Isomer production ratio of the ^{112}Cd$(n,\gamma)^{113}$Cd reaction in an *s*-process branching point

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A β-decay unstable isomer with a half-life of 14.1 yr at 264 keV in ^{113}Cd is a branching point in the *s* process, from which a weak branch reaches to a rare tin isotope ^{115}Sn whose astrophysical origin has been an open problem. We have measured γ rays decaying to the ground state or the isomer in the ^{112}Cd$(n,\gamma)^{113}$Cd reaction using high-energy resolution detectors in conjunction with a time-of-flight method. The relative production ratios of the isomer to the total following the neutron capture reactions on ^{112}Cd have been evaluated in an energy region of up to 9 keV, and the spin and parity of several resonances have been assigned.

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I. INTRODUCTION

The astrophysical origin of a rare tin isotope, ^{115}Sn, has remained an open question. Although most nuclides heavier than iron were synthesized by two stellar neutron capture reactions of *s* and *r* processes, the solar abundance of ^{115}Sn cannot be explained by these processes [1–7]. One of the other candidates is photodisintegration reactions in supernova explosions (γ process) [8,9] but the solar abundance of ^{115}Sn is underproduced by the γ-process theoretical calculations. Although proton capture reactions in this mass region has been studied [10], a nucleosynthesis path to ^{115}Sn has not been studied. An isomer with a half-life of 14.1(5) yr at 264 keV in ^{113}Cd is a branching point in the *s* process [1]. The main flow in the *s* process goes on a path of ^{112}Cd$(n,\gamma)^{113}$Cd$^{gd}(n,\gamma)^{114}$Cd. The ^{113}Cd isomer decays to a daughter nucleus ^{113}In and ^{115}Sn is synthesized through a nucleosynthesis flow of ^{113}In$(n,\gamma)^{114}$In$(e,\nu)^{114}$Sn$(n,\gamma)^{115}$Sn [1,6,7]. The cross section of the ^{112}Cd$(n,\gamma)^{113}$Cd^{m} reaction at the thermal energy were measured using nuclear reactors [6,11]. Resonance parameters and cascade γ rays of Cd isotopes were measured with pulse neutrons [12,13]. Hayakawa *et al.* [6] pointed out that the *s*-process abundance of ^{115}Sn depends on the ratio of the ^{112}Cd$(n,\gamma)^{113}$Cd^{m} reaction cross section to the ^{112}Cd$(n,\gamma)^{113}$Cd reaction cross section in the *s* process by calculating a classical steady flow model. However, the ratios at astrophysical energies have not been well measured.

The ratio at thermal energy was measured using neutrons at the JRR-3 nuclear reactor [6]. To explore the ratio in an energy region of keV, we measured γ rays from neutron capture reactions on an enriched ^{112}Cd target with intense neutron pulses provided by the Accurate Neutron-Nucleus Reaction measurement Instrument (ANNRI) in the Materials and Life Science Experimental Facility (MLF) at the Japan Proton Accelerator Research Complex (J-PARC) [7]. The isomer production ratios were evaluated from the intensity ratios of γ rays populating the isomer to γ rays decaying to the total of the ground state and the isomer. The measured ratios were consistent with results calculated using a Hauser-Feshbach statistical model. It was found that the isomer ratio is sensitive to the spin and parity of a compound state formed by a neutron capture. In the previous experiment [7], we measured the ratios of resonances with energies of up to 5 keV. The detection efficiency of resonances in an energy region of keV is constrained by the double-pulse mode for neutron beams, in which two short pulses with a width of 100 ns and an interval time of 600 ns is included in a macropulse provided from an accelerator so that there are two energy components at the same time. The previous study [7] in the energy region of up to 5 keV shows that the *s*-process contribution from the ^{113}Cd isomer to the ^{115}Sn solar abundance is small. However, the statistical model calculation suggested a possibility that the isomer production ratio drastically increases in the high energy region [7]. Thus, it is expected to be measured in an energy region of up to 30 keV, the energy region relevant to the *s* process in the core He burning of massive stars and shell He burning of low mass stars.

