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京都大学臨界集合体実験装置における トリウム装荷加速器駆動システムの実験ベンチマーク

Experimental Benchmarks on Thorium-Loaded Accelerator-Driven System at Kyoto University Critical Assembly

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Preface

These experimental benchmarks were contributed to the Collaborative Works (CW) in the International Atomic Energy Agency (IAEA), as entitled "**Accelerator Driven Systems (ADS) Applications and Use of Low-Enriched Uranium in ADS**," from 2013 to 2014.

The major objective of these benchmarks is to contribute to research and development of ADS through the thorium (Th)-loaded ADS experimental data with the use of 14 MeV neutrons and spallation neutrons generated by 100 MeV protons and tungsten target carried out at the Kyoto University Critical Assembly (KUCA) A-core.

Special thanks are due the KUCA and the fixed-filed alternative gradient (FFAG) accelerator staff and students for support and patience throughout a series of Th-loaded ADS experiments carried out at KUCA.

Cheol Ho Pyeon

January 2015

Keywords:
Th-ADS, KUCA, FFAG accelerator, 14 MeV neutrons, Spallation neutrons

要　旨

この実験ベンチマーク問題は、国際原子力機関（IAEA）において 2013 年から 2014 年にかけて行われた国際共同作業プロジェクト「加速器駆動システムの応用と低濃縮ウラニウムの利用」の一部として採択された「京都大学臨界集合体実験装置におけるトリウム装荷加速器駆動システムの実験ベンチマーク」である。

この実験ベンチマーク問題は、KUCA の A 架台において行われた 14 MeV 中性子（パルス中性子発生装置）および FFAG 加速器から得られる核破砕中性子を用いた実験を通して、ADS の基礎研究の発展に貢献することを目的としている。

最後に、KUCA においてトリウム装荷 ADS 実験を準備および運転にご協力をいただいた KUCA および FFAG 加速器のスタッフ、実験における測定および数値解析に献身的に取り組んだ KUCA 棟に在籍していた学生諸君に心から感謝の意を表します。

卞　哲浩

2015 年 1 月

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5th January, 2015

1. Collaborative Work Specifications

1-1. Introduction

The accelerator-driven system (ADS) was developed for producing energy and for transmuting minor actinides and long-lived fission products. ADS has attracted worldwide attention in recent years because of its superior safety characteristics and potential for burning plutonium and nuclear waste. An outstanding advantage of its use is the anticipated absence of reactivity accidents, provided sufficient subcriticality is ensured. At the Kyoto University Research Reactor Institute [1]-[2], a series of experiments for ADS was launched in fiscal 2003 at the Kyoto University Critical Assembly (KUCA) [3]-[19], with sights on a future plan (**K**umatori **A**ccelerator Driven **R**eactor **T**est Facility **&** Innovation Research **Labo**ratory: Kart & Lab. Project). A new accelerator was attached to the KUCA facility in March 2008, and the high-energy neutrons generated by the interaction of 100 MeV protons with tungsten target was injected into KUCA. The new accelerator is called the fixed-field alternating gradient (FFAG) accelerator of the synchrotron type developed by High Energy Accelerator Research Organization (KEK) in Japan.

At KUCA, by combining a critical assembly of a solid-moderated and -reflected type core with the accelerators, 14 MeV pulsed neutrons generated by D-T (deuteron–tritium) reactions and 100 MeV protons generated from the FFAG accelerator were injected separately into the subcritical system, where the highly-enriched uranium (HEU), thorium (Th) and natural uranium (NU) fuel was loaded together with the reflectors, including the polyethylene (PE), graphite (Gr) and beryllium (Be). The experiments of thorium-loaded ADS were carried out by varying the neutron spectrum and the external neutron source, and were aimed at investigating the sensitivity of thermal neutron flux for thorium capture reactions, and the prompt and delayed neutron behaviors in the subcritical system. The thorium-loaded ADS experiments provided importantly the effects of the neutron spectrum and the external neutron source on both the static and kinetic parameters. In the experiments, for the investigation of the effect of neutron spectrum, the moderator material was varied in fuel region. As the other investigation of the effect, the external neutron source (14 MeV neutrons and 100 MeV protons) was injected separately into the thorium-loaded core varying the moderator. Before the subcritical experiments, the thorium plate irradiation experiment was carried out in the KUCA core, to analyze experimentally the thorium capture and fission reactions in the critical system as the reference of subcritical system, although the probability of ^{232}Th capture and ^{233}U fission reactions could be examined in the subcritical state.

The objectives of this study were to investigate the neutronic characteristics of the thorium-loaded ADS with external neutron source through the static and kinetic experiments, and to evaluate the accuracy of the Monte Carlo analyses through the calculations by the MCNPX [20] code with ENDF/B-VII.0 [21], JENDL/HE-2007 [22]-[23] and JENDL/D-99 [24] libraries.

1-2. Experimental Settings

1-2-1. Description of KUCA core

KUCA comprises solid-moderated and -reflected type-A and -B cores, and a water-moderated and -reflected type-C core. In the present series of experiments, the solid-moderated and -reflected type-A core was combined with a Cockcroft-Walton type pulsed neutron generator and the FFAG accelerator at KUCA.

The A-core (A3/8"P36EU(3)) configuration used for measuring the reaction rates of thorium metal plate is shown in Fig. 1-1. The fuel rods were constructed of a combination of 21 elements that were loaded on the grid plate. The materials used in the critical assemblies were always in the form of rectangular parallelepipe, 2" sq. with thickness ranging between 1/16" and 2". The upper and lower parts of the fuel region were polyethylene reflector layers of more than 500 mm long, as shown in Fig. 1-2. The fuel rod, a 93% enriched uranium-aluminum (U-Al) alloy, consisted of 36 cells of 2 polyethylene plates 1/8" and 1/4" thick, and a U-Al plate 1/16" thick and 2" sq. The functional height of the core was approximately 400 mm.

1-2-2. Description of 14 MeV Pulsed neutron generator

The pulsed neutron generator was combined with the KUCA A-core, where 14 MeV pulsed neutrons were injected into the subcritical system. In the experiments, the deuteron beams were led to the tritium (^{3}H; T) target located outside the polyethylene reflector. At the pulsed neutron generator, the beam peak characteristics are about 0.5 mA intensity, 10 μs pulsed width, 100 Hz pulsed frequency, 25 mm diameter spot size and 1×10^{7} 1/*s* neutron yield.

1-2-3. Description of FFAG accelerator

100 MeV protons generated from the FFAG accelerator were injected onto the tungsten target. The main characteristics are under the following parameters: 100 MeV energy, 0.3 nA intensity, 20 Hz pulsed frequency, 100 ns pulsed width and 40 mm diameter spot size at the tungsten target (50 mm diameter and 9 mm thick). The thickness of target was determined on the basis of previous analyses [11], [18] in the reaction rates for high-energy protons. A level of the neutron yield generated at the target was around 1.0×10^{7} 1/*s* by an injection of 100 MeV protons onto the tungsten target.

1-3. Experimental Results

1-3-1. Indium (In) reaction rate distribution (Th-loaded ADS experiments)

Indium (In) wire 1.5 mm diameter and 600 mm long was set in the axial center position along (16, 17-Q, Y; 14 MeV neutrons) and (13, 14-I, A'; 100 MeV protons) the vertical shown in Figs. 2-1, 2-2, 2-3 and 2-4 for measuring the reaction rate distribution. The experimental results of the In wire were obtained by measuring total counts of the peak energy of γ-ray emittance and normalized by the counts of irradiated Nb (10×10×1 mm) and In foils (10×10×1 mm) emitted from ^{93}Nb(*n*, 2*n*)^{94}Nb (threshold energy 9 MeV) and ^{115}In(*n*, *n*')^{115m}In (threshold energy 0.3 MeV) reactions set in the location of the tritium and tungsten targets, respectively.

