Determination of the Number of ^{232}Th Nuclei in the Sample Using Small Solid Angle Method

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ABSTRACT

The ^{232}Th(*n*, *f*) cross-section data from EXFOR library were collected and analyzed, which show that the measurement results using mono-energetic neutron sources are obviously higher than those using white neutron sources. In order to clarify the existing discrepancies, accurate ^{232}Th(*n*, *f*) cross-section measurements are being planned. The $^{232}Th(OH)_4$ foil sample was prepared. A small solid angle device was designed and installed by which the number of ^{232}Th nuclei in the sample was determined.

I. INTRODUCTION

^{232}Th, the most stable isotope of thorium (natural abundance ~100%), is a fissionable nucleus which plays an important role in the Th-U fuel cycle [1,2]. The study of the ^{232}Th (*n*, *f*) reaction is important in nuclear engineering applications [3-5]. Besides, measurements of this cross section could enhance our understanding in nuclear physics. For example, the special structure of the ^{232}Th(*n, f*) cross sections called the "thorium anomaly" effect could be explained by triple-humped barriers of ^{232}Th [6,7].

However, comparing with other fissile nuclei, such as ^{238}U or ^{235}U, existing measurements of ^{232}Th are much fewer for the (*n*, *f*) reaction. Besides, there is a systematic difference between the measurement results using mono-energetic neutron sources and those using white neutron sources in the MeV region. The experimental cross sections from 1.5 to 15 MeV taken from EXFOR library are shown in Fig. 1 [8]. Due to the update of experimental techniques and apparatus, only the data after 1970s were collected and analyzed. In order to show the overall trend of the experimental data more clearly, error bars were omitted, and the data points are connected with straight lines.

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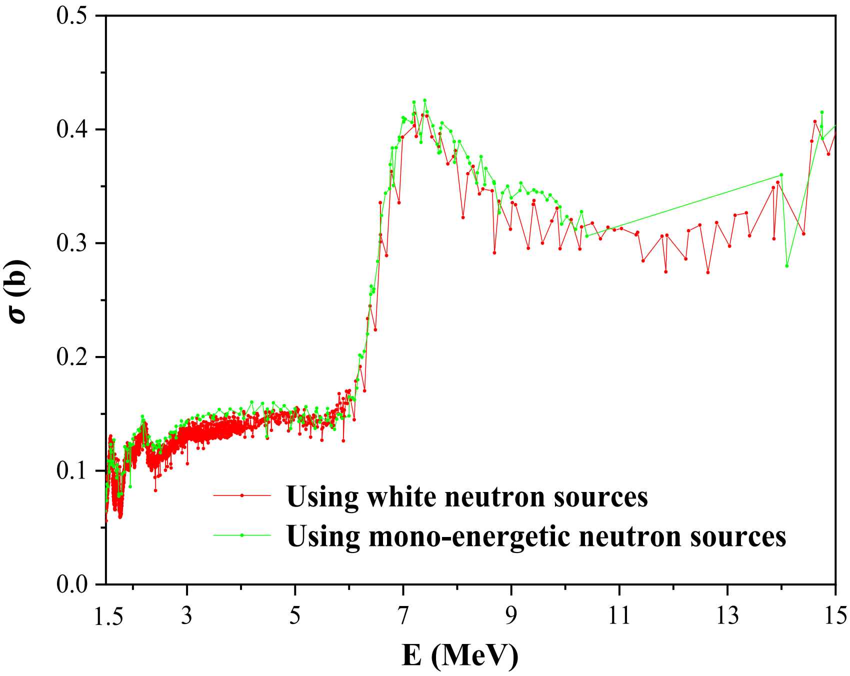

Fig. 1 (Color online) Experimental cross sections of the ^{232}Th(n, f) reaction from 1.5 to 15.0 MeV using white neutron sources and mono-energetic neutron sources. The results were taken from EXFOR library (after 1970s).

From Fig. 1, one could see that the measurement results using mono-energetic neutron sources are obviously higher than those using white neutron sources. The results using mono-energetic neutron sources was 5.47% higher than those using white neutron sources on average, and the percentage was up to 8.16% for 2.0 MeV < E_n < 4.0 MeV. So, in order to clarify the existing discrepancies, accurate ^{232}Th(n, f) cross-section measurements are being planned.

