**Proceedings of the 17th International Symposium on the
Packaging and Transportation of Radioactive Materials
PATRAM 2013
August 18-23, 2013, San Francisco, CA, USA**

# METHODS FOR THE DETERMINATION OF THE QUANTITY OF ACTIVITY WITHIN A TOOL OR SPARE PART PACKAGE

**Gaël Penessot**
EDF/UTO, Noisy le Grand, France

**François Renard**
EDF/UTO, Noisy le Grand, France

**Thierry Miquel**
EDF/UTO, Noisy le Grand, France

## ABSTRACT

According to the regulation of radioactive transport, the package classification depends upon the hazard of the radioactive contents. It is essential to know the nature of the radionuclides that are present within the package, and the overall quantity of activity of the material to be transported.

To achieve the maintenance of its Nuclear Power Plants (NPP), EDF ships contaminated tools or spare parts, to another NPP, or to a maintenance facility. According to their activity, these objects are shipped either in an Industrial Package (as SCO-I or SCO-II), in an Excepted Package, or in a Type A Package. Before the shipment, the consignor must know the overall activity of the package, and the surface contamination on the object, on accessible and inaccessible surfaces. While the measurement of the contamination on accessible surfaces is possible, by wiping a known surface, or by direct measurement, it is often not possible to determine or to measure directly the activity on inaccessible surfaces. Similarly, the overall activity of an object with a complex shape, must be indirectly determined.

The methods of determination of activity should be reliable. They should be user-friendly, and should cover a wide range of possible cases met in an industrial activity. For packages characterization purpose, the methods should be conservative, but too significant over-estimations should be avoided as they lead to cost overruns.

Several methods are described in this paper. These methods are based on the operational measurements that are currently performed during the NPP activities: the radiation levels measurements on the materials, and the surfaces contaminations measurements. These methods imply the precise knowledge of the radionuclides that are present in the facility.

For each method, the domain of validity is presented.

## INTRODUCTION

Électricité De France (EDF) operates 58 Pressurized Water Reactors (PWR), located in 19 Nuclear Power Plants (NPP). Each year, the French NPP carry out more than 3,600 shipments of radioactive tools or spare parts for the need of the maintenance operations. These objects

are contaminated, and are usually transported in IP1, IP2, Type A or Excepted packages. They are various in shape and weight.

The value of the overall activity of these objects, the contamination on the accessible and inaccessible surfaces must be known precisely in order to set up the package, and make sure the shipments fulfill the regulatory requirements. It is yet often difficult to directly measure the activity or the contamination on the whole surface of the objects, that is why indirect methods are used.

This paper present several methods to determine the quantity of activity on a tool or spare part used on a PWR NPP. These methods are meant to be used by the operators in industrial situations.

## PRINCIPLES

### Radionuclides

Radionuclides in a PWR NPP come primarily from the neutron activation of structural materials and corrosion products transported by water in the primary circuit. There are also some traces of fission products induced by the dust on the fuel, or when a serious fuel assembly leak occurs.

$RN_\gamma$: Gamma short lived radionuclides (T<31 years), which generates a measurable dose rate, are quite easy to detect. These are: $^{51}$Cr, $^{54}$Mn, $^{58}$Co, $^{137}$Cs, $^{134}$Cs, $^{60}$Co, $^{65}$Zn, $^{125}$Sb, $^{59}$Fe, and $^{110m}$Ag.

$RN_\beta$: Beta radionuclides are quite hard to detect on the field. These are mainly: $^{55}$Fe, $^{63}$Ni, $^{59}$Ni but also $^{79}$Se, $^{90}$Sr, $^{99}$Tc, $^{107}$Pd, $^{126}$Sn, $^{129}$I, $^{135}$Cs, $^{151}$Sm, $^{10}$Be, $^{14}$C, $^{36}$Cl, $^{41}$Ca, $^{93}$Mo, $^{93}$Zr, $^{94}$Nb, $^{108m}$Ag, $^{121m}$Sn. These activities can be estimated in proportion of the activity of a gamma radionuclide tracer.

