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Measurement of the thermal neutron capture cross section of ^{180}W

W. G. Kang, Y. D. Kim,[*] and J. I. Lee
Department of Physics, Sejong University, Seoul 143-747, Korea

I. S. Hahn and A. R. Kim
Department of Science Education, Ewha Woman's University, Seoul 120-750, Korea

H. J. Kim
Physics Department, Kyungpook National University, Daegu 702-701, Korea

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We measured the thermal neutron capture cross section for the ^{180}W nucleus. There is only one previous measurement with regard to this cross section, and it yielded a value of $30^{+300\%}_{-100\%}$ b. To determine whether ^{181}W is an appropriate low energy neutrino source, the thermal neutron capture cross section should be measured more precisely to estimate the production rate of ^{181}W inside a nuclear reactor. We measured the cross section of ^{180}W using a natural tungsten foil and obtained a value of 22.6 ± 1.7 b.

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Neutrino oscillation experiments such as Super-Kamiokande (SK) [1], K2K [2], SNO [3], and KAMLAND [4] have achieved great progress in understanding the masses and mixing angles of neutrinos. The nonzero masses and large mixing angles of neutrinos have been confirmed, and this fact has stimulated further theoretical models on neutrino masses. The above-mentioned neutrino oscillation experiments detected neutrinos from the sun (SK, SNO), an accelerator (K2K), and nuclear reactors (KAMLAND, CHOOZ). Therefore, the neutrino sources are fixed at their locations, which limits the flexibility of the experiments to some extent. In addition to these neutrino sources, an artificial neutrino source (ANS) has been studied for the calibration of neutrino detectors such as GALLEX [5] and SAGE [6]. The ANS refers to the neutrinos emitted from β decaying nuclei. For example, the neutrinos from the electron capture of ^{51}Cr nuclei are monoenergetic mostly with an energy of 753 keV. Approximately 1 MCi (mega-Curie) of ^{51}Cr was produced by a (n, γ) capture reaction inside a nuclear reactor with enriched ^{50}Cr to calibrate the solar neutrino detector [5,6]. Recently, a ^{37}Ar neutrino source, in the amount of about 0.5 MCi, has been developed [7] and used for SAGE solar neutrino experiment [8].

Besides the calibration of neutrino detectors, a radioisotope neutrino source can be used to study nonstandard neutrino properties and possibly in neutrino oscillation experiments. Recently, two groups, TEXONO [9] and MUNU [10], reported the most stringent upper limits on the neutrino magnetic moments to be $\mu_\nu < 7.4 \times 10^{-11} \mu_B$ and $\mu_\nu < 9.0 \times 10^{-11} \mu_B$, respectively, from the measurements of νe elastic scattering energy spectra of the reactor neutrinos. The neutrino magnetic moment is one of the most fundamental properties of neutrinos, and it is anticipated that the sensitivity can be greatly improved if a strong neutrino source of more than 1 MCi with a proper radioisotope is available.

There are other candidates for neutrino sources besides ^{51}Cr and ^{37}Ar, such as ^{181}W, ^{170}Tm, and ^{147}Pm [11]. The appropriate radioisotope for an ANS should have characteristics such as (1) the decay time should be relatively long, from 10 days to 10 years, to perform an experiment, (2) γ intensities should be low for safety issues, (3) the abundance of mother nuclei should be relatively large for low cost, (4) the thermal neutron capture cross section should be large, and (5) it should be possible to produce a very pure target material for irradiation to reduce the γ activity from contaminants. Among the nuclei, ^{147}Pm is produced from nuclear spent fuel, and other nuclei are produced by the (n, γ) reaction in nuclear reactors. ^{181}W possesses good properties except for the low abundance of ^{180}W at a level of 0.12%. Therefore, enrichment is necessary for tungsten, and the production cost depends on the amount of material required to produce the desired activity, which is usually more than 1 MCi. In this respect, the thermal neutron capture cross section of ^{180}W is an important parameter to know. To date, however, there is only one measurement with regard to the thermal neutron capture cross section for this nucleus, which is $30^{+300\%}_{-100\%}$ b. This value was measured by Pomerance more than 50 years ago [12]. Since the uncertainty of this value is very large, it is necessary to measure the cross section more precisely to determine if ^{181}W is a good candidate for an ANS.

