Misassigned neutron resonances of ^{142}Nd and stellar neutron capture cross sections

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Time-of-flight spectra of the neutron capture events of ^{142}Nd were measured using a spallation neutron source at the Japan Proton Accelerator Research Complex. The first six resonances of ^{142}Nd reported in a previous work were not observed. The experimental results and cross-search of resonance energies in nuclear data libraries suggested that resonances of the impurity nuclide ^{141}Pr have been mistakenly assigned as ^{142}Nd in the previous experiment. To investigate the impact of the nonexistence of the resonances on the *s*-process nucleosynthesis model, the Maxwellian averaged neutron capture cross sections with and without the misassigned resonances were compared.

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In the current nucleosynthesis model, ^{142}Nd is created in the slow neutron capture process (*s*-process). The neutron capture cross section of ^{142}Nd has been significantly important in the study of *s*-process nucleosynthesis. Arlandini *et al.* demonstrated that stellar *s*-process models, in which a low-mass asymptotic giant branch star was modeled, successfully reproduce the solar abundance of ^{142}Nd when improved experimental data of the capture cross section of ^{142}Nd are used for nucleosynthesis calculations [1].

The neutron capture reaction of ^{142}Nd in the energy region relevant to the *s*-process nucleosynthesis is dominated by resolved neutron resonances. The neutron resonance parameters of ^{142}Nd have been measured by different research groups [2–4] and the resonance parameters in evaluated nuclear data libraries such as JENDL-4.0 [5] and ENDF/B-VII.1 [6] are based on their experimental data. Wisshak *et al.* made resonance analysis measurements using a BaF_2 detector array at Karlsruhe and determined resonance capture areas in the energy region from 3.271 to 20.9 keV [4]. Combining Wisshak's results with previous experimental data, resolved resonance parameters were determined in nuclear data evaluations.

However, only a single experimental data set of resonance parameters exists below 2 keV. Resonance parameters in the low-energy region were determined in transmission experiments by Tellier in 1971 [2]. Resonance measurements below 2 keV have not been performed in other facilities since Tellier's experiments. The parameters of the low-energy resonances may affect the Maxwellian averaged cross sections (MACS) which are basic inputs for nucleosynthesis calculations. This motivated us to perform the present time-of-flight (TOF) experiments using a spallation neutron source at the Japan Proton Accelerator Research Complex (J-PARC) [7].

Experiments were carried out with the Accurate Neutron-Nucleus Reaction Measurement Instrument (ANNRI) [8,9] in the Materials and Life Science Experimental Facility of J-PARC. Neutron capture γ rays were detected with a NaI(Tl) detector located at an angle of 90° with respect to the neutron beam axis. The TOF of detected events were measured with a high resolution time digitizer. The details of the experimental instruments and method can be found elsewhere [10]. An isotopically enriched ^{142}Nd sample in the chemical form of Nd_2O_3 was used. The isotopic composition is summarized in Table I. The chemical purity was 99.44% and the net weight of ^{142}Nd was 1.490 g. Because Nd_2O_3 is hygroscopic, the sample was pressed to form a pellet and then heated at 1100 °C for 1 h to remove moisture absorbed in the sample. The diameter of the sample pellet was 10.4 mm and the thickness was 3.2 mm. The sample was placed at a flight length of 27.9 m from the J-PARC spallation neutron source. A 3 GeV pulsed proton beam was injected into a mercury target of the spallation source. The proton accelerator was run at a beam power of approximately 200 kW and at a repetition rate of 25 Hz. The operational mode of the accelerator was the so-called double-bunch mode, in which two 100 ns proton beam pulses separated by 600 ns delay are used for each neutron production cycle [11]. A signal from a beam pulse monitor before the spallation target was used as the start trigger for TOF measurements. The measurement time was 15 h.

Figure 1 shows the obtained TOF spectrum with a TOF time bin of 1 μs. The TOF positions of the first nine ^{142}Nd resonances in a resonance databook by Mughabghab [12] are indicated by the arrows. The resonance parameters of the nine resonances [12] are summarized in Table II. The first seven resonances are based on only Tellier's report [2]. Most of the observed resonances in the TOF spectrum were attributed to

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TABLE I. Isotopic composition (%) of the ^{142}Nd sample.

^{142}Nd	^{143}Nd	^{144}Nd	^{145}Nd	^{146}Nd	^{148}Nd	^{150}Nd
95.7	3.10	0.76	0.15	0.19	0.05	0.05

the impurity isotope ^{143}Nd. Figure 2 is a close-up of the TOF region between 40 and 100 μs. The TOF time bin is 100 ns. Resonances identified as ^{142}Nd and ^{143}Nd are marked with red full and blue open points, respectively. Each resonance has doublet peaks due to the J-PARC double-bunch proton beam operation. The resonance at 1687 eV was observed, but all six resonances below 1687 eV in Table II were not confirmed in the TOF spectra.