$RN_\alpha$: Alpha long lived radionuclides are hard to detect and quantify: $^{241}$Am, $^{238}$Pu, $^{239}$Pu, $^{241}$Pu, $^{243}$Cm, $^{244}$Cm, $^{237}$Np, $^{234}$U, $^{235}$U, $^{238}$U. Generally, we only find some traces.

### Sources

There are two kinds of sources:

- Surface contamination of objects by radionuclides deposition. The dissemination of these products is due to the circulation of the coolant in the primary circuit and in the auxiliary circuits. Tools used for the reactor maintenances during the outages are exclusively contaminated objects. Most of the spare parts removed from the reactors are also contaminated objects.
- Activation of structural materials. The only concerned objects are those subjected to neutron flux, that means objects close to the operating core or spent fuel. Some spare parts, removed within the pressure vessel, may be contaminated and activated objects.

## Measures

Radiological measures on objects or packages are the inputs of the activity evaluation methods. They usually are gamma equivalent dose rate (EDR) measurements, relatively easy to do. Measurements of surface contamination ($A_s$) can also be used.

The maximum values $EDR_{max}$ and $As_{max}$ must be retained to ensure an upper bound of the evaluation.

To measure low activities, proper routines should be used. The background noise values will be deducted in order to use net values. Sensitive and responsive detectors should be used to ensure weak signals detection.

## General method

A modelization is used to evaluate the activity from the measures. Generally, gamma equivalent dose rate is used. The transfer function $TF_i$ is established and reflects the morphology of the package or object, measurement configuration, environment, etc. Generic transfer functions are calculated with an EDF radiation protection determinist code called PANTHERE.

For each radionuclide we have a relationship between the EDR and the activity:

$$TF_i = \frac{EDR_{model}}{a_{i,model}}$$

$$\begin{cases} a_{\gamma,i} = SF \times EDR \times \dfrac{p_{A_\gamma,i}}{\sum_i p_{A_\gamma,i} \times TF_i} \\ a_{\beta,i} = a_{\gamma,y} \times r_{i/y} \\ a_{\alpha,i} = a_{\gamma,y} \times r_{i/y} \end{cases}$$

$p_{A,i}$ is the activity proportion for radionuclide i.

SF is a "Safety Factor" to consider measures uncertainties. In practice, SF=1,25.

The total evaluated activity is:

$$A = \sum_i a_{\gamma,i} + \sum_i a_{\beta,i} + \sum_i a_{\alpha,i}$$

To consider radioactive decay at the measure time:

$$\begin{cases} a_{\gamma,i}(t_m) = SF \times EDR(t_m) \times \dfrac{p_{A_\gamma,i}(t_m)}{\sum_i p_{A_\gamma,i}(t_m) \times TF_i} = SF \times EDR(t_m) \times \dfrac{p_{A_\gamma,i}(t_0) \times e^{(-\lambda i \times \Delta t)}}{\sum_i p_{A_\gamma,i}(t_0) \times e^{(-\lambda i \times \Delta t)} \times TF_i} \\ a_{\beta,i}(t_m) = a_{\gamma,y}(t_m) \times r_{i,y}(t_0) \times e^{(\lambda_y - \lambda_i) \times \Delta t} \\ a_{\alpha,i}(t_m) = a_{\gamma,y}(t_m) \times r_{i,y}(t_0) \times e^{(\lambda_y - \lambda_i) \times \Delta t} \end{cases}$$

