Measurement on the thermal neutron capture cross section of $^{180}\text{W}$

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(Dated: February 1, 2008)

Abstract

We have measured the thermal neutron capture cross section for $^{180}\text{W}$ nucleus. There is only one previous data on this cross section with a value of $30^{+300}\%_{-100}\%$ barn. To consider $^{181}\text{W}$ as a low energy neutrino source, the thermal neutron capture cross section should be measured more precisely to estimate the production rate of $^{181}\text{W}$ inside a nuclear reactor. We measured the cross section of $^{180}\text{W}$ with a natural tungsten foil and obtained a new value of $21.9 \pm 2.5$ barn.

PACS numbers: 25.40.Lw, 14.60.St

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I. MOTIVATION

Neutrino oscillation experiments such as Super-Kamiokande (SK) [1], K2K [2], SNO [3], and the KAMLAND [4], have achieved great progresses in understanding the masses and mixing angles of the neutrinos. The non-zero masses and the large mixing angles of the neutrinos are confirmed and stimulate further theoretical models on the neutrino masses. The above neutrino oscillation experiments detected the neutrinos from the sun (SK, SNO), from accelerator (K2K), and from nuclear reactors (KAMLAND, CHOOZ). Therefore the neutrino sources are fixed in their locations, which limits the flexibility of the experiments to some extent. In addition to these neutrino sources, artificial neutrino source (ANS) has been studied for the calibration of neutrino detectors such as GALLEX [5] and SAGE [6]. The ANS refers the neutrinos from a beta decaying nuclei. For example, the neutrinos from electron capture of $^{51}$Cr nuclei are mostly mono-energetic at the energy of about 750 keV. About 1 MCi(million Curie) of $^{51}$Cr was produced by $(n, \gamma)$ capture reaction inside a nuclear reactor with enriched $^{50}$Cr to calibrate the solar neutrino detector [5, 6].

Besides the calibration of a neutrino detector, the radioisotope neutrino source can be used to study non-standard neutrino properties and possibly for neutrino oscillation experiment. For example, about 1 MCi neutrino source can be used to measure the magnetic moment of the neutrinos with a neutrino detector closely located to the source. Recently two groups, TEXONO [7] and MUNU [8], reported most stringent upper limits on the neutrino magnetic moment as $\mu_\nu < 7.4 \times 10^{-11} \mu_B$ and $\mu_\nu < 9.0 \times 10^{-11} \mu_B$ respectively from the measurements of $\nu e$ elastic scattering energy spectra with the reactor neutrinos. The neutrino magnetic moment is one of the most fundamental properties of neutrinos and it is anticipated that one can improve the sensitivity if a strong neutrino source with a proper radioisotope is available.

There are other candidates for a neutrino source besides $^{51}$Cr, such as $^{181}$W, $^{170}$Tm, and $^{147}$Pm, etc [9]. An appropriate radioisotope for ANS should have characteristics such as; (1) relatively long decay time between 10 days and 10 year to perform an experiment, (2) low gamma intensities for safety issue, (3) abundances of mother nuclei should be relatively large for low cost, (4) thermal neutron capture cross section should be large. Among the nuclei, $^{147}$Pm is produced from nuclear spent fuel, and the others are produced by $(n, \gamma)$ reaction in nuclear reactors. $^{181}$W has good properties except the low abundance of $^{180}$W at the
level of 0.12%. Therefore an enrichment is necessary for tungsten, and the production cost depends on the amount of material needed to produce the desirable activity, which is usually more than 1MCi. In this respect, the thermal neutron capture cross section of $^{180}$W is an important parameter to know. Until now, however, there is only one data on the thermal neutron capture cross section on this nucleus, 30 $^{\pm}300$% −100% barn, measured by Pomerance more than 50 years ago [10]. Since the uncertainty of this data is very large, it is necessary to measure the cross section more precisely to determine if $^{181}$W is a good candidate for ANS.

