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A study of ^{239}Pu production rate in a water cooled natural uranium blanket mock-up of a fusion–fission hybrid reactor

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Abstract
The ^{239}Pu production rate is important data in neutronics design for a natural uranium blanket of a fusion–fission hybrid reactor, and the accuracy and reliability should be validated by integral experiments. The distribution of ^{239}Pu production rates in a subcritical natural uranium blanket mock-up was obtained for the first time with a D-T neutron generator by using an activation technique. Natural uranium foils were placed in different spatial locations of the mock-up, the counts of 277.6 keV γ-rays emitted from ^{239}Np generated by ^{238}U capture reaction were measured by an HPGe γ spectrometer, and the self-absorption of natural uranium foils was corrected. The experiment was analyzed using the Super Monte Carlo neutron transport code SuperMC2.0 with recent nuclear data of ^{238}U from the ENDF/B-VII.0, ENDF/B-VII.1, JENDL-4.0u2, JEFF-3.2 and CENDL-3.1 libraries. Calculation results with the JEFF-3.2 library agree with the experimental ones best, and they agree within the experimental uncertainty in general with the average ratios of calculation results to experimental results (C/E) in the range of 0.93 to 1.01.

Keywords: D-T neutrons, ^{239}Pu production rate, natural uranium blanket mock-up, integral neutronics experiment

(Some figures may appear in colour only in the online journal)

1. Introduction

Hybrid reactors have been considered for decades to be potential tools for exploiting natural nuclear resources in an optimized way [1]. Fusion–fission hybrid reactors have the potential attractiveness of easing the requirement of fusion plasma technology with a low fusion gain Q, and plasma-facing material technology with a low neutron wall loading. As they have the advantage of using spent fuel from pressurized water reactors (PWRs) or natural uranium (NU), subcritical systems without critical safety issues and nuclear nonproliferation, fusion–fission hybrid reactors have attracted comprehensive attention and research [2, 3] and many conceptual designs have been presented [4–8].

