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A METHOD TO IMPROVE DOSE ASSESSMENT BY RECONSTRUCTION OF THE COMPLETE ISOTOPES INVENTORY

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Radiation shielding assessments may underestimate the expected dose if some isotopes at trace level are not considered in the isotopes inventory of the shielded radioactive materials. Indeed, information about traces is not often available. Nevertheless, the activation of some minor isotopic traces may significantly contribute to the dose build-up. This paper presents a new method (Isotopes Inventory Reconstruction—IIR) estimating the concentration of the minor isotopes in the irradiated material at the beginning of the cooling period. The method requires the solution of the inverse problem describing the irradiated material's decay. In a mixture of an irradiated uranium–plutonium oxide shielded by a set-up made of stainless-steel, porous polyethylene plaster and lead methyl methacrylate, the comparison between different methods proves that the IIR-method allows better assessment of the dose than other approximate methods.

INTRODUCTION

Radiation shielding studies are often carried on in two steps. The first step is the determination of emitted radiations by the shielded radioactive materials (the so-called source term). Second, the transport of the emitted radiations from the source to the surroundings is assessed in order to determine the ambient dose equivalent rate. The first step is carried out using a depletion code that simulates the nuclide depletion of the materials and computes the radiations (gamma, beta...) emitted from the radioactive materials. The depletion code solves numerically the generalized Bateman equations that describe the isotopic transmutations during the neutron activation process (successive irradiation and cooling periods). The second step is performed using a code that simulates the transport of radiations. The radiation transport code solves numerically the Boltzmann equation using either a Monte Carlo method or a deterministic one such as the discrete ordinates method. A point kernel method is also used to deal with gamma propagation depending on the shielding study.

The accuracy of dose assessment depends on: the level of details in the isotopic inventory, the accuracy of the radiations spectra estimated by the depletion code, the level of details of the geometrical model used in the radiation transport calculations and finally the numerical accuracy of the transport code. The numerical accuracy of the two main codes (depletion and transport codes) depends on the numerical methods used to solve the corresponding equations (Bateman differential equations and Boltzmann integral) and also on the uncertainties in the nuclear data.

The paper presents a method for improving the assessment of the ambient dose equivalent rate in radiation shielding studies where the radioactive source is a mixture of uranium and plutonium oxides whose nuclide inventory is not sufficiently precise. This nuclide inventory contains often only the so-called ‘principal’ isotopes (with significant quantities). Other isotopes, progenies of the principal isotopes and with very small concentration (‘minor isotopes’), are unknown or neglected. Here chemical impurities at trace level, possibly present in the oxide mixture, are not taken into account.

In some radiation studies, knowing the complete isotopic inventory of the radioactive material is essential: some minor isotopes (progenies of principal isotopes) contribute significantly to the dose equivalent rate⁽¹⁾.

This problem emerges in different radiation shielding contexts. For example, within the Uranium Natural Graphite Gas (UNGG) reactor dismantling framework, the isotopes at trace level (called impurities) that are present at the initial state in the reactor structures (before activation) contribute much to the structure activation during the reactor operation⁽²⁾. Another example concerns the field of maintenance and decommissioning of accelerators and accelerator-driven systems, where irradiation conditions or material initial composition are incomplete or imprecise. For this context, specific characterization methods are developed to calculate with higher precision the radionuclide inventory^(3, 4).

One possible way of taking into account the minor isotopes in a uranium–plutonium oxide mixture (MOX) is to compute the decay of the principal isotopic inventory over a period, long enough to obtain

minor isotopes but short enough not to modify other isotopic quantities. This method has the disadvantage of being very sensitive to the decay-period calibration. The estimated quantities of the minor isotopes and even of the major isotopes in some configurations are dependent on the choice of the decay-period, unlike the method presented here.

In the first section of this paper, an example is given to illustrate that taking into account the minor isotopes is essential to correctly evaluate the dose equivalent rate. In the second section, a new method is described for reconstructing the complete isotopic inventory from an incomplete inventory of a given MOX. Some validation elements are also given. At last in the third section an application shows that the new method improves the assessment of the dose equivalent rate, comparing to other methods.

INFLUENCE OF THE RADIOACTIVE PROGENY ISOTOPES ON DOSE EQUIVALENT RATE

Data

The radioactive material studied here is a 3-year cooled MOX. This kind of materials is commonly found in blankets⁽⁵⁾ and contains ^{241}Am . The complete nuclide inventory of the MOX is given in Table 1 and contains the principal isotopes (main masses) and minor isotopes or progeny isotopes coming from the radioactive disintegration of the principal isotopes. Starting with known masses for (U, Pu, Am), the following of the decay chains enables to obtain the quantities of all progenies.

For the purpose of this paper, an imprecise nuclide inventory that contains only principal isotopes is considered in a given MOX where minor isotopes are neglected. Nuclide inventory is expressed in mass percentages of the (U,Pu) O_2 mixture, Table 1. When the masses of (U, Pu, Am) are known with precision of 10 ppm, the complete inventory obtained by following the decay chains is perfectly stable.

Particles of two kinds are emitted by the (U,Pu) O_2 mixture :

- neutrons emitted by spontaneous fissions and (α , n) reactions on oxygen atom of the mixture; the O-atom mass concentration is 11.84% in the given mixture; and
- gamma photons directly emitted by the mixture disintegration and gamma coming from (n, γ) reactions occurring in the different shielding materials setting around the mixture.

The geometrical set-up of the study case is shown in Figure 1. It is an 1D approximation of a jar containing the MOX. Typical materials and thicknesses encountered in radiation protection problems are considered. The radioactive source is a sphere with

Table 1. Complete and incomplete isotopic inventories of (U,Pu,Am) O_2 .

