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Dating a nuclear event using Nb-95/Zr-95 isotopic activity ratio

Kassoum Yamba[1*], Oumar Sanogo[1], Martin B. Kalinowski[2], Jean Koulidiati[3]

[1]Centre National de Recherche Scientifique et Technologique (IRSAT/CNRST), Ouagadougou, Burkina Faso
[2]Comprehensive nuclear Test-Ban Treaty Organization (CTBTO), Vienna, Austria
[3]Université Joseph Ki Zerbo, Ouagadougou, Burkina Faso

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Abstract : This work investigates the use of the radionuclides Zr-95, Nb-95m and Nb-95 for dating of a nuclear event, for different scenarios of release. Unlike others studies on this issue, we present general formulas (without any simplification) of the isotopic activity ratio Nb-95/Zr-95 taking into account the decay of Nb-95m. Parameters and it uncertainties are well calculated taking into account two databases like ENDF.B.VII.I and LARA/LNHB-CEA. An example of application is made using proposed constants in order to validate our results. Results are very interesting and can be useful for a fast assessment of nuclear event zero time.

Keywords : Zero-time, Isotopic Activity ratio, Nb-95, Zr-95

Datation d'un évènement nucléaire par l'utilisation du rapport d'activité isotopique Nb-95/Zr-95

Résumé : Ce travail porte sur l'utilisation des radionucléides Zr-95, Nb-95m et Nb-95 dans le cadre de la datation des évènements nucléaires, pour différents scénarios de libération radioactive. Sans aucune simplification de calcul, les formules générales des rapports d'activités Nb-95/Zr-95 sont établies tout en tenant en compte la décroissance de l'élément intermédiaire Nb-95m. Des paramètres ainsi que leurs incertitudes sont évalués en considérant deux bases de données à savoir ENDF.B.VII.I and LARA/LNHB-CEA. Afin de valider nos résultats, une application est faite en utilisant des données d'observations réelles du système de surveillance internationale (SSI) de l'Organisation du traité d'interdiction complète des essais nucléaires (OTICE). Les résultats peuvent être utilisés pour une évaluation rapide du temps zéro d'un évènement nucléaire.

Mots clés : Temps-zéro, Rapport d'activité, Nb-95, Zr-95

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[*] **Auteur de Correspondance**
Kassoum Yamba
Centre National de Recherche Scientifique et Technologique (IRSAT/CNRST), Ouagadougou, Burkina Faso
BP 7047 Ouagadougou – Burkina Faso
Phone: +22670032683 E-mail: fairlir@yahoo.fr

1. Introduction

The international monitoring system IMS built as part of verification regime of the Comprehensive Nuclear Test Ban Treaty Organization (CTBTO) comprises four monitoring technologies, namely, Infrasound for atmospheric tests, seismic for underground tests, hydro acoustic for underwater tests and radionuclides for all environments. The Comprehensive Nuclear Test Ban Treaty (CTBT) is not yet interred into force, but the Verification Regime is already almost established. Among the monitoring station established, 80 are radionuclides and of these, 40 are equipped with noble gases detection system. This radionuclides network includes 16 certified radionuclide laboratories. They may evaluate filter samples further if requested [1]. So, the radionuclide technology is complementary to the three waveform verification technologies employed by the CTBTO verification regime. This technology is the only one that is able to confirm whether an explosion detected and located by the others is indicative of a nuclear test.

Fission products Zr-95 and Nb-95 are radionuclides pairs with parent-daughter relationship. Their radioactive decay is an ideally suited chronometer for dating of nuclear events [2][3] and the accuracy of the time assessment can be long until 400 days after the event [4]. Zr95 and Nb95 are non-volatile fission products [5], and are therefore of high interest in the frame of Verification Regime of Comprehensive Nuclear Test Ban Treaty (CTBT).

Zr95 disintegrates by beta minus emissions to excited levels and to fundamental level of Nb-95, and the excited levels of Nb-95 disintegrates (for 97.5%) to the Nb-95 ground state. We can use these two radioactive decay modes to evaluate the age of nuclear event, by the analysis of the isotopic activity ratio Nb-95/Zr-95. But, The zero time can be calculate only when the activities of the pair are determined at a moment at which transient equilibrium has not been established between the two [6]. Thus in this paper, we have considered two kinds of release: a sudden release and a continuous release. In order to diversify data used in the algorithms, two nuclear databases were used, ENDF.B.VII.I [7] and LARA/LNHB-CEA [8].

2. Methodological Approach

We can distinguish two kinds of release. The first case is a sudden release of radionuclides, like the scenario of a nuclear explosion. The second case is a continuous release of radionuclides.

