Study of proton- and deuteron-induced spallation reactions on the long-lived fission product $^{93}$Zr at 105 MeV/nucleon in inverse kinematics

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Spallation reactions for the long-lived fission product $^{93}$Zr have been studied in order to provide basic data necessary for nuclear waste transmutation. Isotopic-production cross sections via proton- and deuteron-induced spallation reactions on $^{93}$Zr at 105 MeV/nucleon were measured in inverse kinematics at the RIKEN Radioactive Isotope Beam Factory. Remarkable jumps in isotopic production originating from the neutron magic number $N = 50$ were observed in Zr and Y isotopes. The experimental results were compared to the PHITS calculations considering both the intranuclear cascade and evaporation processes, and the calculations greatly overestimated the measured production yield, corresponding to few-nucleon-removal reactions. The present data suggest that the spallation reaction is a potential candidate for the treatment of $^{93}$Zr in spent nuclear fuel.

Subject Index D23, D50

1. Introduction

For a long time, the management of radioactive waste from spent fuel has been one of the crucial issues in the sustainable use of nuclear power. Nuclear waste transmutation by accelerator-driven systems (ADS) and fast reactors (FR) was proposed for reduction of high-level radioactive waste.
(HLW), and a great deal of research and development (R&D) has so far been devoted toward the realization of transmutation technology [1]. In the R&D, fundamental nuclear physics plays an essential role in exploring the possible options of nuclear transmutation.

In the present work, we pay attention to a radioisotope, $^{93}$Zr, which is one of the typical long-lived fission products (LLFPs) contained in HLW. It takes a large share of production yield ($6.3 \times 10^{-2}$ per thermal neutron fission of $^{235}$U) [2,3]. It has a half-life of $1.53 \times 10^6$ years and decays through $\beta^-$ emission into the stable nucleus $^{93}$Nb ($Q_\beta = 91$ keV) or its metastable state $^{93m}$Nb ($Q_\beta = 60$ keV), which has a half-life of 16 years. Though the radiotoxicity is limited by its low-energy radiation, its long lifetime increases concerns about the long-term storage of reprocessed waste. For consideration of a reasonable transmutation process to reduce the effective lifetime of $^{93}$Zr, experimental cross section data of the possible reaction are very scarce. Only a few thermal neutron-capture reaction data [4–6] are currently available, but the cross section is relatively small compared to those of the other LLFPs. The efficiency of proton-induced reactions, such as ($p$, $n$), ($p$, $2p$), or ($p$, $pn$) with energies lower than 15 MeV, has also been discussed [7] but it was concluded that those reactions are not so effective for the transmutation of $^{93}$Zr.

Spallation reaction can be an alternative option for nuclear transmutation. Recent experimental studies indicate that proton- and/or deuteron-induced spallation reactions at intermediate energy (100–200 MeV/nucleon) are sufficiently effective for the transmutation of fission products $^{137}$Cs, $^{90}$Sr [8], and $^{107}$Pd [9]. In contrast to neutron capture, the total cross section of spallation reactions is almost independent of nuclear structure. Thus spallation reactions are expected to be more effective for the transmutation of $^{93}$Zr, which has a small neutron-capture cross section.

Although the systematics of the total cross section is well known, measurements of the isotopic-production cross sections are significant. Firstly, information on neutrons emitted through the reaction is important. Highly excited prefragments, which are formed at the first stage of the spallation reaction, emit nucleons or light fragments via the evaporation process. Among them, neutrons can invoke subsequent neutron-capture reactions and hence they are especially important for use in further transmutation processes. For that purpose, it is important to understand the neutron number distribution of residual nuclei and the energy distribution of emitted neutrons. Secondly, the production of long-lived radioactive isotopes through the reaction, which should be minimized in transmutation processes, can be more precisely evaluated if the experimental data are available.

In this paper, we report on the results of the experimental study on spallation reactions of the LLFP $^{93}$Zr. The isotopic-production cross sections through proton- and deuteron-induced spallation reactions on $^{93}$Zr at 105 MeV/nucleon were measured by using the same inverse kinematics technique as in Refs. [8,9]. The experimental results were compared to theoretical model calculations utilizing both intranuclear cascade and evaporation models. Finally, the efficiency of the spallation reaction on $^{93}$Zr for the transmutation process is also discussed.

