

Recent progress in nuclear data measurement for ADS at IMP

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Abstract Recent progress in nuclear data measurement for ADS at Institute of Modern Physics is reviewed briefly. Based on the cooler storage ring of the Heavy Ion Research Facility in Lanzhou, nuclear data terminal was established. The nuclear data measurement facility for the ADS spallation target has been constructed, which provides a very important platform for the experimental measurements of spallation reactions. A number of experiments have been conducted in the nuclear data terminal. A Neutron Time-of-Flight (NTOF) spectrometer was developed for the study of neutron production from spallation reactions related to the ADS project. The experiments of 400 MeV/u ¹⁶O bombarded on a tungsten target were presented using a NTOF spectrometer. Neutron yields for 250 MeV protons incident on a thick grain-made tungsten target and a thick solid lead target have been measured using the water-bath neutron activation method. Spallation residual productions were studied by bombarding W and Pb targets with a 250 MeV proton beam using the neutron activation method. Benchmarking of evaluated nuclear data libraries was performed for D-T neutrons on ADS relevant materials by using the benchmark experimental facility at the China Institute of Atomic Energy.

Dedicated to Joseph B. Natowitz in honour of his 80th birthday

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Zhi-Qiang Chen zqchen@impcas.ac.cn **Keywords** Accelerator-driven systems · Nuclear data measurement · Neutron Time-of-Flight spectrometer · Integral experiment · Evaluated nuclear data

1 Introduction

Since the early 1990s, the accelerator-driven systems (ADS) have been proposed for nuclear waste transmutation and power generation [1, 2]. An ADS system consists of a high-power proton accelerator, a heavy-metal spallation target, and a subcritical core. When high-intensity and high-energy protons bombard a spallation target, very intense neutrons are produced through the spallation process and consequently are multiplied in the subcritical core, which surrounds the spallation target. Due to the excellent safety characteristics and potential for burning minor actinide (MA) and long-lived fission products (LLFPs), ADS has aroused worldwide interest.

In 2011, the Chinese Academy of Sciences (CAS) launched a Strategic Priority Research Program "the Future Advanced Nuclear Fission Energy–ADS Transmutation System." CAS also developed a three-stage "roadmap" for ADS development [3]. In phase I, the ADS program will first mainly focus on the R&D of key technologies in accelerators, spallation targets, and subcritical cores. A research facility, the Chinese Initiative Accelerator Driven System (CIADS), will be designed and built by 2022. In phase II, an ADS demo facility with the thermal power output of hundreds of megawatts is going to be established around 2030. Developing an industrial-scale ADS for commercial applications is the ultimate goal in phase III.

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For the conceptual and engineering designs of the ADS, the nuclear data play a very important role in many areas which include accelerator, spallation target, subcritical core, and nuclear transmutation. In this paper, recent progress in nuclear data measurement for the ADS at the IMP is reviewed.

2 Development of nuclear data measurements for ADS

The nuclear data at intermediate and at higher energies based on nuclear reaction information are a fundamental requirement for spallation target design. Based on cooler storage ring of the Heavy Ion Research Facility in Lanzhou (HIRFL–CSR), nuclear data terminal was established. Figure 1 shows the layout of HIRFL–CSR. HIRFL–CSR consists of a superconducting ECR source, SFC (sector focus cyclotron), SSC (separated sector cyclotron), RIBLL1 (radioactive ion beam line1 in Lanzhou), RIBLL2, CSRm (cooler-storage-ring main ring), and CSRe (cooler-storage-ring experiment ring). The primary beam (up to 1 GeV/u for ¹²C) can be extracted from the CSRm and directly sent to the nuclear data terminal to do experiments.

A number of experiments have been conducted in the nuclear data terminal. The following is a timeline of activities on the nuclear data measurements at the IMP.

2011: Nuclear data experimental terminal was established.