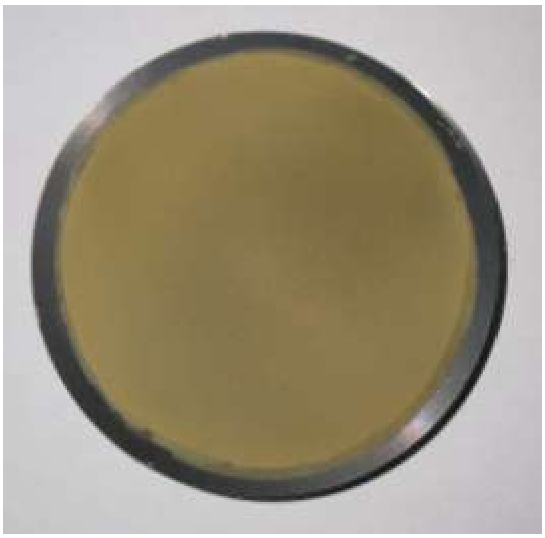

Fig. 2 (Color online) Picture of the ^{232}Th(OH)$_4$ sample.

A $^{232}Th(OH)_4$ foil sample with tantalum backing was prepared by Joint Institute for Nuclear Research in Dubna, and picture of which is shown in Fig. 2. The diameter of the sample is 44.0 mm. Determination of the number of ^{232}Th nuclei is the indispensable step for ^{232}Th(n, f) cross-section measurements. For the α-radioactive material, α-spectrometry is a common technique of nucleus number determination. The decay chain of ^{232}Th is presented in Fig. 3.

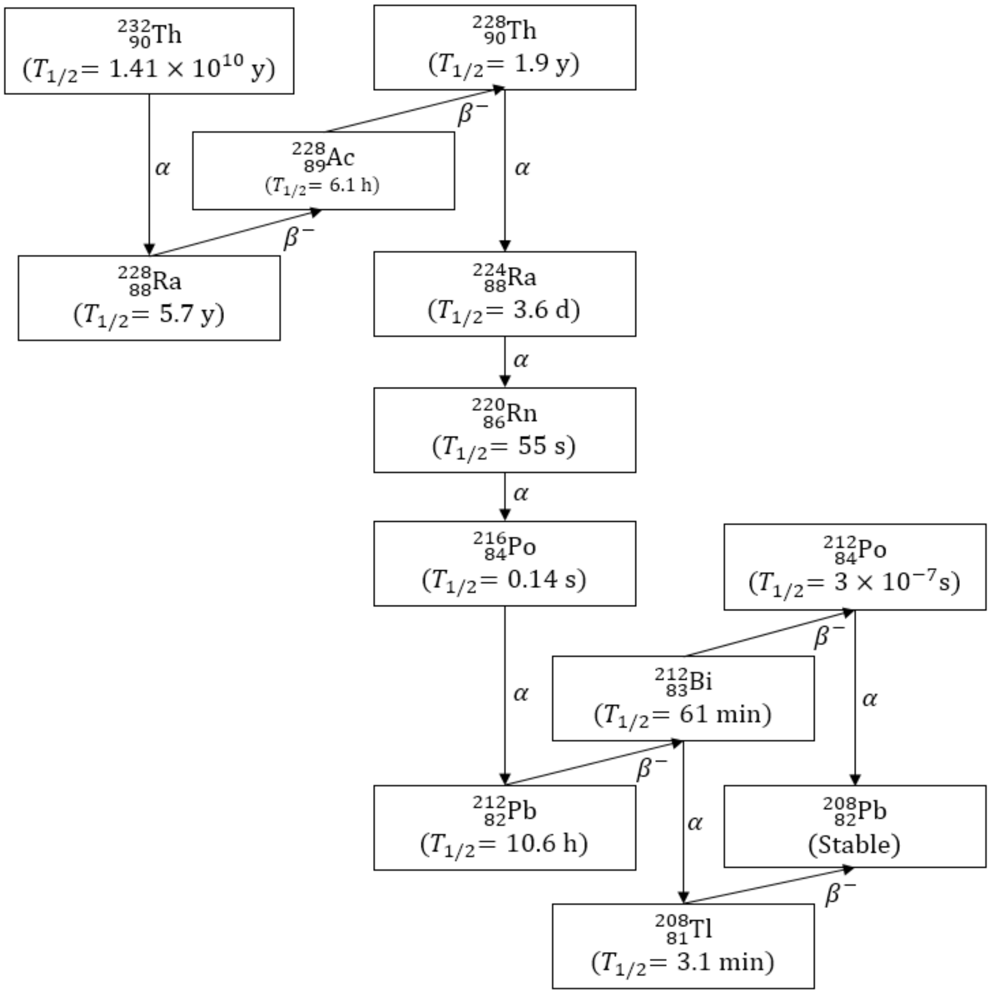

Fig. 3 The decay chain of ^{232}Th.