2.1. Isotopic activity ratio analysis in the case of a sudden release

Radioactive decay of Zr-95, Nb-95m and Nb-95 allows writing the following differential equations of decay in the case of a sudden release (nuclear explosion scenario):

$$System\ A: \begin{cases} \frac{\mathrm{dN_{Zr}(t)}}{\mathrm{dt}} + \lambda_{\mathrm{Zr}} \cdot \mathrm{N_{Zr}(t)} = 0 & (1) \\ \frac{\mathrm{dN_{Nbm}(t)}}{\mathrm{dt}} + \lambda_{\mathrm{Nbm}} \cdot \mathrm{N_{Nbm}(t)} = \mathrm{P_{Zr\to Nbm}} \cdot \lambda_{\mathrm{Zr}} \cdot \mathrm{N_{Zr}(t)} & (2) \\ \frac{\mathrm{dN_{Nb}(t)}}{\mathrm{dt}} + \lambda_{\mathrm{Nb}} \cdot \mathrm{N_{Nb}(t)} = \mathrm{P_{Zr\to Nb}} \cdot \lambda_{\mathrm{Zr}} \cdot \mathrm{N_{Zr}(t)} + \mathrm{P_{Nbm\to Nb}} \cdot \lambda_{\mathrm{Nbm}} \cdot \mathrm{N_{Nbm}(t)} & (3) \end{cases}$$

Where: λ_{Zr} is the decay constant of Zr-95, λ_{Nbm} the decay constant of Nb-95m, λ_{Nb} the decay constant of Nb-95, $P_{Zr\to Nbm}$ the probability of decay of the transition $Zr_{95} \to Nb_{95m}$, $P_{Zr\to Nb}$ the probability of decay of the transition $Zr_{95} \to Nb_{95}$, $P_{Nbm\to Nb}$ the probability of decay of the transition $Nb_{95m} \to Nb_{95}$, t : Time (for the value of each constant, see **Table I, Table II and Table III).**

We need some constants, like cumulative and independent yield fission product, to evaluate the activity ratio r(t) as part of dating of a nuclear event. The following table (**Table I**) shows cumulative and independent fission product (fission induced by thermal neutron) of particles Zr-95, Nb-95, Nb-95m [9, 10].

The following tables (**Table II and Table III**) show peak line, half-life, emission probability (or emission intensity) and decay probability (or Branching) of radionuclides Zr-95, Nb-95, Nb-95m, depending to the databases ENDF.B.VII.I and LARA.LNHB/CEA.

***Table I** : Cumulative and independent yields fission products of Zr95, Nb-95 and Nb-95m .*

	$Cumulative\ Yield\ (\%)$			$Independent\ Yield\ (\%)$		
	Zr_{95}	Nb_{95m}	Nb_{95}	Zr_{95}	Nb_{95m}	Nb_{95}
$ENDF.B.VII$ 2011	6.5027	$6.5052 \cdot 10^{-2}$	6.5029	$1.2724 \cdot 10^{-1}$	$2.4798 \cdot 10^{-5}$	$1.0599 \cdot 10^{-4}$
	± 0.0910	$\pm 0.0911 \cdot 10^{-2}$	± 0.0910	$\pm 0.2036 \cdot 10^{-1}$	$\pm 1.5871 \cdot 10^{-5}$	$\pm 0.67835 \cdot 10^{-4}$

Table II : Peak line, half-life and emission probability depending to the database.

		$Peak\ line\ (keV)$	$half-life\ (days)$	$Emission\ probability\ \%$
$ENDF.B.VII.I$	Zr_{95}	756.730(12)	64.0324(60)	54.38(22)
	Nb_{95m}	235.69(2)	3.60995(3000)	24.8300(7715)
	Nb_{95}	765.800(6)	34.9907(60)	99.810(7)
$LARA-LNHB/CEA$	Zr_{95}	756.729(12)	64.032(6)	54.38(22)
	Nb_{95m}	235.69(2)	3.61(3)	25.1(3)
	Nb_{95}	765.803(6)	34.991(6)	99.808(7)

Table III : Decay probability depending to the database.

	$Decay\ probability\ (or\ Branching)\ (\%)$		
	$Zr_{95} \to Nb_{95}$	$Zr_{95} \to Nb_{95m}$	$Nb_{95m} \to Nb_{95}$
$ENDF.B.VII.I$	98.9198(3118)	1.0802(3118)	94.4(6)
$LARA-LNHB/CEA$	98.90000(31257)	1.08(7)	97.5(1)

2.2. Isotopic activity ratio analysis in the case of a continuous release

The radioactive decay of the three particles Zr-95, Nb-95m and Nb-95 allows writing the following differential equations of decay in the case of a continuous release:

$$System\ B: \begin{cases} \dfrac{\mathrm{dN_{Zr}(t)}}{\mathrm{dt}} + \lambda_{\mathrm{Zr}} \cdot \mathrm{N_{Zr}(t)} = \mathrm{K_{Zr}} & (4) \\ \dfrac{\mathrm{dN_{Nbm}(t)}}{\mathrm{dt}} + \lambda_{\mathrm{Nbm}} \cdot \mathrm{N_{Nbm}(t)} = \mathrm{P_{Zr\to Nbm}} \cdot \lambda_{\mathrm{Zr}} \cdot \mathrm{N_{Zr}(t)} + \mathrm{K_{Nbm}} & (5) \\ \dfrac{\mathrm{dN_{Nb}(t)}}{\mathrm{dt}} + \lambda_{\mathrm{Nb}} \cdot \mathrm{N_{Nb}(t)} = \mathrm{P_{Zr\to Nb}} \cdot \lambda_{\mathrm{Zr}} \cdot \mathrm{N_{Zr}(t)} + \\ \quad \mathrm{P_{Nbm\to Nb}} \cdot \lambda_{\mathrm{Nbm}} \cdot \mathrm{N_{Nbm}(t)} + \mathrm{K_{Nb}} & (6) \end{cases}$$

Where K is the production rate, the others parameters are previously defined.