- Development of nuclear data measurement facility: Neutron Time-of-Flight spectrometer, light charged particles (LCPs) spectrometer, water-bath neutron activation experimental setup, and PISA detection system.
- (2) Study of neutron measurement method.
- (3) "400 AMeV ¹²C + Pb" experiment (thick target, neutron activation method) for a neutronics study of spallation target.

2013:

- (1) "165–350 MeV/u 12 C + H₂O" experiments for LCPs spectrometry test.
- (2) "400 AMeV ¹²C + Pb/W" experiments (thick target, water-bath neutron activation method) for a neutronics study of spallation target.
- (3) "14 MeV n + Ga" experiment for benchmarking of evaluated nuclear data.

2014:

- "250 MeV p + Pb, W" experiments (thick target, water-bath neutron activation method) for the neutronics of spallation target.
- (2) "14 MeV n + W" experiment for benchmarking of evaluated nuclear data.



Fig. 1 (Color online) HIRFL-CSR layout HIRFL-CSR





Fig. 3 (Color online) Comparisons of experimental neutron energy spectrum with GEANT4 calculations for 400 MeV/u ¹⁶O bombarded on a tungsten target at detection angles of 10° to 60° Reproduce with permission from [7]

2015:

- "300 MeV d + Be, Pb" experiments for ranges and neutron energy spectrum measurements.
- (2) "14 MeV n + W (granular), polyethylene, graphite, silicon carbide" experiments for benchmarking of evaluated nuclear data.

2016:

- (1) "14 MeV n + 238 U" experiment for benchmarking of nuclear data.
- (2) "14 MeV n + (W+ 238 U + C + CH₂ combinations) experiments"

2.1 Nuclear data measurement facility for ADS spallation target

The nuclear data measurement facility for ADS spallation target has been constructed, which provides a very important platform for the experimental measurements of spallation reactions, as shown in Fig. 2. The facility consists of a water-bath neutron activation measurement setup, light charged particles spectrometer, Neutron Timeof-Flight spectrometer, PISA detection system, HPGe detector array, liquid scintillator array, and electronics and data acquisition system. The light charged particles spectrometer consists of several telescopes (a plastic detector + a CsI(Tl) detector). The plastic detector is used to measure the TOF and energy loss of the particles, and the remaining energy of the particles is collected in the CsI(Tl) detector. The light charged particles can be identified with the TOF- $\Delta E - E$ technique. The Neutron Time-of-Flight spectrometer consists of several BC501A liquid scintillation detectors. The energy spectrum of the neutron can be obtained by Time-of-Flight method. A thin plastic scintillator detector is placed before the BC501A detector to veto the charged particle entering into the neutron detector. The liquid scintillation neutron detector can be used to measure the quick timing information and excellent neutron-gamma discrimination capability. The water-bath neutron activation measurement setup is a water tank. The target is put in the center of water tank, and the activation foils are located in the water around the target in order to measure the



Fig. 4 (Color online) Neutron productions for 250 MeV protons bombarding on a thick grain-made tungsten target and a solid lead target. Reproduce with permission from [8, 9]

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moderated neutrons by the water. Via capture reaction (n, γ), the stable isotopes composing the detector foils were transmuted into radioactive ones, which were identified by observing the characteristic gamma rays. The measurements were performed by using an HPGe detector array. The proton-induced spallation (PISA) detection system, which was once used for proton-induced spallation studies using the internal beam of the COZY storage ring at Forschungszentrum Jülich Germany [4], was shipped to Lanzhou in 2007.

2.2 Neutron energy spectrum measurements

A Neutron Time-of-Flight (NTOF) spectrometer was developed for the study of neutron production from spallation reactions related to the ADS project. The NTOF spectrometer is capable of measuring neutrons in wide energy and angular ranges. It consists of a beam pickup detector and ten individual neutron detection modules. The beam pickup detector is composed of a BC404 plastic scintillator detector with a dual-PMT readout at both ends and is located upstream from the target. Each of the neutron detection modules is composed of a thin plastic scintillator detector (veto detector) and BC501A organic liquid scintillator detector. The gamma response function is required for energy calibration of an organic liquid scintillator detector by means of gamma sources. The GEANT4 and FLUKA Monte Carlo simulation packages were used to simulate the response function of the detector for standard ²²Na, ⁶⁰Co, and ¹³⁷Cs gamma sources [5]. The simulated results showed a good agreement with experimental data by incorporating the energy resolution function to simulation codes. A method of efficiency calibration for large neutron detectors has also been studied using GEANT4 [6].