1-3-2. Time evolution data of PNS and Noise methods (Th-loaded ADS experiments)

For 14 MeV neutrons, to monitor carefully the prompt and delayed neutron behaviors, each core was set with three ^{3}He detectors (20 mm diameter and 300 mm long) at the positions of #1 at (12, X), #2 at (15, R) and #3 at (18, R) (Fig. 2-1 for Th-PE, Th-Gr, Th-Be, Th-HEU-PE and NU-PE cores), and #1 at (12, R) and #2 at (15, R) (Fig. 2-3 for Th-HEU-5PE and Th-HEU-Gr-PE cores). Through the time evolution of the prompt and delayed neutrons, the prompt neutron decay constant was deduced by the least-square fitting of the time evolution of the neutrons to an exponential function over the time optimal duration. Subcriticality was deduced by the extrapolated area ratio method on the basis of the prompt and delayed neutron behaviors. For 100 MeV protons, neutron detectors (^{3}He detectors; #1, #2 and #3) were set at three positions shown in Fig. 2-2 for Th-PE, Th-Gr, Th-Be, Th-HEU-PE and NU-PE cores: #1 at (18, A), #2 at (15, H) and #3 at (12, H)), and Fig. 2-4 for Th-HEU-5PE and Th-HEU-Gr-PE cores: #1 at (18, H) and #2 at (15, H).

1-3-3. Critical position and Excess reactivity (Th plate irradiation experiment)

The critical state was adjusted by maintaining the control rods in certain positions, and the excess reactivity was attained on the basis of its integral calibration curve obtained by the positive period method.

