Measurement and analysis of the ^{238}U(n, 2n) reaction rate in depleted uranium/polyethylene shells*

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Abstract: In order to check the conceptual design of the subcritical blanket in a fusion-fission hybrid reactor, a depleted uranium/polyethylene simulation device with alternate shells has been established. The measurement of the ^{238}U(n, 2n) reaction rate was carried out using an activation technique, by measuring the 208 keV γ rays emitted from ^{237}U. The self-absorption of depleted uranium foils with different thicknesses was experimentally corrected. The distribution of the ^{238}U(n, 2n) reaction rate at 90° to the incident D^+ beam was obtained, with uncertainty between 5.3% and 6.0%. The experiment was analyzed using MCNP5 code with the ENDF/BVI library, and the calculated results are all about 5% higher than the measured results.

Key words: 14 MeV neutron, ^{238}U(n, 2n) reaction, self-absorption correction, Monte Carlo simulation

PACS: 28.20.-v, 28.41.Ak, 28.52.Av **DOI:** 10.1088/1674-1137/36/7/018

1 Introduction

A fusion-fission hybrid reactor, mainly consisting of a fusion neutron source and a subcritical blanket, is one of the key new technologies which may solve the present energy problems. The fusion reactor core generates 14 MeV neutrons by D-T reactions, and the subcritical blanket is designed to be fueled with natural uranium or spent fuel generated by PWRs (Pressurized Water Reactor) in the form of a UZr alloy, with a coolant of light water; the UZr alloy and light water are designed to be arranged alternately [1, 2].

A hybrid reactor cannot be designed without substantiating its neutron-physics characteristics. The ^{238}U(n, 2n) reaction, with a threshold energy of about 6 MeV and a cross section larger than 1 b between 7.7 MeV and 13.5 MeV, is almost the only reaction except for fission which produces neutrons. In addition, the ^{238}U(n, 2n) reaction is one of the principal means of accumulating ^{232}U, whose decay leads to the formation of ^{208}Tl, an emitter of hard γ rays which degrades the radiation environment in all stages of the fuel cycle [3].

So it is very significant to carry out ^{238}U(n, 2n) reaction rate experiments to check the conceptual design of the subcritical blanket. As polyethylene has a similar neutron moderation character to water, but is more practical in engineering, an alternate depleted uranium /polyethylene-shell simulation device has been established to carry out ^{238}U(n, 2n) reaction rate experiment.

It is common to measure the ^{238}U(n, 2n) cross section using the activation technique, by counting the 208 keV γ rays emitted from ^{237}U [3–6], however, very few papers introduce the measurement of the integral ^{238}U(n, 2n) reaction rate, with interference of the 209.75 keV γ rays produced by the ^{238}U(n, γ) reaction. V. V. Afanas'ev et al. measured the ^{238}U(n, 2n) reaction rate on a model of the blanket of a fusion reactor with a uranium neutron breeder [7]. They counted the 208 keV γ ray using a semiconductor detector, and finally obtained ^{238}U(n, 2n) reaction rate with a relative uncertainty of about 10%. The calculation was performed by the Monte Carlo method, using the BLANK code, and calculations generally differed from the measured results by about 10%.

In this paper, the measurement of the γ spectrum

Received 9 September 2011
* Supported by Chinese Special Project for ITER (2010GB111002)
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was begun 18 d after the end of irradiation to eliminate the interference of the 209.75 keV γ ray, and finally the ^{238}U(n, 2n) reaction rate distribution at 90° to incident D^+ beam was obtained, with uncertainty between 5.3% and 6.0%, much smaller than that of V. V. Afanas'ev's results. Among the highlights of the experiment are the self-absorption correction and counting efficiency determination in conjunction with a ^{243}Am foil with the same radius as activated foils. In addition, the measured ^{238}U(n, 2n) reaction rate distribution was compared with that calculated by MCNP5 code [8] using the ENDF/BVI nuclear data library, for checking the accuracy of the related simulation program and data library.

