Model Analysis of Isotope-Production Cross Sections for Proton- and Deuteron-Induced Reactions on ^{93}Zr

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Measurements of isotope-production cross sections in proton- and deuteron-induced reactions on ^{93}Zr at 50, 105, and 209 MeV/nucleon have been performed so far. The calculation by a combination of INCL4.6 and GEM (INCL4.6/GEM) implemented in Particle and Heavy Ion Transport code System (PHITS) shows remarkable underestimation in the neutron-deficient region of odd-atomic number isotopes, where isotopes are produced predominantly via the evaporation process described by GEM. In contrast, the calculation by INCL++/ABLA07 reproduces their production cross sections successfully. The model analysis by using INCL++/ABLA07 and INCL++/GEM reveals that the reexamination of level density used in GEM is required to improve underestimation seen in the production cross sections calculated by PHITS.

1. Introduction

Treatment of High-Level radioactive Waste (HLW) is one of the crucial issues for nuclear energy developments because of the long-term radiotoxicity of long-lived fission products (LLFPs) included in HLW. Nuclear transmutation technology has received much attention as a possible method of the burden reduction of disposal of HLW by converting LLFPs into stable and/or short-lived nuclei. So far, measurements of isotope-production cross sections in proton- and deuteron-induced reactions on ^{93}Zr at 50 [1], 105 [2], and 209 [3] MeV/nucleon have been performed in order to provide the fundamental data necessary for the design of nuclear transmutation system with spallation reaction. The measured data were compared with the theoretical model calculation by Particle and Heavy Ion Transport code System (PHITS) [4] to validate the reliability of PHITS calculation for use in the design of the optimized transmutation system. From the comparison, remarkable underestimation by PHITS calculation in the neutron-deficient region of odd-atomic number isotopes such as Nb and Y isotopes were seen regardless of incident particle and energy as shown in Fig. 1.

In PHITS calculation, the spallation reaction is described by a two-step process: an excited pre-fragment is formed by hard nucleon-nucleon binary collisions in an Intranuclear Cascade process (INC process), and subsequent de-excitation by emissions of light particles is simulated in an evaporation process. The Liège Intranuclear Cascade model (INCL4.6) [5] and the Generalized Evaporation Model (GEM) [6] are used for the INC process and the evaporation process in PHITS calculations, respectively. The magnitude of the production cross section in the neutron-deficient region is mainly determined in the evaporation process. Therefore, the underestimation is inferred

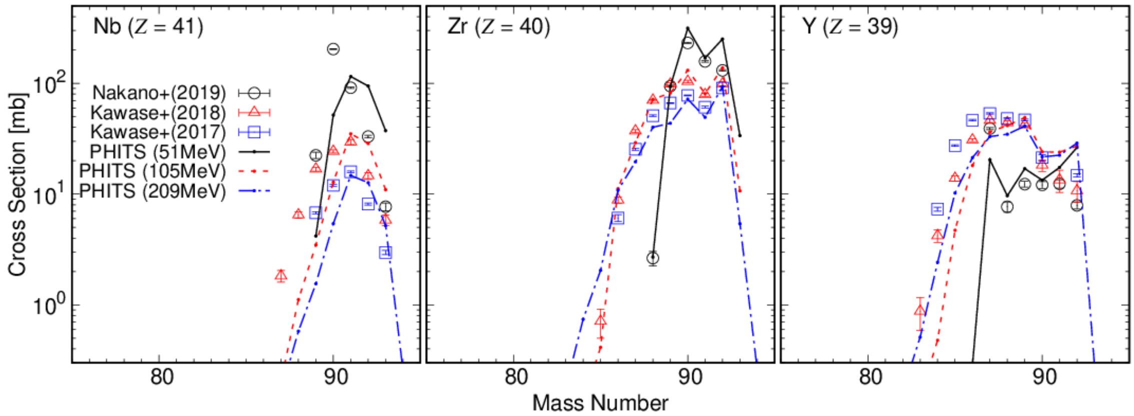

Fig. 1, Isotope-production cross section in proton-induced reaction on ^{93}Zr for Nb (Left), Zr (Center), and Y (Right) isotopes. The circles, triangles, and squares represent the measured data at 51 MeV [1], 105 MeV [2], and 209 MeV [3], respectively. The solid, dashed, and dot-dashed lines indicate the calculated data by PHITS at 51, 105, and 209 MeV, respectively.

to be originated from GEM calculation. In this work, the cause of the underestimation seen in the isotope-production cross sections calculated by PHITS was investigated to achieve a better description of the isotope-production cross sections in the spallation reaction with PHITS.

