the amount of activity corresponding to the shipment to the surface disposal of all the 58 French PWR NPP with a 40-year program of operation, a practical application of the method is presented.

For global quantification of a 40-year operation, we start only from limited detailed calculation made on metallic LLW of Chooz A PWR plant. Only direct activation of stable (no radioactive) chemical element

in their natural isotopic composition is considered. Activation considered came from a neutron spectrum computed by multiplication of 100% nominal power spectrum with a scalar function  $u(t)$  representing the operating cycle history of the NPP.

A radioactive decay like notion is also used to describe the rate of decreasing of the stable parent nuclide. The time corresponding to that decay is replaced by the "effective nominal power time" because this very time is actually the one that is at the origin of the parent nuclide equivalent decreasing. The difference between both types of computation is quantified and a general rule is given to know if the global approximated method is acceptable or not as another uncertainty source.

The first step of the method is based on linearity of activation with respect to initial chemical compositions and allow us to introduce activation matrix.

## ACTIVATION MATRIX

### *Demonstration by superposition principle*

For a given neutron flux history, we know that the multiplication by  $\lambda$  applied to the vector  $X_1$  of masses of target elements induces the same multiplication by  $\lambda$  on each radio nuclide (RN) produced.

We have 
$$f(\lambda X_1) = \lambda f(X_1)$$

For a given neutron flux history, let's consider separately the RN produced by a first  $X_1$  mass vector and by a second  $X_2$  mass vector.

If we activate simultaneously these two vectors (sum of masses of each element), we will get the sum of activities obtained for each radio nuclide.

We have 
$$f(X_1+X_2) = f(X_1)+f(X_2)$$

We recognize a linear operator from  $R^m$  to  $R^p$  where  $m$  is the number of chemical elements (subscript  $j$ ) in their natural isotopic composition and  $p$  is the number of RN (subscript  $i$ ). The matrix of that linear operator will be called "activation matrix"  $(Mat_{i,j})_{i,j}$ .

For a given part of a NPP, this matrix is a function of the neutron flux at 100% nominal power and of the operating history.

The best way to obtain it consists in computing separately the activation of each chemical element at a given density. This allows us to get, per mass unit, each column of the "activation matrix".

The matrix multiplication by mass vector  $X$  of chemical elements gives the activities even if we have to modify the level of an impurity at the end of the study.

This is very useful when you know that the main uncertainty on activation comes from the order of magnitude of impurities which effect on final activity may be very high.

What has been explained about each chemical element can be generalized to each isotope and give a more detailed "activation matrix".

## UNDER FLUX DECREASING OF STABLE “TARGET” COMPUTATION

Radio nuclide generation come from transformation of the stable parent nuclides. As radioactive decreasing, this phenomenon is stochastic and proportional to the quantity of stable parent isotopes.

The only difference is that the time involved is not the real time but the nominal power effective time because the reaction is proportional to the number of neutron, this number being itself proportional to the produced power. Later on, a notion of “effective nominal power time” will allow us to cope with activation dynamics.

As far as we remain at 100% nominal power, there is no difference. The macroscopic effect is an effective nominal power half-life and the analogy make it easy to understand.

## BATEMAN’S EQUATIONS WITH CONSTANT NEUTRON FLUX

Let’s give to stable isotope a peculiar treatment in solving activation equations. We have seen that, because of a same stochastic generation at nuclear scale, any stable isotope behaves under constant neutron flux exactly as if radioactive without flux.

Let’s consider X as the stable isotope vector of dimension m which each component decreases under a neutron flux corresponding to the constant fraction of nominal power u.

$$\frac{dX}{dt} + u \cdot N \cdot X(t) = X(0) \quad (\text{Eq. 1})$$

If we don’t take into account the generation of a stable isotope by activation of an other stable isotope, matrix N (m × m) corresponding to nominal power neutron flux is diagonal.

$$N = \text{diag}[v_j]_{j \in [1, m]} \quad (\text{Eq. 2})$$

Let’s consider V as the radioactive isotope vector which each component decreases without any neutron (matrix L) and also under neutron flux (matrix M) corresponding to the constant fraction of nominal power u. P is a (p×m) matrix and P(.,j) is its column j. If we consider the restriction of V to radioactive nuclides which are direct daughters of a non-radioactive nuclide, L and M are also diagonal.

