Thick-target yields of radioactive targets deduced from inverse kinematics

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The thick-target yield (TTY) of long-lived fission products (LLFP) is an essential quantity and represents basic data for transmutation. In order to evaluate TTY on radioactive targets including LLFP, we suggest a conversion method from inverse kinematics corresponding to the reaction of radioactive beams on stable targets. We demonstrate the method to deduce the TTY from inverse kinematics derived from the theoretical definition. This method is highly applicable in reactions at the energy per nucleon $\epsilon > 20$ MeV/A as practically confirmed by the simulation of the SRIM2008 code. In this paper, we apply the method to the $^{nat}\text{Cu}^{(12}\text{C},X)^{24}\text{Na}$ reaction to confirm availability. In addition, it is applied to the $^{137}\text{Cs} + ^{12}\text{C}$ reaction to reduce $^{137}\text{Cs}$ and to suggest a TTY measurement of the $^{137}\text{Cs}$ induced reaction on a thick $^{12}\text{C}$ target.

Keywords: thick-target yield; nuclear transmutation; radioactive waste; cesium 137

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1. Introduction

Transmutation of long-lived fission products (LLFP) is one of the key technologies to reduce radioactive wastes produced in nuclear power plants\[1\]. Nuclear data, such as cross sections, related to the transmutation is unavoidable for the technology. Indeed, a recent project measures neutron capture cross sections of LLFP and minor actinides\[2\], although experiments of such radioactive targets remain limited due to high radioactivity of the targets.

One candidate method to obtain the data is to utilize the experimental values measured in inverse kinematics. The inverse kinematics were realized by the recent progress of radioactive beams. Radioactive isotopes, including LLFP, are available as a beam in accelerators to obtain nuclear data. For instance, an experiment has been performed at RIBF to obtain cross section data relating to $^{90}$Sr and $^{137}$Cs was performed at RIBF. It is also applied to obtain cross sections deduced from the thick-target method of elastic scattering (e.g., Ref.\[3\]).

In addition to such cross section data, an essential quantity for the transmutation is thick-target yields (TTY)\[4\]. If we require information pertaining to transmutation of LLFP lumps, the TTY plays a key role. However, the TTY to reduce LLFP are nearly impossible to obtain directly in accelerators since the preparation of LLFP lumps as a target is miserable due to high radioactivity.

In this paper, we suggest a new method to estimate TTY and subsequently reduce radioactivity of LLFP lumps using inverse kinematics. The method enable us to evaluate the possibility of transmutation of LLFP lumps.

2. Method

In general, the reaction probability $Y$ is defined as

$$Y = \frac{N_r}{N_i}, \quad (1)$$
where \( N_r \) and \( N_i \) are the numbers of reacted and incident particles. In the case of thick-target matter, the \( Y \) denotes TTY itself of the reaction and the differential reaction probability \( dY \) at an infinitesimal length in the matter \( dx \) [cm] can be described as:

\[
dY = \frac{\sigma \rho N_A}{A_T} dx,
\]

where the cross section \( \sigma \) [cm\(^2\)], the Avogadro constant \( N_A \) [mol\(^{-1}\)], the mass number of the target \( A_T \) [g·mol\(^{-1}\)] and the density \( \rho \) [g·cm\(^{-3}\)]. We can obtain the TTY by integrating the thickness of the target and introducing a stopping power \( S(E) = -\frac{dE}{dx_{\text{eq}}} \) [MeV·g\(^{-1}\)·cm\(^2\)]:

\[
Y(E_{\text{in}}) = \frac{N_A}{A_T} \int_0^{E_{\text{in}}} \sigma(E) \frac{1}{S(E)} dE,
\]

at the incident energy in a laboratory system \( E_{\text{in}} \) [MeV]. The lower limit of energy integration of Eq. (3) corresponds to zero, i.e. the incident particles stop inside of the target. The integration variable and the upper limit can be converted into energy per nucleon \( \epsilon = E/A_P \) with the mass number of the projectile \( A_P \) and its incident energy per nucleon \( \epsilon_{\text{in}} \). The TTY is rewritten as:

\[
Y(\epsilon_{\text{in}}) = \frac{N_A A_P}{A_T} \int_0^{\epsilon_{\text{in}}} \sigma(\epsilon) \frac{1}{S(\epsilon)} d\epsilon,
\]

which leads to:

\[
\frac{dY(\epsilon)}{d\epsilon} = \frac{N_A A_P}{A_T} \sigma(\epsilon) \frac{1}{S(\epsilon)}.
\]

We note that the Eqs. (4) and (5) are available under the condition that all the projectiles stop inside the target, namely, the incident energy \( \epsilon_{\text{in}} \) should be less than the minimum energy to pass through the target.