3. Results and Discussion

3.1. Sudden release scenario

By solving differential equations from the system A, we obtain:

$$A_{Zr}(t) = A_{Zr}(0) \cdot e^{-\lambda_{Zr} \cdot t} \qquad (7)$$

$$A_{Nbm}(t) = A_{Nbm}(0) \cdot e^{-\lambda_{Nbm} \cdot t} + P_{Zr\to Nbm} \cdot A_{Zr}(0) \cdot \frac{\lambda_{Nbm}}{\lambda_{NBm} - \lambda_{Zr}} \cdot \left(e^{-\lambda_{Zr} \cdot t} - e^{-\lambda_{Nbm} \cdot t}\right) \qquad (8)$$

$$\begin{aligned} A_{Nb}(t) = {} & A_{Nb}(0) \cdot e^{-\lambda_{Nb} \cdot t} + A_{Zr}(0) \\ & \cdot \left(\frac{\lambda_{Nb} P_{Zr\to Nb}}{\lambda_{Nb} - \lambda_{Zr}} + \frac{\lambda_{Nbm} \lambda_{Nb} P_{Zr\to Nbm} P_{Nbm\to Nb}}{(\lambda_{Nbm} - \lambda_{Zr})(\lambda_{Nb} - \lambda_{Zr})}\right) \\ & \cdot \left(e^{-\lambda_{Zr} \cdot t} - e^{-\lambda_{Nb} \cdot t}\right) + (A_{Nbm}(0) \frac{\lambda_{Nb} P_{Nbm\to Nb}}{\lambda_{Nb} - \lambda_{Nbm}} \\ & - A_{Zr}(0) \cdot \frac{\lambda_{Nbm} \lambda_{Nb} P_{Zr\to Nbm} P_{Nbm\to Nb}}{(\lambda_{Nbm} - \lambda_{Zr})(\lambda_{Nb} - \lambda_{Nbm})}) \\ & \cdot \left(e^{-\lambda_{Nbm} \cdot t} - e^{-\lambda_{Nb} \cdot t}\right) \qquad (9) \end{aligned}$$

The activity ratio $r(t) = \frac{A_{Nb}(t)}{A_{Zr}(t)}$ of radionuclides Nb95 and Zr95 is given by:

$$\begin{aligned} r(t) & \\ = {} & \frac{A_{Nb}(0)}{A_{Zr}(0)} e^{(\lambda_{Zr} - \lambda_{Nb})t} \\ & + \left(\frac{\lambda_{Nb} P_{Zr\to Nb}}{\lambda_{Nb} - \lambda_{Zr}} + \frac{\lambda_{Nbm} \lambda_{Nb} P_{Zr\to Nbm} P_{Nbm\to Nb}}{(\lambda_{Nbm} - \lambda_{Zr})(\lambda_{Nb} - \lambda_{Zr})}\right) \left(1 - e^{(\lambda_{Zr} - \lambda_{Nb})t}\right) \\ & + \left(\frac{A_{Nbm}(0)}{A_{Zr}(0)} \frac{\lambda_{Nb} P_{Nbm\to Nb}}{\lambda_{Nb} - \lambda_{Nbm}} - \frac{\lambda_{Nbm} \lambda_{Nb} P_{Zr\to Nbm} P_{Nbm\to Nb}}{(\lambda_{Nbm} - \lambda_{Zr})(\lambda_{Nb} - \lambda_{Nbm})}\right) \left(e^{(\lambda_{Zr} - \lambda_{Nbm})t} \right. \\ & \left. - e^{(\lambda_{Zr} - \lambda_{Nb})t}\right) \qquad (10) \end{aligned}$$

From this equation above (see eq.[10]), we notice two important factors, like the initial activities ratios of Zr-95, Nb-95m and Nb-95. We can admit that if the release results from an atmospheric test (direct release), so the initial activities ratios are not nulls and can be rewrite as a function of decay constant and fission yield (see **Table I**). However, if the release results from an underground test, we can assume that Zr-95 diffuses through the earth crust, and the initial activities ratios are nulls because $A_{Nb}(0) = A_{Nbm}(0) = 0$. This kind of situation is already studied by many others authors on the use of these radionuclides pairs.