## First method: Detailed numeric analysis of the object

In this method, the complete and detailed numeric simulation of the object directly gives the transfer function TF. In operation, an EDR measurement on specific points around the object allows the operator to calculate its activity.
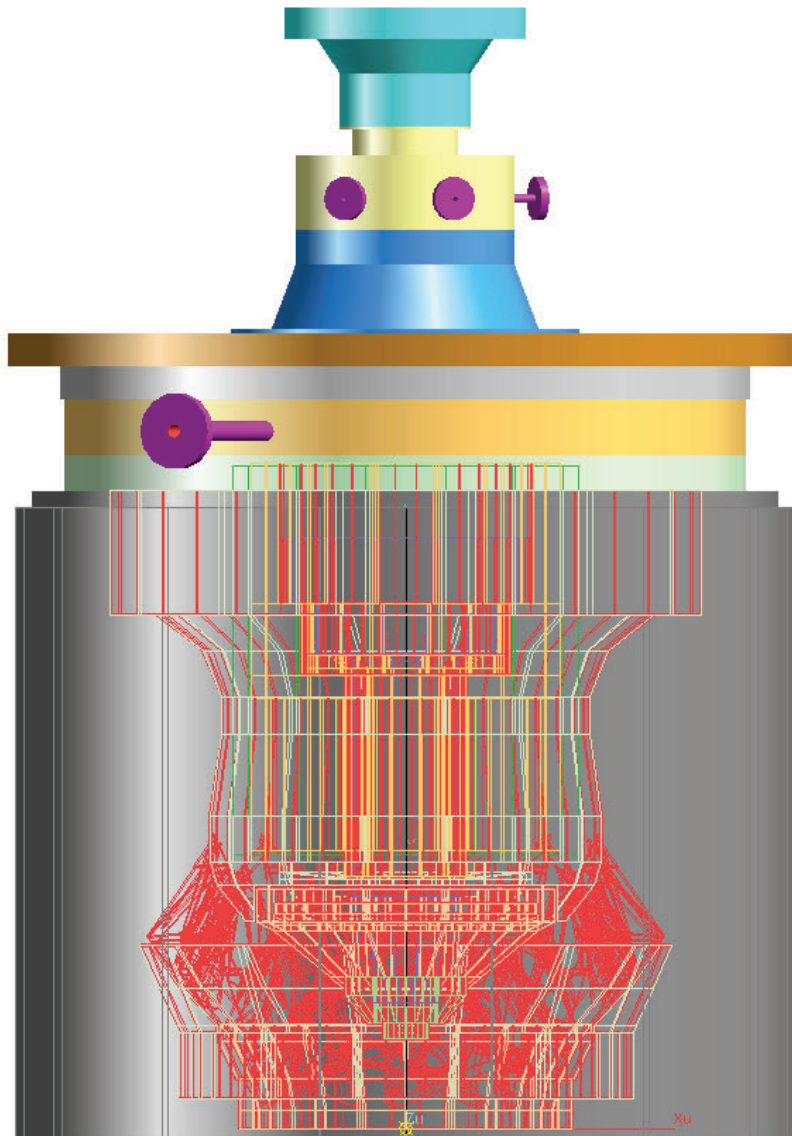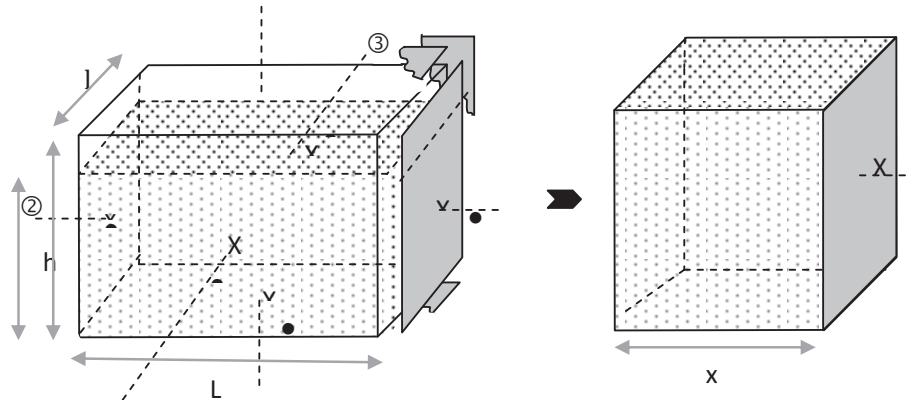


**Figure 1: primary coolant pump and shielding**

## Second method: EDR measures on package sides

The second method is based on dose rate measures on the package sides. It is not suitable for massive objects

In this case, we consider a package full of contaminated materials. The dose rate measure is realized from the outside of the package. The aim is to position the detector on the center of the package wall in order to have a contact measure. From this specific point, the package can be assimilated to a cube with an equivalent volume $V_0$. This equivalence allows to generalize the approach.