2. Method

INCL++/ABLA07, which is a combination of C++ version of the Liège Intranuclear Cascade model [7] as the INC process and ABLA07 [8] as the evaporation process, successfully reproduces the isotope-production cross section, especially in the neutron-deficient region of odd-atomic-number isotopes. Therefore, the comparison between ABLA07 and GEM may provide information on the cause of the underestimation in the PHITS calculation. Figure 2 shows the measured isotope-production cross sections in the proton-induced reaction on ^{93}Zr at 105 MeV [2] together with the calculations by INCL4.6/GEM implemented in PHITS and INCL++/ABLA07. The circles, the solid lines, and the dashed lines correspond to the measured data, the PHITS calculation, and the INCL++/ABLA07 calculation, respectively. As mentioned above, INCL++/ABLA07 reproduces the production cross sections in the neutron-deficient region of odd-atomic number isotopes well. While, PHITS fails to predict them. In GEM implemented in PHITS, charged particle emissions are enhanced from the original GEM. To remove the effect of this enhancement, the calculation by using INCL4.6 and original GEM was performed, and the result is shown by the dot-dashed lines in Fig. 2. The underestimation was improved by using the original GEM, but still two times underestimated. Prior to detailed comparisons between ABLA07 and original GEM, we examined the difference in isotope production caused by INC calculations with different codes. The calculation by using INCL++ and original GEM was performed as shown by the dotted lines in Fig. 2. In the regions of interest, the effect by the INC process is negligible. Therefore, we have performed the comparison between ABLA07 and GEM by INCL++/ABLA calculation and that by INCL++ and original GEM (INCL++/GEM).

For the comparison, the ^{93}Zr + p reaction at 105 MeV was chosen. Since the two-step process

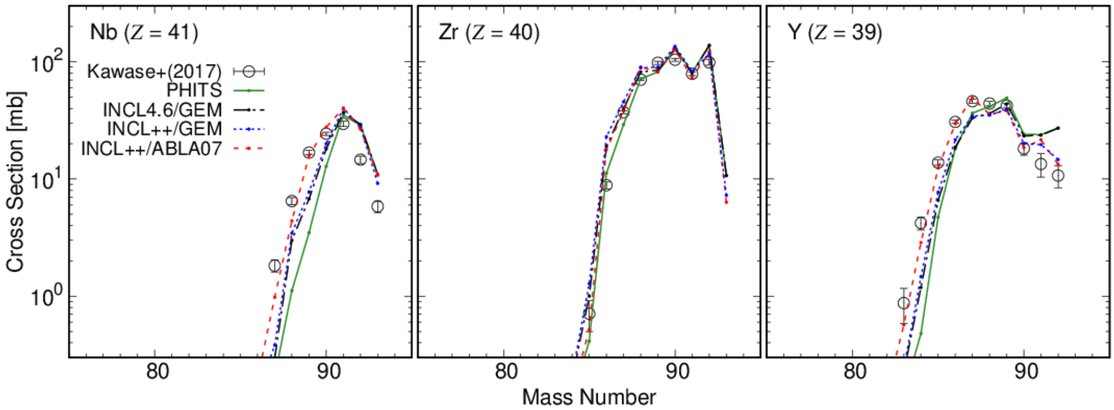

Fig. 2, Isotope-production cross section in proton-induced reaction on ^{93}Zr at 105 MeV for Nb (Left), Zr (Center), and Y (Right) isotopes. The circles represent the measured data [2], and the lines indicate the calculated data by PHITS (solid), INCL4.6/GEM (dot-dashed), INCL++/GEM (dotted), and INCL++/ABLA07 (dashed).

is employed, the de-excitation process depends on the mass and atomic number of pre-fragment and its excitation energy. Monte Carlo calculations of the ^{93}Zr + p reaction at 105 MeV were performed 10^7 times by using INCL++/ABLA07 and INCL++/GEM. For each calculation, the atomic number, the mass number, and the excitation energy of the pre-fragment formed in the INC process described by INCL++ were recorded. Then the evaporation processes were simulated by using the two evaporation codes, ABLA07 and GEM, and the atomic number, the mass number, and the kinetic energy of both the residual nuclei and emitted particles were recorded. The recorded data were analyzed event-by-event to clarify the difference in particle emissions between ABLA07 and GEM.

