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Cross sections for nuclide production in proton- and deuteron-induced reactions on ^{93}Nb measured using the inverse kinematics method

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Abstract. Isotopic production cross sections were measured for proton- and deuteron-induced reactions on ^{93}Nb by means of the inverse kinematics method at RIKEN Radioactive Isotope Beam Factory. The measured production cross sections of residual nuclei in the reaction ^{93}Nb + p at 113 MeV/u were compared with previous data measured by the conventional activation method in the proton energy range between 46 and 249 MeV. The present inverse kinematics data of four reaction products (^{90}Mo, ^{90}Nb, ^{88}Y, and ^{86}Y) were in good agreement with the data of activation measurement. Also, the model calculations with PHITS describing the intra-nuclear cascade and evaporation processes generally well reproduced the measured isotopic production cross sections.

1. Introduction

The disposal of high-level radioactive waste is one of the crucial issues concerning nuclear power plants. Long-term radioactivity of long-lived fission products (LLFPs) is a large factor of the issue. Research and development of the methods to reduce their half-lives and/or radiotoxicity by nuclear reactions are strongly desired. However, experimental nuclear reaction data of LLFPs are not sufficient to find an optimum pathway of nuclear transmutation because of considerable difficulty in both manufacturing and handling of LLFP targets. To overcome this situation, a new research program has recently been launched on a series of cross section measurements of residues produced in proton and deuteron induced spallation reactions on LLFPs (^{79}Se, ^{93}Zr [1], ^{107}Pd [2], ^{126}Sn and ^{135}Cs) using inverse kinematics at RIKEN Radioactive Isotope Beam Factory (RIBF) [3]. Using the inverse kinematics method, one can measure production yields of residual nuclei over a wide range of atomic and mass numbers including stable nuclei, while the production yields of stable isotopes cannot be measured in principle by conventional activation methods.

To confirm the consistency between the two methods, a measurement of proton and deuteron induced production cross sections on stable nucleus ^{93}Nb was performed using the same inverse kinematics method as in Refs. [1,2]. Niobium-93 was chosen because the experimental data of activation cross sections for proton-induced reactions on ^{93}Nb are available over a wide range of incident energy [4]. Therefore, intercomparison between the data measured by means of the two methods is suitable to confirm the reliability of the inverse kinematics method.

In the present work, the isotopic production cross sections for the reactions induced by ^{93}Nb projectiles on proton and deuteron at 113 MeV/u are derived and

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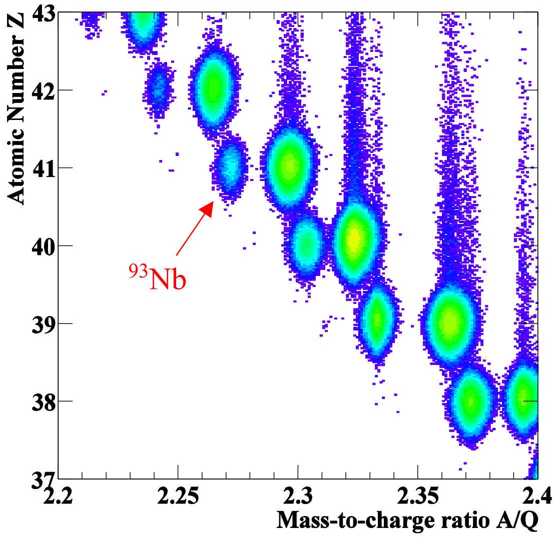

Figure 1. Two-dimensional plot of the atomic number Z and the mass-to-charge ratio A/Q of secondary beam particles in BigRIPS.

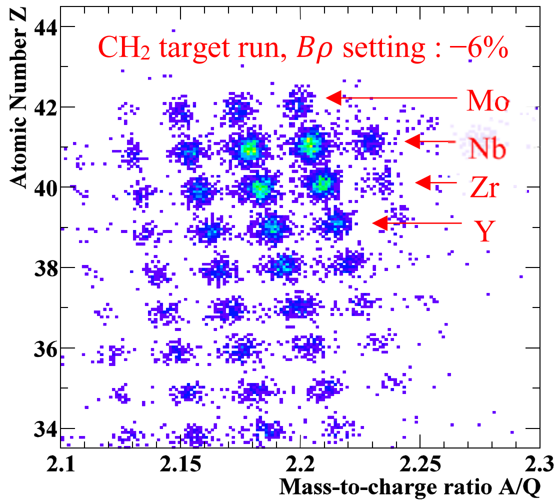

Figure 2. Two-dimensional plot of the atomic number Z and the mass-to-charge ratio A/Q of the reaction fragments in the ZeroDegree Spectrometer.