2. Experiment

The experiment was carried out at the RIKEN RI Beam Factory (RIBF). Secondary beams including $^{93}$Zr were produced by in-flight fission of a $^{238}$U primary beam at 345 MeV/nucleon using a 3 mm thick $^9$Be production target. The secondary beam produced was separated and identified event-by-event by using the BigRIPS in-flight separator with the $Bp$–TOF–$\Delta E$ method [10]. The momentum acceptance of BigRIPS was limited to $\pm 1\%$ by the slit at the momentum-dispersive focal plane F1. Figure 1 shows the correlation plot of the mass-to-charge ratio $A/Q$ and the proton number $Z$ for the secondary beam. The $A$ and $Z$ resolutions for $^{93}$Zr were 0.16 (FWHM) and 0.40 (FWHM),
respectively. Thanks to the excellent resolving power of BigRIPS, the objective isotope $^{93}\text{Zr}$ was unambiguously distinguished out of the cocktail beams. The tail in the vertical direction is due to pile-up signals in an ion chamber installed at the double-achromatic focal plane, but it does not affect the final cross sections at all. The typical intensity of the $^{93}\text{Zr}$ beam was $7.1 \times 10^3$ counts per seconds (cps) and the purity was 13.6% in front of the secondary target.

The secondary beams bombarded CH$_2$ (179.2 mg/cm$^2$), CD$_2$ (218.2 mg/cm$^2$), and natural C targets (226.0 mg/cm$^2$) installed at the F8 focal plane, which is located downstream of BigRIPS. The beam energy was 105 MeV/nucleon at the center of the secondary targets. Fragments produced through the spallation reaction were momentum-analyzed and identified event-by-event by using the ZeroDegree Spectrometer (ZDS) [11] with a similar method to that in BigRIPS. Since the momentum acceptance of the ZDS is limited to $\pm 3\%$, measurements were done for five different momentum settings ($\Delta (B\rho)/B\rho = -9\%, -6\%, -3\%, 0\%, \text{ and } +3\%$) for each target in order to cover the produced isotopes with a wide range of $A/Q$.

Figure 2 shows a correlation plot of $A/Q$ and $Z$ for the $\Delta (B\rho)/B\rho = -6\%$ run in the ZDS used for particle identification. The $A$ and $Z$ resolutions for $^{90}\text{Zr}$ were 0.24 (FWHM) and 0.42 (FWHM), respectively. For background estimation, measurements with an empty target, i.e., the target frame, were also carried out.

The charge state distribution at the ZDS was determined from the particle trajectory in the beamline. The charge state ratios of the $^{93}\text{Zr}$ beam at the F11 focal plane at the end of the ZDS were 88.6%, 11.3%, and 0.1% for the fully stripped ($Q = Z$), the hydrogen-like ($Q = Z - 1$), and the helium-like ($Q = Z - 2$) charge states, respectively.

3. Results and discussion

The isotopic-production cross sections via the spallation reactions using proton- and deuteron targets are shown in Fig. 3. The circles and diamonds indicate proton-induced cross sections ($\sigma_p$) and deuteron-induced cross sections ($\sigma_d$), respectively. The contributions of protons and deuterons in
Fig. 2. Correlation plot of the proton number $Z$ and the mass-to-charge ratio $A/Q$ in the ZeroDegree spectrometer.

Fig. 3. Isotopic-production cross section as a function of mass number for each isotope in the experimental acceptance: (a) Nb ($Z = 41$), (b) Zr ($Z = 40$), (c) Y ($Z = 39$), (d) Sr ($Z = 38$), (e) Rb ($Z = 37$), and (f) Kr ($Z = 36$).

the CH$_2$ and CD$_2$ targets were extracted by subtracting those of carbon and background deduced from the C-target and empty-target runs. The error bars show only the statistical uncertainties. The systematic uncertainties are from two factors. One is the target thickness, which is less than 2%, and
Fig. 4. Comparison of the charge-unchanging ($\Delta Z = 0$) cross sections with the result of $^{107}$Pd + $p, d$ reactions at 118 MeV/nucleon [9]. Cross sections are normalized by the one-neutron-removal cross section for each data set.

another is the charge state distribution in the ZDS. Since the charge state distributions for isotopes except for Zr were not measured, they were estimated by calculations using the LISE++ code [12] and their uncertainties are 5% at most.