For quantitative discussion, expected capture yields under the present experimental conditions were calculated from resonance parameters in JENDL-4.0 that are based on Ref. [2] for the first six resonances. The isotopic composition of the sample in Table I was input in the calculations. The doublet beam pulse structure and neutron self-shielding effect in the sample were also taken into account. The $1/E$ incident neutron spectrum and statistically fluctuating continuum background mimicking the present experiments were assumed. Previous measurements show that the energy dependence of the incident neutron spectrum can be described approximately by $1/E$ in this energy region [11]. The estimated capture yield TOF spectrum with a TOF bin of 1 μs is shown in Fig. 3(a). Figure 3(b) shows the contribution of ^{142}Nd to the estimation. The vertical lines indicate the TOF positions of the first eight resonances. As seen in the estimated TOF spectrum, some of the resonances cannot be clearly identified because of impurity isotope resonances, the background, and the doublet peak structure; but at least the first two (218.6 and 235.0 eV) and possibly the third (636.4 eV) resonances should be observed. However, the first three resonances did not appear in the present experiments.

In further investigation cross-searching the resonance energies in evaluated nuclear data libraries, we found that the energies of the first six resonances in Tellier's report are very close to those of ^{141}Pr as listed in Table II. This improbable coincidence suggests that ^{141}Pr possibly existed as an impurity

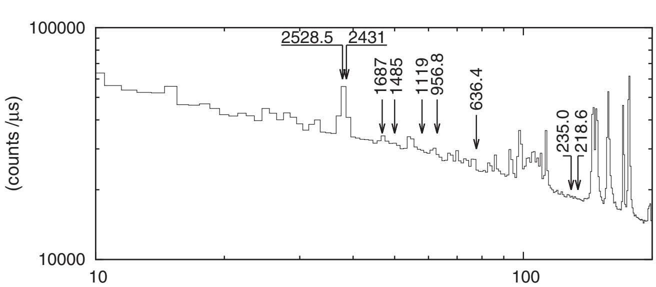

FIG. 1. TOF spectrum of the ^{142}Nd sample. The TOF time bin is 1 μs. The TOF position of the first nine resonances of ^{142}Nd in Ref. [12] are indicated by arrows.

TABLE II. Resonance parameters of the first nine resonances of ^{142}Nd, and resonances of ^{141}Pr close to the ^{142}Nd resonances in Ref. [12].

^{142}Nd					^{141}Pr
E (eV)	J	l	$g\Gamma_n$ (eV)	$g\Gamma_n\Gamma_\gamma/\Gamma$ (meV)	E (eV)
218.6 ± 0.2		(1)	0.00090 ± 0.00005		218.7 ± 0.3
235.0 ± 0.2		(1)	0.00080 ± 0.00005		235.2 ± 0.3
636.4 ± 0.6		(1)	0.00150 ± 0.00025		635.8 ± 0.5
956.8 ± 1.0		(1)	0.0018 ± 0.0004		956.8 ± 1.0
1119 ± 1		(1)	0.00145 ± 0.00030		1119.5 ± 1.0
1485 ± 2		(1)	0.00100 ± 0.00025		1484.0 ± 1.5
1687 ± 2	(3/2)	(1)	0.051 ± 0.010	29.4	
2431.0 ± 2.4	(3/2)	(1)	0.029	21	
2528.5 ± 2.5		0	9.7 ± 0.3	22	

in the sample of Tellier's experiments and the resonances of ^{141}Pr were misassigned as ^{142}Nd. They used isotopically enriched samples for seven Nd isotopes with two different thicknesses but chemical impurities in the samples were not described. We made an additional experiment on a ^{141}Pr sample. All six resonances were observed in the TOF spectrum as shown in Fig. 4.