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

II. EXPERIMENTAL SETUP

An irradiation area at HANARO research reactor facility in Korea was used for the measurement. At HANARO, a neutron irradiation facility for BNCT (Boron Neutron Capture Therapy) consists of a water shutter, a fast neutron and gamma ray filter, a liquid nitrogen cooling system, a beam collimator, and shieldings [11]. 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 50.1mm X 50.1mm X 0.138mm (6.644g) to measure the Cd ratio at the same time of irradiation. The thickness of the foil was calculated by dividing the mass of the foil by the area with a density of 19.25$g/cm^3$. One foil was sandwiched by two Cadmium foils of about 1mm thickness. The thermal neutron flux was previously measured at this beam line as about 7-8×10$^8$ neutrons/sec/cm$^2$.

Table [II] shows the informations of stable isotopes in natural tungsten foil [12]. The idea 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 which were measured with much smaller errors. This method has the advantage of cancellation of potential systematic errors from thermal neutron flux, flux profile, foil thickness, foil size, and irradiation time etc. Neither the absorption effect
inside the tungsten foil contributes to the errors in the final cross section since all the stable tungsten isotopes will see the same amount of the neutron flux regardless of the absorption. As shown in the table, there are γs with sufficiently long half-lives for $^{185}$W, $^{187}$W, and $^{181}$W, from which we can obtain the production rates of the three radioisotopes. There are two more γs from $^{187}$W decay with significant gamma intensities, 134.2 keV and 479.5 keV, but these gammas were not used in current analysis. 134.2 keV gamma is not separable from 136.3 keV gamma from $^{181}$W and 479.5 keV gamma was overlapped with other unidentified gammas.

TABLE I: Stable isotopes in natural tungsten. $T_{1/2}$, $E_{\gamma}$, and $I_{\gamma}$ (gamma intensity) are for the produced radioisotopes by $(n, \gamma)$ reaction. The values in the parentheses in $I_{\gamma}$ indicates the associated errors.

| Isotope | Abun. | $\sigma$ | $T_{1/2}$ | $E_{\gamma}$ | $I_{\gamma}$ |
|---------|-------|---------|-----------|--------------|--------------|
|         | (%)   | (barn)  | (day)     | (keV)        | (%)          |
| $^{180}$W | 0.12  | $30^{+90}_{-30}$ | 121.2     | 136.3        | 0.0311(10)   |
|          |       |         |           |              |              |
|          |       | 21.9 ± 2.5$^{b}$ | 152.3     |              | 0.083(3)$^{e}$ |
| $^{184}$W | 30.6  | $1.76 \pm 0.09$ | 75.1      | 125.4        | 0.0192(3)    |
| $^{186}$W | 28.4  | $39.5 \pm 2.3$ | 0.988     |              |              |
|          |       |           |           | 551.5        | 5.08(17)     |
|          |       |           |           | 618.4        | 6.28(21)     |
|          |       |           |           | 685.8        | 27.3(9)      |
|          |       |           |           | 772.9        | 4.12(13)     |

$^{a}$Reference [10].
$^{b}$This work.
$^{c}$Reference [13].
$^{d}$Reference [14].
$^{e}$The gamma intensity of this level is written as 0.0083 by mistake in all the existing database including NNDC database, Table of Isotopes (8th Ed.), Nuclear Data Sheets [15], and NUDAT. The correct value, 0.083, was reported in the paper of [16]. We reported this mistake to NNDC database group, and it is corrected in NNDC database at the moment of this writing.

We irradiated both the enclosed and open foils for 5 hours at the BNCT facility, and the irradiated foils are 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 enough counts from the decay of
FIG. 1: HPGe spectrum of tungsten foils with (lower) and without (upper) Cd enclosure.

