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Analytical model for release calculations in solid thin-foils ISOL targets

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ABSTRACT

A detailed analytical model has been developed to simulate isotope-release curves from thin-foils ISOL targets. It involves the separate modeling of diffusion and effusion inside the target. The former has been modeled using both first and second Fick's law. The latter, effusion from the surface of the target material to the end of the ionizer, was simulated with the Monte Carlo code MolFlow+. The calculated delay-time distribution for this process was then fitted using a double-exponential function. The release curve obtained from the convolution of diffusion and effusion shows good agreement with experimental data from two different target geometries used at ISOLDE. Moreover, the experimental yields are well reproduced when combining the release fraction with calculated in-target production.

1. Introduction

Isotope Separation On Line (ISOL) is a powerful technique to produce radioactive ion beams (RIB) which are used for experimental studies in many fields, including nuclear physics, astrophysics and nuclear medicine [1].

In the ISOL method isotopes are produced through the interaction of a high-energy light-particle beam (typically protons) with a high-Z target material, in which they are usually thermalized. These isotopes are subsequently released through diffusion in the material and effusion from the target-container volume towards an ion source where they are ionized and then extracted in an ion beam. Mass purification happens in a dedicated magnetic separator downstream the ion source.

Currently-used solid targets consist of a cylindrical container which is filled with pellets or thin foils, constituting the material in which radioactive nuclei are produced (see Fig. 1).

Maximizing the RIB intensity at the experimental station is a major objective for any ISOL facility. This intensity I is described by the following formula derived from [3]:

$$I = \int \phi(E, x)\sigma(E)\rho(x)N_A/A \; dE \; dx \; \epsilon_{Diff} \; \epsilon_{Eff} \; \epsilon_{Ion} \; \epsilon_{MS}, \quad (1)$$

where $\sigma(E)$ is the reaction cross section for the production of a specific isotope. This cross section depends on the target nuclide, with mass A, and the energy E and type of the incoming particle. The target density is $\rho(x)$, while N_A denotes the Avogadro number. The other four terms in Eq. (1) represent the efficiencies related to diffusion out of the target material (ϵ_{Diff}), effusion through the target voids and transfer line (ϵ_{Eff}), ionization (ϵ_{Ion}) and mass separation (ϵ_{MS}). An accurate description of the processes involved is crucial not only for predicting the outward isotope flux but also for target optimization. This paper reports on our approach where we model separately the in-target production, diffusion and effusion processes which are then combined to obtain an overall isotope-release curve in thin-foils targets. The developed model is benchmarked with experimental data from two different solid tantalum-target geometries operated at ISOLDE and compared with previous calculations [4].

2. Methodology

A first step towards target optimization for high RIB intensity is to accurately model the release of isotopes from the location where they are created to the ion source. In this section our release model for a solid thin-foils target is described. It includes diffusion inside the target material, effusion in the vacuum of both the target container and transfer line, and radioactive decay.

2.1. Diffusion

The produced isotopes migrate from inside the solid material to the surface where they desorb. In the case of a thin-foils target, the

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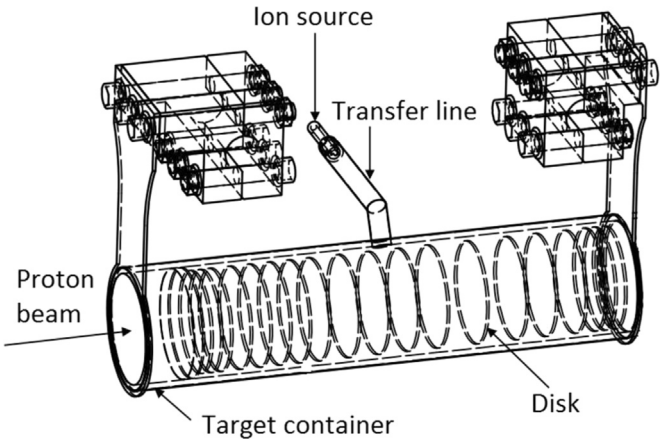

Fig. 1. Schematic representation of a standard solid ISOL target. Picture extracted from [2].