2 The experimental method

In this experiment, the ^{238}U(n, 2n) reaction rate is obtained using the foil activation technique. The depleted uranium foils were irradiated in the neutron flux $\phi(s^{-1})$ for a period of time T. After irradiation, the foils were cooled for a period of time τ before being counted with an HPGe spectrometer for a time t. The relationship between the 208 keV γ ray peak counts N_γ and the ^{238}U(n, 2n)^{237}U reaction rate R (reaction per source neutron per ^{238}U atom) is given by Eq. (1):

$$N_\gamma = N_8 \cdot \phi \cdot R \cdot b \cdot \eta \cdot \frac{1}{\lambda}(1-e^{-\lambda T})e^{-\lambda \tau}(1-e^{-\lambda t}), \quad (1)$$

where N_8 is the ^{238}U atom number of the activated foil, b is the 208 keV γ ray branching ratio of ^{237}U, η is the counting efficiency of HPGe γ spectrometer to 208 keV γ rays, and λ is the decay constant of ^{237}U.

As the neutron flux changes during the irradiation and uranium absorbs γ ray severely in the foils, the correction factor K for the variation of the flux ϕ and $A(d)$ for the self-absorption of the foil thickness d(cm) were brought in, which will be introduced later, then Eq. (1) changes to Eq. (2):

$$R = N_\gamma \cdot \lambda / [N_8 \cdot \overline{\phi} \cdot b \cdot \eta \cdot K \cdot A(d) \cdot (1-e^{-\lambda T})e^{-\lambda \tau}(1-e^{-\lambda t})], \quad (2)$$

where $\overline{\phi}$ is the average neutron flux (s^{-1}).

3 The experimental setup

The alternate shells are composed of 3 depleted uranium (99.58% ^{238}U, ρ=18.8 g/cm^3) shells and 3 polyethylene (ρ=0.95 g/cm^3) shells, with radius of 13.1, 18.1, 19.4, 23.35, 25.4, 30.0, 35.0 cm from inside to outside. The D beam gets through the drift tube, and bombards the tritium target located at the center. Depleted uranium foils are located in the vertical measuring canal, fixed with depleted uranium and polyethylene blocks and kept identical with materials outside the canal. The apparatus and foil-arrangements are shown in Fig. 1.

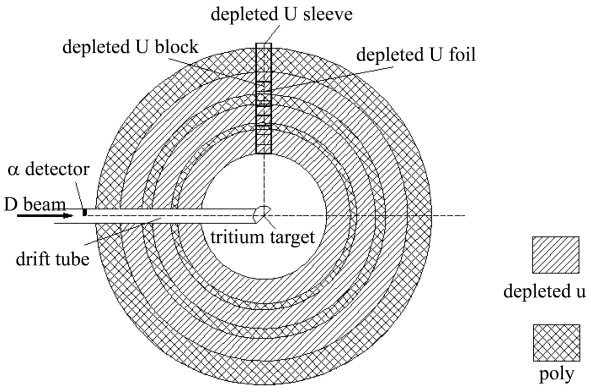

Fig. 1. Schematic view of the experimental setup.

γ rays were measured with an HPGe spectrometer consisting of an ORTEC GEM60P detector with a sensitive volume of 250 cm^3 and an ORTEC GAMMAVISION analyzer. The system has an energy resolution of 1.87 keV and a relative efficiency of 60% at 1.33 MeV. In this paper, γ sources are all placed at a distance of 2.560 cm away from the surface of detector.

4 The experimental procedure

4.1 Irradiation with D-T neutrons

The depleted uranium samples, with a radius of 1.2 cm and a thickness of 0.015–0.019 cm, has the same isotopic abundance and density as the depleted uranium shells. The samples were irradiated on the PD-300 accelerator neutron generator for about 7 h, with an average neutron yield of $(1.52\pm0.038)\times10^{10}$ s^{-1}. Neutrons were produced by T(d, n)^{4}He reactions with a D^+ beam with an average energy of 134 keV. During the irradiation, the neutron yield was measured with the associated α particle method. An Au-Si surface barrier detector was installed at 178° to incident D^+ beam, giving out associated α particle counts ($N_\alpha(i)$) for a series (n) of time intervals $\Delta\tau(n\Delta\tau = T)$, and the time interval was set to be 10 s. Neutron yield is proportional to the α particle counts with a relationship of $\Phi_n = k_0 N_\alpha$. The parameter k_0 is determined by the radius of the Au-Si detector, the distance of the detector from the tritium target and the anisotropy correction [9], and k_0 is calculated to be 1.313×10^7