To measure the capture cross section, we irradiated natural tungsten foils in a thermal neutron irradiation facility and measured the γ's from the ^{181}W decay. In a natural tungsten foil, ^{184}W and ^{186}W are more abundant, and the capture cross sections can be measured with sufficiently small uncertainties. Therefore, we can obtain the ^{180}W capture cross section by a comparison of the measured activities of ^{185}W and ^{187}W. In this report, we describe the measurement of the capture cross section of the ^{180}W$(n, \gamma)^{181}$W reaction with a considerably smaller error than that in the previous measurement.

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[*] ydkim@sejong.ac.kr

An irradiation area at the HANARO research reactor facility in Korea was used for the measurement. At HANARO, the neutron irradiation facility for the boron neutron capture therapy (BNCT) consists of a water shutter, a fast neutron and γ ray filter, a liquid nitrogen cooling system, a beam collimator, and shieldings [13]. The cadmium ratio (Cd ratio) at this facility is known to be larger than 100 at the position of irradiation. We prepared two identical tungsten foils (99.9% pure) with a size of $50.1 \times 50.1 \times 0.138$ mm (6.644 g) to measure the Cd ratio simultaneously with the irradiation. The thickness of the foil was calculated by dividing the mass of the foil by the area, considering a density of 19.25. One foil was sandwiched between two cadmium foils of approximately 1-mm thickness. The thermal neutron flux that was previously measured at this beam line was approximately $(7 \sim 8) \times 10^8$ neutrons/cm^2 s.

Table I shows the information on the stable isotopes in the natural tungsten foil [14]. The underlying concept is that we can obtain the capture cross section of ^{180}W with respect to the capture cross sections of ^{184}W and ^{186}W. This method has the advantage of eliminating potential systematic errors from the thermal neutron flux, flux profile, foil thickness, foil size, irradiation time, etc. Furthermore, the absorption effect of the tungsten foil does not contribute to the errors in the final cross section, because all the stable tungsten isotopes will experience the same amount of neutron flux regardless of the absorption. As shown in Table I, there are γ's with sufficiently long half-lives for ^{185}W, ^{187} W, and ^{181}W, and we can obtain the production rates of the three radioisotopes from them. There are two more γ's from ^{187}W decay with significant γ intensities, 134.2 and 479.5 keV; however these γ's are not used in the current analysis. The 134.2-keV γ is not separable from the 136.3-keV γ of ^{181}W, and the 479.5-keV γ overlaps with other unidentified γ's.

TABLE I. Stable isotopes in natural tungsten. $T_{1/2}$, E_γ, and I_γ (gamma intensity) correspond to the radioisotopes produced by the (n, γ) reaction. The values in parentheses for I_γ indicate the associated errors.

Isotope	Abun. (%)	σ (barn)	$T_{1/2}$ (day)	E_γ (keV)	I_γ (%)
^{180}W	0.12	30^{+90}_{-30} [a]	121.2	136.3	0.0311(10)
		22.6 ± 1.7 [b]		152.3	0.083(3) [c]
^{184}W	30.6	1.76 ± 0.09 [d]	75.1	125.4	0.0192(3)
^{186}W	28.4	39.5 ± 2.3 [e]	0.988	551.5	5.08(17)
				618.4	6.28(21)
				685.8	27.3(9)
				772.9	4.12(13)

[a] Reference [12].

[b] This work.

[c] The γ intensity of this level is written as 0.0083 by mistake in all the existing databases including the NNDC database, *Table of Isotopes* (8th ed.), Nuclear Data Sheets [17], and NUDAT. A correct value of 0.083 was reported in Reference [18]. We reported this mistake to the NNDC database group, and it presently stands corrected in the NNDC database.

