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Irradiation history and resulting isotope decay scheme influence on yttrium gamma activity

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Spallation neutrons produced during QUINTA setup irradiation react with ^{89}Y samples producing a set of gamma active yttrium isotopes. These reactions are used to determine neutron fields inside setup. The measured gamma activity used for isotope production determination is influenced by several phenomena. This work is focused on two of them - on irradiation history (IH) and decay scheme. After introductory explanation of the both effects some QUINTA experiment results are shown as the illustrative examples.

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1. Introduction

The activation detectors based on yttrium are used together with others for neutron production studies during QUINTA setup experiments. The basic reactions taken into account at yttrium sample activation are of type ^{89}Y(n,xn). The resulting isotopes are ^{88}Y, ^{87}Y, ^{86}Y and ^{85}Y and x=2, … ,5 respectively. The reaction yield or in other words determination of production rate of resulting isotope is based on sample gamma spectra measurements. Each isotope can be identified by the energy of emitted gamma quanta i.e. by energy of the peak in the gamma spectrum.

Table 1. ^{89}Y(n,xn) reaction basic data.

Reaction	**Produced Isotope**	**T1/2**	**Reaction Threshold [MeV]**	**γ-line Energy [keV]**	**γ-line Intensity [%]**
^{89}Y(n,2n)	^{88}Y	106.65d	11.5	898.042	93.7
				1836.063	99.2
^{89}Y(n,3n)	^{87}mY	13.37h	20.8	380.79	78
	^{87}Y	79.8h	20.8	388.53	82.00
				484.805	89.7
^{89}Y(n,4n)	^{86}Y	14.74h	32.7	1076.64	82.00
^{89}Y(n,5n)	^{85}Y	2.68h	42.6	231.67	84.00
		4.86h	42.6	231.67	22.8

2. Irradiation history effect on sample activity

To make the isotope quantities comparable between themselves the measured values have to be corrected for several effects. These corrections are described in many gamma spectrometry textbooks and manuals (for example nice introduction of Gordon Gilmore [1]). We use standardly needed spectrometry correction during all experiments. More specifically our way of correction description is shown at [2,3]. Here we concentrate on two specific corrections important in our case: correction for beam instability during irradiation and production of isomeric states.

Assigning symbol I_k to isotope k production rate per gram of sample one has:

$$I_k = \frac{N_A \sigma_k}{A} \tag{2.1}$$

where N_A - Avogadro's number, number of nuclei in one gram-atom;
A – parent isotope atomic number, gram-atom, [g];
σ_k – reaction cross section, [cm^2];
I_k – isotope k production rate, [cm^2/g].

In our case the I_k is the final parameter looked for. Implicitly this rate is per one beam deuteron. For discussion purpose we'll use an intermediate parameter $N_k(t_{ir})$, the resulting isotope k number of nuclei at the end of irradiation.

Deuteron beam produced by NUCLOTRON is a sequence of pulses of intensity about 10^8-10^{10} deuterons and frequency 0.125 Hz. The NUCLOTRON staff gives us the pulse intensity - φ_i and time of the pulse appearance – t_i. The *i-th* pulse produces $\frac{N_A \sigma_k}{A}\varphi_i = I_k \varphi_i$ nuclei of isotope k in a gram of sample material. As the isotope k decays, the *i-th* pulse contribution to the isotope k quantity at the end of irradiation is only $I_k \varphi_i e^{-\lambda_k (t_{ir} - t_i)}$. The total isotope k quantity at the end of irradiation amounts for sample with mass m:

$$N_k(t_{ir}) = mI_k \sum_{i=1}^{ir} \varphi_i e^{-\lambda_k (t_{ir} - t_i)} \tag{2.2}$$

where λ_k is the isotope k decay constant, index i numbers the beam pulses (ir is index number of last pulse) and $t_{ir} - t_i$ is the time from *i-th* pulse appearance till the end of irradiation. This formula will be referred to as a variable beam formula. In case of constant beam and regular pulses the number of particles in the *i-th* pulse can be expressed as $\varphi = \phi \frac{\Delta t}{t_{ir}}$ with ϕ being the beam integral, and Δt the time from the beginning of one pulse till beginning of the next pulse. For $t_{ir} >> \Delta t$ the (2.2) formula becomes:

$$N_k(t_{ir}) = mI_k \frac{\phi}{t_{ir}} \int_0^{t_{ir}} e^{-\lambda_k (t_{ir} - t)} dt = mI_k \phi \frac{1 - e^{-\lambda_k t_{ir}}}{\lambda_k t_{ir}} \tag{2.3}$$