1-4. References

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Appendix

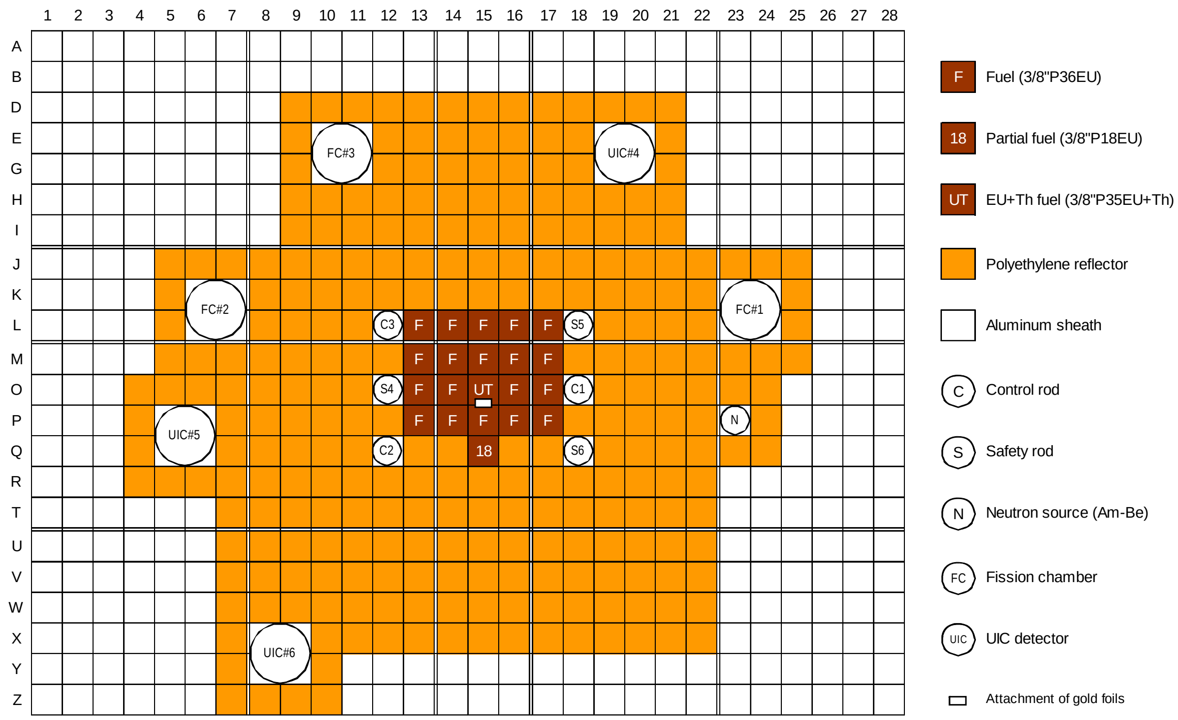

Fig. 1-1 The general view of KUCA core configuration

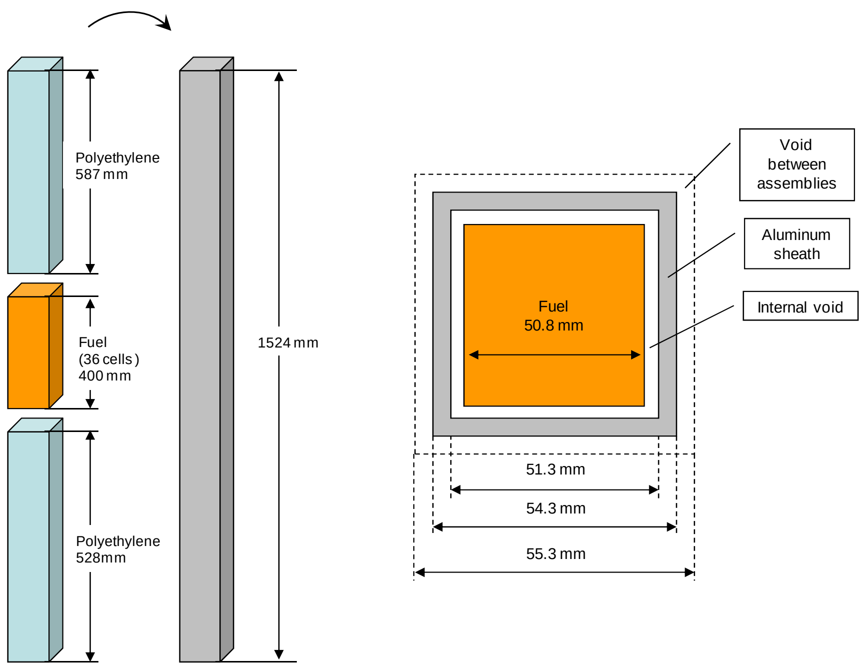

Fig. 1-2 Description of fuel assembly at KUCA

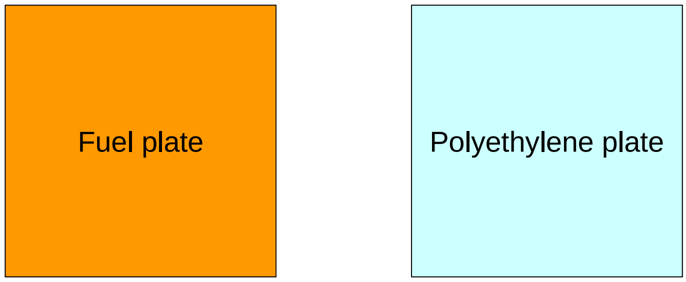

Fig. 1-3 Description of fuel (HEU, Th and NU) and polyethylene plates

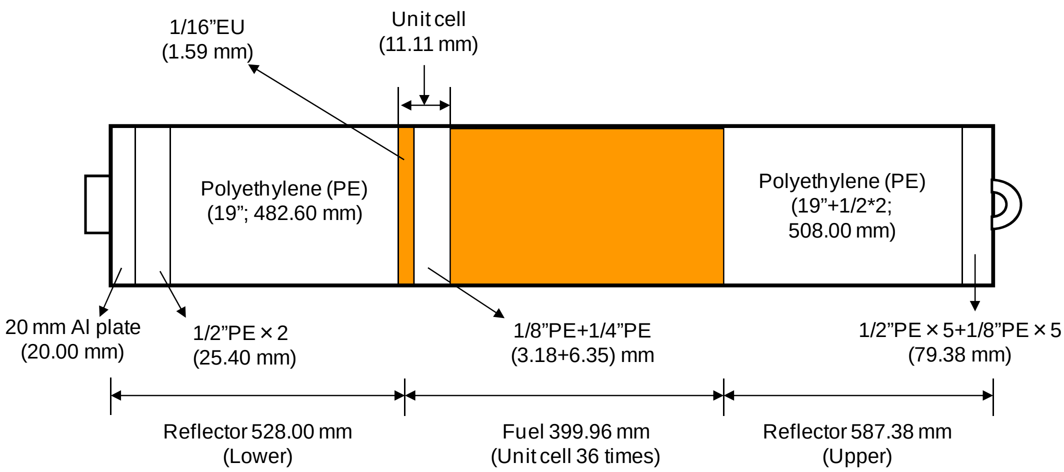

Fig. 1-4 Fall sideways view of fuel assembly “F” shown in Fig. 1-1

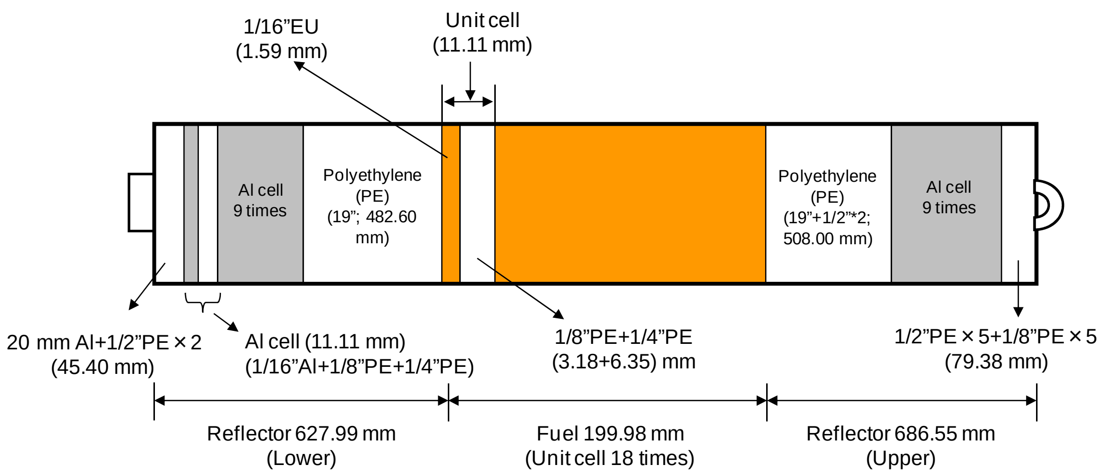

Fig. 1-5 Fall sideways view of partial fuel assembly “18” shown in Fig. 1-1

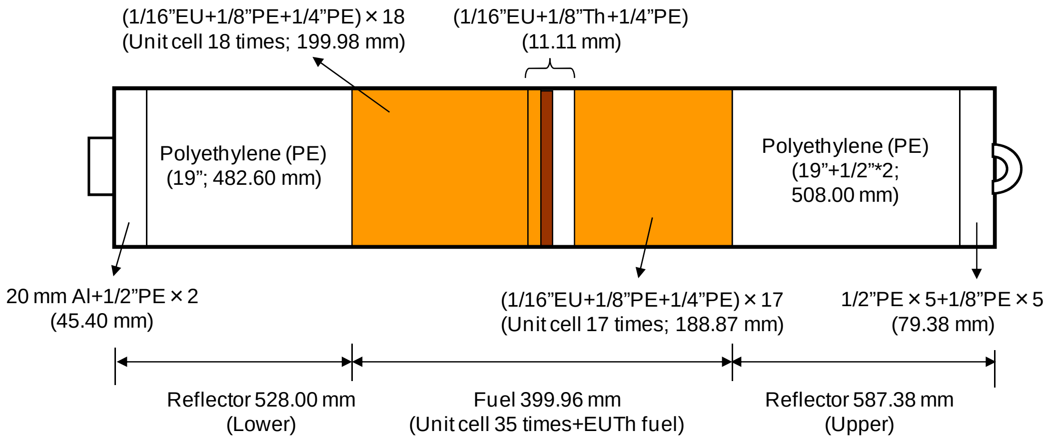