$^{187}$W. The irradiated tungsten foils were measured with a low background HPGe detector located at 700 meter deep underground at Yangyang laboratory of Dark Matter Research Center (DMRC) in Korea. The relative efficiency of the HPGe detector is 100%, and the detection efficiencies are measured with absolute intensity sources before, and compared with GEANT4 simulations [17].

III. RESULTS

Figure 1 shows the HPGe spectra of two tungsten foils with and without cadmium enclosure. The Cd ratio has been estimated by the ratio of the $\gamma$s from $^{187}$W peaks, which are dominant in these spectra. The Cd ratio was obtained to be 245±10, which guarantees the activities we obtain from the open foil has little contribution from non-thermal neutrons.

Figure 2 shows the measured HPGe spectra of gamma peaks at the energies of 136.3, 152.3 keV ($^{181}$W), and 125.4 keV ($^{185}$W). The upper spectrum was obtained with a measurement of 5.4 days starting 25.0 days after the irradiation. Even after 25 days, there are continuous high background events below 400 keV. This was due to the bremsstrahlung photons from the beta 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 observe 125.4, 136.3, and 152.3 keV gamma peaks clearly. We also show the spectra obtained at 81 days (middle spectrum) and 993 days (lower spectrum) since the end of the irradiation period. The data obtained at 81 days after the irradiation was used to get the counts of the
136.3 keV peak of $^{181}$W since the 25 days data still has about 10 % counts from 134.2 keV peak of $^{187}$W. We also confirmed that the half-lives of these peaks are consistent with the expectation of $^{181}$W and $^{185}$W. The bremsstrahlung background was reduced significantly in the spectrum of last data.

Figure 2 BOTTOM 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 of Figure 2 TOP. The 479.5 keV peak is overlapped with another peak and is not used in the analysis. Figure 3 shows the count rates of 152.3 keV and 125.4 keV $\gamma$s obtained at various measurement times after the irradiation. We obtained the half-lives of $^{185}$W and $^{187}$W as $72.4 \pm 1.3$ day and $118.3 \pm 5.5$ days respectively. The half-life of $^{185}$W is slightly off from the value in the database.

Table II: The measured net counts of the $\gamma$s from irradiated tungsten sample with HPGe detector. Data are for a measurement after 25 days from irradiation. $\eta$ is detection efficiency and R is production rate.

| $E_\gamma$ (keV) | Counts     | $\eta$ (%) | R $\times 10^5$/sec | R$_{av}$      |
|------------------|------------|-------------|---------------------|---------------|
| $^{181}$W        | 136.3      | 7644 ± 614  | 12.2                | 4.27 ± 0.92   | 4.14 ± 0.62   |
|                  | 152.3      | 21208 ± 650 | 13.4                | 4.04 ± 0.83   |               |
| $^{185}$W        | 125.4      | 111357 ± 984| 10.9                | 77.0 ± 15.5   | 77.0 ± 15.5   |
| $^{187}$W        | 551.5      | 330 ± 33    | 8.5                 | 1942 ± 439    | 1993 ± 221    |
|                  | 618.4      | 388 ± 36    | 8.0                 | 1967 ± 439    |               |
|                  | 685.8      | 1615 ± 60   | 7.5                 | 2002 ± 413    |               |
|                  | 772.9      | 235 ± 28    | 7.0                 | 2077 ± 488    |               |

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

$$ R = F \frac{m \alpha A_0}{w} \sigma. $$

(1)

Here $F$ is the neutron flux, $m$ is the total mass of the foil, $\alpha$, $w$, and $\sigma$ are the capture cross section, mass number, and the relative abundance of target isotope, respectively. $A_0$ is the Avogadro’s number. The thermal neutron capture cross section of $^{180}$W can be obtained
FIG. 2: TOP : Energy spectra of the tungsten foil in the energy region of $^{181}\text{W}$ and $^{185}\text{W}$ gamma peaks obtained after 25 days (upper), 81 days (middle), 993 days (lower, multiplied by 100 for easy comparison) since the end of irradiation period. BOTTOM : same as TOP in the energy region of $^{187}\text{W}$ obtained after 25 days.