The best way to illustrate the constant beam formula applicability or rather irradiation history influence is to create variable beam to constant beam formulae ratio (IHR).

$$IHR = \frac{N_k(t_{ir})_{VB}}{N_k(t_{ir})_{CB}} = \frac{\lambda_k t_{ir}}{\phi\left(1 - e^{-\lambda_k t_{ir}}\right)} \sum_{i=1}^{ir} \varphi_i e^{-\lambda_k (t_{ir} - t_i)} \tag{2.4}$$

The indexes *VB* and *CB* refer to variable beam and constant beam respectively.

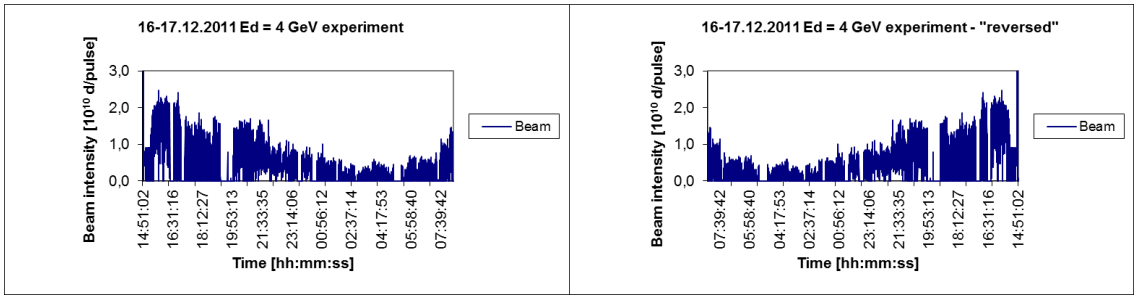

Fig. 1. QUINTA setup 16-17.12.2011 experiment beam run (left) and "reversed" beam run (right). Reversed means the first pulse on left graph becomes the last one on the right.

For illustration of irradiation history effect one experiment was selected - QUINTA setup experiment 16-17.12.2011. To make the effect more spectacular a mirror run was created – named "reversed" – and compared with the real one (see Fig.1). In both cases the deuteron beam integral was 3.37×10^{13} and irradiation time 17:31:00 [hh:mm:ss].

Table 2. 16-17.12.2011 experiment parameter IHR as the measure of irradiation history influence on yttrium isotope production

Beam run\isotope	^{88}Y	^{87m}Y	^{87}Y	^{86}Y	^{85}Y
Beam run averaged (constant)	1.00	1.00	1.00	1.00	1.00
Real beam run	1.00	0.84	0.97	0.86	0.55
Reversed beam run	1.00	1.18	1.03	1.16	1.83

The above displayed example shows very strong dependence on irradiation history for short-lived isotopes and rather weak for long-lived ones compared to irradiation time. Therefore it is very important to assess the average uncertainty on $N_k(t_{ir})$ caused by irradiation history. The IH contribution to isotope production uncertainty does not depend on φ_i uncertainty ($\Delta\varphi_i$) nor on t_i uncertainty (Δt_i). More sensible assessment is like:

$$\Delta N_k\left(t_{ir}\right)_{IH} = \left|\frac{N_k\left(t_{ir}\right)_{VB} - N_k\left(t_{ir}\right)_{CB}}{N_k\left(t_{ir}\right)_{CB}}\right| = \left|IHR - 1\right| \tag{2.5}$$

3. Influence of the isotope decay scheme on measured production

Despite of ^{87m}Y being an isomeric state of the same nucleus ^{87}Y their de-excitation and decay process can be described as if they are different isotopes. Isotope ^{87m}Y will be indexed as "1" and the ^{87}Y one as "2". Both isotopes are produced in ^{89}Y(n,3n) reaction and then both of them decay as in Fig. 2. The ^{87m}Y transits first into ^{87}Y and then decays.