Fig. 1-6 Fall sideways view of partial fuel assembly "UT" shown in Fig. 1-1

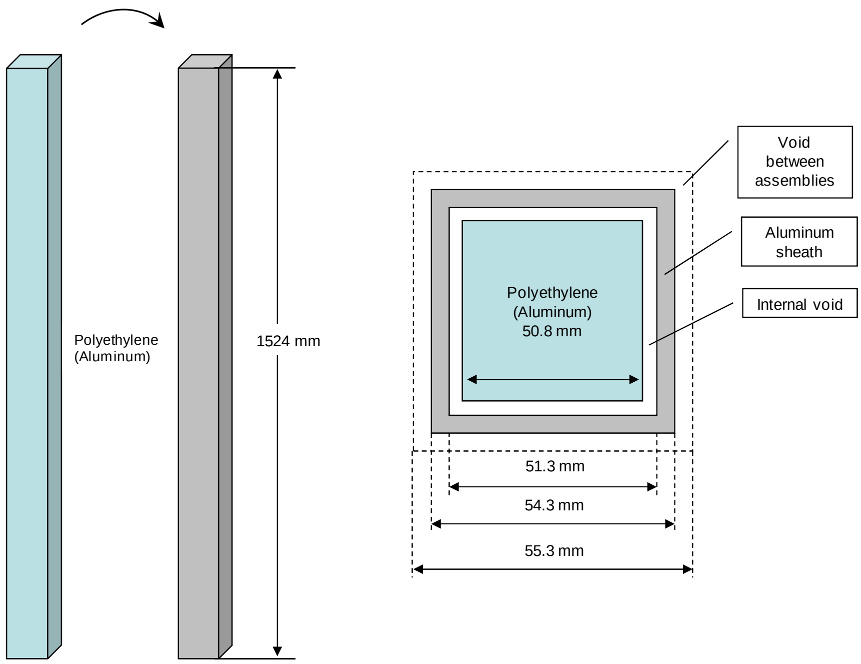

Fig. 1-7 Description of polyethylene reflector at KUCA

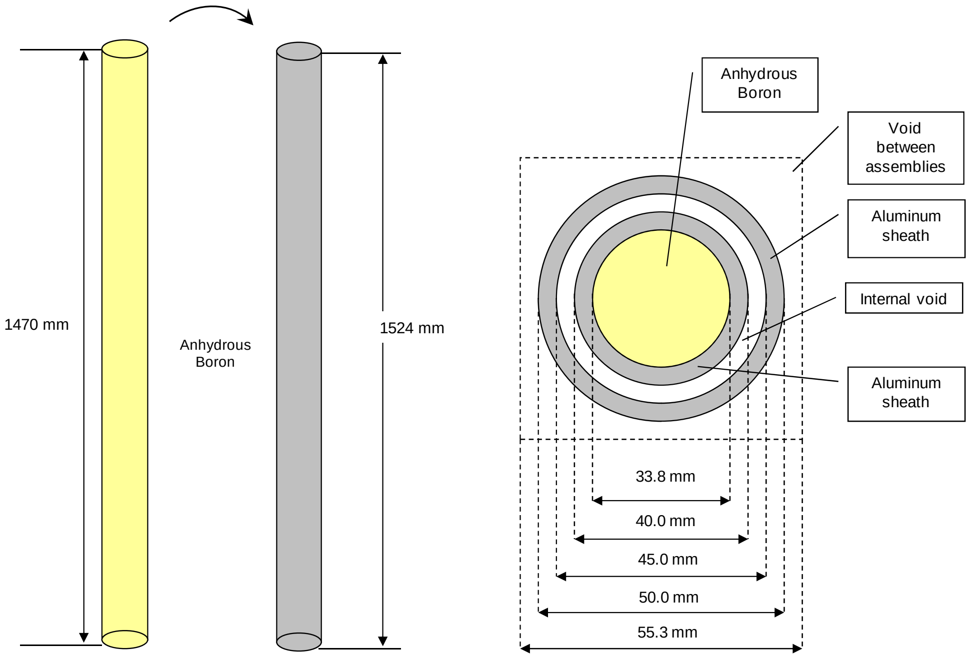

Fig. 1-8 Description of control (safety) rod at KUCA

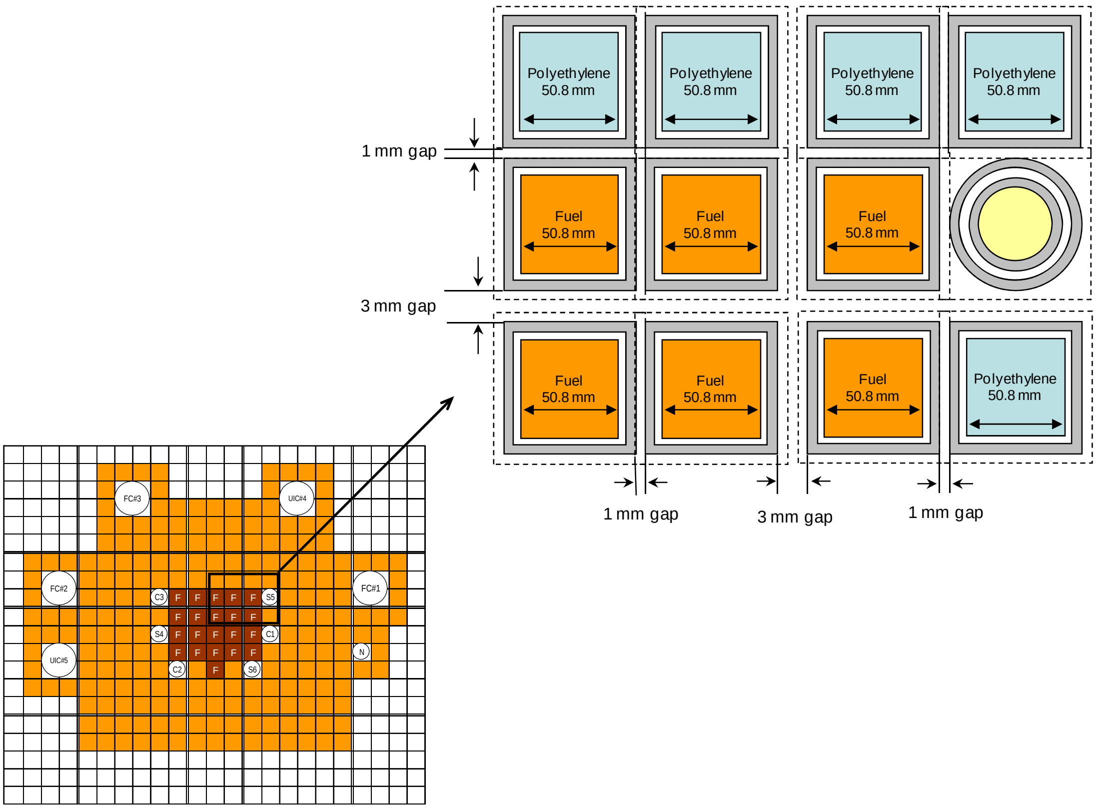

Fig. 1-9 Description of fuel assembly, polyethylene reflector and control rod at KUCA

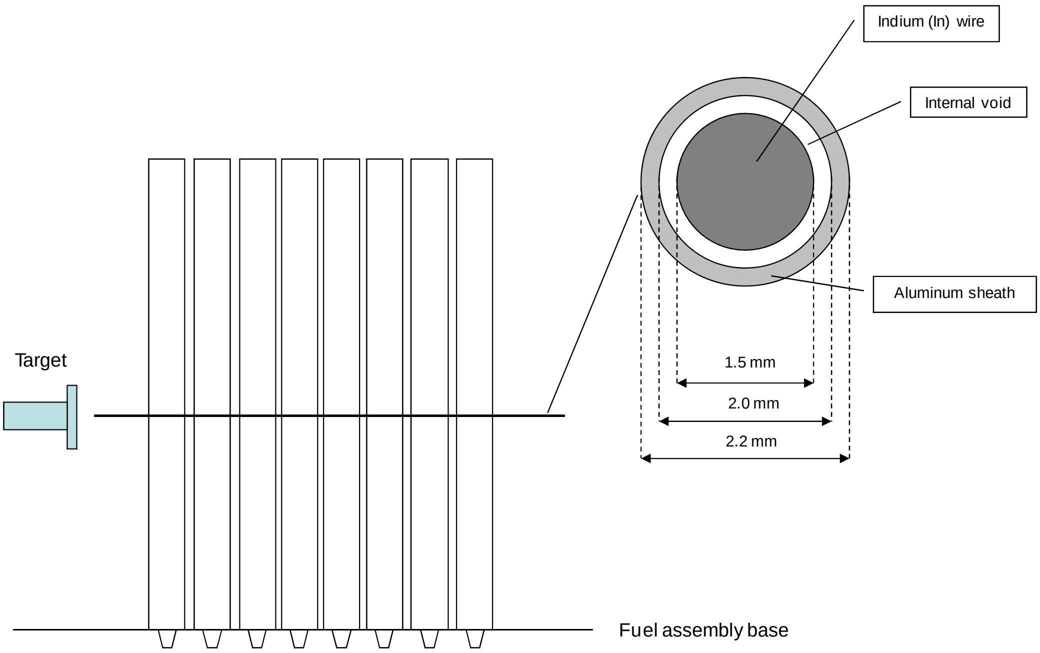

Fig. 1-10 Setting of Indium (In) wire

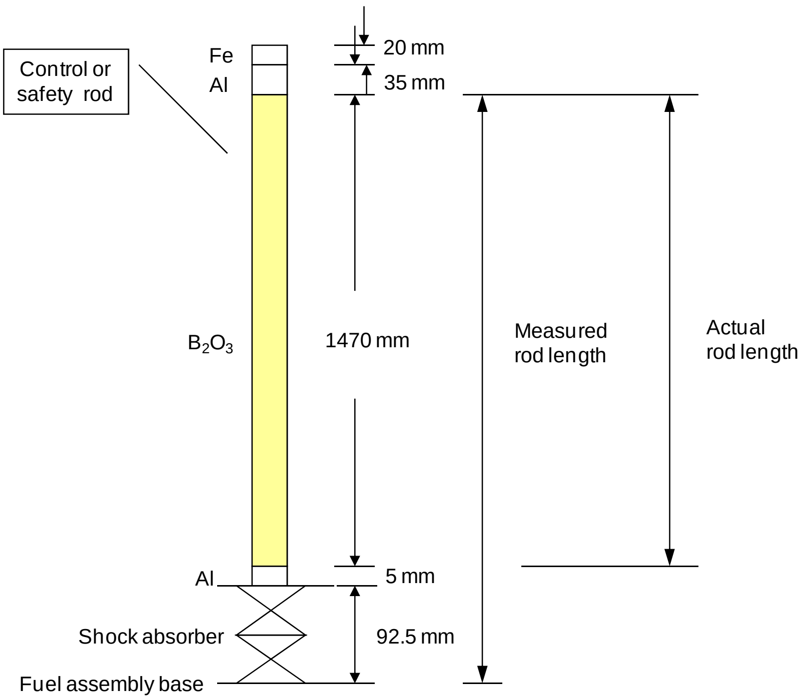

Fig. 1-11 Actual position of control (safety) rod
(Actual position = Measured position – 97.5 mm)

