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Cross section measurement of residues produced in proton- and deuteron-induced spallation reactions on ^{93}Zr at 105 MeV/u using the inverse kinematics method

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Abstract. Isotopic production cross sections in the proton- and deuteron-induced spallation reactions on ^{93}Zr at an energy of 105 MeV/u were measured in inverse kinematics conditions for the development of realistic nuclear transmutation processes for long-lived fission products (LLFPs) with neutron and light-ion beams. The experimental results were compared to the PHITS calculations describing the intra-nuclear cascade and evaporation processes. Although an overall agreement was obtained, a large overestimation of the production cross sections for the removal of a few nucleons was seen. A clear shell effect associated with the neutron magic number $N = 50$ was observed in the measured isotopic production yields of Zr and Y isotopes, which can be reproduced reasonably by the PHITS calculation.

1. Introduction

Treatment of the long-lived fission products (LLFPs) produced in nuclear reactors has been an important issue because of the long-term risk in radioactive waste management. Therefore, some sort of treatment method to transform the LLFPs into short-lived and/or low-radiotoxic materials is strongly desired and nuclear transmutation technology is one of the promising candidates for that. However, the reaction data of LLFPs required for the design of an optimum pathway of the transmutation process are quite scarce at this moment. One of the reasons is that there is considerable difficulty in both manufacturing and handling of LLFP targets, which are needed for the conventional measurement in normal-kinematics conditions such as the activation method.

Zirconium-93 is one of the major LLFP nuclei produced in nuclear reactors [1]. It has a half-life of 1.53×10^6 years and decays into a stable nucleus ^{93}Nb or its meta stable state ^{93m}Nb with a half-life of 16 years through β^- emission ($Q_\beta = 91$ keV and 60 keV for ^{93}Nb and ^{93m}Nb, respectively). Despite the importance of ^{93}Zr in the waste treatment due to its long lifetime, the experimental cross section data of the possible reactions in transmutation process are very scarce. Only the thermal neutron capture reaction data [2–4] are currently available. The possibility of the low-energy (< 15 MeV) proton-induced reactions was also discussed [5] but it was mentioned that those reactions are not so promising for the transmutation of ^{93}Zr. For these reasons, the study of spallation reactions with neutrons and light-ions at relatively higher energies has been desired to explore alternative nuclear transmutation processes.

In the present work, we have applied the inverse-kinematics method to measurements of the isotopic production cross sections of the spallation reactions. In this method, the nuclide to be measured and the probe are the

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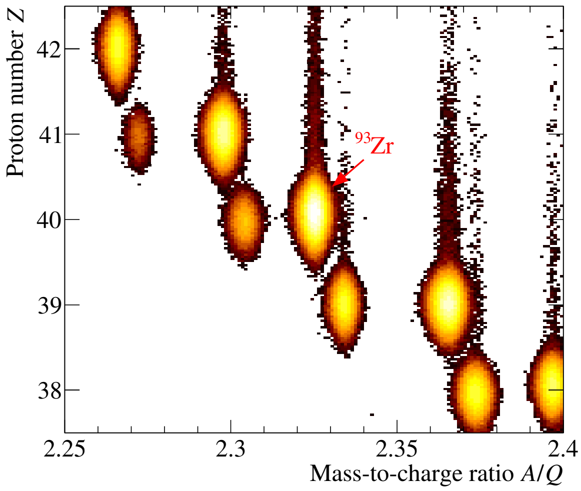

Figure 1. Two-dimensional plot of the proton number Z and the mass-to-charge ratio A/Q of secondary-beam particles in the BigRIPS.

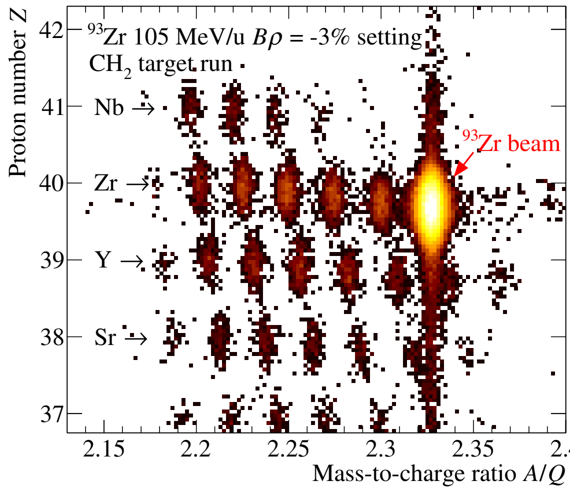

Figure 2. Two-dimensional plot of the proton number Z and the mass-to-charge ratio A/Q of the reaction fragments deduced using the ZeroDegree spectrometer.