compared with the previous proton-injection data of activation measurements in the proton energy range from 46 to 249 MeV [4]. Moreover, the measured production cross sections are compared with model calculations using by Particle and Heavy-Ion Transport code System (PHITS) [5] describing the intra-nuclear cascade and evaporation processes in order to investigate the applicability of the reaction models to the prediction of isotopic production cross sections of residual nuclei in proton- and deuteron-induced reactions.

2. Experiment

The experiment was performed at RIKEN RIBF. The experimental setup and procedure were essentially the same as in Refs. [1,2,6].

The secondary beam containing ^{93}Nb was produced using in-flight fission of a ^{238}U primary beam at 345 MeV/u, caused by bombarding ^{9}Be production target located at the entrance of the BigRIPS in-flight separator [3]. The secondary beam was separated and identified by using the BigRIPS. The particle identification was performed event-by-event using detectors installed in the BigRIPS beamline via the TOF$-B\rho-\Delta E$ method [7,8]. The particle identification plot of the secondary beam is shown in Fig. 1. Here the vertical and horizontal axis correspond to the atomic number Z and the mass-to-charge ratio A/Q, respectively. The obtained resolution were 0.40 (FWHM) in Z and 0.23 (FWHM) in A, which are sufficient for clear identification of ^{93}Nb ions. The energy of ^{93}Nb at the center of the secondary target was 113 MeV/u and its purity which means the ratio of the number of ^{93}Nb ions to that of all ions in front of the secondary target was 4.4%. The contribution of the isomer state ^{93m}Nb ($E = 0.0308$ MeV, $T_{1/2} = 16.12$ years) included in the ^{93}Nb beam was considered to be small.

Then the secondary beam irradiated the secondary targets CH_2 (179.2 mg/cm^2), CD_2 (217.8 mg/cm^2), and natural carbon (226.0 mg/cm^2) placed at the entrance of the ZeroDegree Spectrometer (ZDS) [3]. The residual nuclei produced by nuclear reactions were identified event-by-event using ZDS by the technique similar to the method used in the particle identification at BigRIPS. The momentum acceptance of ZDS is limited to less than $\pm 3\%$. In order to measure the wide range of residual nuclei, five different magnetic rigidity (B_ρ) settings ($\Delta(B_\rho)/B_\rho = -9\%$, -6%, -3%, 0%, and $+3\%$) were used. Here $\Delta(B_\rho)/B_\rho$ means the B_ρ value relative to that of the secondary beams. The particle identification plot of residual nuclei produced by nuclear reaction is shown for CH_2 target run with the setting of -6% in Fig. 2. The resolutions for ^{90}Nb were 0.53 (FWHM) in Z and 0.26 (FWHM) in A and reaction fragments are identified unambiguously.

The isotopic production cross sections of residual nuclei were derived by subtracting the background contribution of carbon and empty frame from residue yields measured in the CH_2 and CD_2 target runs. The proton induced cross section (σ_p) is given by

$$\sigma_p = \frac{1}{2}\left\{ \frac{1}{A_{\mathrm{CH_2}} T_{\mathrm{CH_2}}} \left(\frac{Y_{\mathrm{CH_2}}}{C_{\mathrm{CH_2}} B_{\mathrm{CH_2}}} - \frac{Y_{\mathrm{E}}}{C_{\mathrm{E}} B_{\mathrm{E}}} \right) - \frac{1}{A_C T_C} \left(\frac{Y_C}{C_C B_C} - \frac{Y_E}{C_E B_E} \right) \right\}, \tag{1}$$

where B is the number of ^{93}Nb projectiles and Y is that of detected residual nuclei, and T is the areal density of secondary targets. The correction factors A and C are used to correct for the lost events by the limitation of the acceptance and charge state exchange. The acceptance was limited by the horizontal positions at ZDS. In addition, the particle identification is not performed correctly except for the particles in fully-stripped states. The subscripts CH_2, C, and E denote individual runs with the CH_2 target, the C target, and the empty frame target, respectively. For deuteron induced cross section (σ_d), the subscript CH_2 is replaced by CD_2 in Eq. (1). The systematic error is 1% for uncertainties of the corrections for charge state exchange and less than 2% for those of thickness of the secondary targets.