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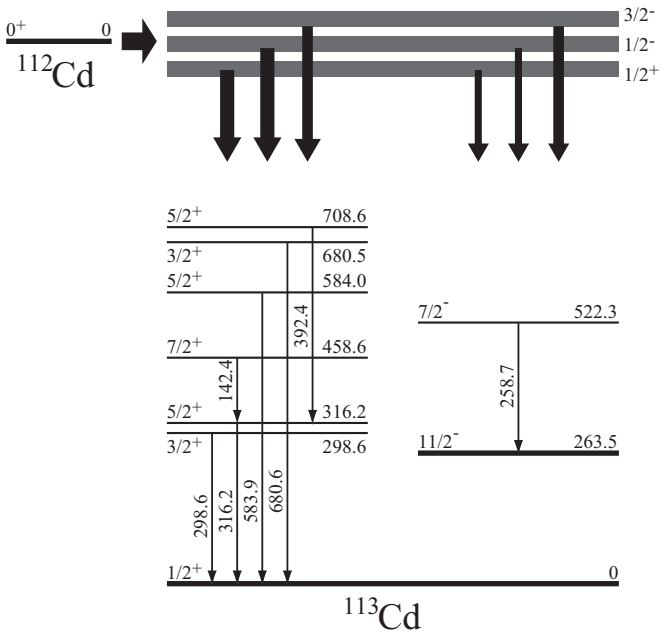

FIG. 1. Schematic view of neutron capture reactions on ^{112}Cd and a partial level scheme of ^{113}Cd. Excited states on ^{113}Cd are populated by neutron capture reactions on ^{112}Cd and they decay to the ground state or the isomer at 264 keV. The isomer production ratio depends on the spin and parity of an excited state. When an *s*-wave capture on ^{112}Cd occurs, excited states with $J^\pi = 1/2^+$ states are populated and subsequently strongly decay to the ground state, but the branching ratio to the isomer is low. In the case of *p*-wave capture, the $J^\pi = 1/2^-$ or $3/2^-$ state is populated. The isomer branching ratio increases with increasing the spin of the initial exited state.

There are experimental techniques to measure precisely the spin and parity of a resonance state, for example, measurements of angular distribution of neutron capture γ rays [14] and a method using polarized neutron beam [15]. The previous study [7] shows that the isomer production method could constrain the possible spin and parity of a resonance. In the present paper, we report the isomer production ratios evaluated through measuring γ-ray intensity ratios following neutron capture reactions using ANNRI. We have used neutron pulses with a single-bunch mode, in which there is only one pulse per each macro pulse. We have measured γ rays decaying to the ground state or the isomer following the ^{112}Cd$(n,\gamma)^{113}$Cd reaction using high-purity germanium (HPGe) detectors in conjunction with a time-of-flight (TOF) method. The relative branching ratios have been measured in the energy region of up to 9 keV.

II. EXPERIMENTAL PROCEDURE

The total energy measurement method is one of the powerful methods to evaluate isomer production ratios [16]. The maximum energy of γ rays corresponding to the neutron separation energy of ^{113}Cd is approximately 6.54 MeV. In contrast, the energy difference between the ground state and the isomer in ^{113}Cd is only 264 keV (see Fig. 1). Under such condition, γ-ray spectroscopy using γ-ray detectors with high energy resolution is effective. The present experiment was performed using ANNRI installed in the BL04 neutron beam line of the MLF at J-PARC. The details of the ANNRI were described in the previous papers [17,18]. High flux pulsed neutrons were generated by spallation reactions on a mercury target with high energy proton beams. The proton beams with an energy of 3 GeV and an average power of 140 kW were injected into the mercury target at a repetition rate of 25 Hz. The proton beam was operated with a single bunch mode with a width of 100 ns. The ANNRI has a high purity germanium (HPGe) detector array, which consists of two cluster HPGe detectors, eight coaxial HPGe detectors, and bismuth germanate (BGO) Compton-suppression detectors. The total detection efficiency is $3.64 \pm 11\%$ at 1.33 MeV [17]. Individual cluster detectors have seven HPGe crystals and a typical energy resolution of a crystal is approximately 3.5 keV at 1.33 MeV. A ^{112}Cd metal foil enriched to 98.27% was set at the center of the detector array. A neutron beam collimator with a diameter of 6 mm was used and thereby the beam diameter on the target is approximately 6 mm. This detector system is located 21.5 m downstream from the neutron source. The energies of neutrons were measured using a time-of-flight (TOF) method. When at least one crystal detected a γ ray, that time and the energies of γ rays measured by individual crystals were recorded event by event in a list mode. In the present study, we used the two cluster HPGe detectors with BGO Compton-suppression detectors for data analysis.