3. Results and Discussion

Figure 3 shows the branching ratios of proton and neutron emissions from ^{93}Nb* (Left), ^{92}Zr* (Center), and ^{92}Y* (Right) pre-fragments as a function of excitation energy. The dashed and the solid lines represent GEM calculation and ABLA07 calculation, respectively. In the GEM calculation, proton emission branching ratios in ^{93}Nb* and ^{92}Y* are twice as large as those in the ABLA07 calculation over the whole excitation energy. Conversely, the ABLA07 calculation shows a larger proton emission branching ratio in ^{92}Zr* than that in the GEM calculation. The branching ratios of other charged particle emission from ^{93}Nb* and ^{92}Y* are almost the same as those of proton emission. This tendency leads to the underestimation seen in the isotope-production cross sections of odd-atomic number isotopes. In the GEM calculation, the large proton emission branching ratio in Nb pre-fragment enhances the decays from Nb isotopes to Zr isotopes, but the decays from Zr isotopes to Y isotopes are suppressed due to the small proton emission branching ratio in Zr pre-fragments. As a result, the odd-atomic number isotopes have smaller yields in the evaporation stage in the GEM calculation, resulting in the underestimation in the neutron-deficient region of odd-atomic number isotopes.

Next, we investigate the reason why the differences are seen in the branching ratios calculated

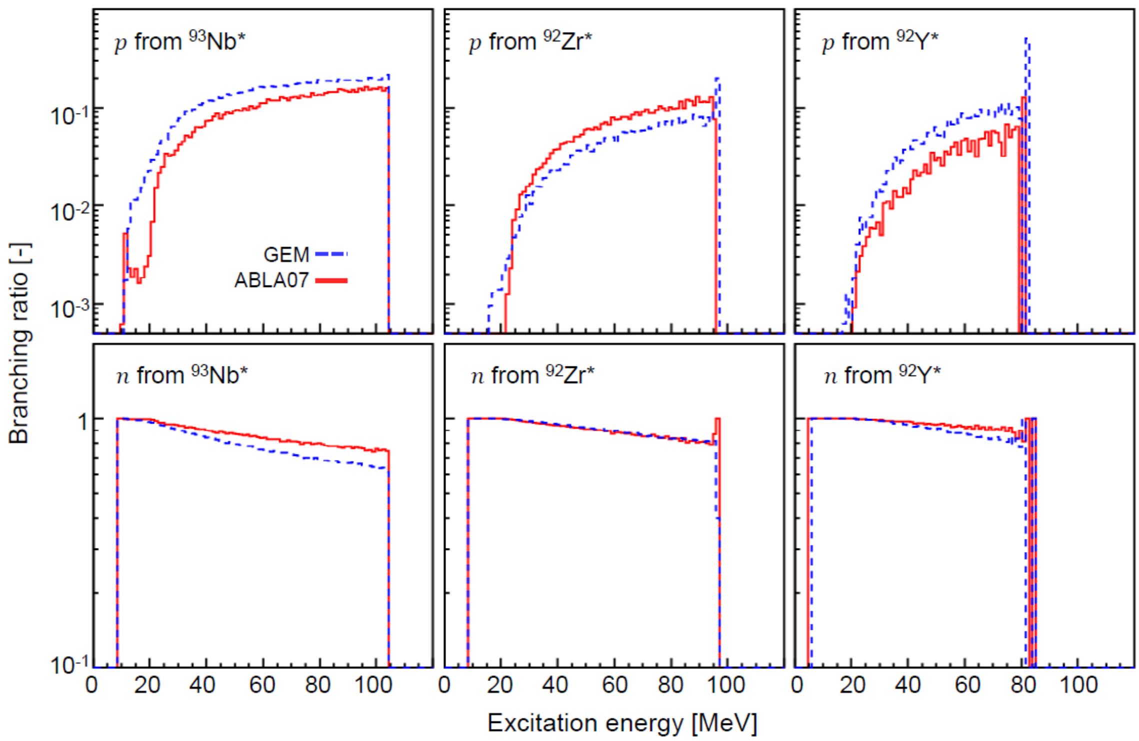

Fig. 3, Branching ratios for proton (Upper) and neutron (Lower) emissions from ^{93}Nb* (Left), ^{92}Zr* (Center), and ^{92}Y* (Right) pre-fragments calculated by GEM and ABLA07 codes as a function of the excitation energy of pre-fragment. The dashed and solid lines correspond to the GEM and ABLA07 calculations, respectively.