The niobium production shown in Fig. 3(a) corresponds to the charge-increasing reactions $^{93}$Zr($p, xn$) and $^{93}$Zr($d, xn$). In this channel, $\sigma_p$ is approximately twice as large as that of $\sigma_d$. This behavior is consistent with the preceding experimental results obtained in the measurements on $^{136}$Xe (500 $A$ MeV) [13], $^{137}$Cs (185 MeV/nucleon) [8], $^{90}$Sr (185 MeV/nucleon) [8], and $^{107}$Pd (196 and 118 MeV/nucleon) [9].

For lower $Z$ isotopes than Zr ($Z = 40$), in contrast, $\sigma_d$ surpasses $\sigma_p$ and the ratio $\sigma_d/\sigma_p$ grows with a decrease in $Z$. This is attributed to two causes. One is the difference in the excitation energy of prefragments formed by the intranuclear cascade process. Since the total kinetic energy of deuterons is twice as large as that of protons, the deuteron-induced reaction results in higher excitation energy in the residual nuclei, followed by the release of more nucleons. Another is the difference of the two-nucleon ($NN$) scattering cross sections: $\sigma_{pn}$ is about twice as large as $\sigma_{pp}$ and $\sigma_{nn}$. Thus, more protons are likely to be scattered in the deuteron-induced case.

Remarkable jumps are found in the production of isotopic chains between $^{90}$Zr and $^{91}$Zr and between $^{89}$Y and $^{90}$Y. The jumps can be interpreted qualitatively by the existence of the magic number $N = 50$. $^{91}$Zr and $^{90}$Y have relatively small neutron separation energies ($S_n$) and easily lose a neutron, while the large $S_n$ of the magic nuclei $^{90}$Zr and $^{89}$Y suppresses further emission of neutrons. Figure 4 shows the comparison of charge-unchanging ($\Delta Z = 0$) cross sections with those of $^{107}$Pd + $p, d$ reactions at 118 MeV/nucleon [9]. The horizontal and vertical axes show the change of neutron number ($\Delta N$) and isotopic-production cross sections normalized by the one-neutron-removal cross section. For the $^{107}$Pd case, unlike the $^{93}$Zr case, the cross sections show a nearly monotonic decrease with a decrease in $\Delta N$ and no jump is seen. Here, both $^{93}$Zr$_{53}$ and $^{107}$Pd$_{61}$ are proton-even neutron-odd nuclei and also the reaction energies are close to each other. The only major difference is the existence of the magic number $N = 50$ at $^{90}$Zr. Thus we conclude that the enhancement in the isotope production at $^{90}$Zr is due to the effect of the magic number $N = 50$. 

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This is the first clear observation of the magic number effect in proton- and deuteron-induced spallation reactions, thus proving the advantage of the inverse kinematic method: it enables measurement of the yield of stable residues. By the conventional activation method, observation of this effect is quite difficult because the magic nuclei in the vicinity of a nonradioactive target are normally stable and therefore their production yield cannot be obtained by γ-ray detection after beam irradiation. It should be noted that this effect should be remarkable in rather low-energy conditions in which the excitation energies of residual nuclei formed after dynamical processes are comparable to the nucleon separation energy of magic nuclei. For example, in the case of the $^{136}$Xe + $^p$ reaction at 500.4 MeV [13], the energy is too high compared to the nucleon separation energy and therefore no magic effect was observed at either $Z = 50$ or $N = 50$.

In Fig. 3, the experimental results are compared to the model calculations by using the Particle and Heavy Ion Transport code System (PHITS) 2.88 [14]. The spallation reactions have been well described as a two-step process, namely, the formation of prefragments via the intranuclear cascade process and the deexcitation process of the prefragments by evaporation of light particles. In the present work, the cascade and evaporation processes were described by the Liège Intranuclear Cascade model (INCL) [15] and the generalized evaporation model (GEM) [16], 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- and deuteron-induced production cross sections, respectively.

At first glance, the overall behavior of the isotopic cross section seems to be well reproduced by the PHITS calculations. The jumps in the production cross section at the magic number for Zr and Y isotopes are also in reasonable agreement. In GEM, the shell effect is included naturally by using the realistic mass table from the evaluated data [17] and the comparison shows that the description in GEM is also reasonably good for the isotopes neighboring magic nuclei, in which the nucleon separation energy undergoes a significant variation.