The initial activities ratios, if non-nulls, can be rewrite by making an approximation, as follow:

$$\frac{A_{Nb}(0)}{A_{Zr}(0)} = \frac{\lambda_{Nb} Y_{Nb}}{\lambda_{Zr} Y_{Zr}} \qquad (11)$$

$$\frac{A_{Nbm}(0)}{A_{Zr}(0)} = \frac{\lambda_{Nbm} Y_{Nbm}}{\lambda_{Zr} Y_{Zr}} \qquad (12)$$

Where: Y_{Zr} is the cumulative fission yield of fission of Zr-95, $Y_{Nbm}$is the independent fission yield of fission of Nb-95m, and $Y_{Nb}$is the independent fission yield of fission of Nb-95 (for the value of each constant, see **Table I**).

In order to make simplifications, and make it easier of using, formula_7 of the activity ratio can be rewrite as follow, where K_1, K_2, K_3 a_1 and a_2 are constants that are well evaluated in our work and given in **Table IV**.

Table IV : Some constants used for a fast and accurate assessment of zero time in the case of continuous nuclear reactor release

	Sudden Release		Continuous Release	
	ENDF.B.VII.I	LARA	ENDF.B.VII.I	LARA
K_1	−2.20610(978)	−2.20650(714)	−1.2055(107)	−1.2058(79)
K_2	1.2358(3594)e^{-3}	1.2761(856)e^{-3}	7.0084(493970)e^{-5}	7.2374(129470)e^{-5}
K_3	2.20490(977)	2.20520(714)	2.20490(977)	2.20520(714)
a_1	−8.98450(355)e^{-3}	−8.98430(355)e^{-3}	−8.98450(355)e^{-3}	−8.98430(355)e^{-3}
a_2	−1.81185(1596)e^{-1}	−1.81183(1596)e^{-1}	−1.81185(1596)e^{-1}	−1.81183(1596)e^{-1}

$$r(t) = K_1 \cdot e^{a_1 t} + K_2 \cdot e^{a_2 t} + K_3 \qquad (13)$$

As we can remark, it is not possible to solve analytically the time t from equation of r(t). So the age of release is derived by the numerical solving of equation 7 for t. We can use Newton's solving method to evaluate the time t numerically.

Thus, we introduce a new function F(t) as follows:

$$F(t) = K_1 \cdot e^{a_1 t} + K_2 \cdot e^{a_2 t} + K_3 - r \qquad (14)$$

Where: $F(t)$: is the Newton's function, r is the measured activity ratio at the reference time by gamma spectrometry.

The differential of F(t) can be write by:

$$F'(t) = K_1 a_1 \cdot e^{a_1 t} + K_2 a_2 \cdot e^{a_2 t} \qquad (15)$$

According to the Newton's solving method, we have:

$$t_{i+1} = t_i - \frac{F(t_i)}{F'(t_i)} \qquad (16)$$

By assuming the time (t) is depending to the following parameters K_1, K_2, K_3, a_1, a_2 and r, the uncertainty of the time (t) is given by:

$$\Delta t = \sqrt{\left(\frac{\partial t}{\partial K_1}\right)^2 (\Delta K_1)^2 + \left(\frac{\partial t}{\partial K_2}\right)^2 (\Delta K_2)^2 + \left(\frac{\partial t}{\partial K_3}\right)^2 (\Delta K_3)^2 + \left(\frac{\partial t}{\partial a_1}\right)^2 (\Delta a_1)^2 + \left(\frac{\partial t}{\partial a_2}\right)^2 (\Delta a_2)^2 + \left(\frac{\partial t}{\partial r}\right)^2 (\Delta r)^2} \qquad (17)$$

Where:

$$\frac{\partial t}{\partial K_1} = \frac{\frac{\partial F}{\partial K_1}}{\frac{\partial F}{\partial t}} = \frac{e^{a_1 t}}{F'(t)} = \frac{e^{a_1 t}}{K_1 a_1 \cdot e^{a_1 t} + K_2 a_2 \cdot e^{a_2 t}} \tag{18}$$

$$\frac{\partial t}{\partial K_2} = \frac{\frac{\partial F}{\partial K_2}}{\frac{\partial F}{\partial t}} = \frac{e^{a_2 t}}{F'(t)} = \frac{e^{a_2 t}}{K_1 a_1 \cdot e^{a_1 t} + K_2 a_2 \cdot e^{a_2 t}} \tag{19}$$

$$\frac{\partial t}{\partial K_3} = \frac{\frac{\partial F}{\partial K_3}}{\frac{\partial F}{\partial t}} = \frac{1}{F'(t)} = \frac{1}{K_1 a_1 \cdot e^{a_1 t} + K_2 a_2 \cdot e^{a_2 t}} \tag{20}$$

$$\frac{\partial t}{\partial a_1} = \frac{\frac{\partial F}{\partial a_1}}{\frac{\partial F}{\partial t}} = \frac{tK_1 e^{a_1 t}}{F'(t)} = \frac{tK_1 e^{a_1 t}}{K_1 a_1 \cdot e^{a_1 t} + K_2 a_2 \cdot e^{a_2 t}} \tag{21}$$

$$\frac{\partial t}{\partial a_2} = \frac{\frac{\partial F}{\partial a_2}}{\frac{\partial F}{\partial t}} = \frac{tK_2 e^{a_2 t}}{F'(t)} = \frac{tK_2 e^{a_2 t}}{K_1 a_1 \cdot e^{a_1 t} + K_2 a_2 \cdot e^{a_2 t}} \tag{22}$$

$$\frac{\partial t}{\partial r} = \frac{\frac{\partial F}{\partial r}}{\frac{\partial F}{\partial t}} = \frac{-1}{F'(t)} = \frac{-1}{K_1 a_1 \cdot e^{a_1 t} + K_2 a_2 \cdot e^{a_2 t}} \tag{23}$$