[d] Reference [15].

[e] Reference [16].

We irradiated both the enclosed and open foils for 5 h at the BNCT facility, and the irradiated foils were left for 12 days or more to reduce the activity of the foils. Even though the half-life of ^{187}W is only 1 day, we still have sufficient counts from the ^{187}W decay. The irradiated tungsten foils were measured with a low-background 100% efficiency high-purity germanium (HPGe) detector located underground at a depth of 700 m in the Yangyang laboratory of the Dark Matter Research Center (DMRC) in Korea.

The Cd ratio was estimated by the ratio of the γ's from the ^{187}W peaks with and without a cadmium enclosure. The obtained ratio was 245 ± 10, which guarantees that the activities in the open foil have little contribution from nonthermal neutrons.

The top of Fig. 1 shows the measured HPGe spectra of the γ peaks at the energies of 136.3, 152.3(^{181}W), and 125.4 keV (^{185}W). The upper spectrum was obtained for a measurement period of 5.4 days, starting 25.0 days after the irradiation. Even after 25 days, there were continuous high background events below 400 keV. This occurrence of events was due to

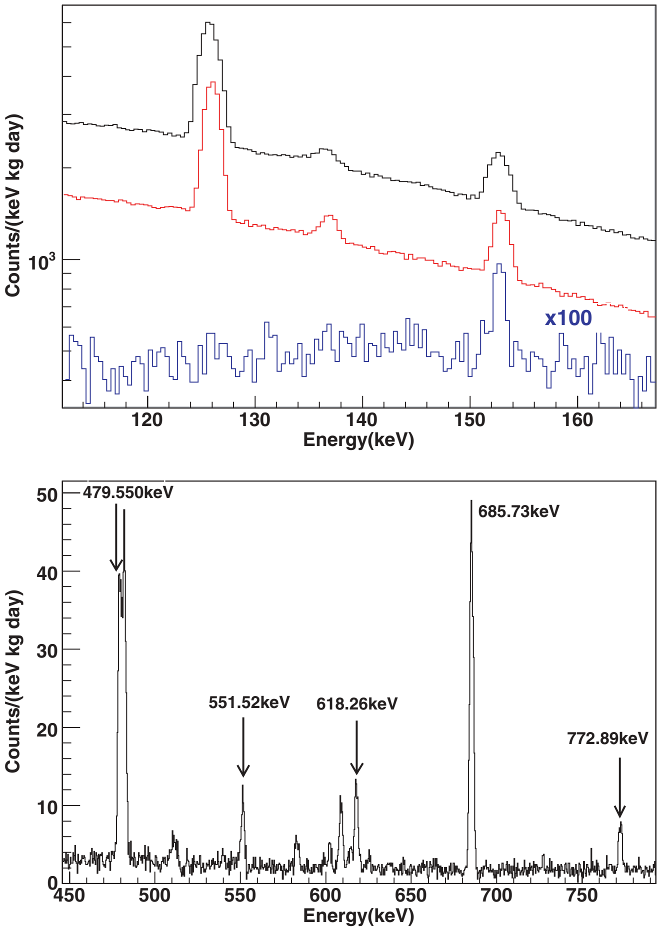

FIG. 1. (Color online) Top panel: Energy spectra of the tungsten foil in the energy region of ^{181}W and ^{185}W γ peaks obtained after 25 days (upper), 81 days (middle), and 993 days (lower, multiplied by 100 for easy comparison) from the end of the irradiation period. Bottom panel: Same as top, but in the energy region of ^{187}W obtained after 25 days.

TABLE II. Net counts of the γ's from irradiated tungsten foil as measured with HPGe detector. Data are for the measurement after 25 days from irradiation. η is the detection efficiency of the γ's, $\delta\eta$ is the error in efficiencies relative to that of 152.3 keV γ (see the text), and R is the production rate.