$$\frac{dV(t)}{dt} + [L + u \cdot M] \cdot V(t) = u P \cdot X(t) \quad (\text{Eq. 3})$$

$$u P \cdot X(t) = u \sum_{j=1}^m X_j(t) P_{.,j} \quad (\text{Eq. 4})$$

The solution X(t) is :

$$X(t) = \exp[-u N \cdot t] \cdot X(0) \quad (\text{Eq. 5})$$

The solution of  $V(t)$  could be verified nevertheless :

$$V(t) = u \sum_{j=1}^m X_j(0) [L + u \cdot (M - \nu_j \cdot I)]^{-1} \left\{ e^{-u \nu_j t} I - \exp[-t(L + u \cdot M)] \right\} P_{\cdot, j} \quad (\text{Eq. 6})$$

Considering only the activation of isotope  $j$ , we have:

$$V_j(t) = u X_j(0) \cdot [L + u \cdot (M - \nu_j \cdot I)]^{-1} \left\{ e^{-u \nu_j t} I - \exp[-t(L + u \cdot M)] \right\} P_{\cdot, j} \quad (\text{Eq. 7})$$

Let's consider  $\Delta t$  the time of activation under constant neutron flux.

We have, in the neighborhood of zero :

$$V(\Delta t) = u \Delta t \cdot \sum_{j=1}^m X_j(0) \cdot P_{\cdot, j} + o(\Delta t) \quad (\text{Eq. 8})$$

Let's introduce what we can call the impulse matrix of dimension  $p \times m$  which is independent of  $L$ ,  $M$  and  $N$  matrixes.

$$\lim_{\Delta t \rightarrow 0} \frac{V(\Delta t)}{\Delta t} = u \cdot \mathfrak{S} \cdot X(0) \quad (\text{Eq. 9})$$

$$\mathfrak{S} = P \quad \text{and} \quad \mathfrak{S}_{\cdot, j} = P_{\cdot, j} \quad (\text{Eq. 10})$$

## BATEMAN'S EQUATIONS WITH A VARIABLE POWER LEVEL

### *Exact solution*

Let's introduce now what we can call the "effective nominal power time" at time  $t$ .

$$\Theta(t) = \int_0^t u(\tau) d\tau$$

Considering only the activation of isotope  $j$ , we have:

$$V_j(t) = X_j(0) \int_0^t u(\tau) \exp[-\nu_j \Theta(\tau)] \exp\{-(t-\tau)L - [\Theta(t) - \Theta(\tau)]M\} d\tau \cdot \mathfrak{S}_{\cdot, j} \quad (\text{Eq. 11})$$

Considering only a radioactive nuclide (i) which is the direct daughter of a non-radioactive nuclide:

$$V_{i,j}(t) = \mathfrak{S}_{i,j} \cdot X_j(0) \int_0^t u(\tau) \exp\{-\nu_j \Theta(\tau) - (t-\tau)\lambda_i - [\Theta(t) - \Theta(\tau)]\mu_i\} d\tau \quad (\text{Eq. 12})$$

**Approximated solution**

Considering only the activation of isotope j, we have:

$$\hat{V}_j(t) = X_j(0) \int_0^t u(\tau) \exp\{-(t-\tau)L\} d\tau \cdot \mathfrak{S}_{..j} \quad (\text{Eq. 13})$$

$$\hat{V}(t) = \left[ \int_0^t u(\tau) \exp\{-(t-\tau)L\} d\tau \right] \cdot \mathfrak{S} \cdot X(0) \quad (\text{Eq. 14})$$

Considering only a radioactive nuclide (i) which is the direct daughter of a non-radioactive nuclide:

$$\hat{V}_{i,j}(t) = \mathfrak{S}_{i,j} \cdot X_j(0) \int_0^t u(\tau) \exp\{-(t-\tau)\lambda_i\} d\tau \quad (\text{Eq. 15})$$

$$\hat{V}_i(t) = \left[ \int_0^t u(\tau) \exp\{-(t-\tau)\lambda_i\} d\tau \right] \sum_{j=1}^m \mathfrak{S}_{i,j} \cdot X_j(0) \quad (\text{Eq. 16})$$

**Error quantification of approximated solution for a RN direct daughter of a non-radioactive nuclide**

In the general case, let's introduce the following approximation: (Eq. 17)

$$\hat{V}_{i,j}(t) - V_{i,j}(t) = \mathfrak{S}_{i,j} \cdot X_j(0) \int_0^t u(\tau) \exp\{-(t-\tau)\lambda_i\} \left[ 1 - \exp\{-(\nu_j \Theta(\tau) - \mu_i [\Theta(t) - \Theta(\tau)])\} \right] d\tau$$

$$0 \leq \hat{V}_{i,j}(t) - V_{i,j}(t) \leq \hat{V}_{i,j}(t) \text{Max}(\nu_j, \mu_i) \Theta(t) \quad (\text{Eq. 18})$$

We can now give the maximum error induced by neglecting, both decreasing under neutron flux of stable parent isotope and decreasing under flux of the RN which has just been generated by activation.