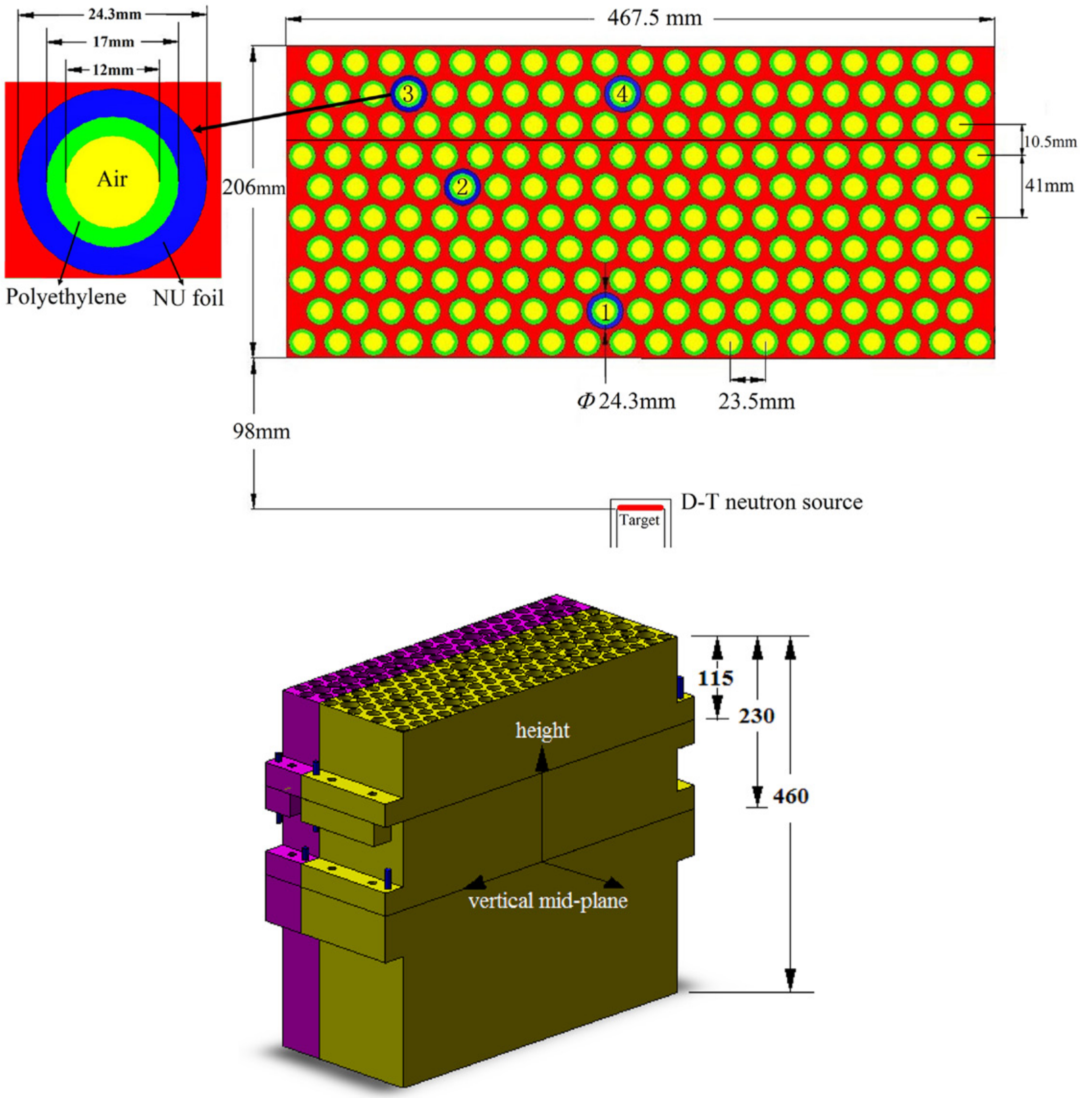

Figure 1. Schematic view of the natural uranium blanket mock-up.

The fusion–fission hybrid energy reactor (FFHER), which was proposed and designed at the China Academy of Engineering Physics (CAEP), is one of the planned advanced reactors applying fusion technologies to solve the present energy crisis [9, 10]. The feasibility of the FFHER design depends on the reliability of the modeling program and relevant nuclear data libraries, and some benchmarks and blanket mockups must be established to carry out corresponding integral neutronics experiments. In the conceptual design of the FFHER proposed in China, the subcritical blanket is designed to be fueled with UZr alloys of natural uranium [9] or spent fuel generated by PWRs [4] or thorium [11] with a coolant of light water. The ^{239}Pu, acting in the role of nuclear fuel in the FFHER blanket conceptual design, is generated from the ^{238}U(n, γ)^{239}U reaction followed by successive two beta decays in a natural uranium (or spent fuel unloaded from PWRs) blanket. The reliability of the breeding performance of ^{239}Pu in the conceptual design depends on the prediction accuracy of the calculation of ^{238}U(n, γ) reaction rates in the subcritical blanket.

There have been extensive numerical studies in the field of fusion–fission (hybrid) reactors throughout the world [12, 13]. Aiming at assessing the conceptual design of the FFHER subcritical blanket, a series of benchmarks with depleted uranium and polyethylene shells was established by our group to carry out integral experiments [14, 15]. Yan *et al* [14] measured the distribution of ^{238}U(n, γ) reaction rates in some alternate depleted uranium/polyethylene shells with an experimental uncertainty of 3.5–3.7%. The simulations and measurements agree within 5% for the ^{238}U (n, γ) reaction rate and within 1% for the total neutron capture rate of ^{238}U using the MCNP code [16] with the ENDF/B-VI.8 libraries. Yang *et al* [17] measured the distribution of ^{238}U(n, γ) reaction rates in a metallic depleted uranium shell with 10.6 cm wall thickness with a T(d, n) source. The experiment was simulated using the MCNP code with the ENDF/B-VI.8 library, and the calculation-to-experiment ratios are 0.972–1.034 for ^{238}U (n, γ) reaction rates. In order to provide support for the feasibility research in the subcritical blanket conceptual design, more integral experiments based on some blanket mock-ups should

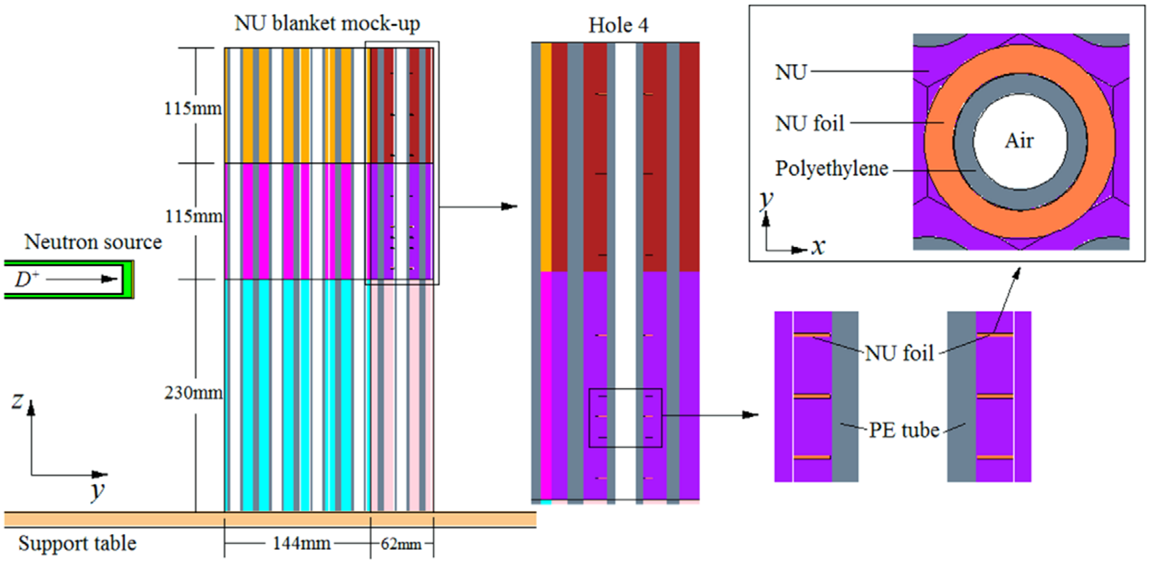

Figure 2. Cross-sectional view of the model of the experiment.