Complete inventory		Incomplete inventory	
Isotope	Mass (%)	Isotope	Mass (%)
^{234}U	5.7834×10^{-04}	^{234}U	5.7835×10^{-04}
$^{235}\text{F}\text{U}$	1.6500×10^{-01}	^{235}U	1.6500×10^{-01}
^{238}U	$6.6000 \times 10^{+01}$	^{238}U	$6.6001 \times 10^{+01}$
^{238}Pu	1.1708×10^{-02}	^{238}Pu	1.1708×10^{-02}
^{239}Pu	9.1708×10^{-04}	^{239}Pu	9.1710×10^{-04}
^{242}Pu	3.0573×10^{-03}	^{242}Pu	3.0574×10^{-03}
^{241}Am	$1.7516 \times 10^{+01}$	^{241}Am	$1.7516 \times 10^{+01}$
$^{242\text{M}}\text{Am}$	$1.2317 \times 10^{+00}$	$^{242\text{M}}\text{Am}$	$1.2317 \times 10^{+00}$
^{243}Am	$3.1591 \times 10^{+00}$	^{243}Am	$3.1592 \times 10^{+00}$
^{242}Cm	3.2062×10^{-03}	^{242}Cm	3.2063×10^{-03}
^{237}Np	8.2956×10^{-02}	^{237}Np	8.2958×10^{-02}
^4He	1.6162×10^{-03}		
$^{242\text{F}}\text{Am}$	1.5910×10^{-05}		
^{239}Np	2.7201×10^{-06}		
^{238}Np	2.3055×10^{-07}		
^{233}U	3.6937×10^{-08}		
^{230}Th	4.1289×10^{-09}		
^{233}Pa	2.7172×10^{-09}		
^{234}Th	9.5781×10^{-10}		
^{231}Pa	4.7850×10^{-10}		
$^{235\text{M}}\text{U}$	1.8474×10^{-12}		
^{231}Th	6.7088×10^{-13}		
$^{229\text{F}}\text{Th}$	1.5283×10^{-13}		
^{226}Ra	5.3821×10^{-14}		
$^{234\text{M}}\text{Pa}$	3.2305×10^{-14}		
$^{234\text{F}}\text{Pa}$	1.6848×10^{-14}		
^{227}Ac	1.4439×10^{-14}		
O	$1.1824 \times 10^{+01}$	O	$1.1824 \times 10^{+01}$
Sum	$1.0000 \times 10^{+02}$		$1.0000 \times 10^{+02}$

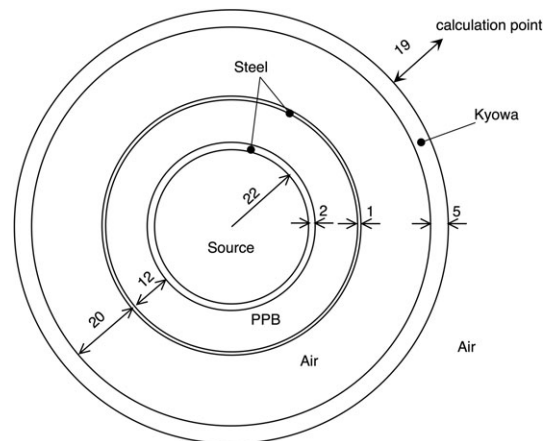


Figure 1. Geometric set-up (dimensions given in cm).

22 cm radius made of (U,Pu) O_2 mixture, surrounded by a stainless-steel sphere of 2 cm thickness. A close radiation shield sheet, made up of porous

polyethylene plaster (called PPB)⁽⁶⁾ of 12 cm thickness is inserted in a stainless-steel container of 1 cm thickness surrounding the source. Another radiation shield sheet made up of transparent lead methyl methacrylate⁽⁶⁾ (Kyowa Glass) of 5 cm thickness is placed at a distance of 20 cm from the close radiation shield (PPB). The set-up is surrounded by air (200 cm thickness). Dose equivalent rate is calculated at 19 cm from the Kyowa Glass shield.

Method to compute the ambient dose equivalent rate

The ambient dose equivalent $H^*(10)$ is the operational quantity for area monitoring^(7, 8) in the framework of external exposure to penetrating radiations. Ambient dose equivalent $H^*(10)$ due to neutron and photon radiations emitted by the radioactive material is calculated by the particles transport codes. Particle flux (particles number/cm²/s) is first calculated by the code and then $H^*(10)$ ($\mu\text{Sv/h}$) is obtained by multiplying the flux by flux-to-ambient dose equivalent conversion coefficients ($\mu\text{Sv cm}^2$)⁽⁹⁾ tabulated in response code libraries.

The computation of neutrons and gamma (directly emitted by the oxide mixture) from the (U,Pu)O₂ mixture isotopic inventory is performed by DARWIN evolution code⁽¹⁰⁾ within a 195 energy group structure. Decay chains and nuclear constants libraries used in the calculation come from the JEFF-3.1 Nuclear Data Library⁽¹¹⁾, that describes the radioactive decay chains of 3849 isotopes.

DARWIN code calculates the total intensity of neutron source, coming from spontaneous fissions and (α, n) reactions on oxygen atom of the mixture. To take into account induced fissions by neutrons coming from spontaneous fissions, this intensity is

multiplied by $1/(1-k_{\text{eff}})$, where k_{eff} is the multiplicative coefficient characteristic of the mixture. The neutrons spectra are distributed according to a Watt-Cranberg spectrum with coefficients 1.012 and 2.249⁽¹²⁾.

Then the dose equivalent rate is obtained by two different computations. The dose due to neutrons and gamma coming from (n, γ) reactions taking place in the different radiation shields is calculated from the neutron source with the 1D discrete ordinates (S_N) code SN1D^(13, 14). The dose due to gamma photons directly emitted by the mixture is calculated in 195 energy groups with NARMER⁽¹⁵⁾ (point kernel attenuation method with build-up factors) from the gamma source. The contribution of each energy group to the dose is also evaluated by the code.

Results

The total neutron sources intensities from complete and incomplete inventories described in Table 1 are calculated and results correspond to the same value 1.3×10^8 n/s. Photon sources from complete and incomplete inventories are also close. They are respectively 1.15×10^{15} and 9.94×10^{14} γ /s. The photon spectra of the complete and incomplete inventories differ, as shown in Figure 2, above the incident energy 0.9 MeV:

- between 0.9 and 1.1 MeV, the spectrum curve of the complete inventory presents two bumps (10^{11} γ /s) due to ²³⁸Np, whereas the intensity of the incomplete inventory is about 6×10^7 γ /s; and
- between 1.3 and 2 MeV, the emission intensity of the complete inventory is on average 10^5 γ /s, whereas the intensity of the incomplete inventory is null.

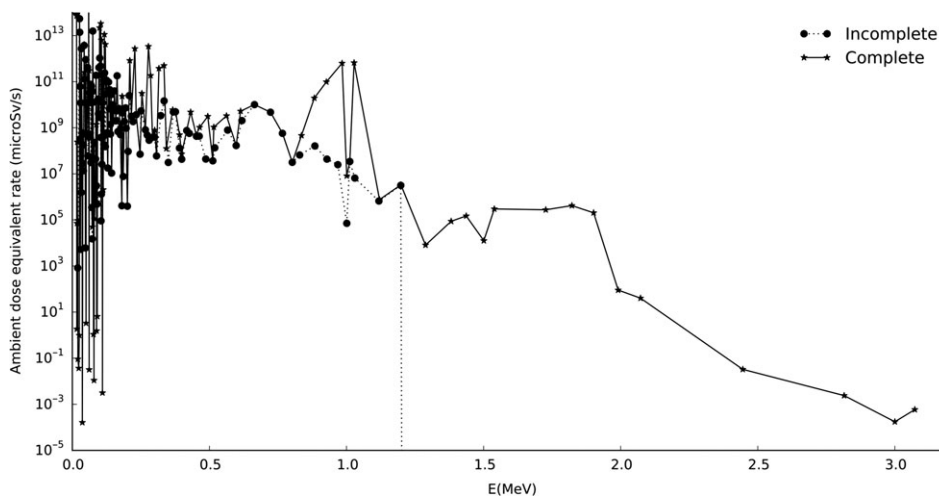


Figure 2. Complete and incomplete gamma emissions spectra.