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, as shown clearly in the Sr isotopes. This can be attributed to the fact that the GEM used in the PHITS calculations does not consider the competition between particle and γ-ray emissions in the evaporation process. Ricciardi et al. [18] have discussed the role of the γ-ray emission in the deexcitation steps of the nuclide production in fragmentation reactions and demonstrated that the inclusion of γ-ray emission is primarily responsible for the smearing of the even–odd staggering appearing in the evaporation model calculation. Secondly, the calculation gives larger cross sections for larger $A$ than the experimental ones and smaller cross sections for lower $A$. In particular, the isotope productions near the target nuclei $^{93}$Zr are greatly overestimated.

To make this overestimation clear, the total production cross section of the observed nuclides with the same mass number $A$ is plotted as a function of mass number $A$, proton number $Z$, and neutron number $N$ in Fig. 5. The solid and dashed lines show the result of the PHITS calculations in which only the production cross sections for the measured isotopes are integrated. In Figs. 5(a) and (b), 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 calculations. These experimental findings suggest 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. In the INCL model, the excitation energy of the prefragment is determined from the energy conservation law...
Fig. 5. Dependence of production cross sections as functions of mass number $A$, proton number $Z$, and neutron number $N$: (a), (c), (e) for proton-induced cross sections, (b), (d), (f) for deuteron-induced cross sections.

Fig. 6. Decomposition of the mass number distribution of $\sigma_p$ by impact parameter $b$.

(see Sect. II C in Ref. [19]). Therefore this discrepancy requires close reexamination of the cascade process itself.

Figure 6 shows the decomposition of the mass number distribution of $\sigma_p$ by impact parameter $b$ in the PHITS calculations. Here the reactions with $b$ larger than 5 fm, which is comparable to the radius of the $^{93}$Zr nucleus used in INCL [15,19], are considered as “peripheral” collisions. The red dashed line and the blue dash-dotted line show the contributions of nonperipheral and peripheral collisions, respectively. Obviously, the peripheral collision largely contributes to the few-nucleon-removal cross section, in which the major discrepancy exists. In the INCL model [15], a Woods–Saxon density distribution with a diffused surface is used as the initial spatial nucleon distribution. The nucleons in the nuclear peripheral region have energy close to the Fermi energy and knocking out such nucleons results in much smaller excitation energies of the residual nuclei than those expected from quantum-mechanical models such as shell models, according to Ref. [20].

To evaluate this surface effect in INCL, we calculated the excitation energy distributions and the isotopic yields while setting the diffuseness parameter in the spatial nucleon density to zero as an
Fig. 7. Comparison to the calculation with uniform spherical nucleon density (INCL-Uniform): (a) excitation energy distribution of the prefragment $^{92}$Zr formed after the cascade process, (b) isotopic production originating from the decay of the prefragment $^{92}$Zr.

extreme case. In this case, the spatial nucleon density is uniform from the center to the nuclear radius and the momentum distribution at the surface is as uniform as in the central part of the nucleus. Figure 7(a) shows the excitation energy of the prefragment $^{92}$Zr formed after the cascade process with the original INCL model (INCL-WS) and the INCL with zero diffuseness assumption (INCL-Uniform). In the INCL-Uniform case, the cross section in the low excitation energy region suffers a significant decrease from INCL-WS, as expected, and the average excitation energy slightly increases. As a result, as shown in Fig. 7(b), the $^{92}$Zr yield originating from the evaporation process of the excited prefragment $^{92}$Zr drastically drops and the yield of small-$A$ isotopes slightly grows. This change coincides with a trend that lessens the disagreement of the mass number distribution shown in Fig. 5(a). Although this simple modification in the peripheral region can improve the reproduction in this case, modification of the model should, nevertheless, be discussed carefully with consideration of systematic experimental data of isotopic-production cross sections and energy distributions of emitted particles. This kind of improvement was attempted with a fuzzy surface plus skin model [20] but the improvement was not satisfactory. Further improvement of the model is needed.