in this experiment. The correction factor K for the variation of the neutron flux is given by Eq. (3):

$$K = n\cdot\frac{e^{\lambda\Delta\tau}-1}{1-e^{-\lambda T}}\cdot\sum_{i=1}^{n}N_\alpha(i)\cdot e^{-\lambda(T-i\Delta\tau)}/\sum_{i=1}^{n}N_\alpha(i), \quad (3)$$

and this correction is found not to exceed 0.1%.

4.2 The counting efficiency determination and self-absorption correction

The counting efficiency of the HPGe detector and self-absorption correction were determined in conjunction with a ^{243}Am foil with the same radius as the activated foils in this paper, as the ^{239}Np (α decay product of ^{243}Am) emits 209.75 keV γ rays with a (3.42±0.05)% [10] probability which have a similar energy as the 208 keV γ rays. The α-decay rate of the ^{243}Am foil can be precisely measured using low-geometry α-counter, and by using the ^{243}Am foil, complicated point-surface transition of point γ sources can be avoided.

The HPGe detector has nearly the same counting efficiency for these two γ rays (differing within 0.5% from interpolation of the counting efficiency curve measured with different γ sources). A counting efficiency of η=0.063±0.0015 was obtained of the HPGe detector to the 208 keV γ ray at a distance of 2.560 cm.

Uranium absorbs these two γ rays similarly (total cross section of 450.7 b [11] for the 209.75 keV γ ray and 459.8 b [11] for the 208 keV γ ray). Counts of γ rays emitted from the back surface of foils decrease exponentially with the thickness (d) of foils. ^{243}Am foil was placed upon depleted uranium foils of a different thickness to measure this exponential decay. The measured and fitted results are shown in Fig. 2, and the fitted normalized emitting ratio function $k'(d)$ of 209.75 keV γ rays is given by Eq. (4),

$$k'(d) = a_0\exp(-a_1 d) = (1.002\pm0.028)e^{-(24.51\pm1.81)d}, \quad (4)$$

where a_0 and a_1 are fitted parameters.

The normalized emitting ratio function $k(d)$ of 208 keV γ rays can be calculated as $k(d) = a_0\exp(-459.8a_1d/450.8)$. As each depleted uranium foil is homogeneously activated in the setup, the self-absorption correction factor $A(d)$ can be expressed as Eq. (5):

$$A(d) = \frac{1}{d}\int_0^d k(x)\mathrm{d}x. \quad (5)$$

The self-absorption correction was simulated by MCNP5 with the ENDF/BVI.8 photoatomic data library, and compared with the experimental results as shown in Fig. 3. From the figure, it can be seen that the calculated absorption correction for the 208 keV γ ray is 1.4% lower than the experimental value when the thickness is 0.02 cm, which may be caused by the non-uniformity and curl of the uranium foils.

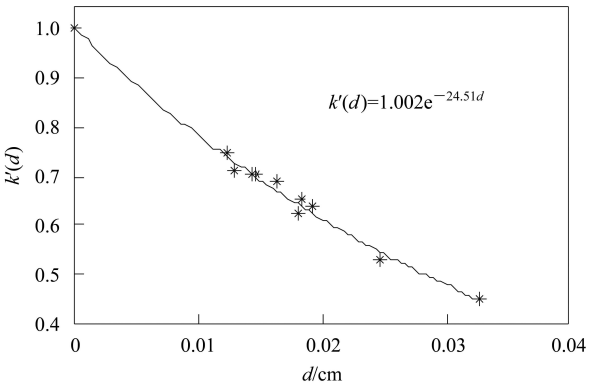

Fig. 2. The measured and fitted results of exponential decay.