Table 2. Complete and incomplete inventories calculated dose.

Ambient dose equivalent ($\mu\text{Sv/h}$)	Isotopic inventory	
	Incomplete	Complete
n	9.33×10^1	9.33×10^1
(n, γ)	4.4	4.4
γ	2.22×10^1	6.71×10^3
Total	1.20×10^2	6.81×10^3

For the complete inventory, the gamma rays emitted between 0.9 and 1.1 MeV come from progenies of the principal isotopes appeared during the 3-year cooling. Despite their small intensity (10^{11} γ /s on average, compared with the total intensity 10^{15} γ /s), these gamma rays significantly increase the gamma-dose rate because of their rather high energy.

Minor isotopes do not contribute to the neutron dose. Indeed neutron dose rate depends exclusively on the principal isotopes.

The gamma-dose rate due to gamma coming from (n, γ) reactions is also the same for both inventories. Moreover, their contribution to the total gamma-dose rate is negligible.

The gamma-dose rate due to gamma directly emitted by the (U,Pu)O₂ mixture disintegration increases when the inventory is complete. The total dose computed with the complete inventory is 50 times higher (see Table 2).

Using the NARMER code, the contribution of each gamma energy group to the gamma-dose rate is performed. The two gamma-energy groups that contribute the most to the gamma-dose are 0.95, 0.995 MeV and 1.022, 1.1 MeV, as shown in Figures 3 and 4. A cross-check with DARWIN results that calculate each isotope contribution to the gamma-spectrum proves that ²³⁸Np, the progeny isotope (minor isotope), has the major contribution to the gamma-dose rate ($\sim 94\%$).

METHOD TO RECONSTRUCT THE COMPLETE ISOTOPIC INVENTORY FROM THE INCOMPLETE INVENTORY

The example presented in Introduction shows that it is possible to improve ambient dose calculation by taking into account all the progeny isotopes of the principal isotopes in a radioactive material such as (U,Pu)O₂.

Consider a radioactive material that has decayed from time $t_0 = 0$ to time t for which the isotopic inventory of minor isotopes is unknown. The method consists in determining the isotopic inventory of the radioactive material at t_0 by tracking back in

time the radioactive decay chains, and then in making the decay calculation with all the decay chains over the interval $(t-t_0)$. This decay calculation yields the complete isotopic inventory at time t . Formally the method presented here relies on solving a well-posed inverse problem⁽¹⁶⁾ when the cooling time is known. The method and its limits are described below.

Approach for solving the radioactive decay inverse problem

The direct problem of radioactive decay is briefly introduced before moving to the inverse problem.

Direct problem

The time evolution of atom densities is governed by the following generalized Bateman equations⁽¹⁷⁾ written here in the case of radioactive decay:

$$\frac{dN_i(t)}{dt} = -\lambda_i N_i(t) + \sum_{k=1}^{i-1} \lambda_{ki} N_k(t), \quad (1)$$

where $N_i(t)$ is the atom density of isotope i at time t , λ_i is the total decay constant of isotope i , λ_{ki} is the disintegration rate of isotope k towards isotope i , it can be expressed as $\lambda_{ki} = \lambda_k b_{ki}$, where b_{ki} is the decay branching ratio of isotope k towards isotope i , with $\sum_i b_{ki} = 1$.

It is always possible to order the isotopes of the decay chain in such a way that the calculation of the i th isotope uses only formerly calculated isotopes. Then whatever the radioactive or stable isotope i is, the isotope i 's quantity can be determined by an analytical calculation:

$$N_i(t) = \alpha_{ii}(t) N_i(0) + \sum_{k=1}^{i-1} \alpha_{ik}(t) N_k(0), \quad (2)$$

where the quantity $\alpha_{ii}(t)$ is expressed as $e^{-\lambda_i t}$, $\alpha_{ii}(t) \geq 0$, and $\alpha_{ik}(t) = \sum_{l=k}^i P_{li} e^{-\lambda_l t}$, $\alpha_{ik}(t) \geq 0$, where P_{li} is expressed as $P_{li} = \frac{\lambda_l}{\lambda_i - \lambda_l} \prod_{j=1}^{i-1} \frac{\lambda_j}{\lambda_j - \lambda_l} \forall l < i$ and $l \neq j$.

The summand $\alpha_{ii}(t) N_i(0)$ is the remaining quantity at time t of initial quantity $N_i(0)$ of isotope i . The other summand $\sum_{k=1}^{i-1} \alpha_{ik}(t) N_k(0)$ is the sum of all direct or non-direct precursors (named isotopes k) contributions to the isotope i 's quantity at time t .

Inverse problem

In order to calculate the initial quantity $N_i(0)$ of an isotope i at time $t = 0$ from its quantity $N_i(t)$ ($i = 1 \dots n$)

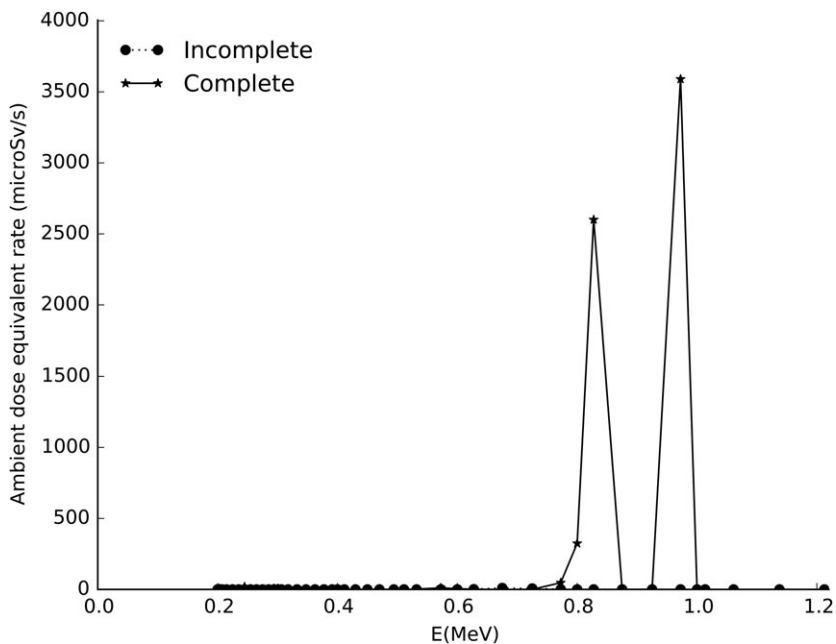


Figure 3. Gamma-dose equivalent rate of complete and incomplete inventories as function of the energy.