In Fig. 3, the experiment of 400 MeV/u ¹⁶O bombarded on a tungsten target that was performed using a NTOF spectrometer with BC501A detectors (12.7 cm diameter and 12.7 cm in length) is shown [7]. The combination of the light output spectra of the veto detector and the BC501A scintillator detector was utilized for the separation of charged particle events from non-charged particle



Fig. 5 (Color online) Spallation residual products for 250 MeV protons bombarding on tungsten and lead targets Reproduce with permission from [10]



Fig. 6 (Color online) Benchmarking of evaluated nuclear data for ADS with neutron nuclear data benchmark experiment setup at CIAE

(neutron and gamma-ray) ones. The neutron and gammaray events can be discriminated by using a two-gate integration method. The anode signal of the BC501A scintillator detector was divided into two pulses and sent to QDCs with total and slow gates. The neutron energy

Table 1Summary ofbenchmark experiments forADS relevant materials withD-T neutrons by IMP

spectrum is obtained from the TOF spectra with normalizing by unit solid angle and incident ion numbers. The comparisons of the experimental neutron energy spectrum at detection angles ranging from 10° to 60° with GEANT4 and with QMD model calculations are shown in Fig. 3. GEANT4 is a Monte Carlo transport code developed in CERN for simulating the passage of particles through matter. In GEANT4, users have abundant choices of physics models to handle the interactions of particles with matter over an extended energy range. Spallation reactions are usually described in two stages: the intra-nuclear cascade and de-excitation. The BIC, INCL, Bertini, and OMD models were embedded in GEANT4 to calculate the first stage of the reactions. The experimental neutron spectra shape is well reproduced by the simulations. The experimental data have been normalized to the simulated one at 10°. The experimental results show that the whole system of the NTOF spectrometer works well, and the data analysis procedure is established.

2.3 Water-bath neutron activation measurements

Neutron yields for 250 MeV protons incident on a thick grain-made tungsten target and a thick solid lead target have been measured using the water-bath neutron activation method as shown in Fig. 4 [8, 9]. A 250 MeV proton beam of HIRFL–CSR was extracted from the CSRm and sent to the nuclear data terminal to irradiate the target, which was located approximately in the center of de-ion