With regard to α counters, the grid ionization chamber (GIC) has a wide range of applications because of the big detection efficiency (~50%). However, both the limited energy resolution and the peak tailing effect restrict its usage [9]. A typical ^{232}Th α spectrum measured by GIC is shown in Fig. 4. The strong asymmetry and tailing of the peaks can be observed from the spectrum. The big tailing of α peaks from the daughter nuclides of ^{232}Th would introduce significant error when determining the counts of the α peak from ^{232}Th. So, other techniques should be used instead of the GIC method.

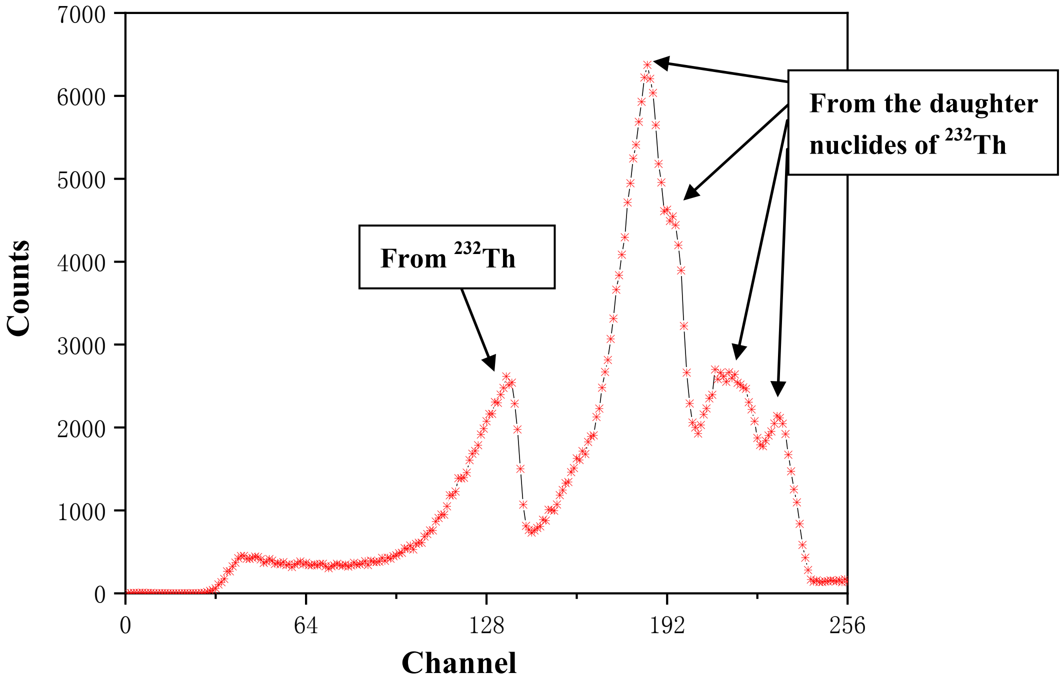

Fig. 4 (Color online) Typical ^{232}Th α spectrum measured by the grid ionization chamber.

In order to reduce the tailing effect of the peaks, the small solid angle method with semiconductor detector was adopted [10]. The emitted α particles within small solid angles would have shorter tracks in the material, which lead to less energy losses and lower tailings of the peaks. Thus, we took the method of small solid angle to determine the number of ^{232}Th nuclei. The details of the experiments will be shown in the next section.

II. DETAILS OF EXPERIMENTS

A small solid angle device was designed and installed for the determination of the number of ^{232}Th nuclei. The device consists of the vacuum chamber, vacuum pumping system, electronic equipment, and digital data-acquisition (DAQ) system.

A CAD drawing of the vacuum chamber showing the main structure is presented in Fig. 5. As Fig. 5 shows, there are three anti-scattering baffles, a sample holder, a diaphragm, and a Au-Si surface barrier semiconductor detector inside the vacuum chamber. The sample holder, detector, diaphragm and anti-scattering baffles are connected by three screw rods which are mounted at the lid of the vacuum chamber, and all of them are centered with a common symmetry axis. In addition, their positions are fixed by nuts on the screw rods.