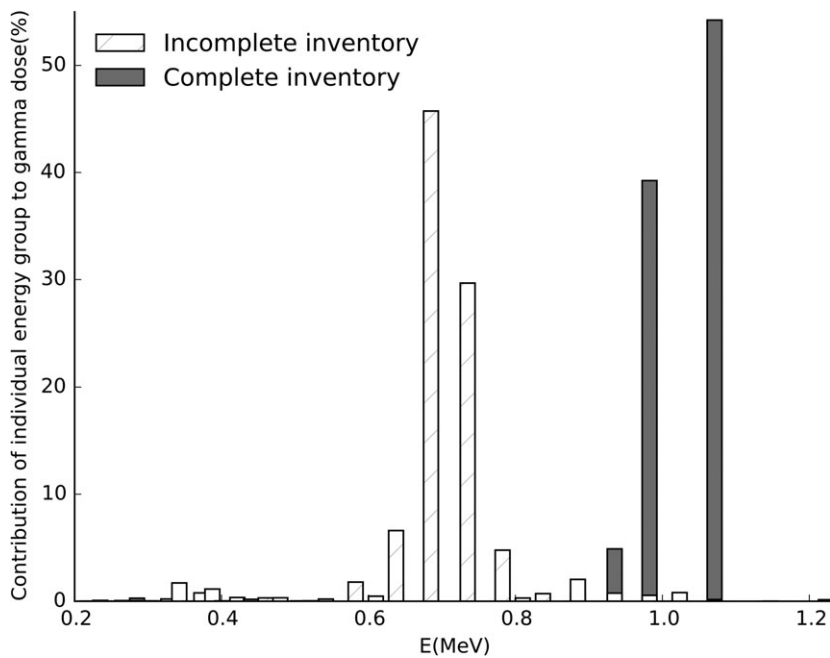


Figure 4. Contribution of individual energy groups to the gamma-dose equivalent rate with complete and incomplete inventories.

at time t , equation (2) is written in matrix form as follows:

$$N^t = A \times N^0, \quad (3)$$

which expands as

$$\begin{bmatrix} N_1(t) \\ \vdots \\ N_n(t) \end{bmatrix} = \begin{bmatrix} \alpha_{11} & 0 & 0 \\ \vdots & \ddots & 0 \\ \alpha_{n1} & \cdots & \alpha_{nn} \end{bmatrix} \times \begin{bmatrix} N_1(0) \\ \vdots \\ N_n(0) \end{bmatrix}, \quad (4)$$

where each coefficient $\alpha_{ik}(t)$ stands for the unit (or elementary) weight (or importance) of precursor k to isotope i at time t .

The incomplete inventory available at time t called ‘observable’ is composed of the quantities of so-called principal isotope i $N_i(t)$ ($i = 1..n$). The initial quantities $N_i(0)$ are calculated under the following assumption:

Assumption: All initial precursors (at time $t = 0$) of ‘principal’ isotopes must be part of the observable at time $t > 0$.

The method presented here is based on the determination of $\alpha_{ik}(t)$.

The n isotopes from the ‘observable’ are ordered by following their emerging order described in the decay chain; then n direct decay calculations are performed by calling n -times the DARWIN solver in the order previously mentioned; each run of rank i ($i = 1, \dots, n$) corresponds to a decay calculation of t duration from an initial unit atom of i th isotope ($N_i(0) = 1$) in the ‘observable’. The elements $N_i(t)$ ($i = 1, \dots, n$) of the final quantity vector N^t are the coefficients $\alpha_{ik}(t)$ looked for; these elements correspond to the i th line of the evolution matrix A . When all the isotopes of the ‘observable’ are processed, the resulting A matrix is a lower triangular matrix.

Then equation (3) is inverted by browsing all the lines of A matrix in increasing order.

If $\alpha_{ii}(t) \neq 0$ at decay time t , whatever the isotope i of the ‘observable’, the initial inventory is given by

$$N_i(0) = \frac{1}{\alpha_{ii}(t)} \left[N_i(t) - \sum_{k=1}^{i-1} \alpha_{ik}(t) N_k(0) \right], \quad (5)$$

Limitation of the method: If the isotope i of the ‘observable’ has a very short lifetime with respect to the decay time t , it cannot be computed as a part of the initial material but has appeared during the disintegration of its precursors. Then the problem is ill posed and further *a priori* information is needed. Indeed in this specific case, $\alpha_{ii}(t) = 0$ and its quantity at time t is written as follows:

$$N_i(t) = \sum_k^{i-1} \alpha_{ik}(t) N_k(0). \quad (6)$$

Reconstruction of the complete isotopic inventory

After the first step detailed in the previous paragraph, a decay calculation of t duration is performed with DARWIN to get the complete nuclide inventory. This reconstructed inventory at time t includes minor isotopes arisen during the radioactive decay and the principal isotopes quantities that are identical to those of the incomplete inventory.

Validation

Validation uses a uranium oxide fuel enriched up to 3.7% and previously irradiated up to 45 GWd/t that constitutes a commonly encountered situation. The principal isotopes are the dominant ones in terms of mass concentration in the mixed oxide. The reference composition results from a 3 year cooling interval after the irradiation phase. The reference composition contains 444 isotopes.

Three ‘observables’ that contains only fractions of principal isotopes are extracted: the first one, called Observable 1, contains 153 principal isotopes. The second one, called Observable 2, contains 102 isotopes. The third one, called Observable 3, contains only 72 isotopes (Table 3). For each observable, the gamma sources are calculated using DARWIN with and without the reconstruction of the complete inventory.

When compared to the gamma-spectrum computed with the 444 isotopes, gamma-spectra of Observables 1, 2 and 3 show significant deviations (Table 3). The reconstruction method enables to obtain the correct gamma-spectrum for Observable 1 (‘Completed Observable 1’), and gamma spectra much closer to the reference spectrum for Observables 2 and 3.

The ambient dose equivalent rate $H^*(10)$ due to gammas is also computed for each composition. The doses are systematically higher when the observable is reconstructed.

APPLICATION

The radioactive material studied is a mixed uranium and plutonium oxide, having decayed for a cooling time of 3 years. The isotopic inventory (with O-atom mass concentration equal to 11.84% in the mixture) at time t is incomplete, i.e. it contains only the dominant isotopes (Table 4). The masses of isotopes of the incomplete inventory are known with precision of 10 ppm but printed with only few digits. The

Table 3. Gamma spectra and H*(10) due to gammas at $t_{ref} = 3$ years, for different representative observables of real composition.

