Spallation reaction study for long-lived fission products in nuclear waste

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Abstract. Spallation reaction for the long-lived fission product ^{107}Pd has been studied for the purpose of nuclear waste transmutation. The isotopic-distribution cross sections on both proton and deuteron were obtained at 118 MeV/nucleon in inverse kinematics at the RIKEN Radioactive Isotope Beam Factory. A large cross-section difference was found between the proton and deuteron results for the light-mass products. The data were compared with the SPACS semi-empirical parameterization and the PHITS calculations including both the intranuclear cascade and evaporation processes. In addition, the potential of spallation reaction for transmutation of ^{107}Pd is discussed.

1 Introduction

The nuclear power is considered to be a potential candidate for the environmental sustainability in the economic activity. The nuclear safety and security are, however, matters of concern for the use of a nuclear power plant. In particular, the management on the high-level radioactive waste (HLW) in the spent fuel produced from the nuclear power plant has received much attention.

Two main components exist in HLW. One is long-lived fission products (LLFPs), and the other is minor actinides (MA). To reduce their high radioactivities and to make resource recycling from the spent fuel, research and development have been devoted to the partitioning and transmutation technology in recent years [1]. The MA transmutation has been extensively studied in a concept of using the accelerator-driven system for recycling. In contrast, the study for the LLFP transmutation has been slowed.

However, the transmutation on LLFP has attracted much attention in recent years. ^{137}Cs and ^{90}Sr are known to be heat generator in nuclear reactor and they carry large radioactivities due to their relative short half-lives of 30 years [2]. The palladium metal belongs to platinum group, which is useful materials in HLW for economic use. However, the Pd metal contains a typical LLFP nucleus, ^{107}Pd, which has a half-life of 6.5×10^6 years [2]. The reduction of the radioactivity of ^{107}Pd is helpful for the use of the palladium metal. It is essential to find effective reactions for the LLFP transmutation. However, the reaction data for LLFP nuclei are currently very scarce.

In order to obtain fundamental data for LLFP transmutation, systematic studies on spallation reactions on both proton and deuteron have been performed at RIKEN for various LLFP nuclei, such as ^{107}Pd [3], ^{137}Cs [4], ^{90}Sr [4] and ^{93}Zr [5]. In these studies, the inverse kinematics technique was applied, namely the LLFP beams were used and proton/deuteron-induced reactions were conducted by using proton and deuteron targets. The technique enables a direct measurement on the reaction products by using advanced RI facilities, and a systematic study on the target

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dependence of reactions. The high-quality data obtained in the present work are critical to verify the spallation models as well as for a possible design on the LLFP transmutation system.

2 Experiment

The experiments were performed at RIKEN Radioactive Isotope Beam Factory, operated by RIKEN Nishina Center and the Center for Nuclear Study, University of Tokyo. The secondary beams including LLFP nuclei were produced by fission of U beam at 345 MeV/nucleon by impinging on a 1-mm-thick beryllium target in the first stage of the BigRIPS in-flight separator [6]. The second stage of BigRIPS was used to identify the particles in the secondary beams by measuring the time of flight (TOF), the magnetic rigidity ($B\rho$), and the energy loss (ΔE) on an event-by-event basis [7, 8]. The atomic number Z and mass-to-charge ratio A/Q were deduced from the ΔE-TOF and TOF-$B\rho$ correlations, respectively. The detailed information on BigRIPS settings can be found in Refs. [3–5].

CH_2, CD_2 [9] and carbon targets are used to conduct the secondary reactions. The targets were located at the entrance of the ZeroDegree spectrometer [6]. The data were accumulated also by using the target frame without target material (empty-target) to obtain the background contributions from the beam-line materials.

The data were taken at different reaction energies. The beam energies were both 185 MeV/nucleon for ^{137}Cs [4] and ^{90}Sr [4] in front of the secondary targets. For ^{107}Pd, the data were taken at both 196 and 118 MeV/nucleon [3]. For ^{93}Zr, the data were taken at around 105 MeV/nucleon [5]. The present work focuses on the results for ^{107}Pd at 118 MeV/nucleon.