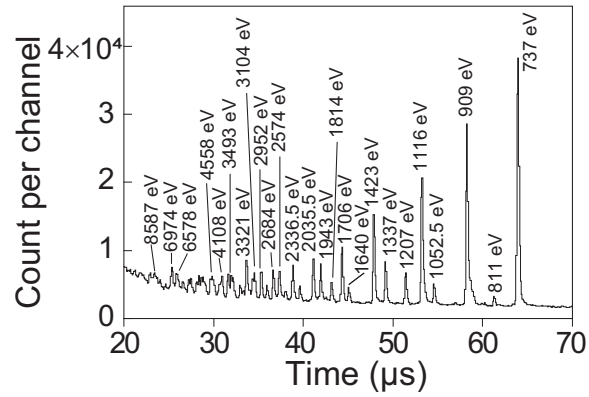

FIG. 2. Typical TOF spectrum measured in the present experiment using a single-bunch mode, in which only a single pulse is included in a single macropulse. The time is recorded when at least one HPGe detector measured a γ ray event by event. The energies indicate the relevant resonance energies on the ^{112}Cd$(n,\gamma)^{113}$Cd reaction.

III. RESULT AND DISCUSSION

In the present experiment, the ^{112}Cd enriched target was used but it included ^{113}Cd with a fraction of 0.6%. Because of its extremely large thermal neutron capture reaction cross section of 20 650 barn, the contribution of the ^{113}Cd$(n,\gamma)^{114}$Cd reaction is observed at the thermal energy. In contrast, resonances in ^{112}Cd dominate in an energy region higher than the thermal energy. Figure 2 shows a typical TOF spectrum that presents clearly neutron capture resonances in ^{112}Cd. Because

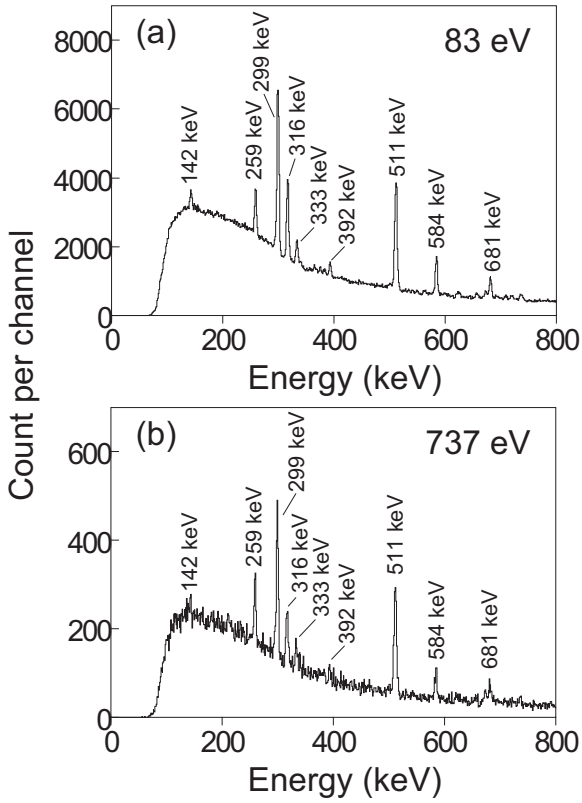

FIG. 3. Energy spectra gated on 83 eV (a) and 737 eV resonances (b). γ rays radiated from neutron capture reactions on ^{112}Cd are clearly observed.

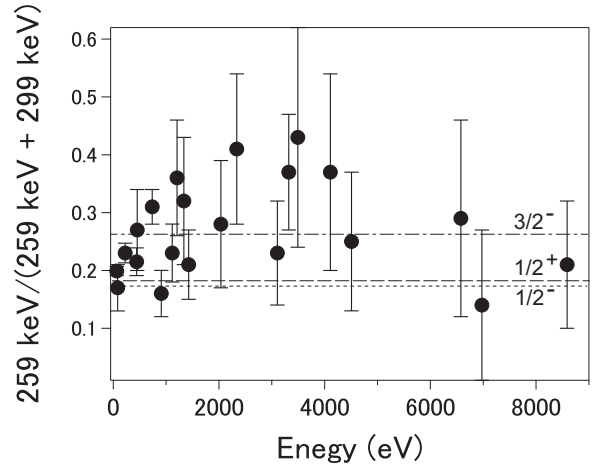

FIG. 4. γ-ray intensity ratios of 259 keV decaying to the isomer to the sum of 259 and 298 keV decaying to the ground state. These values are expected to be approximately proportional to the isomer production ratios. The dashed line represents the expected ratio of resonances with a spin and a parity of $J^\pi = 1/2^+$ calculated using a statistical model. The dotted line and dot-dashed lines represent those of $J^\pi = 1/2^-$ and $J^\pi = 3/2^-$ resonances, respectively.