**Error from practical computation of solutions for RN which are direct daughter of a non-radioactive nuclide and at nominal power**

u(t) being now equal to 1, we can compute then:

$$0 \leq \mathfrak{S}_{i,j} \cdot X_j(0) \int_0^{\Delta t} \exp(-\nu_j \tau) d\tau - V_{i,j}(\Delta t) \leq \Delta t (\lambda_i + \mu_i) \cdot \mathfrak{S}_{i,j} \cdot X_j(0) \int_0^{\Delta t} \exp(-\nu_j \tau) d\tau \quad (\text{Eq. 19})$$

$$0 \leq \mathfrak{S}_{i,j} \cdot X_j(0) \Delta t - \mathfrak{S}_{i,j} \cdot X_j(0) \int_0^{\Delta t} \exp(-\nu_j \tau) d\tau \leq \nu_j \Delta t \mathfrak{S}_{i,j} \cdot X_j(0) \Delta t \quad (\text{Eq. 20})$$

$$0 \leq \mathfrak{S}_{i,j} X_j(0) \Delta t - V_{i,j}(\Delta t) \leq \Delta t (v_j + \lambda_i + \mu_i) \mathfrak{S}_{i,j} X_j(0) \Delta t \quad (\text{Eq. 21})$$

We can deduce from these calculus the computational relative error magnitude:

$$\varepsilon_{relative} \approx \Delta t (v_j + \lambda_i + \mu_i) \quad (\text{Eq. 22})$$

If plant operating history is cut into  $\Delta t$  intervals with  $u(t)$  constant on each of them,  $\Delta t$  being chosen small enough to compute the integral, we have a classical result which quantification of error is well known:

$$V_{i,j}(t) \cong \Delta t \sum_{k=1}^n u_k \exp\{-v_j \Theta[(k-1)\Delta t] - \lambda_i (n-k)\Delta t - \mu_i [\Theta(n\Delta t) - \Theta(k\Delta t)]\} \mathfrak{S}_{i,j} X_j(0)$$

$$\hat{V}_{i,j}(t) \cong \Delta t \sum_{k=1}^n u_k \exp[-\lambda_i (n-k)\Delta t] \cdot \mathfrak{S}_{i,j} X_j(0) \quad (\text{Eq. 24})$$

$\Delta t$  being chosen small enough compared to the dynamic of decreasing of radioactive nuclide (i) (simple radioactive decreasing or under flux decreasing) and small enough compared to the dynamic of under neutron flux decreasing of stable isotope (j), we have:

$$V_{i,j}(t) \cong \sum_{k=1}^n u_k \exp\{-v_j \Theta[(k-1)\Delta t] - \lambda_i (n-k)\Delta t - \mu_i [\Theta(n\Delta t) - \Theta(k\Delta t)]\} V_{i,j}(\Delta t)$$

$$\hat{V}_{i,j}(t) \cong \sum_{k=1}^n u_k \exp[-\lambda_i (n-k)\Delta t] \cdot V_{i,j}(\Delta t) \quad (\text{Eq. 26})$$

In all cases, the approximated form gives a very simplified expression of the linear operator which transform the RN vector after a time  $\Delta t$  at nominal power into the RN vector after a time  $t$  with an operating history composed of several intervals at constant power:

$$\hat{V}(t) \cong \left\{ \sum_{k=1}^n u_k \exp[-L(n-k)\Delta t] \right\} \cdot \hat{V}(\Delta t) \quad (\text{Eq. 27})$$

### CHOOZ A P.W.R. PLANT : FROM REAL OPERATING HISTORY TO AN OTHER ONE

Let's introduce the generalized half life  $\tau$  :

$$\tau_j = \frac{\ln 2}{v_j} \quad \text{and} \quad \mu_i \leq v_j \quad (\text{Eq. 28})$$

The calculation which does not neglect the effect of under-flux decreasing of stable isotope has been performed with respect to  $\tau$  which is the “half life like” computed for the maximum neutron flux calculated on each part of the PWR reactor and on each of the 3 energy groups. If we need to have a less pessimistic calculation, we can take into account the fact that the maximum flux is at least divided by 5 on about 75% of the volume of the PWR reactor.