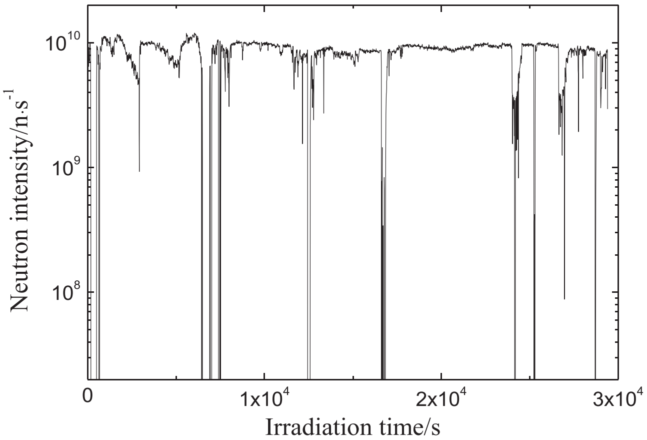

Figure 3. Time dependence of the D-T neutron intensity.

be carried out. For the present study, the neutronics experiments have been extended to include blanket materials and coolant to replicate the blanket structure in a mock-up. Similar experiments have previously been performed by European groups on mock-ups of tritium producing fusion reactor blankets [18–20]. Therefore, some fusion–fission blanket mock-ups should be established and ^{239}Pu production rates can be obtained in more detail. In addition, no experimental studies have been reported so far on a water cooled natural uranium blanket mock-up. To this end, a natural uranium mock-up, based on the materials, structure and characteristics of neutron transportation and energy amplification in the conceptual design, was established by CAEP.

Apparently, our group was the first to measure the distribution of ^{239}Pu production rates in a water cooled natural uranium blanket mock-up of a fusion–fission hybrid reactor with a D-T neutron generator. Annular natural uranium foils were placed in different spatial locations of the blanket mock-up, the counts of 277. 6 keV γ rays emitted from ^{239}Np generated by ^{238}U capture reaction was measured by a HPGe γ spectrometer, the self-absorption of natural uranium foils was corrected and the detection efficiency of the annularity natural uranium foils was calibrated. The distribution of ^{239}Pu production rates in four holes of the mock-up was obtained and the experimental uncertainty was estimated. Experimental results were compared with simulated results by the SuperMC code [21, 22] with ENDF/B-VII.0 [23], ENDF/B-VII.1 [24], JENDL-4.0 [25, 26], JEFF-3.2 [27] and CENDL-3.1 [28] libraries, and a discussion of the results is also presented.

2. Experiment

2.1. Method

^{239}Pu production rates were obtained using the neutron activation method in the experiment. The ^{239}Pu is generated from the ^{238}U$(n,\ \gamma)^{239}$U reaction followed by two successive β decays, and the schematic diagram is shown below.

$$^{238}\mathrm{U} \xrightarrow{(n,\gamma)} {}^{239}\mathrm{U} \xrightarrow[\beta\ \mathrm{decay}]{T_{1/2}=23.54\ \mathrm{min}} {}^{239}\mathrm{Np} \xrightarrow[\beta\ \mathrm{decay}]{T_{1/2}=2.355\,\mathrm{d}} {}^{239}\mathrm{Pu}$$

The half life of ^{239}Pu is about 24110 years and the nuclide can be regarded as relatively stable. ^{239}Np emitted 277.6 keV γ rays with a half life of about 2.335 d. The ^{239}Pu production rate R, defined as the reaction probability normalized for a source neutron and a ^{238}U atom in foils in the experiment, can be determined by counting the number of 277.6 keV γ rays. The natural uranium foils were irradiated in the neutron flux Φ (s^{-1}) for a period of time T_i. After irradiation, the foils were cooled for a period of time T_c before being counted with a HPGe spectrometer for a time T_m. The relationship between the 277.6 keV γ ray peak counts n and ^{239}Pu production rate R is given by equation (1) [17]:

$$R = \frac{n}{N \cdot \Phi \cdot \varepsilon \cdot \eta \cdot F}, \tag{1}$$

where N is the number of ^{238}U atoms in the activated foil, ε is the 277.6 keV γ ray branching ratio of ^{239}Np decay, η is the