To discuss more about Tellier's measurements, we calculated transmission TOF spectra using Tellier's experimental setup parameters. The flight path length (53.4 m), the sample isotopic composition (Table III) and thickness, and the beam pulse width (50 ns) were input into the calculations. Resonance parameters in JENDL-4.0 were used. The calculated TOF spectrum of the ^{142}Nd sample is shown in Fig. 5(b). Figure 5(a) is a transmission TOF of a very thin ^{141}Pr sample. The vertical dashed lines are the TOF position of the six resonances reported as ^{142}Nd by Tellier. The six TOF positions match those of ^{141}Pr resonances. Resonances of impurity isotopes are dominant in the TOF spectrum of the

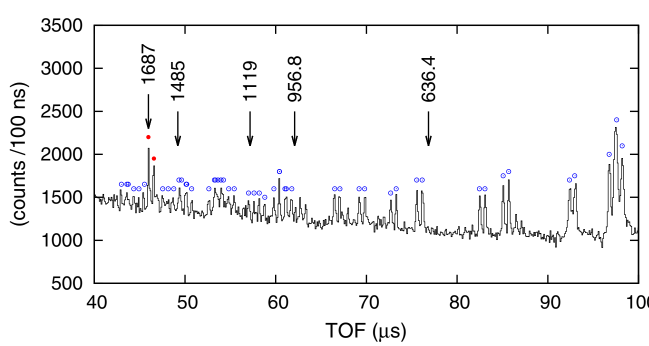

FIG. 2. (Color online) Close-up of the TOF spectrum of the ^{142}Nd sample. The TOF time bin is 100 ns. The TOF position of resonances of ^{142}Nd in Ref. [12] are indicated by arrows. Resonances identified as ^{142}Nd and impurity ^{143}Nd are marked with the red full and blue open points, respectively. Each resonance has doublet peaks due to the double-bunched pulse beam.

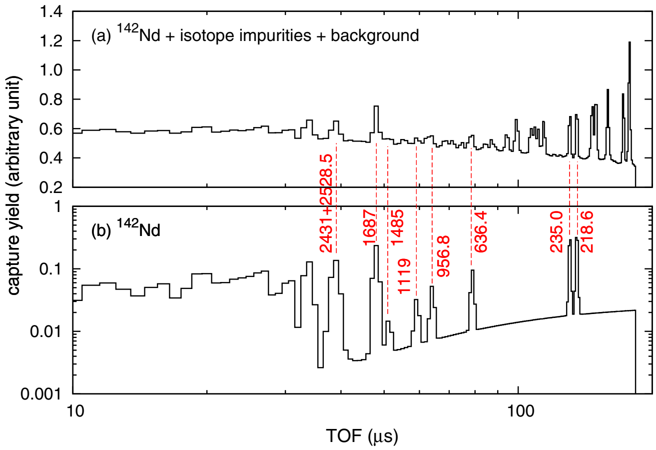

FIG. 3. (Color online) Estimated capture yield TOF spectra under the present experimental conditions. Resonance parameters of JENDL-4.0 were used in the estimation. The TOF bin width is 1 μs. The $1/E$ incident neutron spectrum was assumed. (a) Nd isotope impurities in the sample (Table I) and statistically fluctuating background are included in the calculation. (b) Contribution of ^{142}Nd to the calculation is shown and the red vertical lines indicate the TOF positions of the ^{142}Nd resonances in the low-energy region.

^{142}Nd sample. The comparison of the two TOF spectra reveals that the six resonances of ^{141}Pr reside in TOF regions where the impurity resonances do not exist. Other ^{141}Pr resonances are covered by Nd isotope impurity resonances. Only the six resonances of ^{141}Pr can be observed in the transmission spectrum when ^{141}Pr is included as an impurity in the sample.

Next, we removed the six suspicious resonances of ^{142}Nd from the input resonance data and, instead, added ^{141}Pr impurity to the transmission calculation. After trial calculations changing the amount of ^{141}Pr, we found that 0.35% of ^{141}Pr can consistently reproduce the peak intensities of the six removed ^{142}Nd resonances. Figure 6 shows a comparison of the modified calculation (red dashed) with the original spectrum (black solid). The dip depths of the original and ^{141}Pr-included calculations are in good agreement except for the resonance of 1484 eV. The disagreement at 1484 eV may be caused from

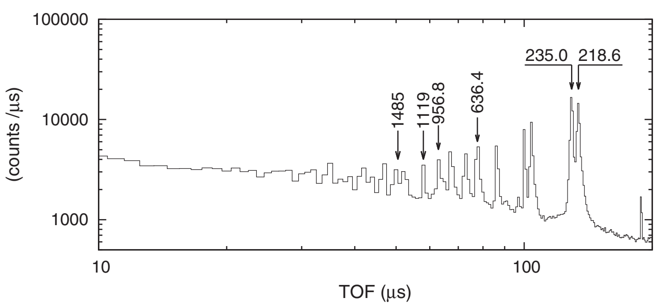

FIG. 4. TOF spectrum of the ^{141}Pr sample. The TOF time bin is 1 μs. The TOF position of the first six resonances of ^{142}Nd in Ref. [12] are indicated by arrows.