A Au-Si surface barrier semiconductor detector (optimum bias voltage is 40 V from experiment) with an active area of about 100 mm^2 was used, and a diaphragm with a radius of 3.50 mm determined the sensitive area. The edge of the diaphragm was processed into the sloping shape in order to prevent α particles from scattering into the detector from the edge.

The α particles could be scattered from the inner chamber wall and a fraction of the scattered particles may arrive at the sensitive area of the detector. In order to avoid the scattering particles, three anti-scattering baffles were installed. The anti-scattering baffle is the thin metallic plate with a central aperture situated in parallel with the source and the detector plane, and it could stop particles flying towards or scattering away from the inner chamber wall.

The sample holder is placed at the bottom. Four nuts and bolts are used to attach the tantalum backing of the $^{232}Th(OH)_4$ sample onto the holder. Besides, rubberized fabric was pasted along the edge of the sample to reinforce the attachment.

The sample-to-diaphragm distance can be changed by adjusting the position of nuts near the sample holder and the diaphragm. The distance could be measured by an electronic digital caliper, and the range of the adjusted distance is from 10 to 50 mm. For every measurement, the adjustment was very careful so that the sample and diaphragm are flat and their planes are parallel. For the present measurement, the holder-to-diaphragm distance was 30.01 mm, and the thickness of the tantalum backing was 0.11 mm. So, the sample-to-diaphragm distance was 29.90 mm.

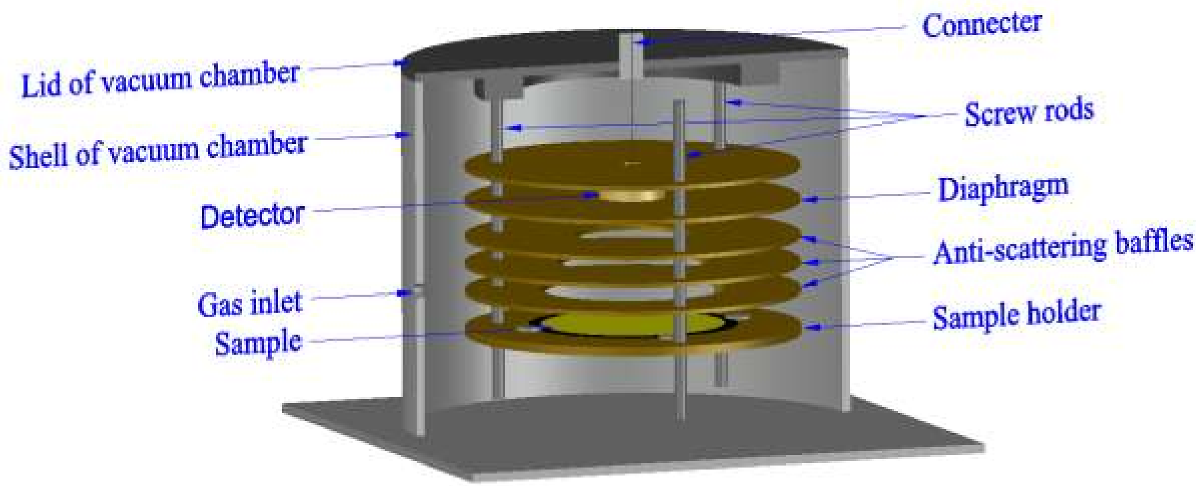

Fig. 5 (Color online) CAD drawing of the small solid angle device.

The electronic equipment and the digital data-acquisition (DAQ) system is shown in Fig. 6. A data-acquisition software was developed using the LABVIEW to control the DAQ system, to process the measurement results and to save the waveform data onto the hard disk of the personal computer (PC).

Au-Si surface barrier detector → MESYTEC MPR-1 charge integrating preamplifier → OVE-832-007 Data Acquisition Card → PC

Fig. 6 Block diagrams of the electronic equipment and the DAQ system.

The experimental process was as follows: 1) foreground measurement of α events from ^{232}Th for about 350 h, and 2) background measurement using tantalum film for about 167 h.

III. RESULTS

The measured energy spectra of α particles are shown in Fig. 7, from which, one could notice that the background is quiet weak. However, the tailing of the peaks from the daughter nuclides would increase the total counts of the ^{232}Th peak. An exponential function was used to fit the tailing and then subtracted from the ^{232}Th peak region. The proportion of the fitting counts to the total ^{232}Th peak counts is 4.81%. One fifth of the fitting counts in the region is taken as the uncertainty from the fitting method.