$E_{min} - E_{max}$ (MeV)	$\gamma/(s \text{ tIHM})$ Reference composition, 444 isotopes	Energy group contri- bution (%)	$\gamma/(s \text{ tIHM})$ Observable 1, 153 isotopes	$\gamma/(s \text{ tIHM})$ Completed Observable 1, 234 isotopes	$\gamma/(s \text{ tIHM})$ Observable 2, 102 isotopes	$\gamma/(s \text{ tIHM})$ Completed Observable 2, 181 isotopes	$\gamma/(s \text{ tIHM})$ Observable 3, 72 isotopes	$\gamma/(s \text{ tIHM})$ Completed Observable 3, 142 isotopes
1.00E-02–2.00E-02	5.0716E+13	0.33	4.9820E+13	5.0708E+13	4.6730E+13	4.7770E+13	3.1758E+13	3.2798E+13
2.00E-02–3.00E-02	1.5157E+14	0.98	1.4765E+14	1.5137E+14	1.4992E+12	5.2139E+12	1.0651E+12	1.0683E+12
3.00E-02–4.50E-02	8.1053E+14	5.26	4.6208E+14	8.1039E+14	4.1598E+14	7.6430E+14	2.9166E+13	3.6274E+14
4.50E-02–6.00E-02	3.8243E+13	0.25	3.8139E+13	3.8229E+13	3.8136E+13	3.8226E+13	1.2460E+13	1.2511E+13
6.00E-02–7.00E-02	1.6671E+12	0.01	1.6455E+12	1.6670E+12	1.6376E+12	1.6591E+12	1.3679E+09	2.2859E+10
7.00E-02–1.00E-01	8.2564E+13	0.54	8.2301E+13	8.2561E+13	8.2137E+13	8.2385E+13	1.0195E+12	1.2669E+12
1.00E-01–1.50E-01	5.2187E+14	3.39	5.2088E+14	5.2186E+14	5.1975E+14	5.2070E+14	1.5466E+11	1.1027E+12
1.50E-01–2.00E-01	1.7043E+13	0.11	1.7034E+13	1.7042E+13	7.2385E+11	7.3194E+11	2.8069E+09	6.7237E+09
2.00E-01–3.00E-01	2.2966E+13	0.15	2.2503E+13	2.2957E+13	2.0490E+13	2.0944E+13	7.5808E+11	1.2121E+12
3.00E-01–4.00E-01	5.9055E+12	0.04	5.5834E+12	5.8877E+12	1.0768E+12	1.3810E+12	4.8725E+11	5.4850E+11
4.00E-01–4.50E-01	7.5560E+13	0.49	7.1959E+13	7.5550E+13	2.2178E+12	5.8085E+12	2.4401E+08	7.4297E+08
4.50E-01–5.10E-01	7.7275E+13	0.50	7.7212E+13	7.7213E+13	5.3003E+13	5.3003E+13	5.2097E+13	5.2097E+13
5.10E-01–5.12E-01	7.1152E+14	4.62	0.0000E+00	7.1152E+14	0.0000E+00	7.1152E+14	0.0000E+00	4.6534E-01
5.12E-01–6.00E-01	8.4522E+14	5.49	8.4482E+14	8.4521E+14	8.4481E+14	8.4521E+14	8.2556E+14	8.2556E+14
6.00E-01–7.00E-01	8.0496E+15	52.26	3.4928E+15	8.0494E+15	3.3980E+15	7.9547E+15	3.3908E+15	7.5228E+15
7.00E-01–8.00E-01	3.0472E+15	19.78	3.0452E+15	3.0458E+15	3.0403E+15	3.0409E+15	2.9702E+15	2.9702E+15
8.00E-01–1.00E+00	4.0222E+14	2.61	3.8703E+14	4.0221E+14	3.7416E+14	3.8934E+14	3.0217E+14	3.0217E+14
1.00E+00–1.33E+00	3.2007E+14	2.08	2.5133E+14	3.2005E+14	2.5130E+14	3.2001E+14	9.6657E+13	9.6657E+13
1.33E+00–1.50E+00	1.2260E+14	0.80	1.1008E+14	1.2259E+14	1.0688E+14	1.1939E+14	1.0471E+14	1.0472E+14
1.50E+00–1.66E+00	1.2154E+13	0.08	6.8326E+12	1.2154E+13	5.2354E+12	1.0557E+13	0.0000E+00	4.4618E+06
1.66E+00–2.00E+00	3.0706E+12	0.02	1.9888E+10	3.0705E+12	1.6093E+10	3.0666E+12	0.0000E+00	1.2592E+07
2.00E+00–2.50E+00	3.1652E+13	0.21	1.7849E+08	3.1653E+13	0.0000E+00	3.1653E+13	0.0000E+00	4.5348E+07
2.50E+00–3.00E+00	5.0935E+11	0.00	0.0000E+00	5.0911E+11	0.0000E+00	5.0911E+11	0.0000E+00	8.0989E-01
3.00E+00–3.50E+00	5.3885E+10	0.00	0.0000E+00	5.3885E+10	0.0000E+00	5.3885E+10	0.0000E+00	2.3726E-01
Total sources								
$\gamma/(s \text{ tIHM})$	1.5402E+16	100	9.6349E+15	1.5400E+16	9.2040E+15	1.4969E+16	7.8191E+15	1.2288E+16
H*(10) ($\mu\text{Sv/h}$)	5.7635E+07		3.7305E+07	5.7626E+07	3.6701E+07	5.7018E+07	3.4551E+07	5.0578E+07
0.95 Confidence interval	[5.7066E+07, 5.8204E+07]		[3.6943E+07, 3.7667E+07]	[5.7058E+07, 5.8195E+07]	[3.6344E+07, 3.7057E+07]	[5.6456E+07, 5.7580E+07]	[3.4213E+07, 3.4888E+07]	[5.0080E+07, 5.1077E+07]

Units: $\gamma/(s \text{ tIHM})$ means gamma number per second and per ton of initial heavy metal.

Table 4. Incomplete inventory of mixed oxide (U,Pu)O₂ at time *t*, incomplete inventory after a 2- and a 3-year cooling, and reconstructed inventory of mixed oxide (U,Pu)O₂ at time *t* obtained by reconstructing the complete inventory.