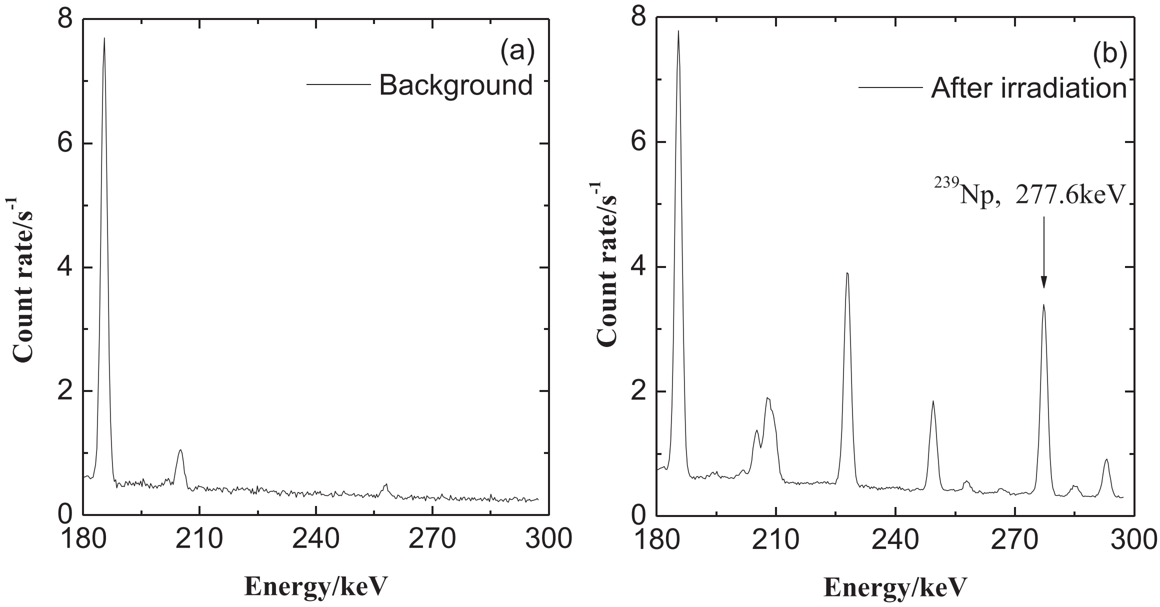

Figure 4. The recorded γ spectrum of background and irradiated NU foils, (a) the recorded background spectrum of a NU foil; (b) the gamma spectrum of a irradiated NU foil after a cooling time of more than 10 h.

Table 1. Analysis of relative standard uncertainty (%).

Neutron flux	Detection efficiency	Self-absorption correction	Branching ratio	^{238}U atom number	Counting statistics
2.5	3.4	2	0.69	0.5	0.4–4.1

detection efficiency of the HPGe γ spectrometer to 277.6 keV γ rays, and F is the time factor, which can be derived by equation (2):

$$F = \frac{\lambda_1}{\lambda_2(\lambda_1 - \lambda_2)}(1 - e^{-\lambda_2 T_i})e^{-\lambda_2 T_c}(1 - e^{-\lambda_2 T_m}) + \frac{\lambda_2}{\lambda_1(\lambda_2 - \lambda_1)}(1 - e^{-\lambda_1 T_i})e^{-\lambda_1 T_c}(1 - e^{-\lambda_1 T_m}), \quad (2)$$

where λ_1 is the decay constant of ^{239}U and λ_2 is the decay constant of ^{239}Np.

As the neutron flux changes during the irradiation and uranium absorbs γ rays severely in the foils, the correction factor K for variation of the flux Φ and $A(d)$ for self-absorption of foil thickness d (cm), which were introduced by Feng *et al* [11] in detail, were brought into the experiment. Equation (1) then changes to equation (3):

$$R = \frac{n \cdot K}{N \cdot \overline{\Phi} \cdot \varepsilon \cdot \eta \cdot F \cdot A(d)}, \quad (3)$$

where $\overline{\Phi}$ is the average neutron flux (s^{-1}) and equals to $\Phi \cdot K$.

2.2. Natural uranium blanket mock-up

According to the design of the FFHER subcritical blanket, energy amplification was mainly supplied by ^{235}U, ^{238}U and ^{239}Pu. In order to reflect the ^{239}Pu production rates in the subcritical blanket accurately, a natural uranium blanket mock-up was designed by the China Academy of Engineering Physics. Polyethylene (PE), which is easy to process and has

Table 2. The distribution of ^{239}Pu production rate R ($\times 10^{-29}$ atom^{-1} n^{-1}) and uncertainty (%).

	Hole 1		Hole 2		Hole 3		Hole 4	
Height/cm	R	Uncertainty	R	Uncertainty	R	Uncertainty	R	Uncertainty
0	43.1	4.8	33.7	4.8	14.9	5.3	21.4	4.8
1	46.0	4.8	33.3	4.8	12.6	5.1	25.3	4.8
3	51.5	4.8	37.6	4.8	13.9	5.4	27.2	4.8
4	48.9	4.8	36.3	4.8	17.4	5.2	23.0	4.8
5	46.8	4.8	36.1	4.8	14.0	5.0	24.9	4.9
8	41.3	4.8	30.9	4.8	12.4	5.1	22.7	4.8
12	32.8	4.8	26.6	4.8	10.2	5.5	18.4	4.8
16	23.6	4.8	19.7	4.8	8.5	6.3	12.0	4.8
20	13.0	4.9	10.8	4.9	4.9	6.0	7.0	4.9

a similar feature to water in moderation of neutrons, was used in the mock-up with different volume to control the ratio of natural uranium to water in the experiment. A schematic diagram of the natural uranium blanket mock-up is shown in figure 1.