in GEM and ABLA07. In GEM and ABLA07, the decay width of emitted particle j is calculated from the inverse cross section and the level density based on the Weisskopf-Ewing formalism [9]:

$$\Gamma_j \propto \int_V^{E_x-Q} \sigma_{\rm inv}(\varepsilon)\frac{\rho_{\rm fin}(E_x-Q-\varepsilon)}{\rho_{\rm init}(E_x)}\varepsilon d\varepsilon, \tag{1}$$

where $E_x, Q, V,$ and ε represent the excitation energy of the parent nucleus, the Q-value of the reaction, the Coulomb barrier between the daughter nucleus and the emitted particle, and the kinetic energy of the emitted particle. Also, $\sigma_{\rm inv}$ denotes the inverse cross section corresponding to the cross section of the inverse process, that is, the incident of the emitted particle into the daughter nucleus. The variables $\rho_{\rm init}$ and $\rho_{\rm fin}$ correspond to the level densities of the parent and the daughter nucleus, respectively. Here, both or either $\sigma_{\rm inv}$ or ρ can be the cause of the underestimation. As the first step, we checked the behavior of $\sigma_{\rm inv}$ with attention to the difference between the odd- and even-atomic number isotopes. Figure 4 shows the $\sigma_{\rm inv}(\varepsilon)$ implemented in GEM and ABLA07. The inverse cross section of the proton emission from ^{93}Nb*, which corresponds to the proton-induced reaction on ^{92}Zr, is drawn by the dashed lines. That of proton emission from ^{92}Zr*, which corresponds to the proton-induced reaction on ^{91}Y, is displayed by the dotted lines. We found that there is no appreciable odd-even effect on the inverse cross section used in both the calculations. Therefore, it is confirmed that the inverse cross section has no dependence on the even-odd of pre-fragment. This strongly implies that the level densities implemented in GEM is the

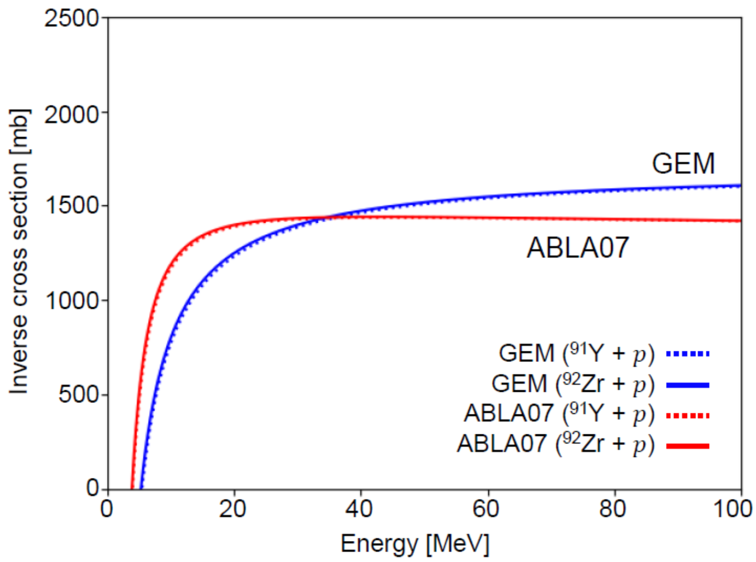

Fig. 4, Inverse cross sections calculated by GEM and ABLA07 as a function of the kinetic energy of the emitted particle. The dotted and dashed lines show the inverse reactions of proton emission from pre-fragments of ^{92}Zr* (^{91}Y + p) and ^{93}Nb* (^{92}Zr + p), respectively.

cause of the remarkable underestimation in the Nb and Y isotopes, and should be reconsidered to resolve the underestimation as discussed in Ref. [10].

4. Conclusion

The isotope-production cross section calculated by INCL4.6/GEM implemented in PHITS shows remarkable underestimation in the neutron-deficient region of odd-atomic number isotopes such as Nb and Y isotopes. The cause of the underestimation was investigated by comparing the calculations by INCL++/GEM and INCL++/ABLA07. From the calculated branching ratios, the proton emissions from the isotopes with the odd-atomic number are greatly enhanced in GEM calculation compared to ABLA07 calculation, resulting in the underestimation of isotope-production cross sections in the neutron-deficient region of odd-atomic number isotopes. Since the inverse cross sections in GEM and ABLA07 show no difference between neighboring nuclei with odd and even atomic numbers, the enhanced proton emission can be originated from the level density in GEM and the modification of the level density may improve the underestimation. In the future, the improvement of the level density in GEM, *e.g.* modification of the pairing correction, is expected.

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