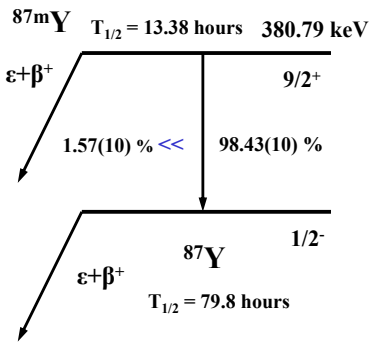

Fig. 2. The ^{87m}Y isomeric transition and decay scheme. The isomer state gamma transition to ground state is dominant in this case.

Each sample activity is measured two times, usually. The first measurement is done as soon as possible after the irradiation to "catch" the short-lived isotopes and then follows the second one, much longer, to measure the long-lived. In case of ^{87}Y the first measurement is performed while the ^{87m}Y decays yet. Neglecting the ^{87m}Y effect leads to discrepancy between series 1 and series 2 results of ^{87}Y.

Assuming constant deuteron beam and constant neutron flux φ one can express the mentioned isotopes quantity dependence on time with two sets of equations.

$$\left.\begin{aligned} \frac{dN_1}{dt} &= mI_1\varphi - \lambda_1 N_1 \\ \frac{dN_{2\,from1}}{dt} &= \lambda_1 N_1 - \lambda_2 N_{2\,from1} \\ \frac{dN_{2direct}}{dt} &= mI_2\varphi - \lambda_2 N_{2direct} \end{aligned}\right| \; t \le t_{irr} \tag{3.1}$$

$$\left.\begin{aligned} \frac{dN_1}{dt} &= -\lambda_1 N_1 \\ \frac{dN_{2\,from1}}{dt} &= \lambda_1 N_1 - \lambda_2 N_{2\,from1} \\ \frac{dN_{2direct}}{dt} &= -\lambda_2 N_{2direct} \end{aligned}\right| \; t > t_{irr} \tag{3.2}$$

$$N_2 = N_{2\,from1} + N_{2direct} \tag{3.3}$$

Here λ_1 and λ_2 are the decay constants, $N_{2direct}$ - nuclei quantity produced directly from (n,3n) reaction, $N_{2\,from1}$ - produced from N_1 decay. The first set refers to irradiation period and the second to post-irradiation decay. Term $mI_{1,2}\varphi$ describes direct production rate of isotope *1,2* while $\lambda_1 N_1$ describes both isotope *1* decay rate and isotope *2* production rate from isotope *1* decay.

Solving the above equation sets one gets for irradiation time t_{ir}:

$$
\begin{aligned}
N_1(t_{ir}) &= mI_1\phi\frac{\left(1-e^{-\lambda_1 t_{ir}}\right)}{\lambda_1 t_{ir}} \\
N_{2\,from1}(t_{ir}) &= mI_1\phi\left[\frac{1-e^{-\lambda_2 t_{ir}}}{\lambda_2 t_{ir}} - \frac{e^{-\lambda_2 t_{ir}}-e^{-\lambda_1 t_{ir}}}{(\lambda_1-\lambda_2)t_{ir}}\right] \\
N_{2direct}(t_{ir}) &= mI_2\phi\frac{\left(1-e^{-\lambda_2 t_{ir}}\right)}{\lambda_2 t_{ir}} \\
N_2(t_{ir}) &= N_{2\,from1}(t_{ir}) + N_{2direct}(t_{ir})
\end{aligned}
\tag{3.4}
$$

For the time after irradiation, t_+ the formulae look like:

$$
\begin{aligned}
N_1(t_+) &= N_1(t_{ir})e^{-\lambda_1 t_+} \\
N_{2\,from1}(t_+) &= N_1(t_{ir})\frac{\lambda_1}{(\lambda_1-\lambda_2)}\left(e^{-\lambda_2 t_+}-e^{-\lambda_1 t_+}\right) \\
N_2(t_+) &= N_2(t_{ir})e^{-\lambda_2 t_+} + N_1(t_{ir})\frac{\lambda_1}{(\lambda_1-\lambda_2)}\left(e^{-\lambda_2 t_+}-e^{-\lambda_1 t_+}\right)
\end{aligned}
\tag{3.5}
$$