projectile and the target, respectively, and hence there is no need to handle a large amount of radioactive materials. Moreover, it has a crucial advantage that one can measure the production yields over a wide range of atomic and mass numbers regardless of their lifetimes. For example, the reaction yield of stable isotopes cannot be measured in principle through the traditional activation method. The inverse-kinematics method has been successfully utilized in the precedent measurements of the spallation reactions [6,7].

A new experiment was performed for the measurements of isotopic production cross sections on an LLFP nucleus ^{93}Zr through the proton- and deuteron-induced spallation reactions. In this article, the experimental result on the isotopic production cross sections will be discussed in a comparison with theoretical model calculations.

2. Experiment

The experiment was carried out at the RIKEN RI Beam Factory (RIBF) [8]. Secondary beams including ^{93}Zr (105 MeV/u) were produced through in-flight fission of a ^{238}U primary beam at 345 MeV/u using a ^{9}Be production target. The produced secondary beam was separated and identified event-by-event by using the BigRIPS in-flight separator with the $B\rho$-TOF-ΔE method described in Ref. [9]. Figure 1 shows a two-dimensional plot of the mass-to-charge ratio A/Q and the proton number Z for the secondary-beam particles which are used in the particle identification (PID) process. The A and Z resolution for ^{93}Zr were 0.16 (FWHM) and 0.40 (FWHM), respectively. A locus corresponding to ^{93}Zr is thus unambiguously identified. The tail to the vertical direction is due to the pile-up events in the ion chamber which was used to measure ΔE but it does not affect the final cross sections at all. The purity of ^{93}Zr was 13.6% in front of the secondary target.

The secondary beam bombarded CH_2 (179.2 mg/cm^2), CD_2 (218.2 mg/cm^2) [10], and natural carbon target (226.0 mg/cm^2) installed at the F8 focal plane. Fragments produced through the spallation reaction were identified event-by-event by using the ZeroDegree Spectrometer (ZDS) [11]. Since the momentum acceptance of ZDS is limited to $\sim \pm 3\%$, the measurement was carried out for 5 different momentum settings ($\Delta(B\rho)/B\rho = -9\%$, -6%, -3%, 0%, and $+3\%$) for each target in order to accept produced isotopes in a wide range of A/Q. For example, Fig. 2 shows a correlation plot in ZDS used for the identification of the reaction products for CH_2 target runs. The A and Z resolution for ^{90}Zr were 0.24 (FWHM) and 0.42 (FWHM), respectively, and as a result, the isotopes were well separated to each other.

3. Results and discussion

The isotopic production cross sections via the spallation reactions were obtained for the residual nuclei in the atomic number range from Nb ($Z = 41$) to Kr ($Z = 36$) by the above-mentioned data analysis. Figure 3 shows the measured data of representative four isotopes (Nb,Zr, Y, and Sr). The crosses and the diamonds indicate that the reaction was induced by the proton-injection and the deuteron-injection, respectively. The contributions of protons and deuterons in the CH_2 and CD_2 targets were obtained by subtracting reaction yields measured in C-target and empty-target runs. The error bars show only the statistical uncertainties.

The niobium production shown in Fig. 3(a) corresponds to charge-exchange channels ^{93}Zr(p, xn) and ^{93}Zr(d, xn). In this channel, the proton-induced cross section (σ_p) is approximately twice larger than that of deuteron-induced one (σ_d). This behavior is consistent with the preceding results obtained in the measurements on ^{137}Cs (185 MeV/u), ^{90}Sr (185 MeV/u) [7], and ^{136}Xe (500A MeV) [6].