In Figs. 5(c) and (d), the reproduction of the proton number distribution seems rather good. At the same time, in Figs. 5(c) and (f), the overestimation in few-nucleon removal is clearly seen in the neutron number distribution. This fact suggests that the branching ratio of proton and neutron emission in evaporation from low excited prefragments should be reexamined simultaneously with the excitation energy distribution. Better neutron number distribution with little change in the proton number distribution could be achieved if the average excitation energy is larger and the branching ratio of proton emission is smaller. Since the branching ratio is determined by the decay width calculated with the inverse reaction cross section and the level density of the residual nucleus in the evaporation model, these quantities used in the GEM should be carefully reexamined. In common with Figs. 3 and 4, the jump at magic number $N = 50$ is quite significant. Hence the shape of the neutron number distribution is probably an important clue to the improvement of the evaporation model.
Table 1. Production summary of long-lived products after the spallation reactions.

| product | $T_{1/2}/\text{yr}$ | $\sigma_p/\text{mb}$ | $\sigma_d/\text{mb}$ | daughter       |
|---------|---------------------|----------------------|----------------------|----------------|
| $^{92}\text{Nb}$ | $3.47 \times 10^7$ | 14.6(11)             | 7.4(8)               | $^{92}\text{Zr}$ (stable) |
| $^{81}\text{Kr}$ | $2.29 \times 10^5$ | 3.3(7)               | 8.4(7)               | $^{81}\text{Br}$ (stable) |
| $^{81}\text{Rb}$ | 4.572             | 1.5(7)               | 15.2(8)              | $^{81}\text{Kr}$       |
| Total   |                    | 19.4(15)             | 31.0(13)             |                 |

To evaluate the effectiveness of the spallation reaction for the transmutation process, the integral cross section is a good measure. The integral cross sections for proton- and deuteron-induced reactions were determined to be 936(11) mb and 1127(9) mb, respectively, by integrating all the measured production cross sections in Fig. 3. The total reaction cross sections in the PHITS calculations are 998 mb and 1313 mb, respectively, slightly (7% and 17%) larger than the experimental integral cross sections. One possible reason for the overestimation of deuteron-induced cross section is that the present measurement is blind to elastic and inelastic breakup of deuterons with low excitation energy, which does not cause any decay of $^{93}\text{Zr}$ nuclei. They are included in the total reaction cross section [21] used in the PHITS calculations.

Though the spallation cross sections are about half of the neutron-capture cross section of 2.239 b in JENDL-4.0 [3], the spallation reaction may still be promising for the transmutation of $^{93}\text{Zr}$ because the transmutation yield is also proportional to the beam flux. By the same discussion as in Ref. [9], the spallation reaction can offer a one order of magnitude larger transmutation effect than the neutron-capture reaction. Furthermore, the deuteron-induced reaction may be a good candidate because the secondary neutrons emitted through the cascade and evaporation processes can also be used in further transmutation processes, e.g., neutron-capture reactions caused in surrounding materials.

From the standpoint of nuclear waste management, the small production of long-lived radioactive isotopes is preferred. Major products are $^{92}\text{Nb}$ and $^{81}\text{Kr}$. In addition, $^{81}\text{Rb}$ accompanies them because it decays into long-lived $^{81}\text{Kr}$ after its 4.5 hour half-life. Table 1 shows a summary of the long-lived isotope production. The sum of the cross sections of the listed isotopes is less than 3% of the total cross section for both proton- and deuteron-induced cases. Therefore, we can conclude that the spallation reaction can effectively be used to reduce the radioactivity of $^{93}\text{Zr}$ in spent nuclear fuel.

4. Summary

The isotopic-production cross sections of the proton- and deuteron-induced spallation reactions of $^{93}\text{Zr}$ at 105 MeV/nucleon were measured in inverse kinematics. Remarkable jumps in the cross sections were observed at the neutron number $N = 50$ for Zr and Y isotope production. This indicates the importance of the effect of magic numbers in the description of spallation reactions in the intermediate energy region. The overall behavior of the cross section was well reproduced by the PHITS calculations with INCL 4.6 for the intranuclear cascade process and GEM for the evaporation process. However, the even–odd staggering effect was too large and a large overestimation of the few-nucleon-removal channel was also seen. These are probably due to the absence of $\gamma$-ray emission from unbound states in prefragments in GEM and the poor reproduction of the excitation energy distribution of prefragments by INCL. The latter may be refined with more realistic treatment of the intranuclear cascade process, especially in the peripheral region. The present data indicate that the spallation reaction is a promising candidate for the reduction of the radioactivity of $^{93}\text{Zr}$.
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