Transfer functions $TF_i$ are determined for different densities d, wall dimension x and sheet metal thickness e. Determinist EDF's radiation shielding code PANTHERE is used for this part.

$$TF_i(d, x, e) = \frac{EDR_i}{a_i}$$

These results are then correlated through a polynomial form in order to unify the answer.

The different calculation steps are:

a.  **Estimate the equivalent volume dimension x :**

$$\begin{cases} x = \sqrt[3]{V_0} \\ V_0 = L.\, l.\, h.\, T_r \end{cases}$$

**b. Calculate transfer functions for each radionuclide:**

$$\begin{cases} \ln(\text{TF}_i) = \sum_{j=0}^{2} \delta_{ij}.\,d^j = \delta_{2i}.\,d^2 + \delta_{1i}.\,d + \delta_{0i} \\[4mm] d = \dfrac{m}{V_0} \\[4mm] \delta_{ij} = \sum_{k=0}^{4} \chi_{ijk}.\,x^k = \chi_{ij4}.\,x^4 + \chi_{ij3}.\,x^3 + \chi_{ij2}.\,x^2 + \chi_{ij1}.\,x + \chi_{ij0} \\[4mm] \chi_{ijk} = \sum_{l=0}^{4} \varepsilon_{ijkl}.\,e^l = \varepsilon_{ijk4}.\,e^4 + \varepsilon_{ij3}.\,e^3 + \varepsilon_{ij2}.\,e^2 + \varepsilon_{ij1}.\,e + \varepsilon_{ij0} \end{cases}$$

**c. Calculate beta activity and global activity**

$$\begin{cases} a_{\gamma,i}(t_m) = \text{SF} \times \text{EDR}_{max}(t_m) \times \dfrac{p_{A_\gamma,i}(t_m)}{\sum_i p_{A_\gamma,i}(t_m) \times \text{TF}_i} = \text{SF} \times \text{EDR}_{max}(t_m) \times \dfrac{p_{A_\gamma,i}(t_0) \times e^{(-\lambda i \times \Delta t)}}{\sum_i p_{A_\gamma,i}(t_0) \times e^{(-\lambda i \times \Delta t)} \times \text{TF}_i} \\[4mm] a_{\beta,i}(t_m) = a_{\gamma,y}(t_m) \times r_{i,y}(t_0) \times e^{(\lambda_y - \lambda_i) \times \Delta t} \\[4mm] a_{\alpha,i}(t_m) = a_{\gamma,y}(t_m) \times r_{i,y}(t_0) \times e^{(\lambda_y - \lambda_i) \times \Delta t} \end{cases}$$

Finally, the global activity is:

$$A = \sum_i a_{\gamma,i} + \sum_i a_{\beta,i} + \sum_i a_{\alpha,i}$$

A spreadsheet tool has been developed to facilitate the method implementation:



The spreadsheet shows input parameters:

| Parameter | | Value |
|---|---|---|
| Dimension L | (cm) | 190 |
| Dimension l | (cm) | 190 |
| Dimension h | (cm) | 200 |
| Epaisseur paroi e | (cm) | 0,3 |
| Taux remplissage Tr | | 1,00 |
| Masse contenu m | (kg) | 2000 |
| Date retrait exploit. $t_0$ | | 01/01/1995 |
| Date mesure Ddd $t_m$ | | 01/01/1995 |
| BdF | (mSv.h$^{-1}$) | |
| Ded max $_{contact, net}$ | (mSv.h$^{-1}$) | 0,0500 |
| Ded moy. $_{contact, net}$ | (mSv.h$^{-1}$) | 0,0300 |
| Arête équivalente x | (cm) | 193 |
| Densité app. d | ($\sim$ g.cm$^{-3}$) | 0,28 |
| $\Delta t$ | (s) | 0 |

| $RN_{γ,i}$ | $^{110m}$Ag | $^{57}$Co | $^{58}$Co | $^{60}$Co | $^{51}$Cr | $^{134}$Cs | $^{137}$Cs | $^{59}$Fe | $^{54}$Mn | $^{124}$Sb | $^{125}$Sb | $^{65}$Zn |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| $\lambda_i$ (s$^{-1}$) | 3,16E-08 | 2,95E-08 | 1,15E-07 | 4,17E-09 | 2,89E-07 | 1,06E-08 | 7,28E-10 | 1,79E-07 | 2,57E-08 | 3,26E-08 | 7,93E-09 | 3,26E-08 |
| $p_{A_{γ,i}}(t_0)$ (pour 1) | 0,11 | 0,00 | 0,41 | 0,39 | 0,00 | 0,01 | 0,02 | 0,00 | 0,04 | 0,00 | 0,01 | 0,01 |
| Traceur $RN_{γ,y}$ | - | - | - | - | - | - | - | - | - | - | - | - |
| $r_{βγ}(t_0)$ | - | - | - | - | - | - | - | - | - | - | - | - |
| $a_{γ,i}(t_m)$ (GBq) | 3,13E-02 | 0,00E+00 | 1,17E-01 | 1,11E-01 | 0,00E+00 | 2,85E-03 | 5,69E-03 | 0,00E+00 | 1,14E-01 | 0,00E+00 | 2,85E-03 | 2,85E-03 |

$$a_{γ,i}(t_m) = Ded_{max}(t_m) \cdot \frac{p_{A_{γ,i}}(t_0) \cdot e^{(-\lambda_i \cdot \Delta t)}}{\sum p_{A_{γ,i}}(t_0) \cdot FT_i \cdot e^{(-\lambda_i \cdot \Delta t)}}$$

$$a_{β,i}(t_m) = a_{γ,y}(t_m) \cdot r_{i/y}(t_0) \cdot \exp\left[(\lambda_y - \lambda_i) \cdot (t_m - t_0)\right]$$

| | (GBq) | |
|---|---|---|
| $A_γ(t_m)$  $A_β(t_m)$ | 2,85E-01 | 2,63E-01 |
| $A(t_m)$ | 5,48E-01 | |

## Third method: Surface contamination measures

The third method is based on surface contamination measures ($A_s$) on the object itself. It is suitable for massive objects and also for objects made of steel plates.

The surface activity is measured on the objects, either directly or indirectly by wiping. It is based on the detection of β/γ radiations. However, given the levels of concentration, the rate of decay and the energy range considered, the measure finally consists to take into account the main radionuclides called RNγ. The resulting value is expressed in $^{60}$Co Bq equivalent.

The total activity can be obtained multiplying surface by surface activity:

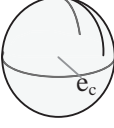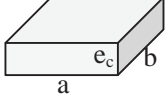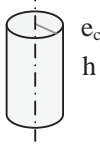$$A\,(Bq) = A_S\left(\frac{Bq}{cm^2}\right) \times S(cm^2)$$

This method is:

- Direct if the surface is known
- Generalized if the surface is estimated

To estimate the surface, we have considered that package content can be "splitted" into small elements with a specific thickness $e_c$.