The mock-up with a lattice cell structure consists of 6 natural uranium (NU) blocks and 390 PE hollow pipes with outer dimensions of 467.5 mm (length) $\times$ 460 mm (height) $\times$ 206 mm (thickness). ^{238}U abundance in the six NU blocks was determined to be between 99.291% and 99.347%. Figure 2 shows a cross-sectional view of the model of the experimental assembly.

36 annular NU foils (^{238}U: 99.319%, ^{235}U: 0.675%) were placed at 0, 10, 30, 40, 50, 80, 120, 160 and 200 mm from

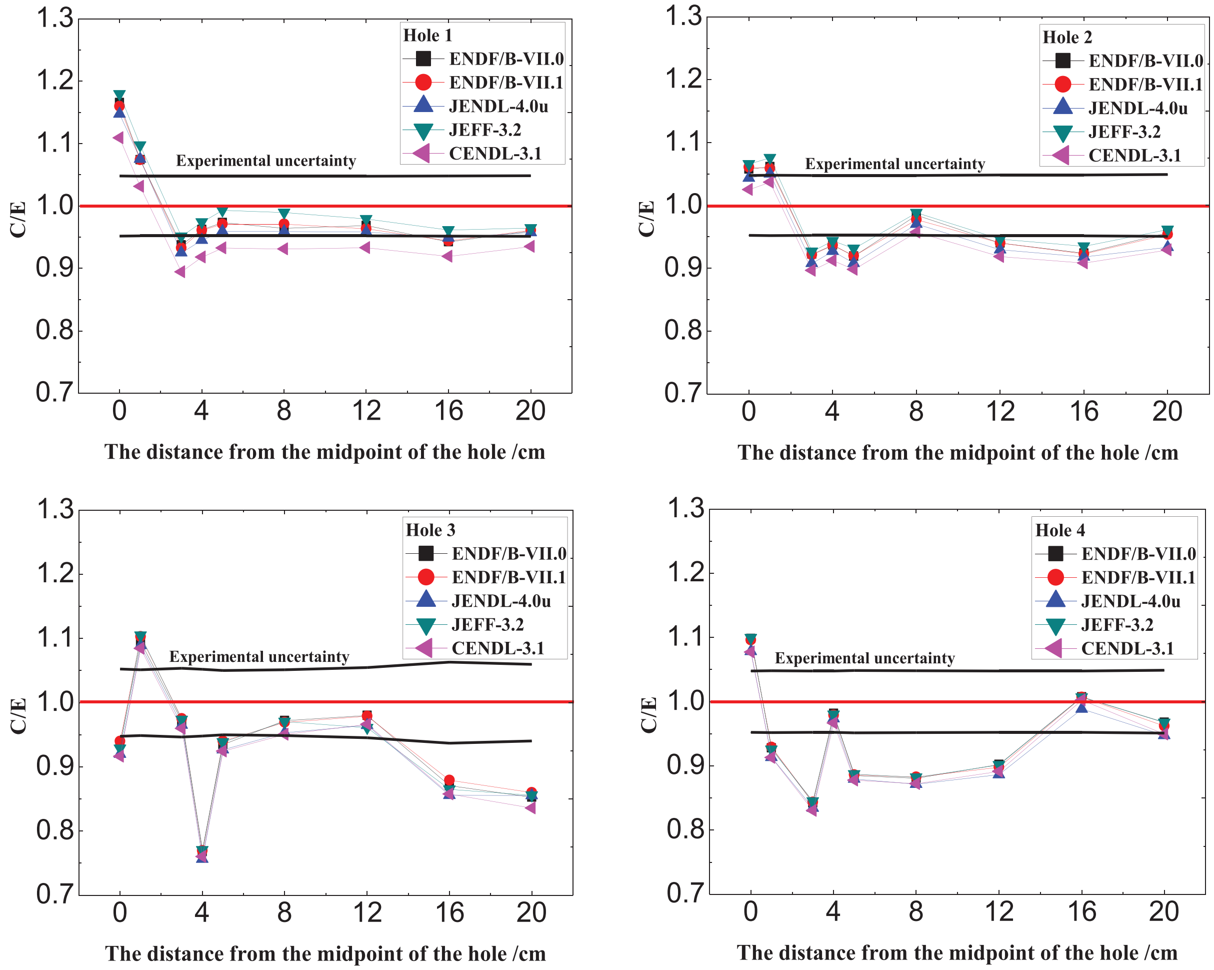

Figure 5. C/E comparison for the ^{239}Pu production rate measurements of the four holes inside the NU mock-up.

Table 3. The average C/E of the four holes for the five evaluated libraries.