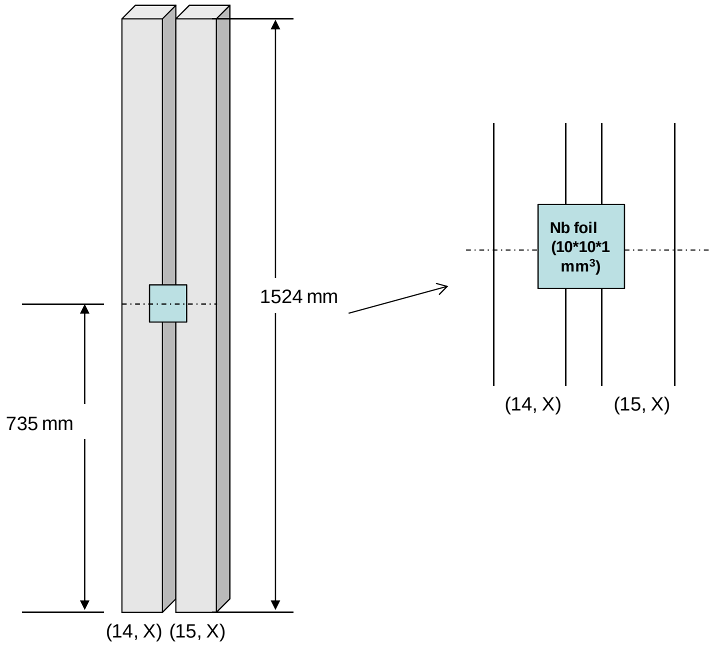

Fig. 1-12　Nb foil attachment near the tritium target in Thorium-loaded ADS with 14 MeV neutrons

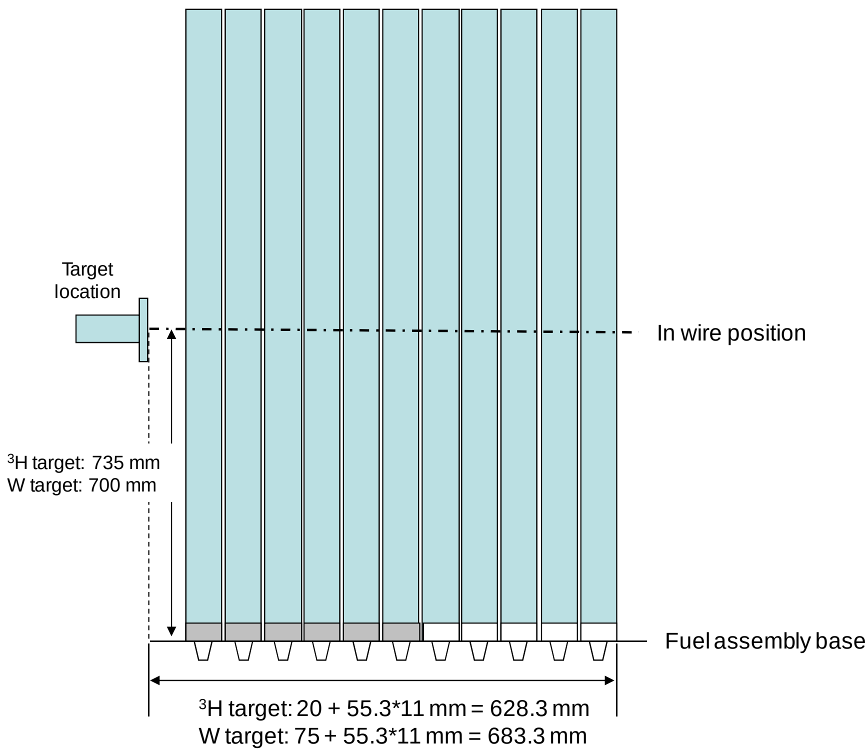

Fig. 1-13　Side view of target and core configuration with 14 MeV neutrons (^{3}H; T target) or 100 MeV protons (tungsten; W target)

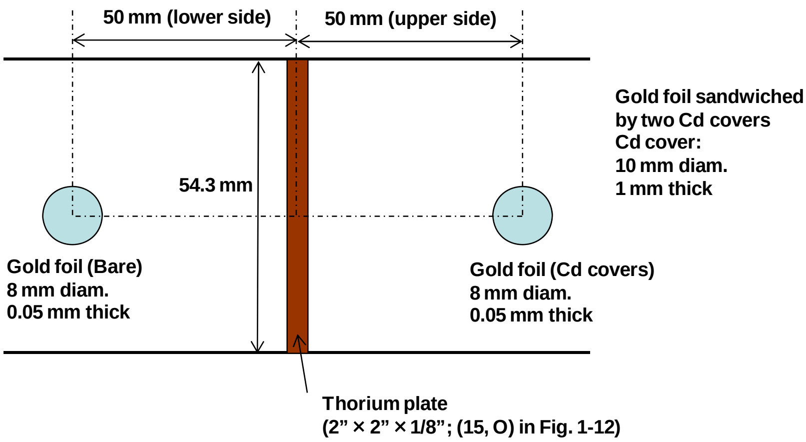

Fig. 1-14 Fall sideways view of UT fuel rod in (15, O; Fig. 1-1)