$$S_c = k(e_c) \times V_c$$

$$S_c = k(e_c) \times m_c/\rho$$

The $k(e_c)$ coefficient is the ratio between the volume and the surface of the object. Some examples are detailed here:

| Object | Surface $S_c$ | Volume $V_c$ | Ratio $k(e_c) = \frac{S_c}{V_c}$ |
|---|---|---|---|
| Sphere  | $4\pi e_c^2$ | $\frac{4}{3}\pi e_c^3$ | $\frac{3}{e_c}$ |
| Parallelepiped (sheet metal)  | $2ab + 2ae_c + 2be_c$ <br> $\approx 2ab$ <br><br> if $\begin{cases} e_c \ll b \\ e_c \ll a \end{cases}$ | $abe_c$ | $\approx \dfrac{2}{e_c}$ |
| Cylinder  | $2\pi e_c(h + e_c) \approx 2\pi e_c h$ <br><br> if $e_c \ll h$ | $\pi e_c^2 h$ | $\approx \dfrac{2}{e_c}$ |

Results show that the following ratio must be used to have an upper bond calculus:

$$k(e_c) = 3/e_c$$

Then, we calculate the activity for each RN:

$$\begin{cases} a_{\gamma,i}(t_m) = SF \times A_s(t_m) \times A_s \times \dfrac{p_{A_\gamma,i}(t_0) \times e^{(-\lambda i \times \Delta t)}}{\Sigma_i\, p_{A_\gamma,i}(t_0) \times e^{(-\lambda i \times \Delta t)}} \\ \\ a_{\beta,i}(t_m) = a_{\gamma,y}(t_m) \times r_{i,y}(t_0) \times e^{(\lambda_y - \lambda_i) \times \Delta t} \\ a_{\alpha,i}(t_m) = a_{\gamma,y}(t_m) \times r_{i,y}(t_0) \times e^{(\lambda_y - \lambda_i) \times \Delta t} \end{cases}$$

Finally:

$$A = \sum_i a_{\gamma,i} + \sum_i a_{\beta,i} + \sum_i a_{\alpha,i}$$

**RESULTS**

Many packages have been modeled to be sure to cover the entire field of application. Calculated contact EDR which reflect measurable signals were used as method inputs. The activity was evaluated and compared to the original activity. This method well renders about activities with a small overestimation:

Example:

| x (cm) | e (cm) | d (g.cm$^{-3}$) | Calculated Activity/Original Activity |
|--------|--------|----------------|----------------------------------------|
| 50 | 0,3 | 0,1 | 109% |
| 50 | 0,3 | 0,4 | 111% |
| 50 | 0,3 | 0,7 | 111% |
| 50 | 0,3 | 1,0 | 110% |
| 50 | 0,3 | 1,3 | 110% |
| 50 | 0,3 | 1,6 | 110% |
| 50 | 0,3 | 1,9 | 110% |
| 50 | 0,3 | 2,2 | 110% |

Activity was determined as precisely as possible on packages containing simple tools with smears. The second method with surface activity As has been applied by this small surface elements finely characterized.

The first method has been applied in parallel.

The comparison is presented here below. The results of the first method by measuring contact EDR are conservative in all cases, with a 1.4 to 9.2 factor.

Methods comparison:



**Figure 2:**

| Activity | | First method (GBq) | Second method (GBq) |
|----------|------------------|--------------------|---------------------|
| RN$\gamma$ activity | A$\gamma$ (t$_m$) | 4,3E-04 | 7,9E-05 |
| RN$_\beta$ activity | A$_\beta$ (t$_m$) | 6,1E-04 | 1,2E-04 |
| Total activity | A (t$_m$) | 1,0E-03 | 1,9E-04 |

## CONCLUSIONS

These methods allow EDF operators to estimate more precisely the activity in a package to be sure to fulfill the regulatory requirements.

These methods are designed to estimate precisely the activity of the package, and they still are conservative.

Once the Transfer Functions are determined by numeric simulation with EDF's code PANTHER, the methods are easy to use by the operators in an industrial situation.

## REFERENCES

[0]    PANTHERE general presentation AB04B020/DU/05008 V2.0 (https://panthere.c-s.fr/AB04B020DU05008_Presentation_Generale_Panthere_1.7.pdf/file_view)