TABLE III. Isotopic composition (%) of the ^{142}Nd sample used in the measurements of Ref. [2].

^{142}Nd	^{143}Nd	^{144}Nd	^{145}Nd	^{146}Nd	^{148}Nd	^{150}Nd
95.0	2.7	1.5	0.3	0.3	0.1	0.1

unknown experimental parameters such as the time resolution function and background, which were not described in Ref. [2].

An amount of ^{141}Pr impurity in the ^{142}Nd sample used in the present experiments was not absolutely determined by the manufacturer. The sample certification sheet provided only an upper limit of 0.3% of Pr. We estimated capture yield TOF spectra including ^{141}Pr impurity in the same way as done for Fig. 3(a). Three calculations changing the ^{141}Pr impurity level from 0 to 0.3% are compared in Fig. 7. Even if 0.3% of ^{141}Pr exists in the sample, only the two resonances (218.7 and 235.2 eV) appear in the TOF spectrum. In the present experiments, the two resonances were not observed. This indicates that the amount of ^{141}Pr impurity was much less than 0.3%.

Finally, to investigate the impact of the present findings to the s-process nucleosynthesis model, the MACS at stellar temperatures were calculated with and without the six resonances. Figure 8 shows MACS calculated using evaluated resonance data in JENDL-4.0 [13]. The open circles and the triangles are calculations with and without the first six resonances, respectively. Table IV shows numerical data of the calculations. The difference between the two calculations is roughly 10% at $kT = 1$ keV but becomes rapidly smaller as kT increases. The effect of the six resonances to MACS is negligible ($<1\%$) at $kT = 8$ keV, a temperature at which the $^{13}C(\alpha,n)^{16}O$ reaction occurs for the s-process nucleosynthesis.

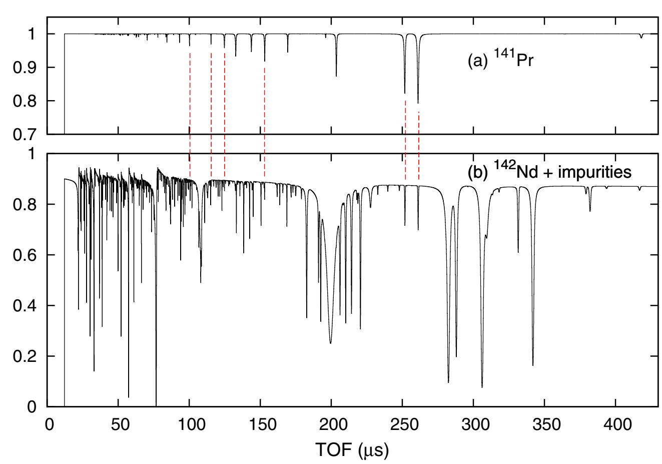

FIG. 5. (Color online) Estimated transmission TOF spectra under the experimental condition of Ref. [2] for (a) a ^{141}Pr and (b) a ^{142}Nd sample with an isotope composition in Ref. [2]. The resonance parameters in JENDL-4.0 were used in the calculation. The ^{142}Nd sample has Nd isotope impurities. The vertical lines indicate the TOF positions of the first six resonances of ^{142}Nd reported in Ref. [2].

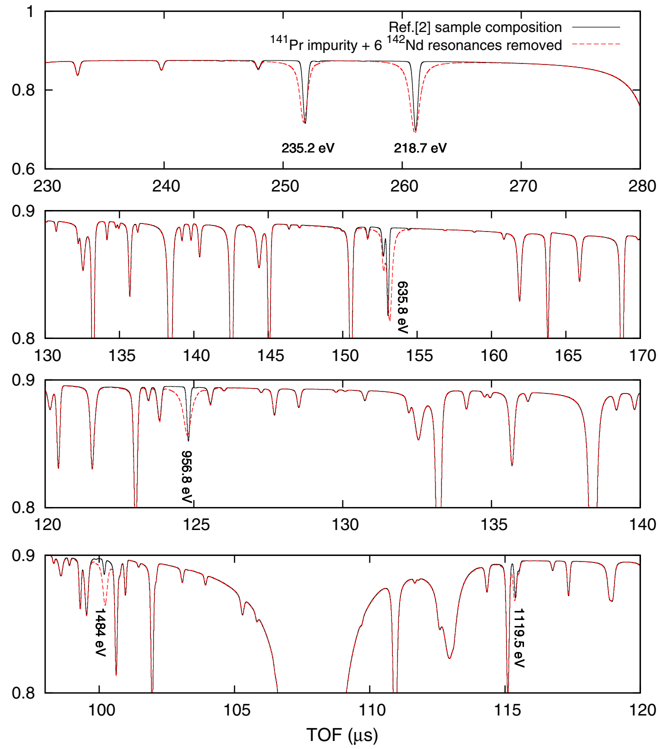

FIG. 6. (Color online) Close-up of the estimated transmission TOF spectra for the same calculation inputs as Fig. 5(b) (black solid line) and for assuming that 0.35% ^{141}Pr impurity exists in the sample and the first six ^{142}Nd resonances reported in Ref. [2] do not exist (red dashed line).