In this paper, we define the projectile \( P \) induced reaction on a target \( T \) as “forward” and its inverse kinematics as “inverse”. Those TTY which are evaluated from Eq. (4) are denoted as \( Y_{\text{for}} \) and \( Y_{\text{inv}} \). We consider the relation between \( Y_{\text{for}} \) and \( Y_{\text{inv}} \) using the inverse kinematics and the common cross section of both reactions. Hence, the ratio \( R(\epsilon) \) between
the differential yields at the same energy $\epsilon$ in both reactions can be expressed as

$$R(\epsilon) \equiv \frac{dY_{\text{for}}(\epsilon)}{dY_{\text{inv}}(\epsilon)} = \frac{A_P^2 S_{\text{inv}}(\epsilon)}{A_T^2 S_{\text{for}}(\epsilon)},$$

(6)

$$dY_{\text{for}}(\epsilon) = R(\epsilon) dY_{\text{inv}}(\epsilon).$$

(7)

This relation suggests that we can evaluate the $Y_{\text{for}}(\epsilon)$ without the direct experiment of the forward kinematics reaction in cases where the $Y_{\text{inv}}(\epsilon)$ and the $R(\epsilon)$ are obtained. Since the stopping power is proportional to the square of the projectile atomic number $Z_P^2$ and the target atomic number $Z_T$ in the high energy region, the heavier projectile is more easily stopped inside the target. This also implies an advantage of the method because the target of the light elements can be thin and prepared more easily.

3. Procedure and Applications

To confirm the procedure, as well as its availability, we show the example of $^{nat}\text{Cu}(^{12}\text{C},X)^{24}\text{Na}$[5], and the TTY of the $^{137}\text{Cs} + ^{12}\text{C}$ reaction for the transmutation.

Indeed, the method is initially applied to find the relation between the $^{nat}\text{Cu}(^{12}\text{C},X)^{24}\text{Na}$ forward and $^{12}\text{C}(^{63,65}\text{Cu},X)^{24}\text{Na}$ inverse kinematics reactions. In order to calculate the $Y_{\text{for}}$ and $Y_{\text{inv}}$, the cross section $\sigma(\epsilon)$ and stopping power $S(\epsilon)$ are necessary. The $\sigma(\epsilon)$ as a function of $\epsilon$ is prepared by the spline fitting of experimental data[5], while the $S(\epsilon)$ is computed using the SRIM2008 code[6]. The $\sigma(\epsilon)$ and $R(\epsilon)$, which is calculated from Eq. (6), are shown in Fig. 1 (a) and (b). The $Y_{\text{for}}$ with $A_T(\text{nat}\text{Cu}) = 63.546$ at 40 and 100 MeV/A are evaluated by Eq. (4) and the results are $Y_{\text{for}}(40) = 0.91 \times 10^{-5}$ and $Y_{\text{for}}(100) = 0.114 \times 10^{-3}$. We can also obtain the TTY of inverse kinematics, $Y_{\text{inv}}(40) = 0.86 \times 10^{-5}$ and $Y_{\text{inv}}(100) = 0.103 \times 10^{-3}$, in the same manner as the $Y_{\text{for}}$.