Using the gamma spectrometry method, we can calculate the activity of these various radionuclides. It is here important to take into account the decay of these particles during the acquisition time. This approach is well describe by using two affiliated radionuclides for dating the sample age. However, our calculations give activities for three radionuclides with affiliation. Formulas allowing to calculate isotopic activities of Zr-95, Nb-95 and Nb-95m using gamma spectrometry are given in eq.[4], [5] and[6], where T is the acquisition time, ccf is the coincidence correction factor, N is the net peak are, P is the emission intensity, and ε the detector efficiency.

$$A_{Zr} = \frac{\lambda_{Zr} N_{Zr} ccf_{Zr}}{\varepsilon_{Zr} P_{Zr} (1 - e^{-\lambda_{Zr} T})} \tag{24}$$

$$\begin{aligned} A_{Nbm} = {} & \frac{\lambda_{Nbm} N_{Nbm} ccf_{Nbm}}{\varepsilon_{Nbm} P_{Nbm} (1 - e^{-\lambda_{Nbm} T})} - P_{Zr\to Nbm} \cdot A_{Zr} \cdot \frac{\lambda_{Nbm}}{\lambda_{NBm} - \lambda_{Zr}} \\ & \cdot \left[\frac{\lambda_{Nbm}}{\lambda_{Zr}} \left(\frac{1 - e^{-\lambda_{Zr} T}}{1 - e^{-\lambda_{Nbm} T}} \right) - 1 \right] \end{aligned} \tag{25}$$

$$\begin{aligned} A_{Nb} = {} & \frac{\lambda_{Nb} N_{Nb} ccf_{Nb}}{\varepsilon_{Nb} P_{Nb} (1 - e^{-\lambda_{Nb} T})} - A_{Zr} \left(\frac{\lambda_{Nb} P_{Zr\to Nb}}{\lambda_{Nb} - \lambda_{Zr}} + \frac{\lambda_{Nbm} \lambda_{Nb} P_{Zr\to Nbm} P_{Nbm\to Nb}}{(\lambda_{Nbm} - \lambda_{Zr})(\lambda_{Nb} - \lambda_{Zr})} \right) \\ & \cdot \left[\frac{\lambda_{Nb}}{\lambda_{Zr}} \cdot \left(\frac{1 - e^{-\lambda_{Zr} T}}{1 - e^{-\lambda_{Nb} T}} \right) - 1 \right] - (A_{Nbm} \frac{\lambda_{Nb} P_{Nbm\to Nb}}{\lambda_{Nb} - \lambda_{Nbm}} \\ & - A_{Zr} \cdot \frac{\lambda_{Nbm} \lambda_{Nb} P_{Zr\to Nbm} P_{Nbm\to Nb}}{(\lambda_{Nbm} - \lambda_{Zr})(\lambda_{Nb} - \lambda_{Nbm})}) \\ & \cdot \left[\frac{\lambda_{Nb}}{\lambda_{Nbm}} \cdot \left(\frac{1 - e^{-\lambda_{Nbm} T}}{1 - e^{-\lambda_{Nb} T}} \right) - 1 \right] \end{aligned} \tag{26}$$

The measured activity ratio ($r = \frac{A_{Nb}}{A_{Zr}}$), at the reference time, by using the gamma ray spectrometry, is given by:

$$\begin{aligned} r = {} & \frac{\lambda_{Nb} N_{Nb} ccf_{Nb} \varepsilon_{Zr} P_{Zr}}{\lambda_{Zr} N_{Zr} ccf_{Zr} \varepsilon_{Nb} P_{Nb}} \frac{(1 - e^{-\lambda_{Zr} T})}{(1 - e^{-\lambda_{Nb} T})} \\ & - \left(\frac{\lambda_{Nb} P_{Zr\to Nb}}{\lambda_{Nb} - \lambda_{Zr}} + \frac{\lambda_{Nbm} \lambda_{Nb} P_{Zr\to Nbm} P_{Nbm\to Nb}}{(\lambda_{Nbm} - \lambda_{Zr})(\lambda_{Nb} - \lambda_{Zr})} \right) \\ & \cdot \left[\frac{\lambda_{Nb}}{\lambda_{Zr}} \cdot \left(\frac{1 - e^{-\lambda_{Zr} T}}{1 - e^{-\lambda_{Nb} T}} \right) - 1 \right] - (\frac{A_{Nbm}}{A_{Zr}} \cdot \frac{\lambda_{Nb} P_{Nbm\to Nb}}{\lambda_{Nb} - \lambda_{Nbm}} \\ & - \frac{\lambda_{Nbm} \lambda_{Nb} P_{Zr\to Nbm} P_{Nbm\to Nb}}{(\lambda_{Nbm} - \lambda_{Zr})(\lambda_{Nb} - \lambda_{Nbm})}) \\ & \cdot \left[\frac{\lambda_{Nb}}{\lambda_{Nbm}} \cdot \left(\frac{1 - e^{-\lambda_{Nbm} T}}{1 - e^{-\lambda_{Nb} T}} \right) - 1 \right] \end{aligned} \tag{27}$$