ENDF/ B-VII.0	ENDF/ B-VII.1	JENDL-4.0u	JEFF-3.2	CENDL-3.1
0.959	0.959	0.948	0.965	0.937

the vertical mid-plane shown in figure 1 for the measurements of ^{239}Pu production rates. The size of the activation foils was 17 mm in inner diameter and 24.3 mm in external diameter with thickness varying between 0.34 mm and 0.46 mm. 390 PE hollow pipes with an inner diameter of 12 mm and an external diameter of 17 mm were placed in the 195 holes to meet the volume ratio of NU to water (2:1) in the design [29]. The length of each PE hollow pipe is 230 mm and the volume ratio of NU to PE is about 2.28:1 in the experiment. As the D-T neutron source is placed at the axis of the mock-up, the neutron property in a 1/4 quadrant of the mock-up is the same as one of the other 1/4 quadrants of the mock-up and the measuring region is selected at the 1/4 quadrant of the mock-up [30].

2.3. ^{239}Pu production rate measurements

The experiment was performed on the PD-300 D-T neutron generator at the Institute of Nuclear Physics and Chemistry, CAEP, and natural uranium foils were irradiated for about 7 h. The distance between the T-Ti target and the front surface of the mock-up was 98 mm. Neutrons were produced by $T(d, n)^4$He reactions with a D^+ beam current of 200 μA and an average energy of 225 keV on the tritium-titanium target (370 GBq). An Au-Si surface barrier detector positioned at 178.2° with respect to the D^+ beam was used to monitor the neutron yield by counting the associated α particles in every 10 s [31, 32]. Figure 3 shows the time dependence of the D-T neutron intensity.

γ rays emitted from NU activation foils were measured by 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 [32]. To obtain acceptable counting statistics, the activated foils were all placed on the surface of the detector aluminum cap during the measurement.

The SuperMC code based on the Monte Carlo method was applied to calibrate the efficiency for an annularity γ source [33]. A series of standard point γ sources, ^{60}Co, ^{133}Ba, ^{137}Cs, ^{152}Eu and ^{241}Am, were used to measure the detection efficiency curve at the axis of the HPGe detector with a source-detector distance of 6 cm. Measured results and calculation results in SuperMC were fit well through adjusting the

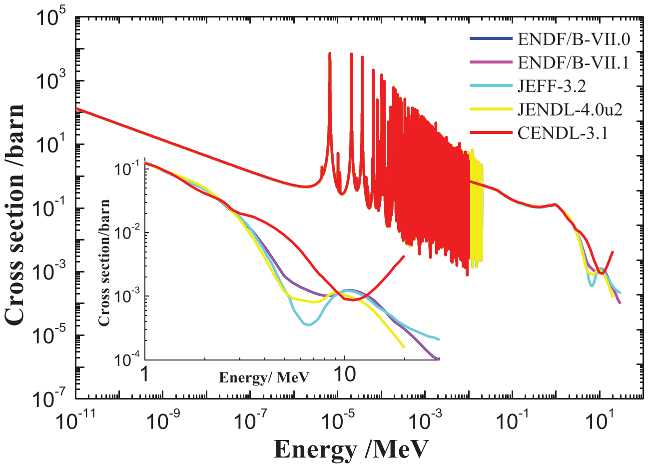

Figure 6. Five evaluated nuclear libraries of ^{238}U(n, γ).

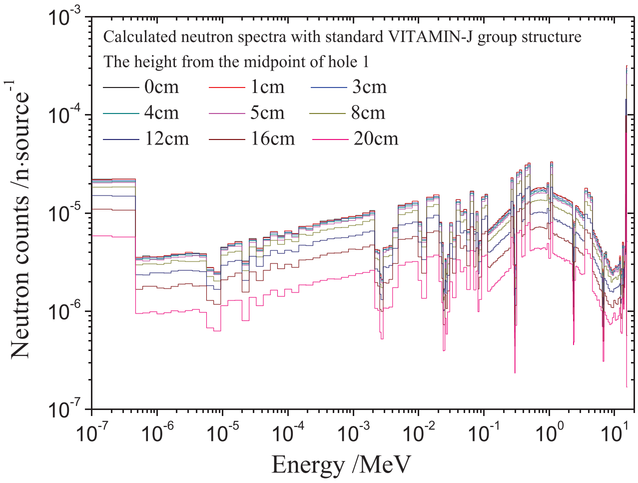

Figure 7. Calculated neutron spectra for the nine NU foils in hole 1.

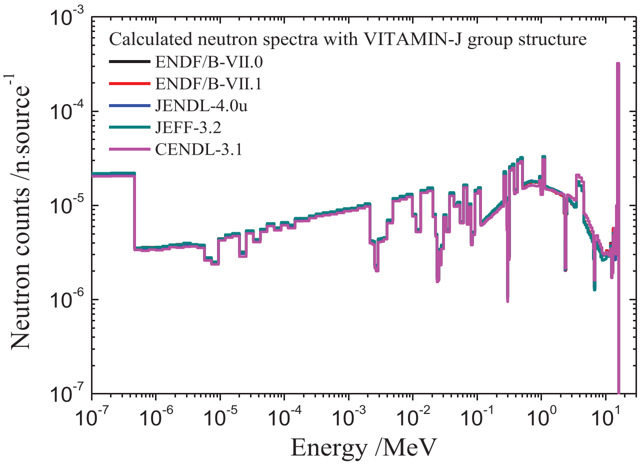

Figure 8. Calculated neutron spectra for the NU foil at 1 cm in hole 1 with five evaluated libraries.

thickness of the dead layer and the diameter of the inner hole of the HPGe detector in the SuperMC model. The calibration method was validated by comparing calculated and experimental full energy peak efficiencies in 277.6 keV using a surface source ^{243}Am (788 Bq) with a diameter of 24 mm. The detection efficiency of the annularity NU foils for the 277.6 keV γ ray was determined to be 11.1% in the experiment.