in the previous experiment using the double-bunch mode [7] two peaks for a single resonance were observed in high energy region, it is difficult to assign exactly resonances in TOF spectra. In addition, pile-up of one of the two peaks of a resonance and one of the pair of another resonance may occurs; in such case a γ-ray spectrum with a gate on a neutron energy is mixed of two γ-ray spectra with different neutron energies. In contrast, in the single-bunch mode only one peak for each resonance is observed. Note that the use of the single-bunch mode causes a disadvantage that the total neutron flux in the single-bunch mode becomes approximately half of the flux in the double-bunch mode because the number of protons in a macro pulse provided from the proton accelerator in the single-bunch mode is approximately half. The energies of the measured resonances are consistent with the previous data [19]. In the present experiment using the single bunch mode, the time resolution becomes better but the neutron flux decreases. We chose a setup to increase the neutron flux but the time resolution becomes low. Thus, the uncertainties of the measured resonance energies are larger than those of the previous data. We use the previous resonance energies [19] to indicate individual resonances in the following discussion.

Figure 3 shows typical γ-ray energy spectra with a gate on 83 or 737 eV neutron energy in the TOF. Most γ rays in these spectra originate from excited states in ^{113}Cd. Although the 333-keV γ ray is observed in some spectra with gates on neutron resonances in ^{112}Cd we cannot assign its origin. A γ ray with an energy of 259 keV that decays to the ^{113}Cd isomer is observed and γ rays decaying to the ground state with energies of 299 and 316 keV are also observed in both spectra. Figure 1 shows a partial level scheme of ^{113}Cd and a schematic view of neutron capture reactions on ^{112}Cd. The γ rays observed in the present experiment are indicated by the arrows in the partial level scheme of ^{113}Cd.

In an energy region from thermal energy to keV, resonance states in ^{113}Cd are populated through neutron capture reactions on the ground state of ^{112}Cd with a spin and parity of $J^\pi = 0^+$. The populated resonance states subsequently decay to the ground state of ^{113}Cd with $J^\pi = 1/2^+$ or the isomer with $J^\pi = 11/2^-$ (see Fig. 1). When s-wave capture reactions with a neutron orbital angular momentum of $l = 0$ on the ground state of an even-even nucleus occur, these reactions, in principle, populate resonances with $J^\pi = 1/2^+$ because of the conservation law of angular momentum. The populated states with $J^\pi = 1/2^+$ strongly decay to the ground state with $J^\pi = 1/2^+$ by the emission of a γ ray or cascade γ rays in the present case. The decay fraction to the isomer with $J^\pi = 11/2^-$ is lower than that to the ground state by several factors or one of magnitude because of the large angular momentum difference of $\Delta J = 5$. In the case of p-wave capture of $l = 1$, resonance states of $J^\pi = 3/2^-$ or $1/2^-$ are populated. When $J^\pi = 3/2^-$ states are populated, the isomer production ratio should be higher than those for $J^\pi = 1/2^+$ and $1/2^-$. In general, the fraction to the isomer increases with increasing the spin of a resonance state.

To evaluate the isomer production ratios as a function of neutron energy, we evaluate the γ ray intensity ratios of 259 keV to the sum of 259 and 299 keV, $R = 259\ \text{keV}/(259 + 299\ \text{keV})$, obtained from individual γ-ray spectra with a gate on a neutron resonance energy in Fig. 4. These ratios are expected to be approximately proportional to the isomer production ratios. To compare the measured ratios with theoretical values we calculated the expected

ratios and neutron capture cross section on ^{112}Cd using the CCONE Hauser-Feshbach statistical model [20]. The neutron capture cross sections were normalized to fit to measured data in the keV energy region [5]. The neutron transmission coefficient was derived by a coupled-channels optical model with the potential form [21]. The potential parameters were fixed so as to reproduce measured data of the total cross section and elastic scattering angular distributions for natural Cd. The ground state of ^{112}Cd was coupled with the $J^\pi = 2^+$, 4^+, and 6^+ levels in the ground-state band with quadrupole and hexadecapole deformation parameters of 0.151 and -0.028, respectively. The levels with excitation energies of up to 1192 keV in ^{113}Cd were adopted from the RIPL-3 database [22]. The excited states above 1192 keV were represented as continuous states by the composite level density formula [23]. The decay γ-rays of $E1$, $M1$, and $E2$ transitions were taken into account. The generalized Lorentzian model is adopted for the $E1$ strength function. The calculated ratio in the case of decaying from $J^\pi = 1/2^+$ states is approximately $R = 0.18$ and the ratio from $J^\pi = 1/2^-$ states is slightly lower than that from $J^\pi = 1/2^+$ states (see Fig. 4). In contrast, the ratio from $3/2^-$ states is approximately $R = 0.27$, which is higher than those from $J^\pi = 1/2^\pm$ states by a factor of approximately 1.5. Note that the branching ratios depend on individual compound states and the measured ratios should be scattered from these calculated ratios.