3.2. Continuous release

By solving these differential equations from system B, we obtain the expressions of isotopic activities given in the following formulas:

$$A_{Zr}(t) = K_{Zr} \left(1 - e^{-\lambda_{Zr} t} \right) \tag{7}$$

$$\begin{aligned} A_{Nbm}(t) = {} & K_{Zr} P_{Zr\to Nbm} \frac{\lambda_{Nbm}}{\lambda_{Nbm} - \lambda_{Zr}} \left(1 - e^{-\lambda_{Zr} t} \right) \\ & + \left(K_{Nbm} - K_{Zr} P_{Zr\to Nbm} \frac{\lambda_{Zr}}{\lambda_{Nbm} - \lambda_{Zr}} \right) \Big(1 \\ & - e^{-\lambda_{Nbm} t} \Big) \end{aligned} \tag{8}$$

$$\begin{aligned} A_{Nb}(t) = {} & K_{Zr} \frac{\lambda_{Nb}}{\lambda_{Nb} - \lambda_{Zr}} \left(P_{Zr\to Nb} + P_{Nbm\to Nb} P_{Zr\to Nbm} \frac{\lambda_{Nbm}}{\lambda_{Nbm} - \lambda_{Zr}} \right) \Big(1 \\ & - e^{-\lambda_{Zr} t} \Big) \\ & + P_{Nbm\to Nb} \frac{\lambda_{Nb}}{\lambda_{Nb} - \lambda_{Nbm}} \Big(K_{Nbm} \\ & - K_{Zr} P_{Zr\to Nbm} \frac{\lambda_{Zr}}{\lambda_{Nbm} - \lambda_{Zr}} \Big) \cdot \left(1 - e^{-\lambda_{Nbm} t} \right) + [K_{Nb} \\ & - K_{Zr} P_{Zr\to Nb} \frac{\lambda_{Zr}}{\lambda_{Nb} - \lambda_{Zr}} - K_{Zr} P_{Nbm\to Nb} P_{Zr\to Nbm} \\ & \cdot \frac{\lambda_{Zr}}{\lambda_{Nb} - \lambda_{Zr}} \frac{\lambda_{Nbm}}{\lambda_{Nbm} - \lambda_{Zr}} - P_{Nbm\to Nb} \frac{\lambda_{Nbm}}{\lambda_{Nb} - \lambda_{Nbm}} \\ & \cdot \left(K_{Nbm} - K_{Zr} P_{Zr\to Nbm} \cdot \frac{\lambda_{Zr}}{\lambda_{Nbm} - \lambda_{Zr}} \right)] \\ & \cdot \left(1 - e^{-\lambda_{Nb} t} \right) \end{aligned} \tag{9}$$

Then, by using these equations above, the activity ratio ($r(t) = \frac{A_{Nb}(t)}{A_{Zr}(t)}$) is given by:

$$\begin{aligned} & r(t) \\ & = \frac{\lambda_{Nb}}{\lambda_{Nb} - \lambda_{Zr}} \left(P_{Zr\to Nb} + P_{Nbm\to Nb} P_{Zr\to Nbm} \frac{\lambda_{Nbm}}{\lambda_{Nbm} - \lambda_{Zr}} \right) + P_{Nbm\to Nb} \\ & \cdot \frac{\lambda_{Nb}}{\lambda_{Nb} - \lambda_{Nbm}} \left(\frac{K_{Nbm}}{K_{Zr}} - P_{Zr\to Nbm} \frac{\lambda_{Zr}}{\lambda_{Nbm} - \lambda_{Zr}} \right) \frac{\left(1 - e^{-\lambda_{Nbm} t} \right)}{\left(1 - e^{-\lambda_{Zr} t} \right)} \\ & + \Big[\frac{K_{Nb}}{K_{Zr}} - P_{Zr\to Nb} \frac{\lambda_{Zr}}{\lambda_{Nb} - \lambda_{Zr}} - P_{Nbm\to Nb} P_{Zr\to Nbm} \cdot \frac{\lambda_{Zr}}{\lambda_{Nb} - \lambda_{Zr}} \frac{\lambda_{Nbm}}{\lambda_{Nbm} - \lambda_{Zr}} \\ & - P_{Nbm\to Nb} \frac{\lambda_{Nbm}}{\lambda_{Nb} - \lambda_{Nbm}} \cdot \left(\frac{K_{Nbm}}{K_{Zr}} - P_{Zr\to Nbm} \cdot \frac{\lambda_{Zr}}{\lambda_{Nbm} - \lambda_{Zr}} \right) \Big] \\ & \cdot \frac{\left(1 - e^{-\lambda_{Nb} t} \right)}{\left(1 - e^{-\lambda_{Zr} t} \right)} \end{aligned} \tag{10}$$