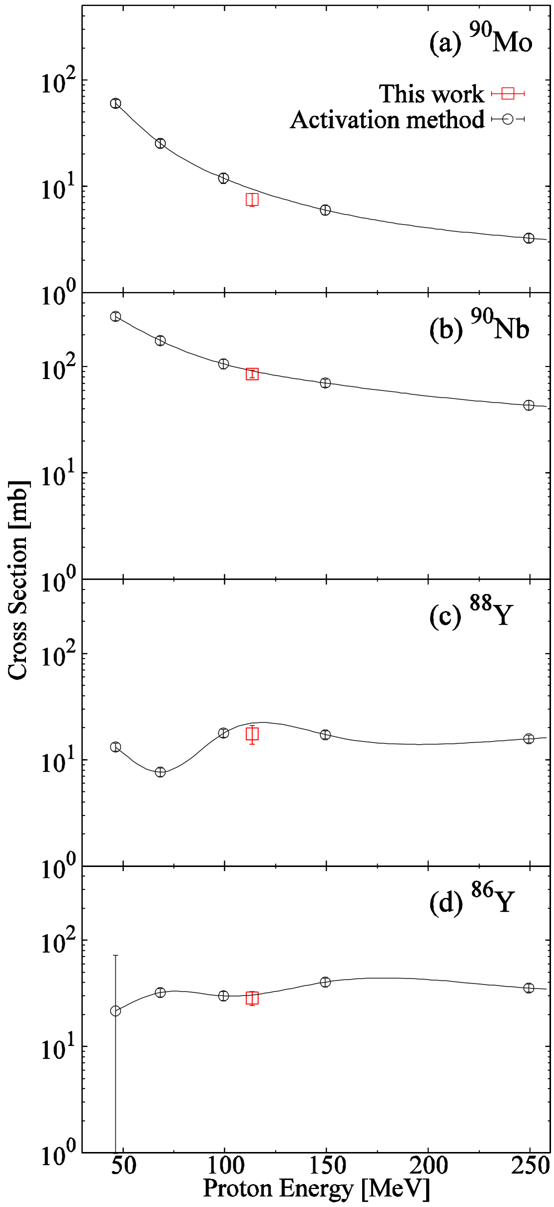

Figure 3. Comparison of production cross sections between the inverse kinematics method and the activation method: (a) ^{90}Mo, (b) ^{90}Nb, (c) ^{88}Y, and (d) ^{86}Y.