The reaction residues were analyzed by the ZeroDegree spectrometer [6] with a momentum acceptance of ±3%. Five $B\rho$ settings, -9%, -6%, -3%, 0%, and +3% relative to the $B\rho$ value of the beam, were applied to cover a wide range of reaction products. The reaction products were identified using again the TOF-$B\rho$-ΔE method in a similar manner to BigRIPS. Examples for particle identifications for both secondary beams in BigRIPS and reaction residues in the ZeroDegree spectrometer are shown in Fig. 1 for the ^{107}Pd beam at 118 MeV/nucleon. Fig. 1(b) displays the reaction products produced from ^{107}Pd on the CD_2 target. Both LLFP beams and reaction products are clearly identified.

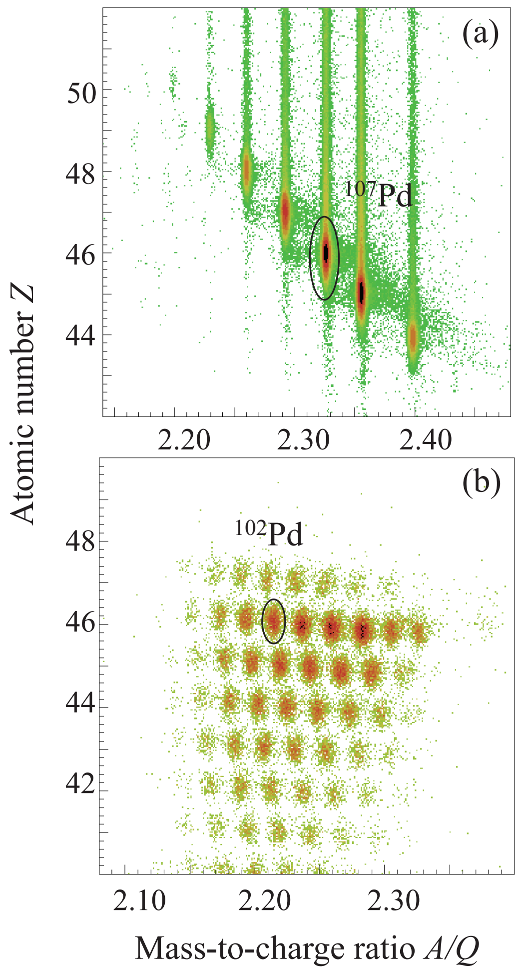

Figure 1. Particle identification plots for (a) the secondary beams in the BigRIPS separator and (b) the reaction products produced from ^{107}Pd and detected by the ZeroDegree spectrometer. Circles indicate the ^{107}Pd beam in (a) and the ^{102}Pd products in (b) to guide the eyes.

3 Results and discussion

Fig. 2 displays the isotopic distribution cross sections on proton and deuteron for ^{107}Pd at 118 MeV/nucleon. The proton- and deuteron-induced cross sections (σ_p and σ_d) were deduced by the respective CH_2 and CD_2 data after the subtraction of the carbon contributions and the contributions from the beam-line materials using the empty-target run.

Fig. 2(a) presents the Ag isotopes, which are produced from the ΔZ=1 reaction. σ_p is found to be larger than σ_d in this channel. Along the Pd and Rh isotopic chains as shown in Fig. 2(b) and (c), both σ_p and σ_d have a small deviation with a wide range and slightly decrease towards the neutron-deficient sides. The Pd and Rh isotopes are produced from the "peripheral" reaction, where the production is dominated mainly by the neutron evaporation [10], resulting in a rather constant distribution . A difference between σ_p and σ_d can be found for Ru, Tc and Mo as shown in Figs. 2(d), (e) and (f). In particular, the difference becomes more significant towards the neutron-deficient side for the light-mass products. These light-mass products are probably produced by the central collision, where the production depends on the energy deposited. More energy could be deposited in the deuteron-induced reaction than that for proton at the same beam energy, because deuteron has two nucleons. More energy deposited will lead to an evaporation of more nucleons in the deuteron-induced reaction as compared with the proton-induced one. Therefore, σ_d becomes much larger than σ_p for the light-mass products.