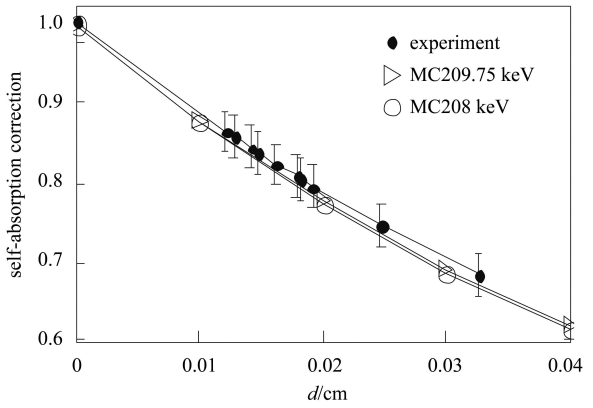

Fig. 3. Compasion of the measured and simulated self-absorption correction.

4.3 Counting the 208 keV γ ray peak

The 208 keV transition occurs with (21.2±0.3)% [12] probability per ^{237}U decay with a half-life of 6.75 d, however, the product nucleus ^{239}Np formed in the neutron capture reaction of ^{238}U emits 209.75 keV γ rays with a half-life of 2.355 d, which causes interference to the counts of the 208 keV γ rays. A cooling period of τ >18 d would eliminate this interference as about 99.5% of the ^{239}Np decays. γ spectrums of an activated foil measured after cooling of 1 d and 18 d are shown in Fig. 4. 208 keV γ ray peak counts were obtained from γ spectrums measured after a cooling time of 18 d.

4.4 Result

As the ^{238}U(n, 2n) reaction rate decreases greatly with the increase in distance to the neutron source, it is difficult to separate the 208 keV γ-ray of foils from

the background in the positions far from the neutron source, only the ^{238}U(n, 2n) reaction rate at 4 positions in the 1st depleted uranium shell was measured, as shown in Table 1, and the relative uncertainty analysis is listed in Table 2.

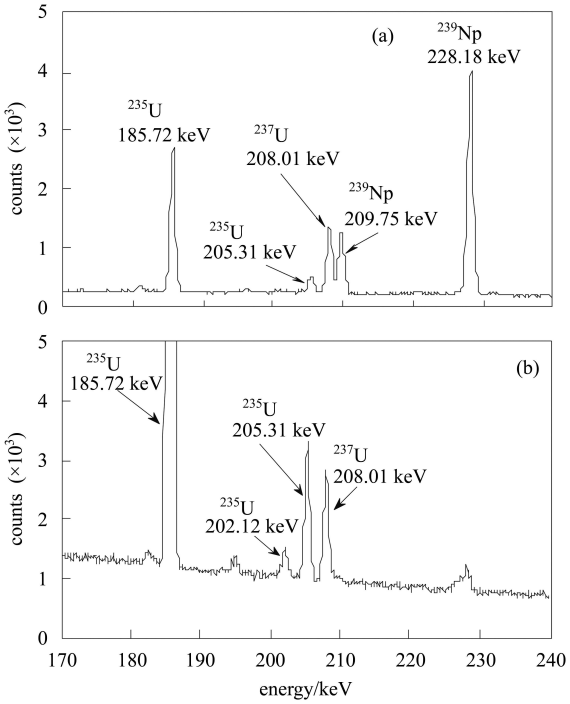

Fig. 4. The γ spectrum of an activated foil measured by an HPGe spectrometer. (a) γ spectrum measured 1 day after irradiation; (b) γ spectrum measured 18 days after irradiation.

Table 1. The measured ^{238}U(n, 2n) reaction rate.

distance from target/cm	^{238}U(n, 2n) reaction rate	uncertainty(%)
13.03	3.92E-28	5.3
13.56	3.51E-28	5.4
15.59	2.09E-28	5.6
17.63	1.30E-28	6.0

Table 2. Analysis of relative standard uncertainty (%).

parameter	value
counting statistics	1.8–3.4
counting efficiency	2.3
self-absorption correction	3.2
neutron flux	2.5
^{238}U atom number	0.5
branching ratio	1.4

5 Analysis and discussion

The ^{238}U(n, 2n) reaction rate was obtained as a function of position in the setup, analysis of the experiment was carried out using MCNP5 with the ENDF/BVI nuclear data library, which is useful for checking the accuracy of the cross section library and assessing the reliability of the conceptual design.