In conclusion, the results of the present TOF experiments suggest that the first six neutron resonances of ^{142}Nd reported in a 1970s experiment are a misassignment of the impurity of ^{141}Pr. These six resonances have been adopted in nuclear data libraries but have to be removed. In MACS calculations, the removal of the six resonances from JENDL-4.0 does not have a large effect on MACS at the s-process important temperature ($kT = 8$ keV). On the other hand, the effect of the removed resonances becomes larger at lower energies and reaches 10% difference at $kT = 1$ keV.

TABLE IV. Calculated Maxwellian-averaged neutron capture cross section of ^{142}Nd. Evaluated resonance data of JENDL-4.0 were used. Calculations with the original JENDL-4.0 parameters and without the first six resonances are compared.

	Maxwellian averaged cross section (mb)	
kT (keV)	JENDL-4.0 original	W/o first 6 res.
1	129	114
2	134	129
3	125	122
5	102	100
8	78.4	77.9
10	68.4	68.1
15	53.2	53.1
20	44.7	44.6
25	39.3	39.2
30	35.6	35.5
35	32.8	32.8
40	30.7	30.7
50	27.8	27.8

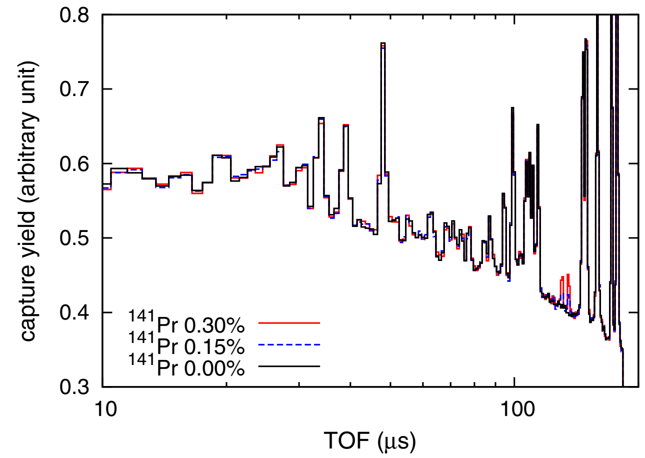

FIG. 7. (Color online) Estimated capture yield TOF spectra under the present experimental condition assuming that the first six ^{142}Nd resonances in Ref. [2] do not exist and ^{141}Pr impurity exists in the sample. The TOF spectra were calculated for ^{141}Pr impurities of 0.3% (red solid), 0.15% (blue dashed), and 0% (black solid). Except for the removed ^{142}Nd resonances and the additional ^{141}Pr impurity, the calculation parameters are the same as those for Fig. 3(a).

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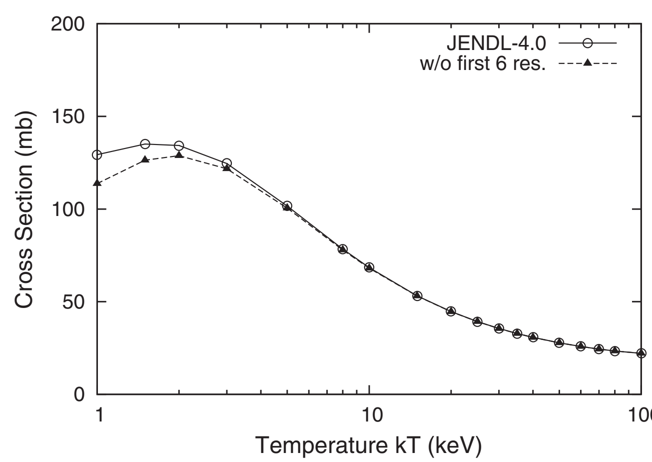

FIG. 8. Calculations of the Maxwellian averaged neutron capture cross section of ^{142}Nd. Evaluated data in JENDL-4.0 were used for the calculations. Calculations with the original JENDL-4.0 (open circle) and without the first six resonances (triangle) are compared.

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