For lower Z isotopes than Zr ($Z = 40$), in contrast, σ_d surpassed σ_p and the ratio σ_d/σ_p grew with decrease in Z. It can be attributed to two reasons. One is the difference of the excitation energy of prefragments formed by the intra-nuclear cascade process. Since the total kinetic energy of deuterons is twice as large as that of protons, deuteron injections deposit larger energy to the residual and a reaction residual with a larger excitation energy releases more nucleons. Another is the difference of the two-nucleon (NN) scattering cross sections: σ_{pn} is twice

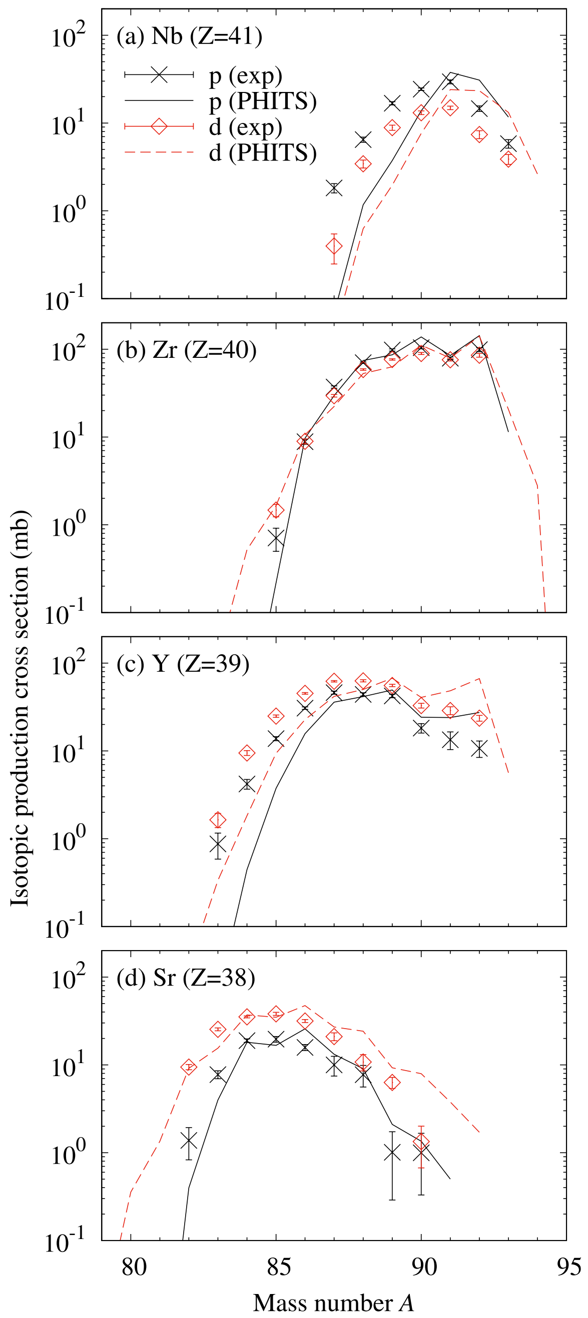

Figure 3. Isotopic production cross sections as a function of mass number for each isotope in the reactions of ^{93}Zr (105 MeV/u) on proton and deuteron: (a) Nb ($Z = 41$), (b) Zr ($Z = 40$), (c) Y ($Z = 39$), and (d) Sr ($Z = 38$).

as large as σ_{pp} and σ_{nn}. Thus more protons are likely to be scattered in the deuteron-injection case.

Also, it is found that there are remarkable jumps in the production of isotopic chain between ^{90}Zr and ^{91}Zr, and between ^{89}Y and ^{90}Y. These can be interpreted qualitatively by the existence of the magic number $N = 50$. ^{91}Zr and ^{90}Y have small neutron separation energies (S_n) and easily lose a neutron while large S_n of the magic nuclei ^{90}Zr and ^{89}Y suppresses further emission of neutrons.

In Fig. 3, the experimental results are compared to the model calculations by using the particle and heavy-ion transport code system (PHITS) 2.82 [12]. The spallation reactions have been well described as a two step process, namely, the formation of prefragments via intra-nuclear cascade process and de-excitation process of the prefragments by evaporation of light particles. In the present work, the cascade and the evaporation processes were described by the Liège Intranuclear Cascade model (INCL 4.6) [13] and the generalized evaporation model (GEM) [14], respectively. The lines in Fig. 3 show the cross sections calculated by using PHITS. The black solid line and the red dashed line correspond to the proton induced and the deuteron induced cross sections, respectively.