The equilibrium level of this activity ratio is given by:

$$r(t \to \infty) = \frac{K_{Nb}}{K_{Zr}} + P_{Nbm\to Nb} \left(\frac{K_{Nbm}}{K_{Zr}} + P_{Zr\to Nbm} \right) + P_{Zr\to Nb} \tag{11}$$

If we assume that $K_{Nbm} = K_{Nb} = 0 \ (only\ K_{Zr} \neq 0)$, then we obtain in these conditions:

$$r(t \to \infty) = P_{Zr\to Nbm} P_{Nbm\to Nb} + P_{Zr\to Nb} \tag{12}$$

By using ENDF.B.VII.I DATA, $r(t \to \infty) = 0.9993951 \pm 0.0042883$. This dot (equilibrium level into the nuclear reactor for example) is taken as 1 in our algorithms and our plotting (see **Figure 1** and **Figure 2**). Thus, for a continuous release scenario, we consider that Nb-95/Zr-95 activity ratio starts from 1.

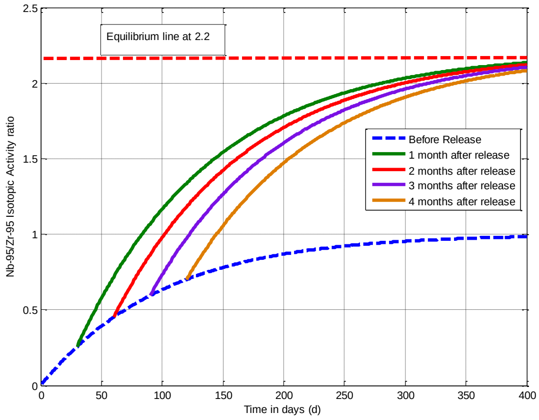

Figure 1*: The dotted line represents the evolution of the activity ratio Nb-95/Zr-95 before the release. This line changes from 0 to 1 (into the reactor for example). The solid lines represent the four first month's release. In this case, the activities ratios change from a number less than one (unknown), until the equilibrium level 2.2049. Thus, for dating a continuous release scenario, one consider that Nb-95/Zr-95 starts from 1 (equilibrium level of Nb-95/Zr-95 activity ratio into the nuclear reactor for example) and reaches its equilibrium level at 2.2049*.

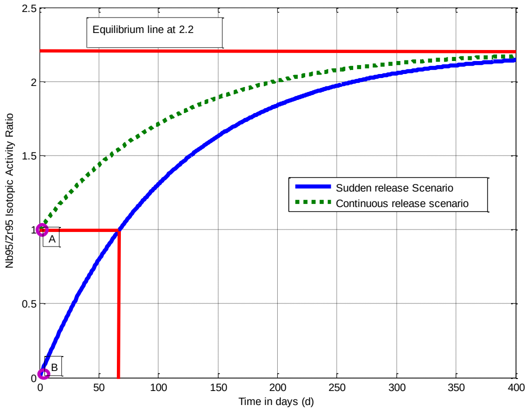

Figure 2*: The dotted line represents the change over time of Nb-95/Zr-95 activity ratio for a continuous release. In this case, the activity ratio starts from 1(see A on the Figure) and reaches the equilibrium level at 2.2049. The solid line represents the evolution of Nb-95/Zr-95 activity ratio for a sudden release scenario. This line starts from 0 (see B on the Figure) and reach its equilibrium level at 2.2049.*

We also use the activity ratio of Nbm95 and Zr95. By noting $r'(t) = \frac{A_{Nbm}(t)}{A_{Zr}(t)}$, we obtain:

$$r'(t) = \frac{\lambda_{Nbm} P_{Zr\to Nbm}}{\lambda_{Nbm} - \lambda_{Zr}} + \left(P_{Zr\to Nbm} - \frac{\lambda_{Nbm} P_{Zr\to Nbm}}{\lambda_{Nbm} - \lambda_{Zr}} + \frac{K_{Nbm}}{K_{Zr}}\right) \frac{(1 - e^{-\lambda_{Nbm} t})}{(1 - e^{-\lambda_{Zr} t})} \qquad (13)$$

In the supposition done above, namely $K_{Nbm} = 0$, then $r'(t)$ is reduced at:

$$r'(t) = \frac{\lambda_{Nbm} P_{Zr\to Nbm}}{\lambda_{Nbm} - \lambda_{Zr}} - \frac{\lambda_{Zr} P_{Zr\to Nbm}}{\lambda_{Nbm} - \lambda_{Zr}} \frac{(1 - e^{-\lambda_{Nbm} t})}{(1 - e^{-\lambda_{Zr} t})} \qquad (14)$$