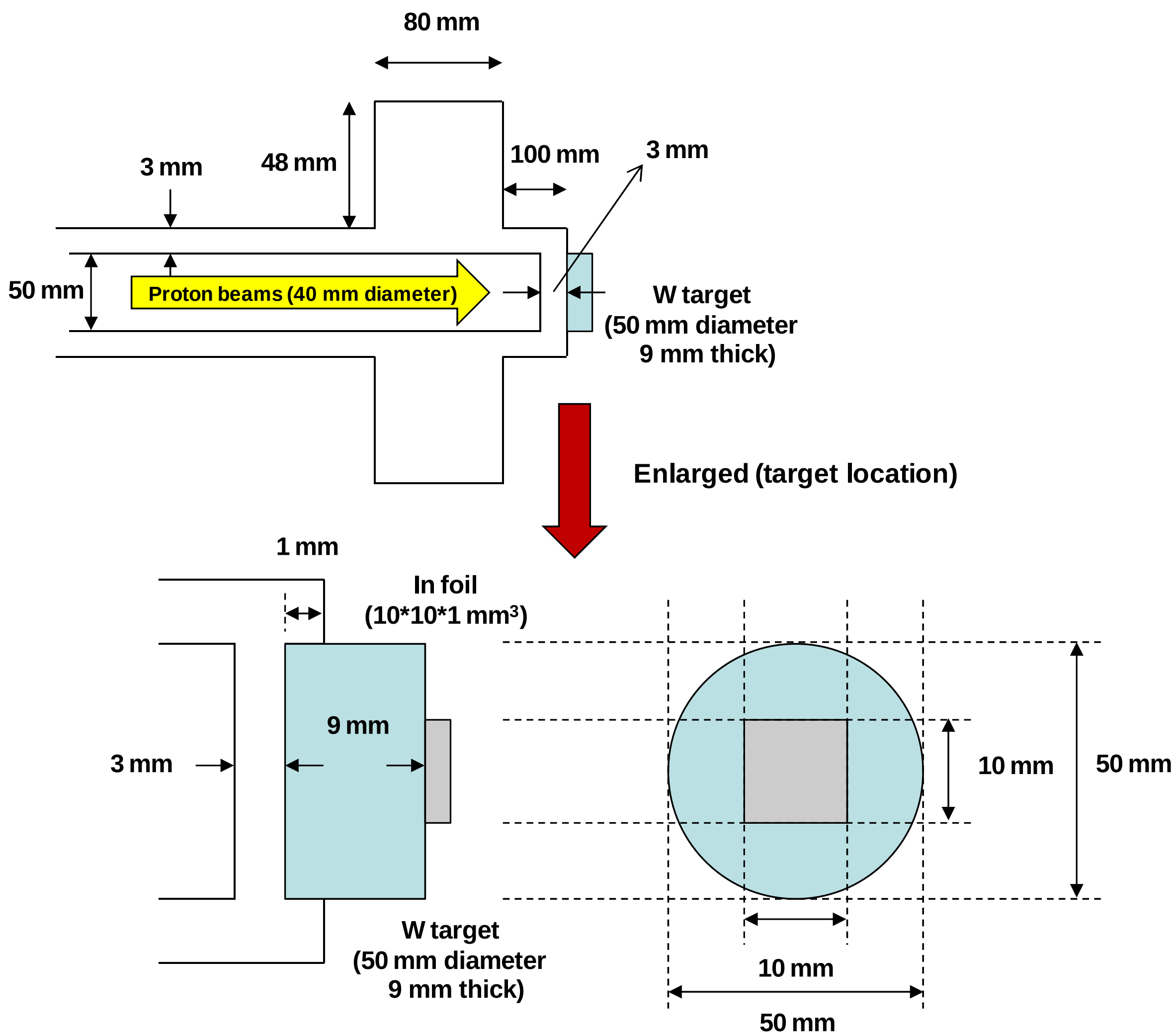

Fig. 1-15 Tungsten target configuration of thorium-loaded ADS experiments with 100 MeV protons (Note that W target was 50 mm diameter and **12 mm** thick in the Th-HEU-5PE and Th-HEU-Gr-PE cores.)