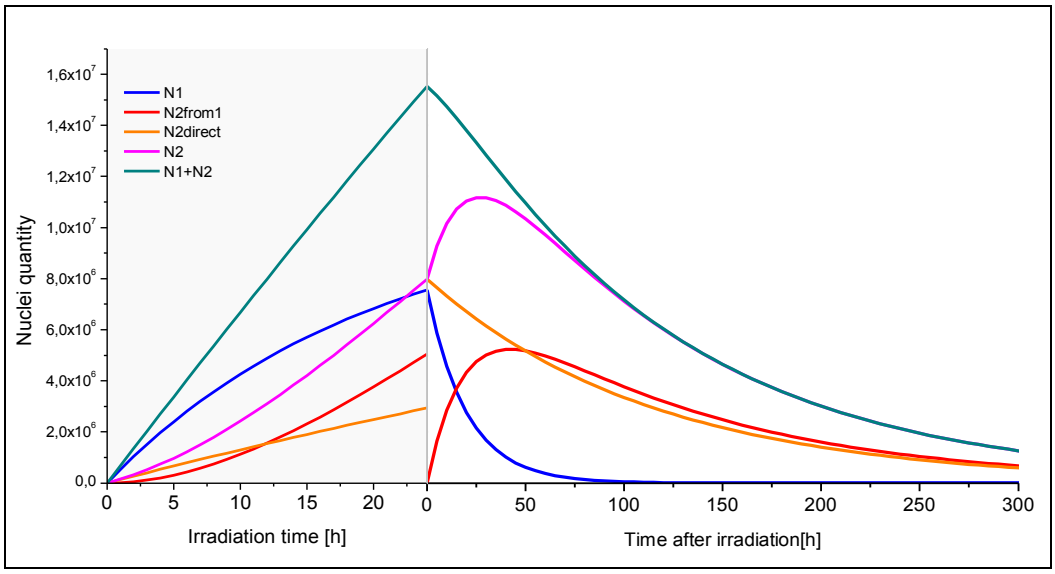

Fig. 3. Time dependence of ^{87m}Y and ^{87}Y quantity during ^{89}Y sample irradiation and after end of irradiation.

As ^{87m}Y and ^{87}Y are the two different states of the same isotope ^{87}Y it makes sense to use function $N_{2total} = N_1 + N_2$ and $I_{2total} = I_1 + I_2$ to describe isotope quantity and production rate.

On the other side the mentioned above quantities of isotope nuclei are related to respective peak area - S corrected for gamma line intensity - I_γ, gamma spectrometer efficiency - ε_p,

coefficient of coincidence – *COI*, real time of measurement t_{real} and the live one – $t_{live,}$ by formula:

$$N_k(t_+) = S\frac{100}{I_\gamma \varepsilon_p COI\left(1-e^{-\lambda_k t_{real}}\right)}\frac{t_{real}}{t_{live}} \tag{3.6}$$

After recalculation the isotope quantities to isotope production rate one gets:

$$\begin{aligned} I_1 &= \frac{N_1(t_+)}{m\phi}\frac{\lambda_1 t_{ir}}{\left(1-e^{-\lambda_1 t_{ir}}\right)}e^{\lambda_1 t_+} \\ I_{2total} &= \frac{N_2(t_+)}{m\phi}\frac{\lambda_2 t_{ir}}{\left(1-e^{-\lambda_2 t_{ir}}\right)}e^{\lambda_2 t_+} + I_1\frac{\lambda_2}{(\lambda_1-\lambda_2)}\left[\frac{\left(1-e^{-\lambda_1 t_{ir}}\right)}{\left(1-e^{-\lambda_2 t_{ir}}\right)}e^{-(\lambda_1-\lambda_2)t_+}-1\right] \end{aligned} \tag{3.7}$$

In case of variable beam run the production process description should be modified. Each beam pulse contribution to $N_k(t_{ir})$ should be treated separately. For the *i-th* pulse, φ_i contributes $mI_1\varphi_i e^{-\lambda_1(t_{ir}-t_i)}$ to $N_1(t_{ir})$ and $mI_1\frac{\lambda_1}{(\lambda_1-\lambda_2)}\left(e^{-\lambda_2(t_{ir}-t_i)}-e^{-\lambda_1(t_{ir}-t_i)}\right)+mI_2\varphi_i e^{-\lambda_2(t_{ir}-t_i)}$ to $N_2(t_{ir})$