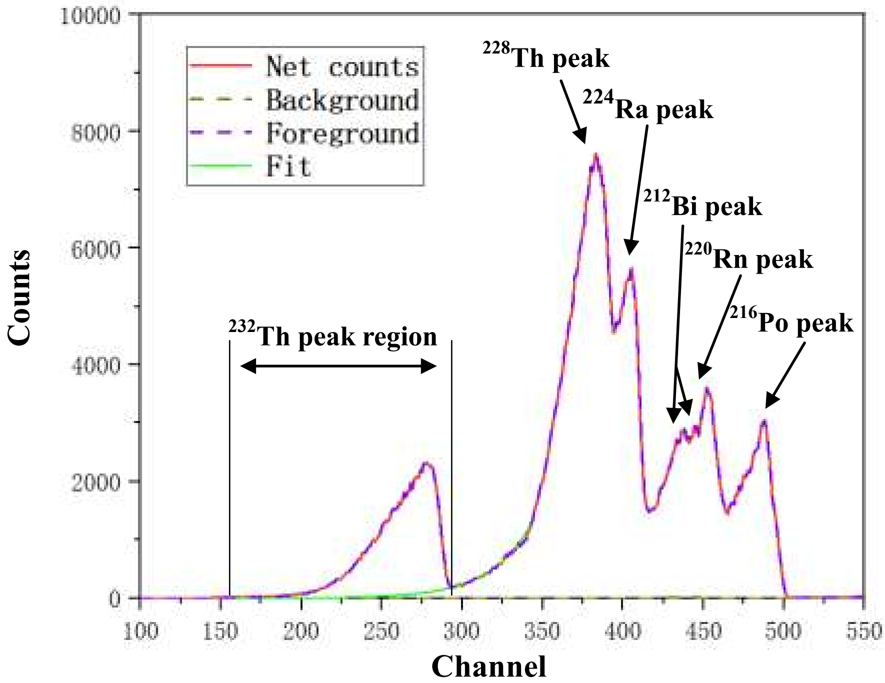

Fig. 7 (Color online) Spectra from measurements.

The number of ^{232}Th nuclei N can be calculated from the following equation:

$$N = \frac{n_{\mathrm{fore}} - n_{\mathrm{fit}} - n_{\mathrm{back}} \times \frac{t_{\mathrm{fore}}}{t_{\mathrm{back}}}}{t_{\mathrm{fore}} \times \varepsilon \times \lambda}, \qquad (1)$$

where n_{fore} and n_{back} are foreground counts and background counts in the ^{232}Th peak region. n_{fit} is the fitting counts in the ^{232}Th peak region. t_{fore} and t_{back} are the durations of foreground and background measurements. ε is the detection efficiency of the small solid angle device calculated through Monte Carlo simulation (back-scattering of α particles was taken into account, and the proportion of the back-scattering α particles to the total α particles is 0.4%), and the value of ε is 0.002460 in the present measurement. λ is the α decay constant of ^{232}Th. From equation (1), the calculated the number ^{232}Th of nuclei is 1.86×10^{19}.

Sources of the error for ε are the uncertainties of geometry (0.75%), Monte Carlo simulation (0.32%), and unevenness of the sample edge (0.30%). Sources of the error for n_{fore} include the uncertainties of ^{232}Th peak region determination (0.19%) and the statistical error (0.35%). Taken from one fifth of the fitting counts, the uncertainty of n_{fit} is 0.96%. The uncertainty of λ is 0.40% [11]. Magnitudes of other sources of errors are so small that they can be ignored. Thus, the total uncertainty of the number of ^{232}Th nuclei is 1.41%.

IV. CONCLUSIONS

In the present work, the ^{232}Th(n, f) cross-section data from EXFOR library were collected and analyzed, and it is found that the measurement results using mono-energetic neutron sources are obviously higher than those using white neutron sources. A $^{232}Th(OH)_4$ foil sample was prepared for the ^{232}Th(n, f) cross-section measurements. In order to determine the ^{232}Th nucleus number in the sample, a small solid angle device was designed and installed. The experiment was conducted, and the measured the number of ^{232}Th nuclei is $1.86\times10^{19}(1\pm1.41\%)$.

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