Sample	Dimension	Angle
Polythene	$10\mathrm{cm} \times 10\mathrm{cm} \times 5\mathrm{cm}$	60°
Gallium	$10 \text{ cm} \times 10 \text{ cm} \times 5 \text{ cm}, 10 \text{ cm} \times 10 \text{ cm} \times 10 \text{ cm},$	60°, 120°
	ϕ 13 cm × 3.2 cm, ϕ 13 cm × 6.4 cm	
Tungsten (block)	$10\text{cm}\times10\text{cm}\times3.6\text{cm}, 10\text{cm}\times10\text{cm}\times7.2\text{cm}$	60°, 120°
Tungsten	9.8 imes 9.9 imes 7.2 cm,	60°, 120°
(Granular)	(granular diameter:1 mm)	
Graphite	ϕ 13 × 2 cm, ϕ 13 × 20 cm	60°, 120°
SiC	ϕ 13 × 2 cm, ϕ 13 × 20 cm	60°, 120°
²³⁸ U	$10 \mathrm{cm} \times 10 \mathrm{cm} \times 2 \mathrm{cm}$,	60°
²³⁸ U	$10 \text{ cm} \times 10 \text{ cm} \times 5 \text{ cm}, 10 \text{ cm} \times 10 \text{ cm} \times 11 \text{ cm}$	60°, 120°
W+U	W: $10 \text{ cm} \times 10 \text{ cm} \times 3.5 \text{ cm}$,U: $10 \text{ cm} \times 10 \text{ cm} \times 2 \text{ cm}$	60°
W+U+C	W: $10 \text{ cm} \times 10 \text{ cm} \times 3.5 \text{ cm}$,U: $10 \text{ cm} \times 10 \text{ cm} \times 2 \text{ cm}$	60°
	C: $10 \text{ cm} \times 10 \text{ cm} \times 2 \text{ cm}$	
W+U+	W: $10 \text{ cm} \times 10 \text{ cm} \times 3.5 \text{ cm}$,U: $10 \text{ cm} \times 10 \text{ cm} \times 2 \text{ cm}$	60°
C+CH2	C: $10 \text{ cm} \times 10 \text{ cm} \times 2 \text{ cm}$, CH2: $10 \text{ cm} \times 10 \text{ cm} \times 2 \text{ cm}$	
U+C	U: $10 \text{ cm} \times 10 \text{ cm} \times 5 \text{ cm}$, C: $10 \text{ cm} \times 10 \text{ cm} \times 10 \text{ cm}$	60°
U+C	U: $10 \text{ cm} \times 10 \text{ cm} \times 5 \text{ cm}$, C: $10 \text{ cm} \times 10 \text{ cm} \times 10 \text{ cm}$	60°
+CH2	CH2: $10 \text{ cm} \times 10 \text{ cm} \times 10 \text{ cm}$	



Fig. 7 (Color online) Neutron leakage spectra measured in the experiment and simulated by the MCNP code for gallium and tungsten targets Reproduce with permission from [11, 12]

light water to produce neutrons. The neutron source strength and flux distribution were derived from the activity of the Au foil. The method for getting the total neutron number was to integrate the thermal neutron flux all over the water-bath volume. Through analyzing the activity of Au foils, the neutron flux distribution in water was obtained. The neutrons slowing down the process shows that the neutrons from tungsten have an average energy lower than neutrons from the lead target. The neutron flux distribution and average neutron production per incident proton have been calculated using GEANT4 code and compared with the experimental data. The simulations described very well the shape of the spatial distribution of neutron flux.

2.4 Spallation residual productions measurement by neutron activation method

Spallation residual productions were studied by bombarding W and Pb targets with a 250 MeV proton beam, as shown in Fig. 5 [10]. The data were obtained by using the off-line gamma spectroscopy method. A total of 28 nuclides in W and 34 nuclides in Pb targets were identified, and their cross sections were determined. Corresponding simulations using the MCNPX transport code were also performed to compare the measurements. In the simulations, eight different models or combinations of intra-nuclear cascade and de-excitation models were introduced. The comparison indicated that most cross sections were fairly and reliably predicted for nuclides in the spallation mass region, but differ greatly for nuclides in the medium and fission mass regions.

3 Benchmarking of evaluated nuclear data libraries with D-T neutrons

The experimental studies of fast neutron scattering are important for the design of nuclear reactors. They play a crucial role for verification of the evaluated nuclear data libraries, especially the elements that are of interest in ADS, fission and fusion reactor technologies. In the neutronics calculations, the uncertainty introduced in the ADS design strongly depends on the calculation code used and the evaluated nuclear data. MCNP (Monte Carlo neutron and photon transport code) is widely used for the benchmarking of evaluated nuclear data libraries. Several evaluated nuclear data libraries, like JENDL-4.0 from Japan, Fig. 8 (Color online) Neutron leakage spectra measured in the experiment and simulated by MCNP code for silicon carbide and graphite targets Reproduce with permission from [13, 14]



ENDF/VBII.1 from USA, JEFF-3.2 from Europe, and CENDL-3.1 from China, have been published in the world. In the design of CIADS, a high-intensity proton beam bombards on a spallation target inside the subcritical core and provides an external neutron source for the subcritical core, as shown in Fig. 6. Tungsten is proposed to be one of the most promising candidates for spallation targets and other structural materials of CIADS project, as well as an important material in fusion devices. The nuclear fuel is ²³⁸U wrapped by silicon carbide. The graphite is considered primary moderator and reflector material. Polyethylene is considered a shielding material. Gallium (Ga) is one of such elements, which can be used as a cooling agent.

