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Decoupling of radionuclide production cumulative cross-sections and TTY: Application to the process $^{nat}\mathrm{Dy}(d,x)^{155}\mathrm{Ho} \rightarrow {}^{155}\mathrm{Dy} \rightarrow {}^{155}\mathrm{Tb}$

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Summary. — Nuclear reaction cross-sections are sometimes presented as cumulative, in the sense that not all production is due to direct reactions, but the decay of a co-produced isomeric state or radioactive father contributes to the total yield of the reaction. In this paper, we discuss one of the scenarios that occur more frequently. Moreover, the results are employed to discuss one of the production routes of the medically relevant ^{155}Tb.

1. – Introduction

A crucial parameter for the production of radionuclides (RNs) is the cross-section (XS), that shall be known for several reasons, such as the optimization of the production process and the determination of the radionuclidic purity (RNP) of the product [1]. It is quite frequent that more than one reaction channel lead to the formation of a RN, some of them involving the decay of other species during irradiation, and this entails a complex dependence of the cumulative XS on the irradiation parameters. In this work we analyze one of the most diffused complex production scheme to determine the dependence of the cumulative XS on the irradiation time. These findings have been applied to the practical case of the production of the medically relevant ^{155}Tb from the decay of ^{155}Dy obtained through the $^{nat}\mathrm{Dy}(d,x)$ reaction.

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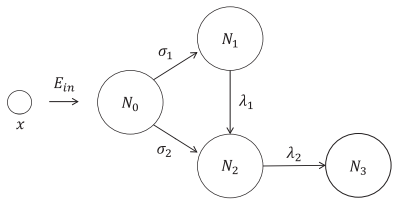

Fig. 1. – Simplified scheme of the production of specie N_2, in which both the direct production and the decay of the co-produced radioactive specie N_1 contribute during irradiation.

2. – Cumulative XS

In the case of direct nuclear reaction of production of a RN, the disintegration rate (D_i [Bq]) is related to the XS ($\sigma_i(E)$ [mb]) through [2]

$$D_i = \lambda_i N_i(t_c) = k\sigma(E)_i \cdot G_i(t_{irr}) \cdot e^{-\lambda_i \cdot t_c} = \frac{\rho dx \cdot N_A \cdot Q}{M \cdot t_{irr} \cdot Ze}\sigma(E)_i \cdot G_i(t_{irr}) \cdot e^{-\lambda_i \cdot t_c} \tag{1}$$

where λ_i is the decay constant [s^{-1}], t_{irr} is the duration of the bombardment [s], t_c is the cooling time [s], that is the time between the End Of the Bombardment (EOB) and the beginning of the measurement, $G_i = (1 - e^{-\lambda \cdot t_{irr}})$. k is a constant that can be calculated as shown in eq. (1), where ρdx is the mass thickness of the target [g/cm^2], N_A is the Avogadro number [mol^{-1}], M is the molar mass [g/mol], Q [C] is the total impinging charge and $Z \cdot e$ is the charge of the single particle of the beam [C].

This is no longer true for more complex production routes. Let us consider the scheme reported in fig. 1 in which the decay of a co-produced isomeric state or radioactive father N_1 contributes to the total yield of production of specie N_2. The equations that describe the production of the RNs 1 and 2 are the following:

$$\frac{\mathrm{d}N_1}{\mathrm{d}t} = k \cdot \sigma_1 - \lambda_1 N_1, \tag{2a}$$

$$\frac{\mathrm{d}N_2}{\mathrm{d}t} = k \cdot \sigma_2 - \lambda_2 N_2 + \lambda_1 N_1. \tag{2b}$$

Considering only the second equation, the solution can be simply derived by integrating eqs. (2) between zero and t_{irr} with the initial condition that $N_1(0) = N_2(0) = 0$:

$$N_2^{EOB} = \frac{k \cdot \sigma_2}{\lambda_2} G_2 + \frac{k \cdot \sigma_1}{\lambda_2 - \lambda_1}\left(G_1 - \frac{\lambda_1}{\lambda_2} G_2\right). \tag{3}$$

By rearranging the terms in the equation, and by multiplying them by λ_2, we obtain the disintegration rate at the EOB:

$$D_2^{EOB} = \lambda_2 \cdot N_2^{EOB} = k \cdot \left[\sigma_2 + \frac{\sigma_1}{\lambda_2 - \lambda_1} \cdot \left(\lambda_2 \frac{G_1}{G_2} - \lambda_1\right)\right] \cdot G_2. \tag{4}$$

From the comparison with eq. (1) calculated at the EOB, we can state that in the case illustrated in fig. 1, the cumulative XS must be intended as a combination of σ_1 and σ_2

with a dependence on t_{irr} as reported in

$$\sigma_2^{cum} = \left[\sigma_2 + \frac{\sigma_1}{\lambda_2 - \lambda_1} \cdot \left(\lambda_2 \frac{G_1}{G_2} - \lambda_1\right)\right]. \tag{5}$$

The Thick-Target Yields (TTY) can be defined as the amount of activity produced on a thick target at the EOB divided by the current of the beam [3]:

$$TTY_2^{EOB} = \frac{N_A}{M \cdot Ze} G_2 \int_{E-\Delta E}^{E} \left[\sigma_2 + \frac{\lambda_1}{\lambda_1 - \lambda_2}\sigma_1 \left(1 - \frac{G_1}{G_2}\right)\right] \left(\frac{\mathrm{d}E}{\rho \mathrm{d}x}\right)^{-1} \cdot \mathrm{d}E, \tag{6}$$

where ΔE is the energy lost in the target and depends on the thickness of the target.