This activity ratio reaches its equilibrium level at:

$$r'(t \to \infty) = \frac{\lambda_{Nbm} P_{Zr\to Nbm}}{\lambda_{Nbm} - \lambda_{Zr}} - \frac{\lambda_{Zr} P_{Zr\to Nbm}}{\lambda_{Nbm} - \lambda_{Zr}} = P_{Zr\to Nbm} \qquad (15)$$

By using ENDF.VII.I, we obtain $r'(t \to \infty) = 0.010802 \pm 0.003118$

So, in order to evaluate the age in the case of a continuous nuclear reactor release , we use the equation describing the isotopic activity ratio in the case of a nuclear explosion, but by replacing respectively $\frac{A_{Nb}(0)}{A_{Zr}(0)}$ and $\frac{A_{Nbm}(0)}{A_{Zr}(0)}$ by $r(t \to \infty)$ (i.e. 0.9993951 ± 0.0042883) and $r'(t \to \infty)$ (i.e. 0.010802 ± 0.003118).

Figures below (**Figure 3** and **Figure 4**) shows the evolution over time of Nb-95/Zr-95 activity ratio for a continuous release and a sudden release.

3.3. Summary

In order to give an overview of our calculations results in this work, **Table IV** summarizes the calculated parameters and its uncertainties. These constants result from simplifications made to obtain a more simple formula of Nb-95/Zr-95 activity ratio (see eq.10).

Our calculation schemes of nuclear event zero time (using Nb-95, Zr-95 and Nb-95m) take into account the analysis of uncertainties. The various parameters analyzed here according to the nuclear databases (like ENDF V.II.I or LNHB/CEA) do not give exactly the same value, especially at the uncertainties. From our analysis, it is important, for an accurate assessment of nuclear event zero time, to use data of ENDF V.II.I and LNHB/CEA, and if need, to make a weighted average for example using least squares method. This could help to get a good value of parameters and could improve the accuracy of the sample age. Furthermore, for a fast assessment of the age, one can use directly parameters proposed in our study.

3.4. Application: An example of a zero time calculation

In order to validate the results of our calculations, we used the measurement results from a Monte Carlo simulation based on the spectrum analysis. The test scenario was an atmospheric nuclear explosion that took place on 08 September 2003 at 17:14:14 UTC. Fifteen days later part of the (unfractionated) debris cloud crossed a virtual IMS aerosol station. Aerosols were collected and measured (using HPGe detector system) in the normal conditions of working of IMS aerosol station [6, 11].

Using Data from ENDF, the sample age is found to be 16.5874 days with an associated uncertainty of

0.2214 days, and using Data from LARA, the age is found to be 16.5903 days with an uncertainty of 0.2213 days. We can remark there not significant difference between both ages. However, the difference time between these two databases in the case of La140 and Ba140 activity ratio dating was so important (about 6 hours) [12].

We can notice that [6] Used the same spectrum data, and found an age of 16.46 days with a standard deviation of 0.26 days. This not significant difference can be explain by some differences between the nuclear data used. For example, the probability of decay from Nb-95m to Nb-95 is used here as 97.5(4) instead 94.4(6).

In order to apply this timing approach to real observations in the case of a continuous release, we have used some measured data in the IMS station at RUP59 where Nb-95 and Zr-95 was observed in one sample. The sample collection stop was on 27 December 2008. The measured activity concentrations are used to estimate the sample age. This allows validating our constants and allows comparing in the case of a continuous release, the sample age between both nuclear databases, ENDF.B.VII.I and LARA/LNHB. From our calculations, using ENDF nuclear data, the sample age is found to be 78.5627 days with an associated uncertainty of 13.0507 days. LARA decay data give a sample age of 78.5360 days with an uncertainty of 13.0442 days.

We can remark that the simplified timing approach proposed in that study works well with realistic conditions, in the case of a nuclear explosion and a continuous release scenario. The various parameters and their uncertainties established in this work are thus validated, and work well for a fast evaluation of a sample age.

4. Conclusion

In this paper, we have study different possible approaches to evaluate the zero time of a nuclear event and the associated uncertainties, using isotopic activity ratio of radionuclides pairs Zr-95 and its daughter Nb-95 taking into account Nbm95 decay. As the nuclear databases do not show always the same values of nuclear constants used for dating of age, it is so careful to take into account data given by nuclear databases, such as ENDF B.V.II.I or LARA-LNHB/CEA. This study can be considered as an additional of the four previous study made by others scientists like [2–4, 6]. The novelty of our study is that formula used for dating the age using Zr95 and Nb95 has been significantly simplified, and all kinds of releases were took into account (atmospheric test scenario, underground test scenario, and continuous release scenario) . This can allow a fast assessment of the sample age using concerned radionuclides.

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6. Disclaimer:

The views expressed by the authors do not necessarily reflect those of the CTBTO Preparatory Commission.

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