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Benchmark experiments on breeding properties of thorium

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ABSTRACT

Thorium is a fertile element that can be applied in the conceptual blanket design of a fusion-fission hybrid reactor. It is essential to validate ^{232}Th nuclear data and obtain the breeding properties of macroscopic thorium assemblies by conducting integral neutronics benchmark experiments. The thorium assemblies with a D-T fusion neutron source consist of a polyethylene shell, depleted uranium shell, and thorium oxide powder cylinder. The ^{232}Th (n, γ), ^{232}Th (n, f), and ^{232}Th (n, 2n) reaction rates in the assemblies are measured by ThO_2 foil activation technique. The experimental uncertainties are about 3.1% for ^{232}Th capture rates, 5.3–5.5% for ^{232}Th fission rates, and 6.8–7.0% for ^{232}Th (n, 2n) reaction rates. From ^{232}Th reaction rates, the fuel and neutron breeding properties of thorium under different neutron spectra are obtained and compared. The experiments are simulated by using the MC code with different evaluated data. The ratios of calculation to experimental values for the reaction rates are analyzed.

1. Introduction

Thorium is a fertile element that can be applied in the conceptual blanket design of a fusion-fission hybrid reactor [1,2]. The actual neutron spectrum in the thorium blanket consists of fast spectrum and thermal spectrum [1,2]. Because ^{232}Th capture cross-section (7.4 barns) at thermal neutron is about 2.7 times larger than that of ^{238}U (2.7 barns), the conversion rate in the Th/U fuel cycle is more than that in the U/Pu fuel cycle, and the neutron economy of thorium is better. Moreover, ^{232}Th capture cross-section at fast neutron is slightly larger than that of ^{238}U, and ^{232}Th is more suitable to breed ^{233}U under fast spectrum. The ^{233}U capture cross-section (45.76 barns) at thermal neutron is smaller than that of ^{239}Pu (270.33 barns), and long-life MA (such as ^{237}Np, ^{241}Am, and ^{242}Cm) and Pu produced in the Th/U fuel cycle are one order of magnitude less than those in the U/Pu fuel cycle. The Th/U fuel cycle is beneficial to prevent nuclear proliferation and reduce the long-life nuclear waste. The reliability of the physical design, such as fissile fuel breeding ratio, for the conceptual blanket depends on the accuracy of nuclear data and code. It is essential to validate the evaluated ^{232}Th nuclear data and study the breeding properties, i.e., fuel and neutron breeding, by integral neutronics benchmark experiment. Only a small number of integral experiments determining the ^{232}Th reaction rates are available, and some large differences exist between the calculation and experimental results [3].

The integral experiments for thorium assemblies with a D-T neutron source were performed at Institute of Nuclear Physics and Chemistry (INPC) [4–11]. The method of measuring integral ^{232}Th reaction rate and its application in an experimental assembly were developed and investigated [4–7].

The breeding properties derived from the ^{232}Th reaction rates are valuable to the breeding ratio in the conceptual blanket based on the Th/U fuel cycle. In the present work, the experiment on breeding properties of representative macroscopic thorium assemblies with a D-T neutron source is carried out and analyzed further. The ^{232}Th reaction rates are simulated by using the MC code with different evaluated data. The breeding properties are comprehensively compared under different neutron spectra.

2. Experimental method

The breeding ratio in the conceptual blanket design of hybrid reactor is more than one [1]. The experiment is used to support the design. The breeding properties of thorium are relevant to the reaction type, cross–section, and neutron spectrum. The fuel breeding is derived from the reaction rate ratio of ^{232}Th capture to fission, and neutron breeding is derived mainly from the ^{232}Th(n,2n) and fission reaction

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rates. The different neutron spectra are constructed by using the macroscopic assemblies in which the nuclides and neutron spectra are relevant to those in the conceptual blanket. The breeding properties under different neutron spectra are obtained by comparing ^{232}Th reaction rates. The activation γ-ray method of off-line measurement of ^{232}Th(n,γ), ^{232}Th (n, f), and ^{232}Th(n,2n) reaction rates is used. The ^{232}Th reaction rates are normalized to one source neutron and one ^{232}Th atom.

The fertile nuclide ^{232}Th(n,γ)^{233}Th capture reaction rate (THCR) indicates the breeding of fissile fuel ^{233}U, i.e., the production rate of ^{233}U (^{233}Pa decay). THCR can be deduced by measuring 311.98 keV γ rays emitted from ^{233}Pa [4,5]. The reaction process is as follows:

$$^{232}Th \xrightarrow{(n,\gamma)} {}^{233}Th \xrightarrow{\beta^-,22.3\,\mathrm{min}} {}^{233}Pa \xrightarrow{\beta^-,26.967d} {}^{233}U$$

The ^{232}Th (n, f) (threshold 0.7 MeV) fission reaction rate (THFR) indicates energy amplification and neutron breeding. THFR is obtained by the fragment yield correction method [6]. To estimate the total fission rate, 151.16 keV γ rays emitted from the decay of ^{85m}Kr, which is one of the fragments of ^{232}Th (n, f) reaction, are measured. The reaction process is as follows:

$$^{232}Th \xrightarrow{(n,f),Y_f} {}^{85m}Kr \xrightarrow{\beta^-,4.48h} {}^{85}Rb$$

The ^{232}Th(n,2n) ^{231}Th (threshold 6.5 MeV) reaction rate (THNR) indicates neutron breeding. THNR is obtained by measuring 84.2 keV γ rays emitted from ^{231}Th [8]. The reaction process is as follows:

$$^{232}\mathrm{Th} \xrightarrow{(\mathrm{n,\,2n})} {}^{231}\mathrm{Th} \xrightarrow{\beta^-,25.52\mathrm{h}} {}^{231}\mathrm{Pa}$$

The ^{232}Th reaction rates are deduced from the measured activity by making appropriate corrections, which include fluctuations of the neutron flux during irradiation, detection efficiency of the HPGe γ spectrometer, self-absorption of gamma rays in the foils, counting statistics, cited value of branching ratio, and ^{85m}Kr yield. From ^{232}Th reaction rates, breeding properties could be obtained and analyzed.