	E_γ (keV)	Counts	η (%)	$\delta\eta$	R ($\times 10^5$/s)	R_{av} ($\times 10^5$/s)
^{181}W	136.3	7644 ± 614	12.0	0.02	4.33 ± 0.38	4.09 ± 0.18
	152.3	21208 ± 650	13.4	0.0	4.04 ± 0.19	
^{185}W	125.4	111357 ± 984	10.4	0.04	78.8 ± 3.5	78.8 ± 3.5
^{187}W	551.5	330 ± 33	9.4	0.10	1765 ± 257	1813 ± 195
	618.4	388 ± 36	8.8	0.10	1789 ± 251	
	685.8	1615 ± 60	8.3	0.10	1820 ± 203	
	772.9	235 ± 28	7.7	0.10	1888 ± 300	

the bremsstrahlung photons from the β decay of ^{185}W to the ground state of ^{185}Re (branching ratio 99.93%), which has a Q value of 433 keV. Even with the bremsstrahlung background, we can still clearly observe the 125.4, 136.3, and 152.3 keV γ peaks. We also show the spectra obtained at 81 days (middle spectrum) and 993 days (lower spectrum) after the completion of the irradiation period. The data obtained at 81 days after irradiation was used to obtain the counts of the 136.3-keV peak of ^{181}W , since the 25-day data still had approximately 10% counts from the 134.2-keV peak of ^{187}W. We obtained the half-lives of ^{185}W and ^{187}W as 72.4 ± 1.3 and 118.3 ± 5.5 days, respectively. The half-life of ^{185}W slightly differs from the value in the database. The bremsstrahlung background was significantly reduced in the spectrum of the last data. The bottom of Fig. 1 shows the peaks of 551.5, 618.4, 685.8, and 772.9 keV (^{187}W) in the higher energy region of the upper spectrum.

Table II shows the measured count rates and related errors. The production rate of radioisotopes from the (n, γ) reaction during the irradiation can be written as

$$R = F\frac{m\alpha A_0}{w}\sigma. \qquad (1)$$

Here, F is the neutron flux, m is the total mass of the foil, and σ, w, and α are the capture cross section, mass number, and relative abundance of the target isotope, respectively. A_0 is Avogadro's number. The thermal neutron capture cross section of ^{180}W can be obtained along with the existing capture cross section data of ^{184}W [15] and ^{186}W [16] as

$$\sigma_{180} = \frac{\alpha_i w_{180} R_{180}}{\alpha_{180} w_i R_i}\sigma_i. \qquad (2)$$

Here, the index i refers to ^{184}W or ^{186}W. The production rate R is calculated from the data as

$$R_i^j = \frac{C_i^j}{\tau_i I_i^j \eta_i^j (1 - e^{-T/\tau_i})(e^{-t_1/\tau_i} - e^{-t_2/\tau_i})}. \qquad (3)$$

Here, C_i^j, I_i^j, and η_i^j are the net count, γ intensity, and detection efficiency of the jth γ of the ith isotope, respectively. R_i^j is the production rate calculated using the net γ count. τ_i is the mean decay time of the produced radioisotope, T is the irradiation time, and t_1 and t_2 are the start and stop times of the HPGe measurement from the end of the irradiation period.

The production rates of each radioisotope are provided in the last two columns of Table II. The errors in the production rates are due to errors in the count statistics, γ intensity [17,19], and detection efficiency. The detection efficiency of the underground HPGe detector was previously studied by comparing the measurements from a calibrated multi-γ radioactive source with GEANT4 simulations [20]. Unfortunately, the efficiencies of the HPGe detector shows discrepancies between the measurements and the simulations, in the amount of about 20% in absolute values for the energy region over 100 keV. However, only relative efficiencies between different γ energies will be important for the ^{180}W cross section, since we obtain it by a comparison to ^{184}W and ^{186}W cross sections. Figure 2 shows the measured (square points) and simulated (circular points) efficiencies with the calibrated source. In this figure, the simulated efficiencies are reduced by 20% to match an efficiency with the measured one at 152.3 keV energy. We have observed that the measured efficiencies are slightly lower (higher) than the simulated efficiencies at energies less (greater) than 152.3 keV. The black histogram shows the simulated efficiency for the