Benchmarking of evaluated nuclear data libraries has been performed for D-T neutrons on ADS relevant materials by using the benchmark experimental facility at China Institute of Atomic Energy (CIAE) in Beijing [11–15]. In Fig. 6, the benchmark experimental facility is shown. The neutron energy generated in the experiment was about 14.8 MeV in the forward direction. A BC501A liquid scintillator counter (neutron detector), which was located behind the concrete wall with a collimated hole, had a size of 5.08 cm in diameter and 2.54 cm in length for detecting leakage neutrons from the sample. Using such a heavy shielding and collimating system, high foreground to background ratio has been achieved. The leakage neutron spectra from the samples were measured by the TOF method using a liquid scintillator detector and compared with the MCNP-4C calculations from ENDF-B-VII.1, JENDL-4.0, and CENDL-3.1 libraries. Table 1 shows a summary of the benchmark experiments for ADS relevant materials with D-T neutrons by the IMP.

In Fig. 7, the neutron leakage spectra measured at 60° and 120° for gallium and tungsten targets are shown. For the gallium target, the measured spectra are rather well reproduced by MCNP-4C simulations with the CENDL3.1, ENDF/B-VII, and JENDL-4.0 evaluated nuclear data libraries, except for the inelastic contributions around En = 10–13 MeV. All three libraries significantly underestimate the inelastic contributions. The inelastic contributions are further studied using the Talys simulation code. The experimental spectra are reproduced reasonably well in the whole energy range by the Talys calculation, including the inelastic contributions. For the tungsten target, in overall, the calculations with the ADS-2.0 and ENDF/B-VII.1 libraries give satisfactory reproductions of the experimentally measured neutron leakage spectra.

In Fig. 8, the neutron leakage spectra measured at 60° and 120° for silicon carbide and graphite targets are shown. This result indicates that the experimental apparatus and the data analyzing procedures are in reasonable shape. For the silicon carbide target, the measured spectra are well reproduced by MCNP calculations with the CENDL-3.1, ENDF/B-VII.1, and JENDL-4.0 evaluated nuclear data libraries, except in the 5-8 MeV range for 20 cm thickness. The discrepancies are mostly considered as caused by the improper evaluation of the angular distribution and secondary neutron energy distribution of the elastic scattering and inelastic scattering in evaluated nuclear data libraries. For the graphite target, the results obtained from a 20-cm-thick sample revealed that the calculation results with CENDL-3.1 and JENDL-4.0 libraries showed good agreements with the experiments conducted in the whole energy region. However, a large discrepancy of approximately 40% was observed below the 3 MeV energy region with the ENDF/B-VII.1 library.

4 Summary

Recent progress in nuclear data measurement for ADS at the IMP is reviewed. The nuclear data measurement facility for ADS spallation target has been constructed, which provides a very important platform for the experimental measurements of spallation reactions. Several experiments about spallation targets (W, Pb) have been performed in the nuclear data terminal. Benchmarking of evaluated nuclear data libraries with D-T neutrons on ADS relevant materials has been carried out. In the future, the high-energy neutron transportation in ADS relevant materials and the neutronics of ADS subcritical target reactor coupling system will be studied. Acknowledgements The author would like to thank Prof. Natowitz for his great help and support in scientific research. From July, 2005– August, 2008, the author worked with Prof. Natowitz as a postdoctoral research associate at Cyclotron Institute, Texas A&M University. From July, 2012–December, 2012, Xing-Quan Liu and Suyalatu Zhang (the author's students) studied in Prof. Natowitz's group. From July, 2014–December, 2014, Xing-Quan Liu and Wei-Ping Lin (the author's students) studied in Prof. Natowitz's group. The author and his wife Hong-Lian Chen also appreciate Prof. Natowitz and his wife Karin who offered great help to them.

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