Here, we note that the $R(\epsilon)$ converges on a constant value at the high energy over 50 MeV/A and that the cross section of $^{nat}\text{Cu}(^{12}\text{C},X)^{24}\text{Na}$ is negligible in the low energy region since such fragmentation reaction requires a large amount of energy. This simple
behavior of $R(\epsilon)$ in Eq. (7) and the small $\sigma(\epsilon)$ allows us to utilize a more convenient conversion method as:

$$Y_{\text{for}}(\epsilon_{\text{in}}) \simeq \tilde{R} Y_{\text{inv}}(\epsilon_{\text{in}}),$$

where $\tilde{R}$ is a constant value of $R(\epsilon)$ at the high energy over 50 MeV/A. We can estimate $Y_{\text{for}}$ from $Y_{\text{inv}}$ and $\tilde{R}_{\text{Cu/C}} = 1.1$ and obtain $Y_{\text{for}}(40) = 0.94 \times 10^{-5}$ and $Y_{\text{for}}(100) = 0.113 \times 10^{-3}$. Indeed, these are in good agreement with values derived from Eq. (4). This conversion method is practically justified using the SRIM2008 code and a negligible cross section at the low energy. If the TTY of $^{12}\text{C}(^{63}\text{Cu},X)^{24}\text{Na}$ and $^{12}\text{C}(^{65}\text{Cu},X)^{24}\text{Na}$ can be measured experimentally, we can confirm the method.

Next, we consider the transmutation reaction of $^{137}\text{Cs}$ induced by $^{12}\text{C}$ as the forward kinematics. The transmutation reaction consists of any channels with the exception of the $^{137}\text{Cs}(^{12}\text{C},X)^{137}\text{Cs}$ reaction. This reaction is a considerable one for the radioactive waste disposal. Its inverse kinematics reaction has the $^{12}\text{C}$ target and the radioactive $^{137}\text{Cs}$ projectile. The $Y^{1\text{hr}}$ to transmute the incident particles bombarding the thick-target
is described as:

$$Y_{tr} = \frac{N_{tr}}{N_i} = \frac{N_i - N_u}{N_i},$$  

(9)

where $N_{tr}$, $N_i$, and $N_u$ are the number of transmuted, incident, and un-transmuted particles, respectively. $N_i$ is a countable number experimentally, and $N_u$ can also be observed through detection of the specific gamma-rays if the projectile is radioactive and can be identified by gamma decay modes. The $Y_{tr}$ of a radioactive projectile is therefore obtainable.

In order to evaluate the $Y_{tr}$ for the transmutation reaction of $^{137}\text{Cs}$, we calculate the $R(\epsilon)$ with Eq. (6) using the SRIM2008 code. The $R(\epsilon)$ of the reaction of $^{137}\text{Cs}$ beam on the $^{12}\text{C}$ target (Cs/C) is shown in Fig. 2. The plateau and convergence of $R(\epsilon)$ can be seen in the high energy region as is the case with the previous example. In this system, the convenient conversion method shown in Eq. (8) will also be available. We can find that the Cs/C system is not a special case since the ratios of two other samples, zirconium (Zr/C) and Uranium (U/C), show similar tendencies.

We expect that the reactions at below 20 MeV/A will contribute little to the transmutation TTY since elastic and inelastic reactions govern at the lower energy region. We estimate the range of $R(\epsilon)$, $1.0 \pm 0.14$, as shown in Fig. 2 and find the relation between $Y_{for}^{tr}(\epsilon_{in})$ and $Y_{inv}^{tr}(\epsilon_{in})$:

$$Y_{for}^{tr}(\epsilon_{in}) = 1.0 \pm 0.14 \times Y_{inv}^{tr}(\epsilon_{in}), \quad (\epsilon_{in} > 20\text{MeV/A}).$$  

(10)

The transmutation TTY of the reaction with $^{12}\text{C}$ beam on the $^{137}\text{Cs}$ lump target can be obtained at approximately 10% uncertainty while accuracy will be improved in more high energy regions.
Figure 2  Ratio of differential yields between cesium and carbon (Cs/C) in comparison with two other samples, zirconium (Zr/C) and Uranium (U/C).

4. Summary

In addition to cross sections, the thick-target yield is one key quantity for transmutation to reduce LLFP. While there is a program to measure cross sections of LLFP and minor actinides, direct measurement remains difficult due to high radioactivity. In light of this, we suggest a new method to estimate the thick-target yield using inverse kinematics. As an example, we show that the thick-target yield of carbon-induced reaction on the cesium lump is estimated from the cesium-induced reaction on a carbon target. More precise analysis will be discussed in the forthcoming papers.

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