3. Experimental assemblies

The experimental assemblies contain a D-T fusion neutron source, thorium sample, polyethylene shell, depleted uranium shell, and ThO_2 powder cylinder.

3.1. D-T fusion neutron source

The neutron generator at INPC produces 14-MeV neutrons using a 225-keV D^+ beam bombarding a T-Ti target. The neutron source is located at the center of the spherical assemblies. An Au-Si surface barrier semiconductor detector is at an angle of 178.2° with respect to the incident D^+ beam in the drift tube, which was used to measure the absolute yield by counting the number of associated α particles [12]. A LabVIEW-based auto-timing counter is used to record the data [13]. The yield of the D-T neutron source is about 3×10^{10} neutrons/s.

3.2. Polyethylene shell

One can assume that the elastic scattering cross-sections of H and C are reliable because they are widely used as standard cross sections [14]. The polyethylene (PE) shell is adopted for measuring the ^{232}Th reaction rates and validating the method. The inner radius (IR) and the outer radius (OR) of the PE shell with a density of 0.95 g/cm^3 are 80 and 230 mm [9], respectively, as shown in Fig. 1. Five slices of ThO_2 (concentration > 99.95%) foils are put in the radial channel at 0° to the incident D^+ beam. The distances of foils to the source are 89.1, 100.3, 111.5, 122.7, and 133.9 mm, respectively. The mass and size of foils are about 4.2 g and ϕ30 mm × 1 mm, respectively.

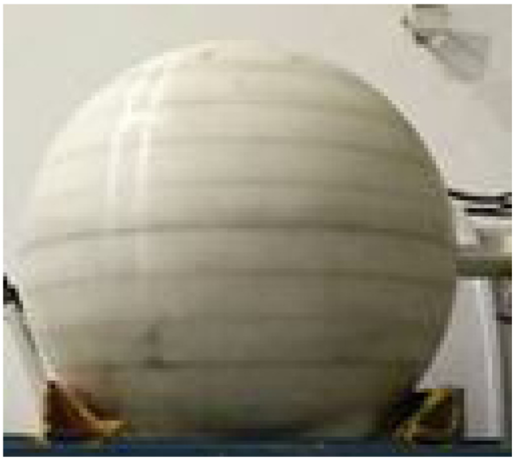

Fig. 1. Polyethylene shell assembly.

3.3. Depleted uranium shell

Thorium is placed in a uranium blanket, and the neutrons released from the U reaction process react with ^{232}Th to produce fissile ^{233}U. The depleted uranium (DU) shell in which fission nuclides are relevant to the blanket fuel is adopted. The IR/OR of the DU (~99.6% ^{238}U, ~0.4% ^{235}U) shell is 131 mm/300 mm [8], as shown in Fig. 2. Six slices of ThO_2 samples are put in the radial channel at 90° to the incident D^+ beam. The distances of the samples to the source are 138, 146, 154, 161, 174, and 187 mm, respectively. ThO_2 samples are in the form of foils, where ThO_2 powder is filled in a plexiglass box with an inner diameter of 18 mm and an outer diameter of 19 mm. The mass of ThO_2 powder is about 0.45 g, and the thickness is about 0.7 mm.

3.4. ThO_2 powder cylinder

Based on thorium oxide powder, the ThO_2 assembly is established, as shown in Fig. 3 [10,11]. ThO_2 powder is filled in a stainless steel/aluminum cylinder container with an IR of 93.4 mm and OR of 96.2 mm. The height of the thorium oxide cylinder is 168.9 mm and the density is 1.5 g/cm^3. The distance between the T-Ti target center and the front end of the experimental assembly is 78.8 mm. Five pieces of ThO_2 foils are mounted at 0° to the incident D^+ beam and are fixed using sample holders consisting of aluminum plate and stainless steel. The distances of foils to the source are 105.8, 126.1, 145.5, 165.0, and 185.5 mm, respectively. The mass of the thorium foils is about 5.0 g, and its size is about ϕ30 mm × 1 mm.

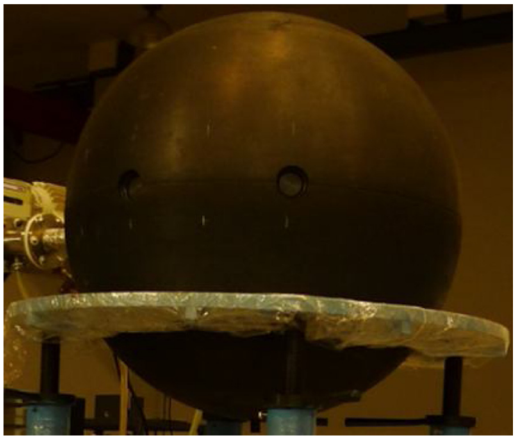

Fig. 2. Depleted uranium shell assembly.