Incomplete inventory		Incomplete inventory		Incomplete inventory		Reconstructed inventory	
		After <i>d</i> = 2 years cooling		After <i>d</i> = 3 years cooling			
Isotope	%Mass	Isotope	%Mass	Isotope	%Mass	Isotope	%Mass
²³² U	6.7439 × 10 ⁻⁰⁶	²³² U	8.9840 × 10 ⁻⁰⁶	²³² U	9.7182 × 10 ⁻⁰⁶	²³² U	6.7439 × 10 ⁻⁰⁶
²³⁴ U	3.3720 × 10 ⁻⁰²	²³⁴ U	3.7186 × 10 ⁻⁰²	²³⁴ U	3.8899 × 10 ⁻⁰²	²³⁴ U	3.3720 × 10 ⁻⁰²
²³⁵ U	6.8339 × 10 ⁻⁰¹	²³⁵ Fu	6.8383 × 10 ⁻⁰¹	²³⁵ Fu	6.8405 × 10 ⁻⁰¹	²³⁵ Fu	6.8339 × 10 ⁻⁰¹
²³⁶ U	3.1323 × 10 ⁻⁰¹	²³⁶ U	3.1385 × 10 ⁻⁰¹	²³⁶ U	3.1417 × 10 ⁻⁰¹	²³⁶ U	3.1323 × 10 ⁻⁰¹
²³⁸ U	7.3900 × 10 ⁺⁰¹	²³⁸ U	7.3900 × 10 ⁺⁰¹	²³⁸ U	7.3901 × 10 ⁺⁰¹	²³⁸ U	7.3900 × 10 ⁺⁰¹
²³⁸ Pu	6.3472 × 10 ⁻⁰⁶	²³⁶ Pu	3.9078 × 10 ⁻⁰⁶	²³⁶ Pu	3.0663 × 10 ⁻⁰⁶	²³⁶ Pu	6.3472 × 10 ⁻⁰⁶
²³⁹ Pu	2.2480 × 10 ⁻⁰¹	²³⁸ Pu	2.2127 × 10 ⁻⁰¹	²³⁸ Pu	2.1953 × 10 ⁻⁰¹	²³⁸ Pu	2.2480 × 10 ⁻⁰¹
²⁴⁰ Pu	7.5890 × 10 ⁺⁰⁰	²³⁹ Pu	7.5886 × 10 ⁺⁰⁰	²³⁹ Pu	7.5884 × 10 ⁺⁰⁰	²³⁹ Pu	7.5890 × 10 ⁺⁰⁰
²⁴¹ Pu	3.0414 × 10 ⁺⁰⁰	²⁴⁰ Pu	3.0408 × 10 ⁺⁰⁰	²⁴⁰ Pu	3.0404 × 10 ⁺⁰⁰	²⁴⁰ Pu	3.0414 × 10 ⁺⁰⁰
²⁴² Pu	1.4546 × 10 ⁺⁰⁰	²⁴¹ Pu	1.3205 × 10 ⁺⁰⁰	²⁴¹ Pu	1.2581 × 10 ⁺⁰⁰	²⁴¹ Pu	1.4546 × 10 ⁺⁰⁰
²⁴¹ Am	6.8763 × 10 ⁻⁰¹	²⁴² Pu	6.8763 × 10 ⁻⁰¹	²⁴² Pu	6.8764 × 10 ⁻⁰¹	²⁴² Pu	6.8763 × 10 ⁻⁰¹
	2.2659 × 10 ⁻⁰¹	²⁴¹ Am	3.5978 × 10 ⁻⁰¹	²⁴¹ Am	4.2150 × 10 ⁻⁰¹	²⁴¹ Am	2.2659 × 10 ⁻⁰¹
		²³⁷ Np	9.3021 × 10 ⁻⁰⁴	²³⁷ Np	1.5473 × 10 ⁻⁰³	²³⁷ Np	5.5413 × 10 ⁻⁰⁴
		⁴ He	9.3097 × 10 ⁻⁰⁵	⁴ He	1.4186 × 10 ⁻⁰⁴	⁴ He	1.2727 × 10 ⁻⁰⁴
		²³⁰ Th	1.9663 × 10 ⁻⁰⁷	²³⁰ Th	3.0211 × 10 ⁻⁰⁷	²³⁰ Th	2.5852 × 10 ⁻⁰⁷
		²²⁸ Th	1.1241 × 10 ⁻⁰⁷	²²⁸ Th	1.5511 × 10 ⁻⁰⁷	²²⁸ Th	7.6371 × 10 ⁻⁰⁸
		²³⁷ U	4.1249 × 10 ⁻⁰⁸	²⁰⁸ Pb	8.2852 × 10 ⁻⁰⁸	²³⁷ U	4.5437 × 10 ⁻⁰⁸
		²⁰⁸ Pb	3.8641 × 10 ⁻⁰⁸	²³⁷ U	3.9301 × 10 ⁻⁰⁸	²⁰⁸ Pb	2.8960 × 10 ⁻⁰⁸
		²³² Th	1.8028 × 10 ⁻⁰⁸	²³² Th	2.7056 × 10 ⁻⁰⁸	²³² Th	2.6975 × 10 ⁻⁰⁸
		²³⁵ Mu	1.5287 × 10 ⁻⁰⁸	²³⁵ Mu	1.5287 × 10 ⁻⁰⁸	²³⁵ Mu	1.5288 × 10 ⁻⁰⁸
		²³¹ Pa	1.3207 × 10 ⁻⁰⁹	²³¹ Pa	1.9828 × 10 ⁻⁰⁹	²³¹ Pa	1.9809 × 10 ⁻⁰⁹
		²³⁴ Th	1.0725 × 10 ⁻⁰⁹	²³⁴ Th	1.0725 × 10 ⁻⁰⁹	²³⁴ Th	1.0725 × 10 ⁻⁰⁹
		²²⁴ Ra	5.7196 × 10 ⁻¹⁰	²²⁴ Ra	7.9115 × 10 ⁻¹⁰	²²⁴ Ra	3.8825 × 10 ⁻¹⁰
		²³³ U	2.4437 × 10 ⁻¹⁰	²³³ U	6.1565 × 10 ⁻¹⁰	²³³ U	1.6196 × 10 ⁻¹⁰
		²¹² Pb	6.5873 × 10 ⁻¹¹	²¹² Pb	9.1144 × 10 ⁻¹¹	²¹² Pb	4.4710 × 10 ⁻¹¹
		²³³ Pa	2.9570 × 10 ⁻¹¹	²³³ Pa	5.0174 × 10 ⁻¹¹	²³³ Pa	1.7562 × 10 ⁻¹¹
		²¹² Fu	6.2463 × 10 ⁻¹²	²¹² Fu	8.6428 × 10 ⁻¹²	²¹² Fu	4.2395 × 10 ⁻¹²
		²³¹ Th	2.7804 × 10 ⁻¹²	²²⁶ Ra	3.9943 × 10 ⁻¹²	²²⁶ Ra	3.4016 × 10 ⁻¹²
		²²⁶ Ra	1.7466 × 10 ⁻¹²	²³¹ Th	2.7813 × 10 ⁻¹²	²³¹ Th	2.7786 × 10 ⁻¹²
		²⁰⁸ Tl	1.1104 × 10 ⁻¹³	²⁰⁸ Tl	1.5364 × 10 ⁻¹³	²⁰⁸ Tl	7.5364 × 10 ⁻¹⁴
O	1.1845 × 10 ⁺⁰¹	O	1.1845 × 10 ⁺⁰¹	O	1.1845 × 10 ⁺⁰¹	O	1.1845 × 10 ⁺⁰¹
Sum	1.0000 × 10 ⁺⁰²	Sum	1.0000 × 10 ⁺⁰²	Sum	1.0000 × 10 ⁺⁰²	Sum	1.0000 × 10 ⁺⁰²

geometrical configuration is the same as that of the first example given in this paper.