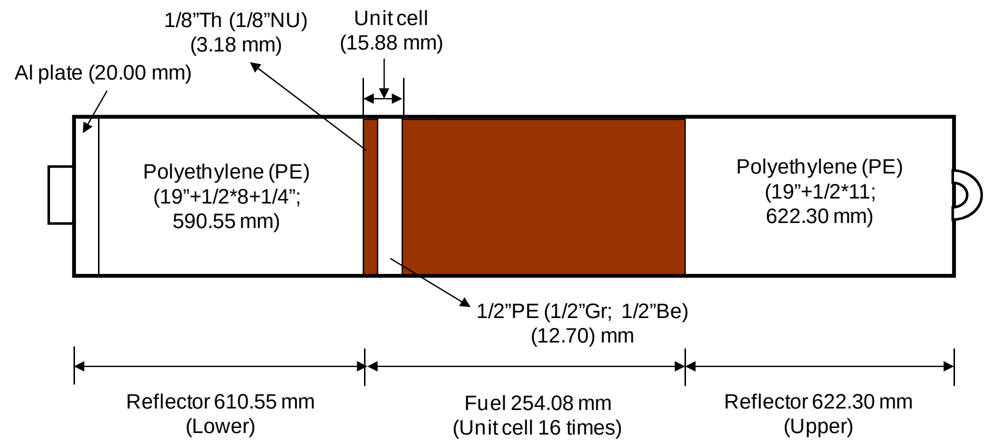

Fig. 1-16 Fall sideways view of fuel assembly Th-PE, Th-Gr, Th-Be and NU-PE

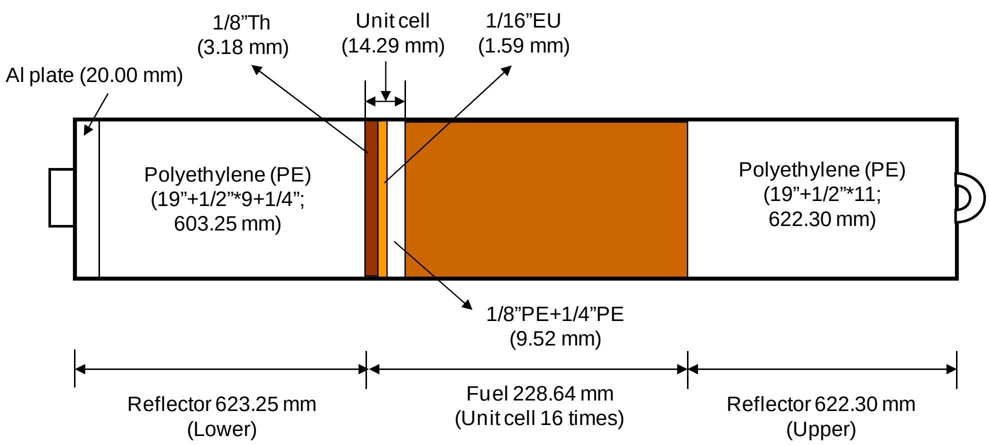

Fig. 1-17 Fall sideways view of fuel assembly Th-HEU-PE

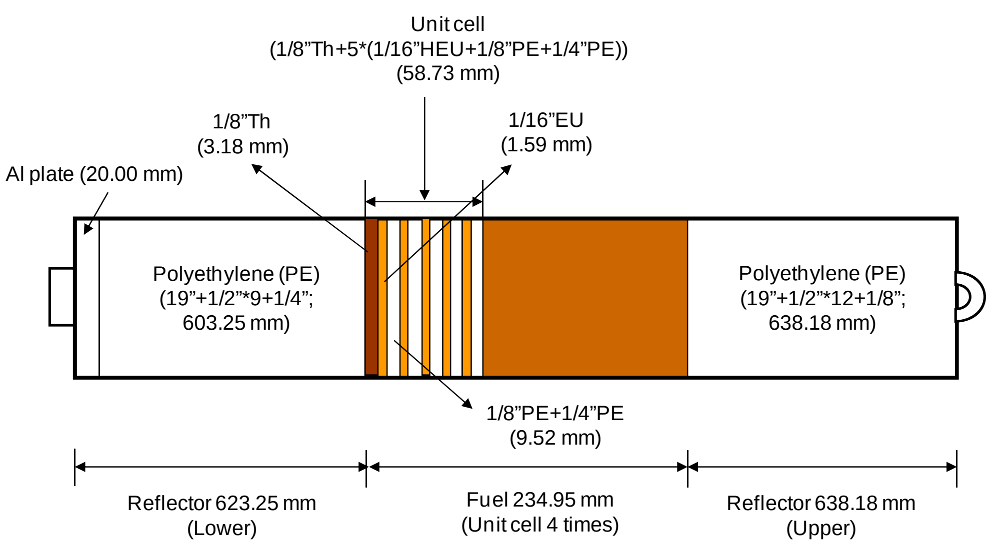

Fig. 1-18 Fall sideways view of fuel assembly Th-HEU-5PE

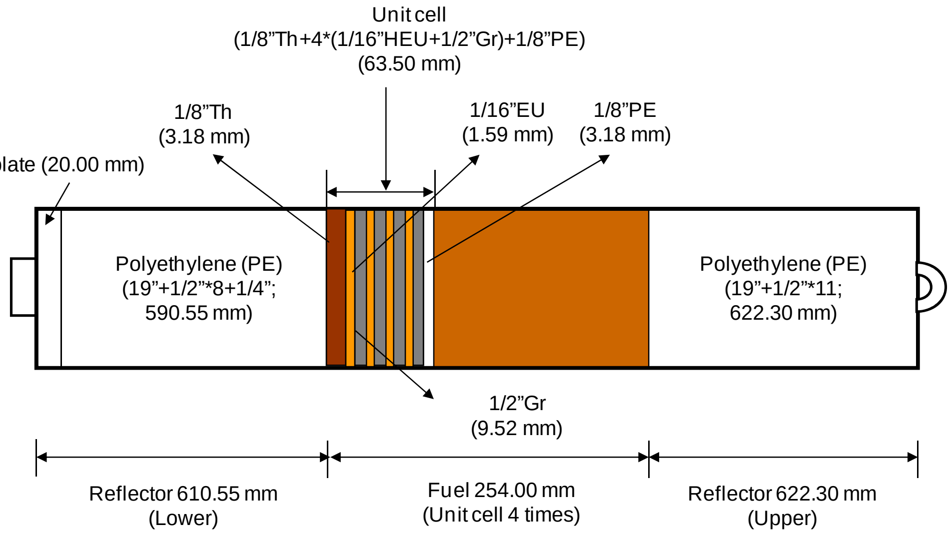

Fig. 1-19 Fall sideways view of fuel assembly Th-HEU-Gr-PE

Table 1-1 Atomic densities of 1/16” thick highly-enriched uranium (HEU) fuel plate (U-Al alloy)

Isotope	Atomic density ($\times 10^{24}/cm^3$)
^{235}U	1.50694×10^{-3}
^{238}U	1.08560×10^{-4}
Al	5.56436×10^{-2}

Table 1-2 Atomic densities of 1/8” thick thorium (Th) fuel plate

Isotope	Atomic density ($\times 10^{24}/cm^3$)
^{232}Th	2.86389×10^{-2}

Table 1-3 Atomic densities of 1/8” thick natural uranium (NU) fuel plate

Isotope	Atomic density ($\times 10^{24}/cm^3$)
^{235}U	3.25792×10^{-4}
^{238}U	4.48577×10^{-2}

Table 1-4 Atomic densities of polyethylene (PE) reflector

Isotope	Atomic density ($\times 10^{24}/cm^3$)			
	1/2” thick plate	1/4” thick plate	1/8” thick plate	19” Polyethylene square rod
H	8.06560×10^{-2}	8.08711×10^{-2}	8.02167×10^{-2}	8.00083×10^{-2}
C	4.03280×10^{-2}	4.04356×10^{-2}	4.01084×10^{-2}	4.00042×10^{-2}

Table 1-5 Atomic densities of graphite (Gr) plate

Isotope	Atomic density ($\times 10^{24}$/cm^3)
C	8.60664×10^{-2}

Table 1-6 Atomic densities of beryllium (Be) plate

Isotope	Atomic density ($\times 10^{24}$/cm^3)
Be	1.22932×10^{-1}

Table 1-7 Atomic densities of control and safety rods

Isotope	Atomic density ($\times 10^{24}$/cm^3)
^{10}B	3.87448×10^{-3}
^{11}B	1.68447×10^{-2}
^{16}O	3.10787×10^{-2}

Table 1-8 Atomic density of aluminum sheath for the core element

Isotope	Atomic density ($\times 10^{24}$/cm^3)
Al	6.00385×10^{-2}

Table 1-9 Atomic densities of ^{93}Nb, ^{112}Cd, ^{115}In and ^{197}Au

Foil (wire)	Isotope	Abundance (%)	Purity (%)	Atomic density ($\times10^{24}/cm^3$)
^{92}Nb	^{92}Nb	100	99.9	5.54947×10^{-2}
Cd	^{106}Cd	1.25	99.99	5.39648×10^{-4}
	^{108}Cd	0.89	99.99	3.91477×10^{-4}
	^{110}Cd	12.51	99.99	5.59564×10^{-3}
	^{111}Cd	12.81	99.99	5.78677×10^{-2}
	^{112}Cd	24.13	99.99	1.10072×10^{-2}
	^{113}Cd	12.22	99.99	5.62419×10^{-3}
	^{114}Cd	28.72	99.99	1.33398×10^{-2}
	^{116}Cd	7.47	99.99	3.53884×10^{-3}
In	^{113}In	4.29	99.99	1.64406×10^{-3}
	^{115}In	95.71	99.99	3.66790×10^{-2}
^{197}Au	^{197}Au	100	99.95	5.90403×10^{-2}

2. Core Configuration

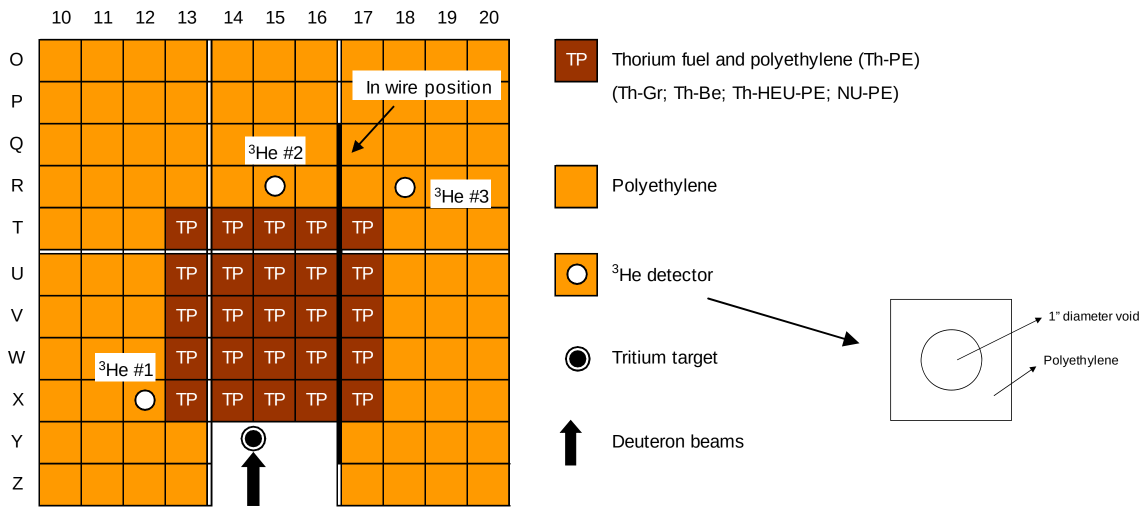

Fig. 2-1 Thorium-loaded ADS core configuration with 14 MeV neutrons (Th-PE, Th-Gr, Th-Be, Th-HEU-PE and NU-PE)

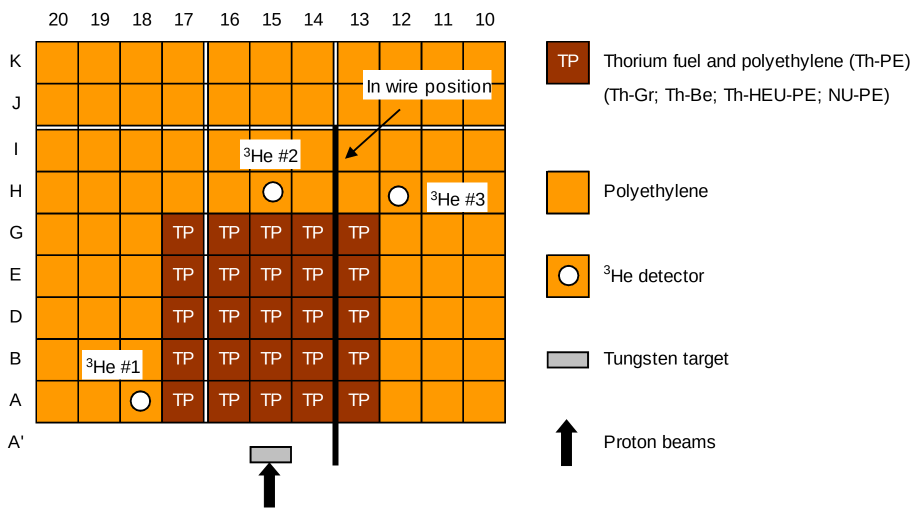

Fig. 2-2 Thorium-loaded ADS core configuration with 100 MeV protons (Th-PE, Th-Gr, Th-Be, Th-HEU-PE and NU-PE)