In that case, the effective nominal power half-life is multiplied by 5, which divided the corresponding error between approximated and exact calculus by 5. In order to reasonably estimate the error, we can select (in the matrix activation row) the major stable element which give the effective nominal power half-life.

It could be demonstrated (but it's rather ... long), that the error calculation may be generalized to chemical element having a given isotopic composition. In that case, the value  $v$  is a weighted average of isotopic values and is dependant on the couple (RN, chemical element).

This precision being given, we can compute the following  $\tau$  (nominal power effective half life) for each couple (RN, chemical element).

Table 1

Radio nuclide	Radioactive half-live (year)	Main chemical element	Minimum $\tau$ nominal power effective half life (year)
CO57	7.43E-01	NI	180.00
SN119M	8.02E-01	SN	427.00
SN119M	8.02E-01	SN	427.00
MN54	8.56E-01	MN	57.30
FE55	2.70E+00	NI	180.00
SB125	2.73E+00	SN	377.00
CO60	5.27E+00	CO	15.70
H3	1.23E+01	CO	15.70
NB93M	1.64E+01	ZR	2180.00
SN121M	5.00E+01	SN	1490.00
NI63	1.00E+02	NI	61.10
MO93	3.50E+03	MO	2520.00
C14	5.73E+03	N	414.00
NB94	2.03E+04	MO	721.00
NI59	7.49E+04	NI	180.00
TC99	2.13E+05	MO	372.00
ZR93	1.53E+06	MO	126.00
BE10	1.60E+06	N	414.00
MN53	3.70E+06	MN	57.30
NB92	3.50E+07	MO	2520.00

The real nominal power effective time of CHOOZ A plant has been of 14.54 year and the planned nominal power effective time of now operating French PWR NPP is of 37.3 year.

Table 2

Chooz A real case	Beginning day	Ending day	Nominal power percentage	Reference 40 y PWR	Beginning day	Ending day	Nominal power percentage
1	0	869	45.30%	1	0	577	78%
2	935	1239	84.20%	2	607	973	100%
3	1283	1589	85.50%	3	991	1357	100%
4	1625	2023	83.80%	4	1375	1741	100%
5	2157	2449	84.00%	5	1759	2125	100%
6	2489	2777	73.60%	6	2143	2509	100%
7	2884	3309	90.10%	7	2527	2893	100%
8	3384	3701	93.70%	8	2911	3277	100%
9	3809	4067	90.90%	9	3295	3661	100%
10	4180	4492	97.50%	10	3716	4082	100%
11	4613	4925	94.40%	11	4100	4466	100%
12	4986	5311	91.30%	12	4484	4850	100%
13	5382	5731	92.70%	13	4868	5234	100%
14	5944	6228	96.50%	14	5252	5618	100%
15	6379	6662	90.00%	15	5636	6002	100%
16	7014	7383	93.00%	16	6020	6386	100%
17	7460	7901	90.00%	17	6404	6770	100%
18	7959	8188	90.00%	18	6788	7154	100%
				19	7172	7538	100%
				20	7598	7964	100%
				21	7982	8348	100%
				22	8366	8732	100%
				23	8750	9116	100%
				24	9134	9500	100%
				25	9518	9884	100%
				26	9902	10268	100%
				27	10286	10652	100%
				28	10670	11036	100%
				29	11113	11479	100%
				30	11497	11863	100%
				31	11881	12247	100%
				32	12265	12631	100%
				33	12649	13015	100%
				34	13092	13458	100%
				35	13476	13842	100%
				36	13860	14226	100%
				37	14244	14610	100%

We have calculated the influence of the neutron flux history when changing the real operating history of Chooz A into a normative 40-year exploitation time. We will use both the approximated activation (eq.



26) and the exact (eq. 25) activation (we only neglect in that case, the under flux decreasing of radio nuclide).

### CURVE TO ILLUSTRATE THE CALCULATION OF ERROR

Each radio nuclide calculation has been performed with respect to variable  $\tau$  varying between 15.7 year (corresponding to Co) and 2520 year (corresponding to Mo) with  $\Delta t = 1$  day and the relative error made when replacing exact calculation (eq. 25) by approximated calculation (eq. 26) has been computed.