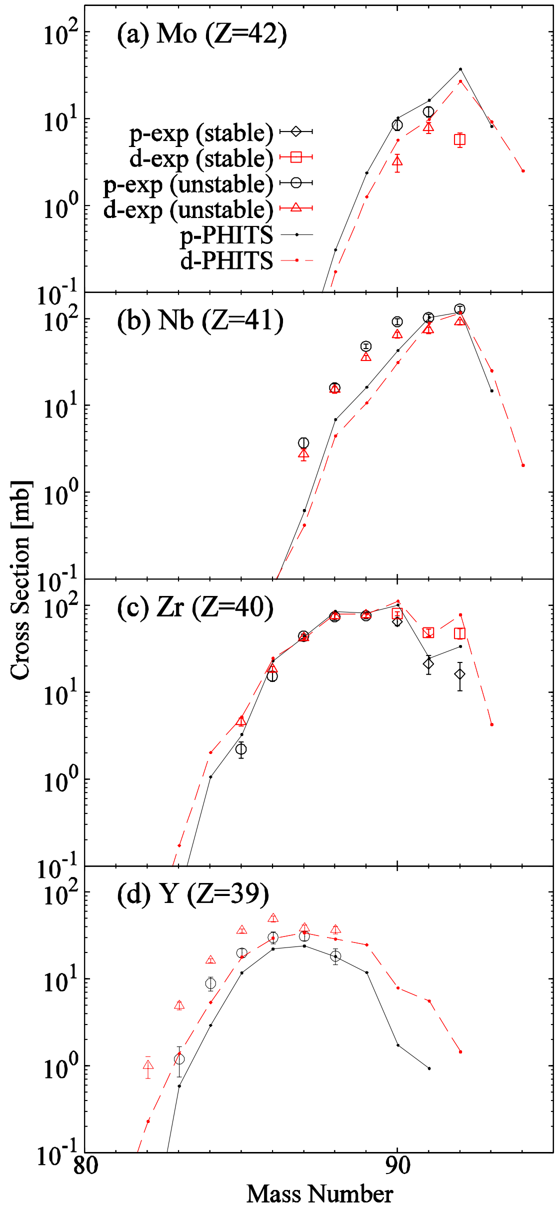

Figure 4. Isotopic production cross section as a function of mass number for each isotope in the experimental acceptance: (a) Mo (Z = 42), (b) Nb (Z = 41), (c) Zr (Z = 40), (d) Y (Z = 39). The data of stable nuclei are denoted by closed symbols.

3. Results and discussion

In Fig. 3, the measured production cross sections of four nuclides (^{90}Mo, ^{90}Nb, ^{88}Y, and ^{86}Y) denoted by open squares are compared with the previous data of activation measurement [4] denoted by open circles. Note that the data of activation measurement are interpolated with the smooth curves drawn by cubic spline function. The error bars include only statistical uncertainties. The present data corresponding to the proton energy of 113 MeV are just on the interpolated curves within statistical errors in the cases of ^{90}Nb and ^{86}Y. Although the data are slightly below the interpolated curves in the cases of ^{90}Mo and ^{88}Y, the curves are within statistical and systematic errors. This result indicates that the experimental data by the inverse kinematics method are consistent with those by the activation method. Thus it was confirmed that the inverse kinematics is a reliable technique to obtain production cross sections of residual nuclei in the proton- and deuteron-induced reactions.

The measured isotopic production cross sections of residual nuclei in both the reactions induced by ^{93}Nb projectiles on proton and deuteron at 113 MeV/u are shown for Mo, Nb, Zr, and Y isotopes in Fig. 4. The black and red symbols denote the data of proton-induced reaction and those of deuteron-induced reaction. The diamonds and squares represent the data of stable nuclei and circles and triangles represent those of unstable nuclei. The error bars include only statistical uncertainties. Compared with the activation method, the experimental result shows the advantage that one can measure a wide range of isotopic production data including stable nuclei by using inverse kinematics method. In Fig. 4, the measured data are compared with model calculations using the PHITS ver.2.76 [5] shown by the solid and dashed lines corresponding to proton and deuteron induced cases, respectively. In the PHITS calculations, the reaction is modelled by two-step processes: the formation of prefragments via intra-nuclear cascade process and the deexcitation process of prefragments by particle evaporation. Both the processes are described by the Liège Intranuclear Cascade model (INCL 4.6) [9] and the generalized evaporation model (GEM) [10], respectively. The overall behavior of measured isotopic cross sections is reproduced reasonably well by the PHITS calculations, but some disagreements are seen between the present measurement and PHITS calculations. The calculation overestimates largely the production of ^{92}Mo via the ^{93}Nb(d,3n) reaction. In addition, relatively large discrepancy exists for ^{92}Zr formed by single proton knockout reactions. On the other hand, the production of neutron-deficient Nb isotopes and the production of Y isotopes in the deuteron case are underestimated. Further work on improvement of the reaction models will be required for reproduction of the experimental cross sections.

4. Summary and conclusions

Isotopic production cross sections of proton- and deuteron-induced reactions on ^{93}Nb at 113 MeV/u were measured by using the inverse kinematics method. The measured production cross sections of residual nuclei were compared with the previous data measured by the activation method. It was confirmed that the inverse kinematics data of four reaction products (^{90}Mo, ^{90}Nb, ^{88}Y, and ^{86}Y) are consistent with the activation data. In addition, the PHITS calculations with INCL 4.6 for the intra-nuclear cascade and GEM for the evaporation process shows overall agreement with the measured isotopic production cross sections of Mo, Nb, Zr, and Y isotopes, but some disagreement are seen. Further theoretical works are necessary to resolve these disagreements.

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