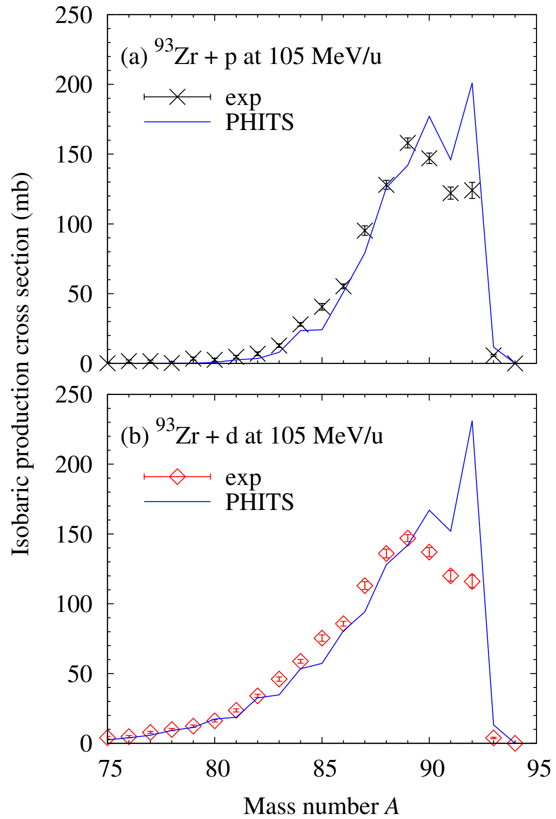

Figure 4. Isobaric distribution of the production cross sections of the residual nuclei measured in the reactions: (a) ^{93}Zr + proton at 105 MeV/u and (b) ^{93}Zr + deuteron at 105 MeV/u.

At first glance, the overall behavior of the isotopic cross section seems to be well reproduced by the PHITS calculations. However, there are two differences between the PHITS calculations and the experimental result. First, the even-odd staggering appearing in the PHITS calculations is not obvious in the experimental result. It can be attributed to the fact that GEM used in the PHITS calculations does not consider γ-ray emission in the evaporation process, which can be expected to smear the staggering by partitioning the excess energy among not only nucleons but also photons. Secondly, the calculation gives larger cross sections for larger A than experimental ones while smaller cross sections for lower A. In particular, the isotope productions near the target nuclei ^{93}Zr are much overestimated.

To make this overestimation clear, the total production cross sections of nuclides having the same mass number A are plotted as a function of A in Fig. 4. The solid lines show the result of the PHITS calculations in which the production cross sections only for the measured isotopes are integrated. It is remarkable that the PHITS calculations considerably overestimate the production of the residual nuclei with $A = 90$–92. In particular, the discrepancy for the residual nuclei with $A = 92$ formed through one-nucleon knockout processes is quite large. On the other hand, the production of isotopes lighter than $A = 90$ is well reproduced by the calculation. These facts suggests that the excitation energy distribution of the prefragments formed after the cascade process is likely to be different from the real one in particular in the low excitation energy part. Moreover, it might be hard to describe appropriately the one-nucleon knockout reaction that mainly occurs near the nuclear surface with the present INCL based on the Fermi-gas model. The large discrepancy near the target nuclei is crucial in the optimization of the reaction path in nuclear transmutation method and therefore some improvement of the calculation models need to be explored in future studies.

4. Summary and conclusions

Isotopic production cross sections of the proton- and deuteron-induced spallation reactions on ^{93}Zr at 105 MeV/u were measured in inverse kinematics condition. The remarkable jumps of the production cross sections were observed at the neutron number $N = 50$. This indicates the importance of the effect of magic numbers in the description of the spallation reactions in the intermediate energy region. The overall behavior of the cross section was well reproduced by PHITS calculations with INCL 4.6 for the intranuclear cascade process and GEM for the evaporation process. However, the even-odd staggering effect was overestimated and also the large discrepancy at the mass region near ^{93}Zr was observed. They are probably due to the absence of the γ-ray emission in GEM and the poor reproduction of the excitation energy distribution of prefragments by INCL, respectively. Further theoretical studies are needed to resolve these inconsistencies.

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