NU foils placed on the surface of the HPGe detector were measured before being irradiated, and no γ ray peak at 277.6 keV was observed in the background spectrum. The recorded background spectrum of a NU foil is shown in figure 4(a). Figure 4(b) shows the measured γ spectrum of the irradiated NU foil after a cooling time of more than 10 h to ensure complete decay of ^{239}U to ^{239}Np. The counts of the γ ray peak at 277.6 keV shown in figure 4(b) were obtained by an ORTEC Gammavision analyzer with an uncertainty of 0.4–4.1%. With the cooling time and the measured time in the HPGe detector, time factor F was calculated with equation (2) for different foils varying between 72.2 s and 2419.4 s.

Our previous work [14, 34] showed that the discrepancy between the simulated and measured self-absorption correction factor for the 277.6 keV γ ray was lower than 1.0% even when the thickness of the depleted foil was 0.5 mm, and that the discrepancy was smaller when the foil was thinner. Consequently, self-absorption corrections $A(d)$ in this experiment were calculated by the SuperMC code employing the ENDFB-VI.8 photoatomic data library and determined to be between 0.769 and 0.824 for 277.6 keV γ rays.

2.4. Results and uncertainty analysis

The reaction rate was deduced from the measured activity by performing the appropriate corrections, which include fluctuations of the neutron flux during irradiation, detection efficiency, self-absorption of γ rays in the NU foils, counting statistics and cited value of the branching ratio. The principal sources of uncertainties associated with the cross sections and their estimated values are given in table 1.

The uncertainties were assumed to be uncorrelated, and the total uncertainties in the reaction rate values could be determined by adding the experimental errors and the uncertainties of nuclear data in quadratic form. The distribution of ^{239}Pu production rates measured based on equation (3) in the four selected holes were listed in table 2.

3. Detailed Monte Carlo simulations and C/E comparison

The experiment has been accurately simulated using the Monte Carlo code SuperMC2.0 with the following nuclear data libraries: ENDF/B-VII.0, ENDF/B-VII.1, JENDL-4.0, JEFF-3.2 and CENDL-3.1. The angle dependences of the D-T source neutron energy and intensity were calculated by the DROSG-2000 code [35]. A great effort was made to render the SuperMC geometry as accurately as possible, including the target chamber, different abundances of ^{238}U in the six NU components, the experimental hall and some related components. The ^{239}Pu production rates were calculated at the measurement positions by using a track-length (F4) tally and tally multiplier card (FMn). Mesh-based windows, a WWP

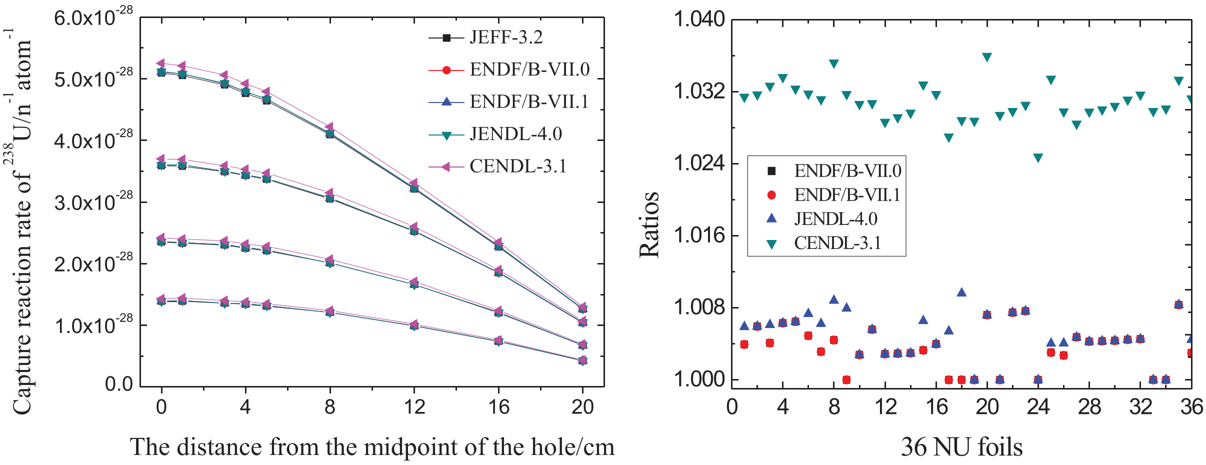

Figure 9. The difference between calculations with the five libraries.

card and WWG card were used to reduce the variance for the complicated mock-up and neutron spectrum. A run of 1×10^9 neutron histories was performed with statistical uncertainties at 1σ lower than 1%. Different discrepancies between the experiment and different evaluated libraries (C/E, the ratio of calculations to experimental results) for the four holes were shown in figure 5.