thickness of each foil is much smaller than its other dimensions. Therefore, diffusion is only considered in the direction perpendicular to the surface of the foil. Fick's second law of diffusion [5]

$$\frac{\partial C(\vec{x}, t)}{\partial t} = \nabla\cdot(D(\vec{x})\nabla C(\vec{x}, t)) \tag{2}$$

is thus simplified to

$$\frac{\partial C(x, t)}{\partial t} = \frac{\partial}{\partial x}\left(D(x)\frac{\partial C(x, t)}{\partial x}\right) \tag{3}$$

where D is the diffusion coefficient of the migrating species in the material and C represents their concentration profile. The temperature dependence of the diffusion coefficient is usually described by an Arrhenius law [6]

$$D = D_0\exp\left[-\frac{\Delta H}{RT}\right] \tag{4}$$

where T is the absolute temperature, ΔH is the activation enthalpy of diffusion and R is the universal gas constant. This relation shows that the diffusion coefficient rises with increasing temperature. For this reason, an ISOL target has to be operated at a high temperature to enhance the diffusion of isotopes. In this study a uniform temperature distribution in the target material was assumed, thereby neglecting possible variations of the temperature profile due to beam-power deposition. Additionally, the initial concentration (C_0) of isotopes inside the target material is considered uniform. These assumptions are crucial to obtain a linear problem, described by

$$\frac{\partial C(x, t)}{\partial t} = D\,\frac{\partial^2 C(x, t)}{\partial x^2}, \tag{5}$$

which is solved analytically by the method of separation of variables, as shown in [5]. The solution is given by

$$C(x, t) = \frac{4C_0}{\pi}\sum_{n=0}^{\infty}\frac{1}{2n+1}\exp\left[-\frac{(2n+1)^2\pi^2 D}{l^2}t\right]\sin\left[\frac{(2n+1)\pi x}{l}\right], \tag{6}$$

where l is the foil thickness. The flux of particles out of the solid target material J is found by applying Fick's first law of diffusion [5]

$$J = -D\frac{\partial C}{\partial x}\bigg|_{x=l}. \tag{7}$$

Inserting (6) in (7) gives

$$J = \frac{4C_0 D}{l}\sum_{n=0}^{\infty}\exp\left[-\frac{(2n+1)^2\pi^2 D}{l^2}t\right]. \tag{8}$$

2.2. Effusion

At the vacuum level of typical ISOL targets, molecular-flow conditions are satisfied [7]. In this regime, an atom desorbed from a surface flies in a straight line until reaching another surface where it can be re-adsorbed.

Effusion, the migration of isotopes under these conditions, was simulated with the Monte Carlo code MolFlow+ [8]. The code tracks particles with mass m, starting from the target-material surface until they exit the ionizer. From MolFlow+ simulations, the distributions for the effusion delay-time and number of collisions can be derived. The Sticking time of particles on surfaces is not implemented in the code, which means that upon interaction the particle is either instantaneously re-desorbed or permanently trapped.

To correct for this effect, the mean sticking time τ of a particle to a surface is introduced in our model.

This parameter is related to the desorption rate r_D of a particle from a surface, given by (see e.g. [9])

$$r_D = n\nu_0\exp\left[-\frac{E_{act}}{k_B T}\right], \tag{9}$$

where n is the number of adsorbate sites per area, ν_0 is a frequency factor, E_{act} is the activation energy for desorption, k_B is the Boltzmann constant and T is the absolute temperature. The bonds between the particle and the surrounding material may be of chemical nature, in which case the activation energy is high, implying a low probability of being re-emitted. Alternatively, weaker van der Waals' interactions lead to a higher re-emission rate.

Since $r_D \propto 1/\tau$, one can write

$$\tau = \tau_0 exp\left[\frac{E_{act}}{k_B T}\right]. \tag{10}$$

Sticking time was included in the model by shifting the particle arrival time by

$$\delta t = t\left(\frac{\tau\,\overline{N_{Coll}}}{\bar{t}}\right), \tag{11}$$

where $\bar{t}$ and $\overline{N_{Coll}}$ (both derived from MolFlow+ output) are respectively the average time which a particle has to travel before exiting the system and the average number of wall collisions.

Frequently, it is desirable to perform release calculations for various isotope masses or for different target temperatures. In this respect, note that both the mass m and temperature T are specified as input parameters in a MolFlow+ calculation. Under the condition that the effusion geometry is at a uniform temperature, the result of this calculation can be employed to derive a delay-time distribution for a different parameter set m' and T'. This procedure, which avoids running multiple lengthy Monte Carlo simulations, is explained in the following paragraph.

In the MolFlow+ setting used in this work, a particle is re-emitted from a surface with the mean particle velocity v according to the Maxwell-Boltzmann distribution [8], given by

$$v = \sqrt{\frac{8RT}{\pi m}}. \tag{12}$$

Since the distance d traveled by a particle to the end of the ionizer only depends on the target geometry one has

$$d = vt = v't' \rightarrow t' = \frac{vt}{v'}, \tag{13}$$