3. – Application: $^{\mathbf{nat}}$Dy(d,x)$^{\mathbf{155}}$Ho$\rightarrow^{\mathbf{155}}Dy\rightarrow^{\mathbf{155}}$Tb

^{155}Tb ($T_{1/2} = 5.32$ d) is a medical relevant radioisotope of terbium. It emits γ-rays with energies suitable for SPECT studies ($E_\gamma = 87$ keV (32%) and 105 keV (25%)) and the absence of β^+/β^- emissions brings reduced radiotoxicity. One of the production routes exploits the so-called precursor technique, in which ^{155}Dy is produced through a nuclear reaction, it is isolated from the non radioisotopic nuclei and, after decay, the produced ^{155}Tb is extracted by using radiochemical methods. To produce ^{155}Dy, the reactions induced by deuterons on natural dysprosium targets may be considered. However, during irradiation, ^{155}Ho is produced, and the decay scheme is the same as the one reported in fig. 1. Simultaneously, ^{157,159}Dy contaminants are co-produced. The terbium isotopes, other than ^{155}Tb, that may be extracted from the dysprosium precursor are ^{157}Tb and ^{159}Tb. Moreover, these two Dy radioisotopes are produced with a reaction scheme similar to the one of ^{155}Dy and are directly linked to ^{157}Ho and ^{159}Ho respectively. Thus, to obtain a precise and meaningful determination of the TTY, all the corrections due to the cumulative XS must be applied. The cumulative XS of these reactions have been measured elsewhere [4] in the 12–32 MeV energetic range, but due to their short decay time, no experimental data of holmium radioisotope XS are available. Nevertheless, nuclear simulations obtained with the EMPIRE 3.2 [5] code may be exploited.

3˙1. *EMPIRE 3.2 simulations*. – The experimental data of the cumulative XS have been compared to the nuclear simulations obtained with the EMPIRE 3.2 nuclear code [5] using default options in the energetic range between 17 MeV and 33 MeV. The XSs of ^{15x}Dy and ^{15x}Ho have been calculated separately and then summed according to eq. (5) and reported in fig. 2(a). At lower energy, the presence of ^{155}Dy has not been detected, thus the theoretical simulations are not supported by experimental data. There is sufficient accordance between the experimental data and the nuclear simulation, therefore it is reasonable to use the simulated data instead of the experimental ones to overcome the absence of data relative to the production of holmium.

3˙2. *Calculation of the cumulated yield*. – The TTYs, calculated through eq. (6), with an irradiation time of 20 h have been reported in fig. 2(b). A target with a thickness of 890 mg/cm^2, has been selected as the result of the optimization that permits to reduce the production of contaminants. This thickness corresponds to a $\Delta E = 15.5$ MeV when the energy of the beam is 33 MeV.

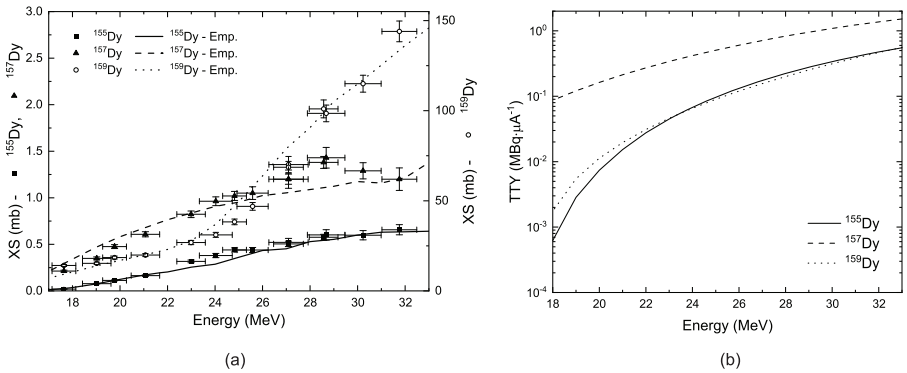

Fig. 2. – (a) Comparison between the experimental XSs [4] and the EMPIRE 3.2 simulations [5]. The left and the right scales refer to ^{155,157}Dy and to ^{159}Dy, respectively. (b) TTY of ^{155,157,159}Dy relative to a 20 hours irradiation of a 890 mg/cm^2 thick ^{nat}Dy target.

After a realistic time of 1800 s, an ideal radiochemical separation, with a 100% separation efficiency, has been performed to remove holmium and terbium from the dysprosium precursor. The extracted dysprosium is left to decay: it has been calculated that the maximum of the activity of ^{155}Tb is obtained after about 38 h, a time after which a second ideal radiochemical separation has been performed to extract the terbium.

After this double step process, ^{155}Tb with RNP of about 99.9% has been obtained, due to the fact that ^{157}Tb decays with a very long decay time, and ^{159}Tb is stable. However, with a realistic deuterons current of 50 μA of a research cyclotron like the one present at GIP ARRONAX [6], the maximum activity that can be obtained is of about 1.45 MBq and it is far too low for any medical application.

4. – Conclusions

In order to know with high precision the RNP of a RN to be employed in nuclear medicine, it is necessary to consider all the reaction channels and decay processes that may contribute. The actual expression of the cumulative XS must be evaluated on a case by case basis, especially to detect any dependence on the parameter of the particular experiment. This has been done for the case of process $^{nat}\mathrm{Dy}(d,x)^{155}\mathrm{Ho}\rightarrow^{155}\mathrm{Dy}\rightarrow^{155}\mathrm{Tb}$. However, when it is not possible, it is paramount to specify all the irradiation information that could be relevant to replicate the result.

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