In the previous study by Liou *et al.* [24], the orbital angular momenta of incident neutrons were assigned by using Bayes's theorem analysis [25]. We summarize the isomer ratio, the spin, and parity of the presently obtained 21 resonances and their previous data in Table I. Most of the presently measured ratios consistent with the previous spin assignments. One of the strongest resonances in the low energy region is the 737-eV resonance that was assigned as the state with $J^\pi = 1/2^+$. However, the isomer ratios obtained in the previous [7] and present experiments indicate that it is the resonance of $J^\pi = 3/2^-$. Furthermore, although the 2336.5-eV resonance was assigned to be $J^\pi = 1/2^+$ it has been assigned to $J^\pi = 3/2^-$ in the present study. The 83-eV resonance was assigned to a p-wave capture [24] but its spin and parity were not evaluated. The present result indicates that this resonance probably is the resonance with $J^\pi = 1/2^+$ or $J^\pi = 1/2^-$. Combining this spin assignment with this previous data [24], the 83-eV resonance can be assigned to be a p-wave resonance with $J^\pi = 1/2^-$.

The ^{113}Cd isomer is a branching point in the s process and the ^{115}Sn abundance depends on the isomer production ratio. The previously measured ratios in an energy region from thermal energy to 5 keV suggest that the s-process contribution to ^{115}Sn is much lower than its solar abundance. The ratios measured in the present study also support the previous result. However, in the present study we have changed the spin assignments of the two resonances from $J^\pi = 1/2^+$ to $J^\pi = 3/2^-$ with the relatively high isomer production ratios. This fact suggests that when the isomer production ratios of other resonances, in particular in an energy region of up to 30 keV, are precisely measured, the s-process contribution may increases and their spin and parity may be changed. Thus, the s-process origin of ^{115}Sn is still an open problem.

TABLE I. Experimental results. Resonance energy, spin, and orbital angular momentum of incident neutrons l are taken from Ref. [19]. R means the isomer production ratio measured in the present study. The spin and parity J^π are newly assigned from the measured isomer production ratios.

Resonance energy (eV)	Spin	l	Present R	Present J^π
66.8	1/2	0	0.199(8)	$1/2^+$
83.2		(1)	0.17(4)	$1/2^-$
226		0	0.230(17)	$1/2^+$
443		0	0.214(24)	$1/2^+$
453		(1)	0.27(7)	
737		0	0.31(3)	$3/2^-$
909	1/2	0	0.16(4)	$1/2^+$
1115	1/2	0	0.23(5)	$1/2^+$
1207		(1)	0.36(10)	$3/2^-$
1337		(1)	0.32(11)	$3/2^-$
1423	1/2	0	0.21(6)	$1/2^+$
2036	1/2	0	0.28(11)	
2336.5	1/2	0	0.41(13)	$3/2^-$
3104	1/2	(0)	0.23(9)	
3321	3/2	1	0.37(10)	$3/2^-$
3493		(1)	0.43(19)	$(3/2^-)$
4108	3/2	1	0.37(17)	$(3/2^-)$
4558	1/2	0	0.25(12)	
6578	1/2	0	0.29(17)	
6975	1/2	0	0.14(13)	
8587	1/2	0	0.21(11)	

IV. SUMMARY

The present study shows measurement of isomer production ratios with high-energy resolution detectors in conjunction with a TOF method is effective for the spin and parity assignments of neutron capture resonances. The isomer ratios are sensitive to, in particular, the spin of resonance states. We have measured the ratios of 21 resonances and assigned the spin and parity of 14 resonances in the ^{112}Cd$(n,\gamma)^{113}$Cd reaction. Among the 14 resonances, we have assigned $J^\pi = 1/2^-$ with p-wave capture for two resonances that were assigned to s-wave resonances in the previous study. We have assigned a state with $J^\pi = 1/2^-$ by combining the previous assignment of p-wave capture and the presently measured ratio. The result shows that the isomer production ratio does not increase drastically in the energy region of $E < 9$ keV, supporting the previous results [6,7] that the s-process contribution from the ^{113}Cd isomer to the solar abundance of ^{115}Sn is relatively minor. However, because the ratios in the energy higher than 9 keV have not been measured, the s-process origin of ^{115}Sn is still an unresolved problem.

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