$$\begin{aligned} N_1(t_{ir}) &= mI_1\sum_{t_i=t_1}^{t_{ir}}\varphi_i e^{-\lambda_k(t_{ir}-t_i)} \\ N_{2\,from1}(t_{ir}) &= mI_1\frac{\lambda_1}{(\lambda_1-\lambda_2)}\sum_{i=1`}^{ir}\varphi_i\left(e^{-\lambda_2(t_{ir}-t_i)}-e^{-\lambda_1(t_{ir}-t_i)}\right) \\ N_{2direct}(t_{ir}) &= mI_2\sum_{i=1}^{ir}\varphi_i e^{-\lambda_2(t_{ir}-t_i)} \\ N_2(t_{ir}) &= N_{2\,from1}(t_{ir})+N_{2direct}(t_{ir}) \end{aligned} \tag{3.8}$$

The formulae describing post-irradiation decay process remain unchanged. Combining (3.5) with (3.9) one gets formulae for isotope production rates:

$$\begin{aligned} I_1 &= \frac{N_1(t_+)}{m}\frac{e^{\lambda_1 t_+}}{\sum_{i=1}^{ir}\varphi_i e^{-\lambda_1(t_{ir}-t_i)}} \\ I_{2total} &= \frac{N_2(t_+)e^{\lambda_2 t_+}}{m\sum_{i=1}^{ir}\varphi_i e^{-\lambda_2(t_{ir}-t_l)}}+\frac{I_1}{(\lambda_1-\lambda_2)}\left[\lambda_1\frac{\sum_{i=1}^{ir}\varphi_i e^{-\lambda_1(t_{ir}-t_i)}}{\sum_{i=1}^{ir}\varphi_i e^{-\lambda_2(t_{ir}-t_l)}}e^{-(\lambda_1-\lambda_2)t_+}-\lambda_2\right] \end{aligned} \tag{3.9}$$

The effect of correction for decay scheme is illustrated on example ^{87}Y production axial distribution got from one of QUINTA experiments – Fig. 4.

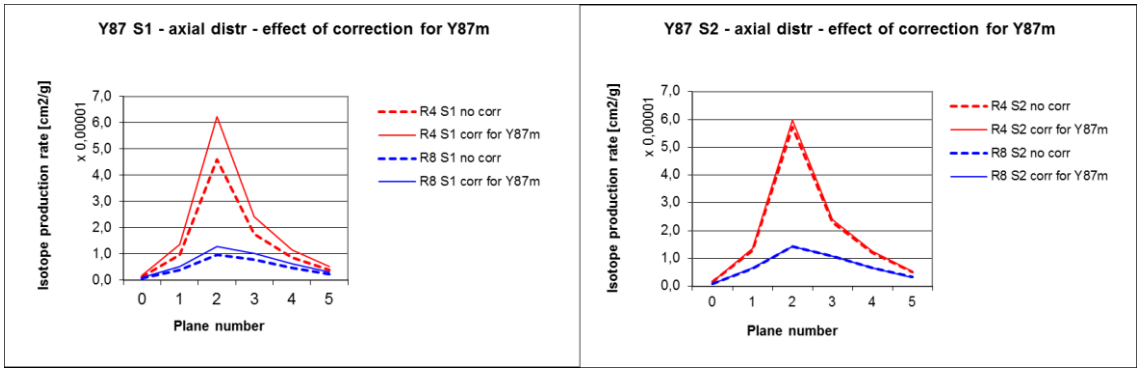

Fig. 4. Experiment QUINTA 2011-03-20-21 6 GeV - ^{87}Y production series 1 and 2 measurement axial distributions as illustration of correction for decay ^{87m}Y→^{87}Y effect. The dashed lines stand for results not corrected results.

Measured gamma spectra were evaluated in the code DEIMOS [5] and consequent analysis was done in EXCEL using the equations mentioned above.

4. Conclusions

As expected the irradiation history effect on measured sample activity depends on resulting isotope half life time. Shorter the half time is larger is the effect of the irradiation history. As concerns ^{89}Y(n,xn) reactions the irradiation history affects the most ^{86}Y, ^{87m}Y and ^{85}Y production.

Influence of decay scheme effect of produced isotope on measured activity depends on time interval between the end of irradiation and start of measurement, on cooling time. Shorter the cooling time is larger is the decay scheme effect and the appropriate correction.

In case of QUINTA experiments where the irradiation times are of order 12-24 hours both effects are necessarily taken into account.

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