Ambient dose equivalent H*(10) is calculated in two steps as explained in the first example of this paper.

Three different ways are used to assess the ambient dose:

- rough (or basic) calculation of the ambient dose, using the incomplete isotopic inventory known at time *t*;
- simple calculation of the ambient dose, using the incomplete inventory decayed from time *t* to time *t* + *d* (*d* small); and
- calculation of the ambient dose with complete isotopic inventory reconstruction (IIR) at time *t*.

The first way to assess the dose (rough calculation) consists in calculating the neutron and photon

sources directly from the incomplete isotopic inventory and then the dose.

The second way to assess the dose (simple calculation) consists in decaying during a short cooling period '*d*' the incomplete inventory with DARWIN. That allows obtaining some radioactive progeny to be taking into account in the radioactive sources determination. Then the dose rate is assessed from these sources. This calculation has the advantage of being simple and fast to implement but can be misleading for two reasons. First the minor isotopes quantities at *t*+*d* after the short cooling time are inaccurate because they have not been calculated with the correct quantities from the beginning. Secondly the principal isotope quantities are also modified especially when their decay rates are high.

The third way to assess the dose rate consists in reconstructing the complete isotopic inventory with

Table 5. Relative difference (expressed in percentage of incomplete inventory quantities) between the principal isotope quantities of reconstructed inventory and incomplete inventory and incomplete inventory decayed d years ($d = 2$ or 3) and incomplete inventory.

Principal isotope	Relative difference between reconstructed and incomplete inventories (%)	Relative difference between incomplete after 2-year decay and incomplete inventories (%)	Relative difference between incomplete after 3-year decay and incomplete inventories (%)
^{232}U	0.00	33.23	44.21
^{234}U	0.00	10.31	15.35
^{235}U	0.00	0.59	0.59
^{236}U	0.00	1.29	1.29
^{238}U	0.00	0.00	0.00
^{236}Pu	0.00	-38.43	-51.65
^{238}Pu	0.00	0.45	0.00
^{239}Pu	0.00	0.00	0.00
^{240}Pu	0.00	0.00	0.00
^{241}Pu	0.00	-8.97	-13.10
^{242}Pu	0.00	-0.29	-0.29
^{241}Am	0.00	56.52	83.48

the method explained in this paper, in order to calculate precisely neutron and gamma sources and the related dose rates.

Table 4 lists the incomplete inventory, the incomplete inventories after a 2- and a 3-year cooling time and the reconstructed inventory obtained with the reconstruction method.

The incomplete inventory is known with precision better than 10 ppm. Sensitivity analysis has shown that these uncertainties do not impact the results of Table 4 and 5: all the masses, even the one of the most minor isotope (Tl 208) are left unchanged when the main isotopes are perturbed with errors of magnitude lower than 10 ppm. However, a complete uncertainty analysis with designs of experiments would be required to provide confidence intervals.

The quantities of each isotope of the incomplete and reconstructed inventories in Table 4 given with two decimals accuracy are the same (cf. Table 5). The accuracy of the method depends only on the uncertainties on the input data, as DARWIN solves with great accuracy the generalized Bateman equations.

On the contrary, the simple calculation with incomplete inventory during a short cooling time yields large differences especially for ^{232}U , ^{236}Pu and ^{241}Am isotopes (cf. Table 5). It demonstrates that this way to assess the dose rate is not accurate and modifies some principal isotopes quantities.

Comparison of neutron and gamma photon emitted by reconstructed and incomplete isotopic inventories

Neutron and gamma sources are calculated using DARWIN. The intensity of the neutron source of the reconstructed inventory is almost identical to that of the incomplete inventory (9.8×10^6 n/s). The neutron

intensity of the incomplete inventory after the 2-year cooling is slightly superior (1.01×10^7 n/s).

The gamma emission intensity of the reconstructed inventory (1.17×10^{13} γ /s) is close to that of the incomplete inventory (1.15×10^{13} γ /s). But the gamma emission spectrum differs between the complete and the incomplete inventory, as shown in Figure 5. The two spectra differ from the 1.2 MeV-energy and up. From this energy the intensity of the reconstructed inventory averages around 10^6 γ /s whereas gamma emissions of the incomplete inventory are null. The gamma rays between 1.2 and 3 MeV in the reconstructed inventory are emitted by minor isotopes by cumulative 3-year decay. Although these rays have a small intensity (around 10^6 γ /s compared with the total gamma emission intensity of 1.17×10^{13} γ /s), they result in a significant increase of gamma-dose, because of the high level of their energies.

The gamma-total emission intensity of the incomplete inventory after a 2- and 3-year cooling period is respectively about 1.8×10^{13} and 2.1×10^{13} γ /s. These values are above the total intensity of the complete and also the incomplete inventories. The gamma-spectrum of the incomplete inventory after 3-year decay is given in Figure 5. This figure shows the small differences between this spectrum and the reconstructed inventory spectrum.

Comparison of ambient dose equivalent rate

Neutron dose rate is the same whether the isotopic inventory is complete or not (cf. Table 6). But this dose rate is higher for the inventory after a 2- or 3-year cooling.