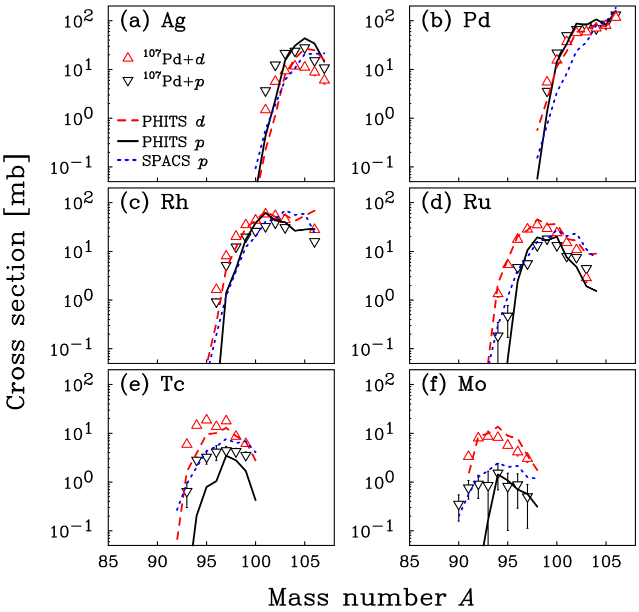

Figure 2. Isotopic distribution cross sections on proton (down triangle) and deuteron (up triangle) produced by ^{107}Pd at 118 MeV/nucleon for the elements (a) Ag, (b) Pd, (c) Rh, (d) Ru, (e) Tc, and (f) Mo. The solid and dashed lines represent the PHITS calculations on proton and deuteron, respectively. The dotted lines display the SPACS calculations on proton.

To have a quantitative understanding on the isotopic distribution cross sections, the data are compared with two spallation models, SPACS and PHITS calculations. SPACS is semi-empirical parameterization [11], which is developed to describe proton- and neutron-induced spallation reactions. The data on proton are reasonably reproduced by SPACS. In general, SPACS overestimated the cross section at the neutron-rich side of the isotopic distribution. Underestimated cross sections were found at the neutron-deficient sides for Ag, Pd and Rh as presented in Figs. 2(a),(b) and (c).

Theoretically, the spallation reaction could be described by a two-step process, namely the nuclear cascade via the nucleon-nucleon collisions and the de-excitation of pre-fragment. A theoretical calculation including these two processes was performed by using the particle and heavy ion transport code system (PHITS) [12]. In the PHITS calculation, the cascade process was simulated by the Intra-nuclear Cascade model of Liège (INCL4.6) [13] and the evaporation processes were made by using the generalized evaporation model (GEM) [14]. The PHITS results present a good agreement with the data as shown in Figs. 2. Underestimation on both the proton and deuteron results at the neutron-deficient side was found for the light-mass ions, such as Ru, Tc and Mo as presented in Figs. 2(d),(e) and (f).

The data are useful to discuss the potential of spallation reaction on the LLFP transmutation. The total spallation reaction cross sections on proton and deuteron were obtained to be 906(9) mb and 1085(8) mb at 118 MeV/nucleon by integrating the isotopic distributions in Figs. 2. These values are about 10% of neutron-capture cross section of 9.2 b [15]. Our data provide a design goal of proton/deuteron flux for the transmutation of ^{107}Pd via spallation reaction. In order to have a similar transmutation effect as neutron-capture reaction, the proton/deuteron flux should be one order magnitude higher than the neutron one.

To evaluate the reduction of the radioactivity after the spallation reactions, the number of long-lived radioactive isotopes created in the reaction is important. After the spallation reaction, most of the products are stable and short-lived isotopes. The main contributions to the radioactivity come from ^{99}Tc, which has a half life of 2.1×10^5 years. Its production cross sections are 3.5 mb and 6.1 mb for proton and deuteron, respectively. As the total spallation cross sections are around 1 b, the number of other LLFP elements is considerably reduced after the spallation reaction.

4 Summary

In summary, proton- and deuteron-induced spallation cross sections were successfully obtained for LLFP nucleus ^{107}Pd at 118 MeV/nucleon. The cross sections on deuteron were found to be much larger than those on proton for the light-mass nuclei because of a large energy deposited in the deuteron-induced reactions. The data are generally reproduced by the SPACS and PHITS calculations. Further development on the spallation models are desired for a further understanding on the spallation reaction mechanism.

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