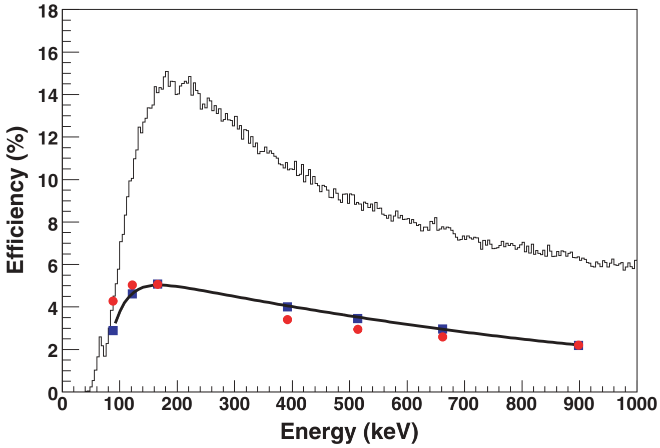

FIG. 2. (Color online) Measured (square points) and simulated (circular points) efficiencies of the HPGe detector with a multi-γ radioactive source. The black histogram is the simulated efficiency for the tungsten foil with the GEANT4 program.

tungsten foil, which decreases for the energies less than 200 keV due to the γ attenuation inside the tungsten foil.

For the production rate in Eq. (3), the simulated efficiencies for the tungsten foil were corrected by the above-mentioned differences between the measurements and the simulations with the calibrated source. The correction factor was 4% for the 125 keV γ of ^{185}W and 10% for γ's of ^{187}W. These correction factors were taken as the uncertainties of the γ efficiencies and are shown in Table II along with the corrected efficiency values. Indeed, the correction improves the consistency between two thermal neutron flux values calculated with ^{184}W and ^{186}W capture cross sections. In the case of ^{181}W and ^{187}W, the production rates obtained with multiple γ's are consistent with each other as expected. The weighted mean of the production rates are provided in the last column of Table II for the three isotopes. Although the thermal neutron flux cancels out in the cross section calculation, we obtained a value of $6.84 \pm 0.28 \times 10^8/\text{cm}^2$ s with the known cross sections of ^{185}W and ^{187}W, which is consistent with the previously reported flux value for the BNCT facility.

The capture cross section of ^{180}W, σ_{180}, is calculated using Eq. (2) with the known cross sections of 1.76 ± 0.09 b for ^{184}W and 39.5 ± 2.3 b for ^{186}W. First we obtained $\frac{\sigma_{180}}{R_{180}}$ with the two cross sections individually since the two cross section data are independent, and the two results are consistent within the uncertainties. Then σ_{180} was obtained by multiplying the weighted mean of two $\frac{\sigma_{180}}{R_{180}}$ values by the production rate of ^{180}W. The final ^{180}W capture cross section thus obtained was 22.6 ± 1.7 b. While our new cross section is consistent with the data obtained by Pomerance [12], the small uncertainty makes it possible to evaluate the feasibility of ^{181}W as a neutrino source; this evaluation was too ambiguous because of the large error in the data obtained by Pomerance.

Although the present result can be improved with a better understanding of the HPGe detector efficiency in the future, it is sufficiently accurate to realistically evaluate the feasibility of ^{181}W as a neutrino source. Considering that ^{181}W enrichment is technically difficult, ^{181}W does not seem to be very attractive. Instead, ^{170}Tm would be a better candidate with respect to the cost; however, the safety issues related the high γ-ray flux and bremsstrahlung photons have to be determined. The measurement method we used in this report can be utilized for other low-abundance isotopes.

In summary, we measured the thermal neutron capture cross section of ^{180}W as 22.6 ± 1.7 b. Although ^{181}W is a good candidate for an artificial neutrino source, the production of ^{181}W would be too expensive considering the thermal neutron capture cross section measured in this work.

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