The detailed configuration of all 6 shells was incorporated into the calculation model. The hall, D beam drift tube, tritium target, measuring canal at 90° direction, depleted uranium foils and blocks were considered. The angular dependences of the source neutron energy and intensity were also considered, which can be calculated by "DROSG-2000" codes [13]. However, the accelerator modules and support base of the shells were not considered, the effects of these neglects on the calculated reaction rate would be very small because the reaction rate was measured in the 1st shell of the setup.

The calculated reaction rate is compared with the measured reaction rate in Fig. 5. As is shown in Fig. 5, the difference between the calculations and the experiments is about 5%, much lower than V. V. Afanas'ev's results, indicating that the cross sections have been greatly improved. However, in our experiment, the calculations are all higher than the experimental results, which are still unsatisfactory.

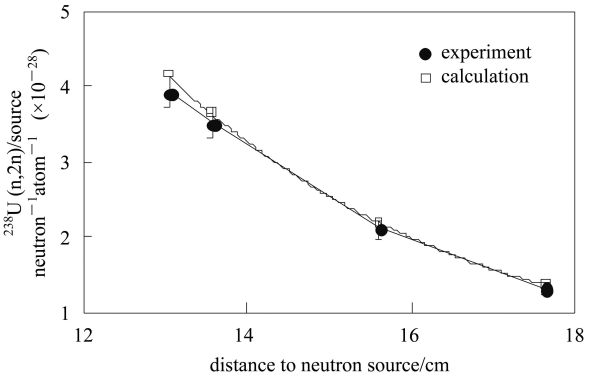

Fig. 5. Comparison of the measured and calculated ^{238}U(n, 2n) reaction rate.

The reaction rate is the integral of the scalar neutron energy spectrum multiplied by the energy dependent reaction cross section over the whole energy interval, as $R = \int \sigma(E)\phi(E)\mathrm{d}E$. In order to find out why the calculations exceed the experimental results, the fractions of the reaction rate at the 1st shell in some energy intervals were calculated with MCNP5, as shown in Fig. 6. The sum of the fractions is normalized to 100%. It is understood from the figure that: the ^{238}U(n, 2n) reaction is mainly(>80%) caused by 13 to 14.1 MeV neutrons. Since the uncertainty of the ^{238}U(n, 2n) reaction cross section is

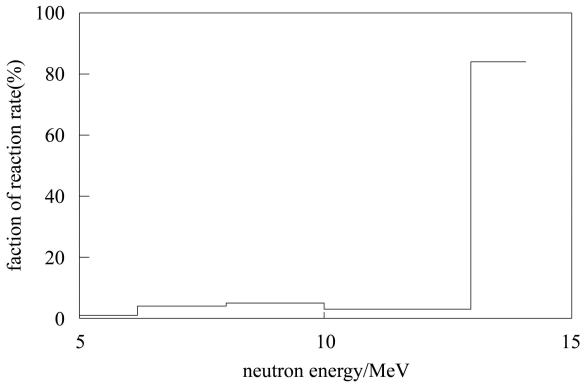

Fig. 6. Fraction of the energy-wise ^{238}U(n, 2n) reaction rate in the 1st depleted Uranium shell.

within 3% for neutrons at 13 and 14.1 MeV, the discrepancy of about 5% may be caused by large uncertainty of the measured reaction rate or a higher neutron flux calculation as a result of higher inelastic scattering of polyethylene or uranium, which should be verified by further experiments.

6 Conclusion

A ^{238}U(n, 2n) reaction rate experiment was carried out on the new-established alternate polyethylene/uranium shells using an activation technique. The ^{238}U(n, 2n) reaction rate distribution was obtained at a 90° direction to the incident D beam, with uncertainty between 5.3% and 6.0%. The results are useful for checking the accuracy of the cross section library and substantiating neutron-physics characteristics of the subcritical reactor. The analysis of the experiment by MCNP5 employing the ENDF/BVI library has revealed an overestimate of about 5% of the ^{238}U(n, 2n) reaction rate, indicating the necessity of further experiments.

We would like to express our sincere thanks to the Accelerator Department of INPC (Institute of Nuclear Physics and Chemistry) for its great efforts to assure the success of the experiment.

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