The average values of C/E of the four holes for the five evaluated libraries in the mock-up were listed in table 3. The calculated results with JEFF-3.2 agree with the measured results the best and agree within the experimental uncertainty in general. The average values of C/E for the four holes are 1.01, 0.975, 0.930 and 0.944, respectively. As hole 3 and hole 4 were far away from the neutron source, the ^{239}Pu production rates in these holes were decreased for the reduced neutrons on the NU foils. Consequently, the statistical fluctuations in experiments or calculations were increased remarkably, and it resulted in the discrepancy between calculations and experiments. On the other hand, the discrepancy would be caused by the neutron spectrum and cross sections.

4. Discussion

The reaction rate is the integral of the neutron energy spectrum multiplied by the energy dependent reaction cross section over the whole energy interval, as shown by $R = \int \sigma(E)\phi(E)\mathrm{d}E$. The cross sections of ^{238}U(n, γ) in the five evaluated libraries were shown in figure 6, and there is some difference in the resonance energy region and continuum energy region. In order to analyze the discrepancy between calculations and experimental results deeply, the neutron spectra on the NU foils with standard group structure VITAMIN-J (175) were calculated with the SuperMC code and the five evaluated libraries.

The scalar neutron energy spectra for the nine NU foils in hole 1 were calculated with the SuperMC code and evaluated library JEFF-3.2. Track-length tally (F4) and E4 card (tally energy) were used and the calculated neutron spectra are shown in figure 7. Neutrons above 10 MeV occupied a great share for each of the foils (17.4–21.1% for hole 1), and some difference in that energy bin was obviously in the five evaluated libraries, as shown in figure 6. For the NU foil at 1 cm in the hole 1, the five evaluated libraries were used to calculate the scalar neutron energy spectrum, but the discrepancies were small, as shown in figure 8.

For observing the influence of the discrepancy of ^{238}U(n, γ) in the five evaluated libraries, the neutron energy spectra on the 36 NU foils were calculated with the JEFF-3.2 library, and the corresponding ^{239}Pu production rates were calculated with the neutron energy spectra and different cross sections of ^{238}U(n, γ) in the five evaluated libraries as shown in figure 9 (left). Figure 9 (right) shows a comparison of the calculated values with the ratio of the calculations using different libraries to those using the JEFF-3.2 library. The calculated results with the JEFF-3.2 library agree with those using the ENDF/B-VII.0, ENDF/B-VII.1 and JENDL-4.0 libraries within an uncertainty of 1%. As the neutron energy spectra calculated with the JEFF-3.2 library were fixed, the calculated ^{239}Pu production rates with the CENDL-3.1 library were almost 3.5% larger than those calculations with the other four evaluated libraries. Consequently, the values of C/E (with the CENDL-3.1 library) were reduced by the larger neutron absorption cross section of ^{238}U as shown in figure 5 and table 3. One more opinion would deduce that the CENDL-3.1 data library gives a systematic higher U-238 capture reaction rate as is to be expected from the cross sections shown in figure 6. The differences of the other libraries, on the other hand, are not so significant.

5. Conclusion

An integral neutronics experiment of the measurement of ^{239}Pu production rates in a water cooled natural uranium blanket mock-up of a fusion–fission hybrid reactor was performed for the first time with a D-T neutron generator using an activation method. Numerical studies have also been performed using the SuperMC code with the ENDF/B-VII.0, ENDF/B-VII.1, JENDL-4.0, JEFF-3.2 and CENDL-3.1 libraries. From the experiment and numerical studies, the following findings have been obtained:

(a) There is a general trend for the underestimation of the ^{239}Pu production rates independent of the nuclear data library. The average ratios of the calculation results (with SuperMC code and JEFF-3.2 library) to experimental results (C/E) are in the range of 0.93–1.01 from the NU blanket mockup experiment for ^{239}Pu production rate measurements. Most of the calculation results agree with the experimental results within the experimental uncertainty.

(b) As the neutron energy spectra with JEFF-3.2 were fixed, the ^{239}Pu production rates for the 36 NU foils were calculated with different cross sections of ^{238}U(n, γ) in the five evaluated libraries and agree within 4%.

(c) The results are useful for checking the accuracy of the cross section in data libraries and substantiating the neutron-physics characteristics of a water cooled natural uranium blanket of a fusion–fission hybrid reactor. The results still revealed that a water cooled natural uranium blanket designed to breed fissile materials with ^{238}U(n, γ) was feasible, and the simulation code SuperMC and the libraries used could be used to design FFHER blanket neutronics effectively.

Valuable experience has been gained in mock-up experiments in order to set up a correct methodology to be adopted in natural uranium blanket neutronics experiments planned in the FFHER design in China, and devoted to the validation of numerical tools used to predict the ^{239}Pu production rate, in particular the need to use appropriate and independent experimental techniques. In future works, uranium fission ionization chambers and NU foils will be used for the fission reaction rate measurements, and the neutron spectra inside and outside of the mock-up will be measured as well.

Acknowledgments

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