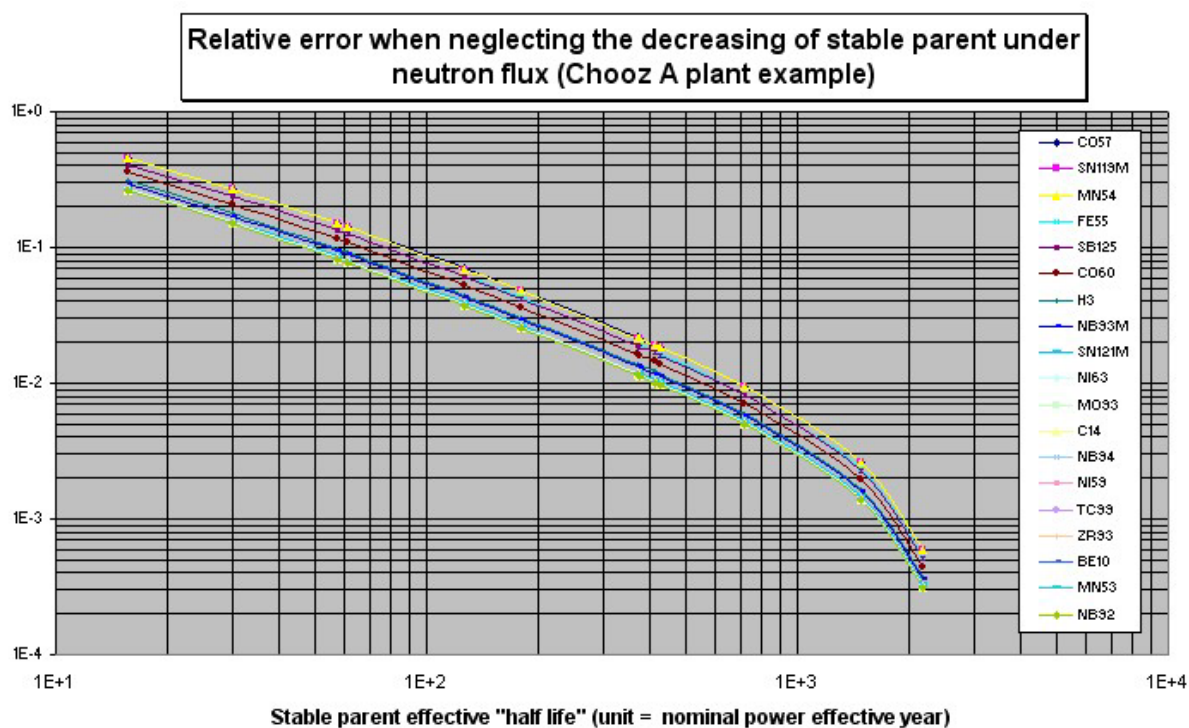


Fig. 1

If we consider the different stable parent chemical element effective "half life" from table 1 associated to each RN, we can see that the relative error may become very low and anyway, give a good order of magnitude for global prevision purposes.

### CONCLUSION AND LESSONS LEARNED

#### Approximated method

Even if we don't know precisely the detailed initial chemical composition, the approximated (Eq. 26) formula allows us to extrapolate from an given operating history to an other.

With this simple method, starting from a single plant detailed computations, a first estimate of the global quantity of Co-57, Sn-119m, Mn-54, Fe-55, Sb-125, Co-60, H-3, Nb-93m, Sn-121m, Ni-63, Mo-93, C-14, Nb-94, Ni-59, Tc-99, Zr-93, Be-10, Mn-53 and Nb-92 from the 58 P.W.R. plant dismantling metallic waste can be given using the approximated method with a sufficient precision for activity inventory previsions for disposal.

### **Exact method**

Otherwise, this exact method (Eq. 25) is particularly useful for any other activation calculation purpose because it needs only a few simple and fast “heavy code” computations at 100% power during a fixed time period. Starting only from these elementary results, a plain electronic sheet may give all full computations and sensibility analysis with respect to initial composition of activated materials and also with respect to detailed operating history.

Such activation computation properties, using approximation or not according to purposes, should improve dismantling activated waste characterization efficiency.

### **REFERENCES**

- 1 Characterization Of Graphite Sleeves From Bugey 1 Edf Plant For Permanent Disposal – Mesurement And Calculation Of Scaling Factors For Difficult-To-Measure Nuclides » by Bernard Poncet - WM'03 Conference, February 2003, TUCSON, Arizona.