with the existing capture cross section data of $^{184}\text{W}$ [13] and $^{186}\text{W}$ [14] as

$$\sigma_{180} = \frac{\alpha_i w_{180} R_{180}}{\alpha_{180} w_i R_i} \sigma_i.$$  \hspace{1cm} (2)

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

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

Here $C_i^j$, $I_i^j$, $\eta_i^j$ are the net count, the gamma intensity, and the detection efficiency of $j$ th gamma from $i$ th isotope respectively, and $R_i$ is the production rate calculated with the net gamma count. $\tau_i$ is the mean decay time of the produced radioisotope, $T$ is the irradiation
FIG. 3: The activity of 125.4 keV (upper) and 152.3 keV (lower) $\gamma$s as a function of time of measurements. The half-lives of $^{181}\text{W}$ and $^{185}\text{W}$ obtained from the data are written in the figure in units of days.

time, and $t_1, t_2$ are the starting and stop time of the HPGe measurement since the end of the irradiation period.

The production rates of each radioisotope are in the last two columns of Table II. The errors in the production rates are due to the errors in count statistics, gamma intensity [15, 18], and detection efficiency. Unfortunately, the efficiencies of the HPGe detector at underground is not accurately measured since we observed discrepancies between calibrated source measurements and the GEANT4 simulations. The conservative estimation of the discrepancies are in the order of 20% for the energy region over 100 keV, and it contributes most for the uncertainties in the production rates. The errors of the abundances of each isotopes are negligible compared with the other errors, so neglected in the analysis. In the case of $^{181}\text{W}$ and $^{187}\text{W}$, the production rates obtained with multiple $\gamma$s are consistent with each other as expected. The weighted mean of the production rates are in the last column of Table II. Though, the thermal neutron flux cancels out in the cross section calculation, we can calculate the thermal neutron flux with the known cross section of $^{185}\text{W}$ and $^{187}\text{W}$. The obtained flux results in $7.6 \pm 0.7 \times 10^8 / \text{cm}^2 / \text{sec}$, which is consistent with the value reported for the BNCT facility previously.

The capture cross section of $^{180}\text{W}$ is calculated using equation 2 with the known cross section of $1.76 \pm 0.09$ barn for $^{184}\text{W}$ and $39.5 \pm 2.3$ barn for $^{186}\text{W}$, and we obtained $24.7 \pm 3.9$ barn and $20.1 \pm 3.2$ barn respectively. The two cross sections are consistent within the uncertainties. The final $^{180}\text{W}$ capture cross section is obtained with the weighted mean of
the two values, and it is $21.9 \pm 2.5$ barn. While our new cross section is consistent with the
data by Pomerance \[10\], the small uncertainty makes it possible to evaluate the feasibility of
$^{181}$W as a neutrino source, which was too ambiguous due to the large error in the Pomerance
data.

IV. DISCUSSION

Though the present result can be improved with a better understanding of the HPGe
detector efficiency in the future, it is accurate enough to evaluate the feasibility of $^{181}$W as a
neutrino source more realistically. Considering that $^{181}$W enrichment is technically difficult,$^{181}$W seem to be not very attractive. Instead, $^{170}$Tm would be a better candidate in the
respect of the cost, but one has to figure out the safety issue related the high gamma-ray
flux, and bremsstrahlung photons. 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 $21.9 \pm 2.5$ barn. Though $^{181}$W is a good candidate for
artificial neutrino source, the production of $^{181}$W would be too expensive considering the
thermal neutron capture cross section measured in this work.

Acknowledgments

This work was supported by the Korea Research Foundation (Grant KRF-2002-070-C00027). The authors thank Dr. M.S. Kim and the staffs of the BNCT facility of HANARO
research reactor for the neutron irradiation and the DMRC group for using the HPGe de-
tector at underground.

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