The dose rate due to gamma coming from (n, γ) reactions is also the same for the reconstructed and

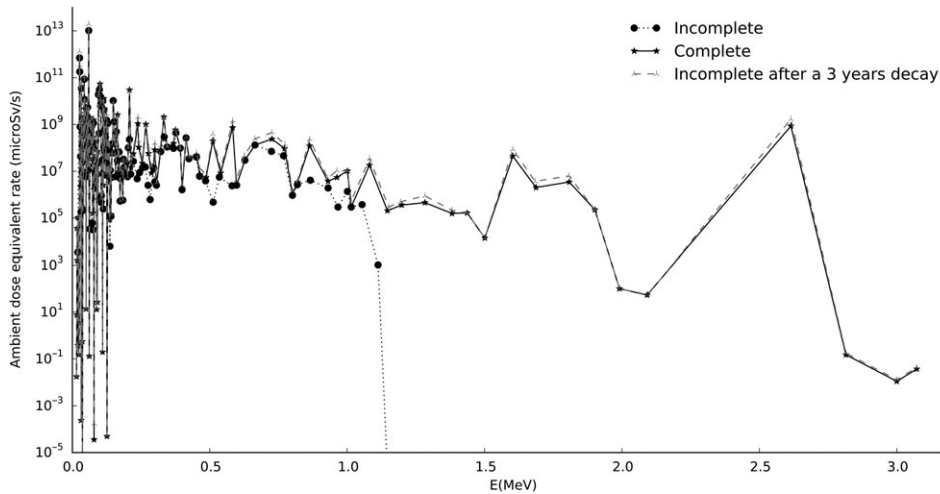


Figure 5. Gamma spectrum related to the three inventories: reconstructed, incomplete and incomplete after a 3 years cooling time.

Table 6. Dose equivalent rate calculated with incomplete and complete inventories.

Ambient dose equivalent rate ($\mu\text{Sv/h}$)	Incomplete inventory at time t	Incomplete inventory after 2-year decay	Incomplete inventory after 3-year decay	Complete inventory at time t
n	$8.63 \times 10^{+00}$	$8.87 \times 10^{+00}$	$9.06 \times 10^{+00}$	$8.63 \times 10^{+00}$
(n, γ)	$1.69 \times 10^{+00}$	$1.73 \times 10^{+00}$	$1.77 \times 10^{+00}$	$1.69 \times 10^{+00}$
γ	4.14×10^{-01}	$4.62 \times 10^{+01}$	$6.35 \times 10^{+01}$	$3.26 \times 10^{+01}$
Total	$1.07 \times 10^{+01}$	$5.68 \times 10^{+01}$	$7.43 \times 10^{+01}$	$4.29 \times 10^{+01}$

the incomplete inventories (as expected since the neutron emission source is identical for the two inventories) and is slightly higher for inventory after 2- or 3-year cooling. This dose is negligible if it is compared with gamma-dose (gamma emitted directly from the composition) for the reconstructed inventory and the 3-year decayed inventory.

The calculation with inventory after a 3-year cooling period (and also to a lesser extent with inventory after 2-year cooling period) overestimates the dose, compared with the reconstructed inventory. The calculation with the reconstructed inventory gives the most accurate estimation of the dose. All the digits of Table 6 remain the same when the incomplete inventory is perturbed by 10 ppm or less, so, in this application case, the results obtained for three computations are statistically different.

The gamma-dose rate (due to gamma emitted directly from the composition) is 78 times higher with the reconstructed inventory. As to the total dose, it is four times higher with reconstructed inventory.

The NARMER code calculates each gamma-energy bin contribution to the gamma-dose. The

energy group between 2.33 and 2.67 MeV contributes the most to the gamma-dose (see Figure 6). From a cross-check with DARWIN results, it is possible to see that the progeny isotope ^{208}Tl contributes the most to the gamma-dose rate. Its contribution amounts to around 94%.

Effect of the shielding configuration

Furthermore, four shielding settings have been studied:

- (1) without shielding;
- (2) with a 3 cm thick stainless-steel shield;
- (3) with a 3 cm thick stainless-steel shield and a 5 cm thick shield of Kyowa Glass; and
- (4) with a 3 cm thick stainless-steel shield, a close radiation shielding sheet made of PPB and a 5 cm thick shield of Kyowa Glass.

The consideration of the reconstructed inventory instead of the incomplete inventory affects only the gamma dose. The relative difference between the gamma dose of complete and incomplete inventory

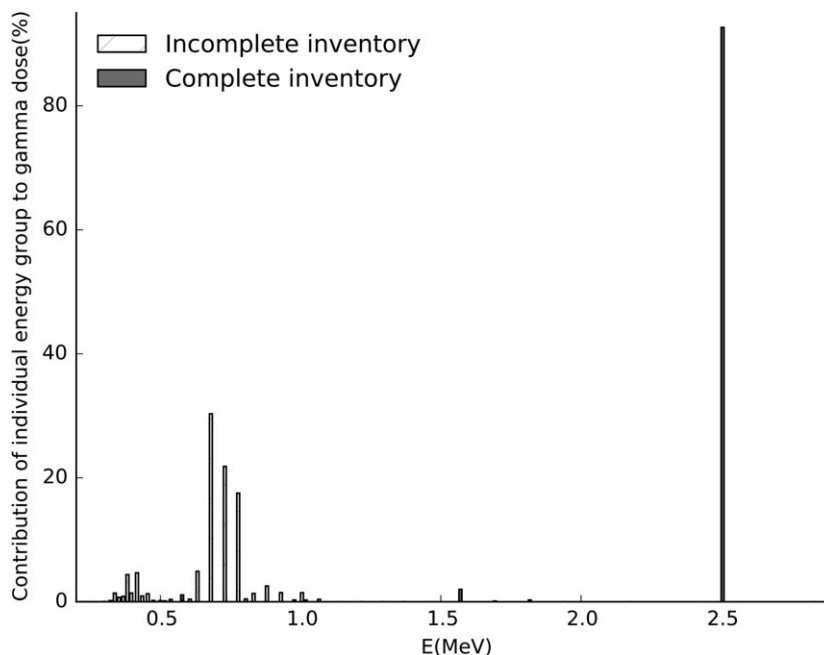


Figure 6. Contribution of each energetic group to the gamma dose with reconstructed and incomplete inventory.

is almost constant and around 98%, whatever the shielding setting is. However the total dose computed with the incomplete inventory is underestimated by 25–75% according to the shielding setting considered (from 1 to 4) because the neutron dose part decreases compared with the gamma dose part. The improvement is much more important with the shielding setting number 4 (total dose four times higher) than with the others shielding settings (total dose 1.5–2 times higher). Indeed the 'Kyowa Glass' shielding stops most of neutron and gamma rays, but not the energetic gamma rays around the 2 MeV-energy emitted by minor isotopes. Close radiation shielding sheet (made of bore) decreases much more the neutron dose (divided by 10) than the gamma-dose (divided by 2), then the gamma-dose contribution to the total dose increases with setting number 4 compared with the other shielding settings.

CONCLUSION

The IIR-method presented in this paper allows calculating more accurately the ambient dose rate produced by radioactive materials that have decayed for a known duration. In typical setups with uranium and plutonium oxide, the dose rate computed with the incomplete inventory is roughly underestimated whereas the dose rates calculated by other approximate methods are strongly dependent on the

methods. A simple resolution of the inverse problem of radioactive decay provides a correct means to complete the inventory and compute better dose rate estimates in most cases. In a future work, sensitivity analysis will enable to compute confidence intervals for the dose and set limits for the application of the new method.

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