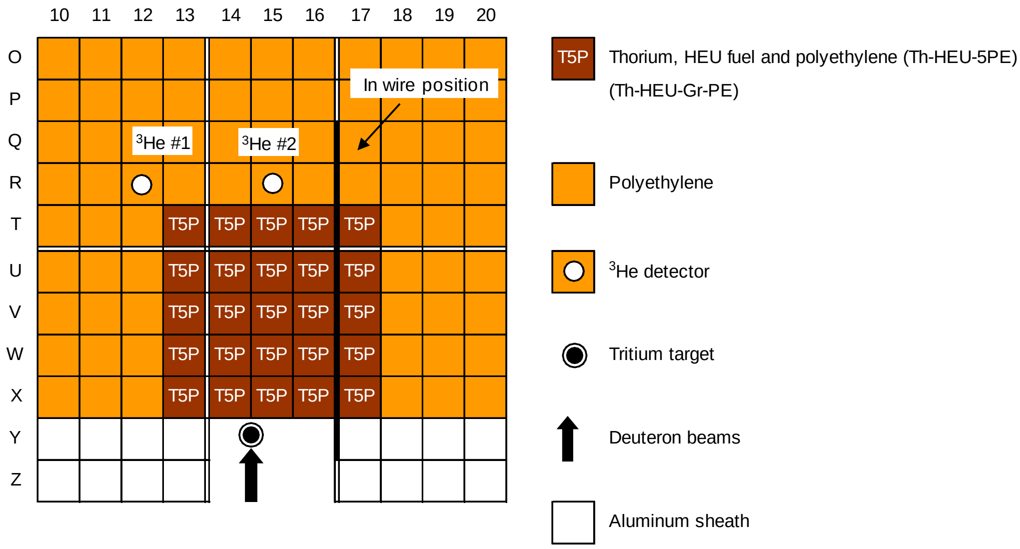

Fig. 2-3 Thorium-loaded ADS core configuration with 14 MeV neutrons (Th-HEU-5PE and Th-HEU-Gr-PE)

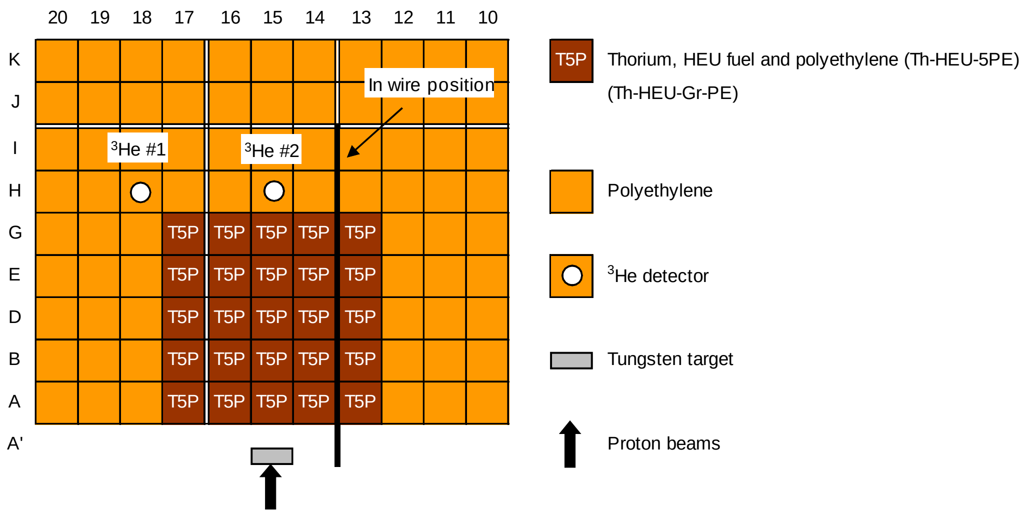

Fig. 2-4 Thorium-loaded ADS core configuration with 100 MeV protons (Th-HEU-5PE and Th-HEU-Gr-PE)

Table 2-1 List of thorium-loaded ADS cores

Core	Cell patter	k_{eff}
Th-PE	1/8”Th+1/2”PE	0.00613
Th-Gr	1/8”Th+1/2”Gr	0.00952
Th-Be	1/8”Th+1/2”Be	0.00765
Th-HEU-PE	1/8”Th+1/16”HEU+3/8”PE	0.58754
NU-PE	1/8”NU+1/2”PE	0.50867
Th-HEU-5PE	1/8”Th+5*(1/16”HEU+3/8”PE)	0.85121
Th-HEU-Gr-PE	1/8”Th+4*(1/16”HEU+1/2”Gr)+1/8”PE	0.35473

Th: Thorium; PE: Polyethylene; Gr: Graphite; Be: Beryllium;
HEU: Highly-enriched uranium; NU: Natural uranium

3. Results of Experiments

3-1. Thorium plate irradiation experiment

Table 3-1 Criticality position at the critical state (Th plate irradiation experiment)

Rod	Rod position [mm]
C1	1201.21
C2	1201.18
C3	567.01
S4	1200.00
S5	1200.00
S6	1200.00
Excess reactivity [%$\Delta k/k$]	0.225

Table 3-2 Measured results of Au foils, Cd ratio and thermal neutron flux

	Parameters	Results in Experiments
Au foil (Bare)	Reaction rate [1/s/cm^3]	$(4.76 \pm 0.02)\times10^8$
Au foil (Cd covered)	Reaction rate [1/s/cm^3]	$(1.62 \pm 0.01)\times10^8$
Cd ratio	-	2.93 ± 0.02
Thermal neutron flux	Neutron flux [1/s/cm^2]	$(5.87 \pm 0.05)\times10^7$

Table 3-3 Nuclides and characteristics of γ-ray measurements of thorium reactions

Reaction	Nuclide	Half-life ($T_{1/2}$)	Emission γ-ray energy [keV]	Emission rate [%]	Yield of fission product [%]
Capture	^{233}Pa	26.95 d	311.9	38.60	-
Fission	^{91}Sr	9.48 h	1024.3	34.01	7.36
	^{92}Sr	2.71 h	1383.9	90.00	6.92
	^{97}Zr	17 h	743.4	94.60	4.43
	^{135}I	6.7 h	1260.5	30.28	5.52
	^{142}La	1.55 h	641.2	48.90	6.54

Table 3-4 Measured results in reaction rates of thorium capture and fission reactions

Reaction	Nuclide	Reaction rate [1/s/cm^3]	Fission rate [1/s/cm^3]
Capture	^{233}Pa	$(5.82 \pm 0.01)\times10^6$	-
Fission	^{91}Sr	$(1.05 \pm 0.01)\times10^4$	$(1.42 \pm 0.04)\times10^5$
	^{92}Sr	$(1.01 \pm 0.01)\times10^4$	$(1.46 \pm 0.05)\times10^5$
	^{97}Zr	$(6.52 \pm 0.04)\times10^3$	$(1.47 \pm 0.02)\times10^5$
	^{135}I	$(8.28 \pm 0.07)\times10^3$	$(1.50 \pm 0.04)\times10^5$
	^{142}La	$(1.14 \pm 0.01)\times10^4$	$(1.56 \pm 0.05)\times10^5$

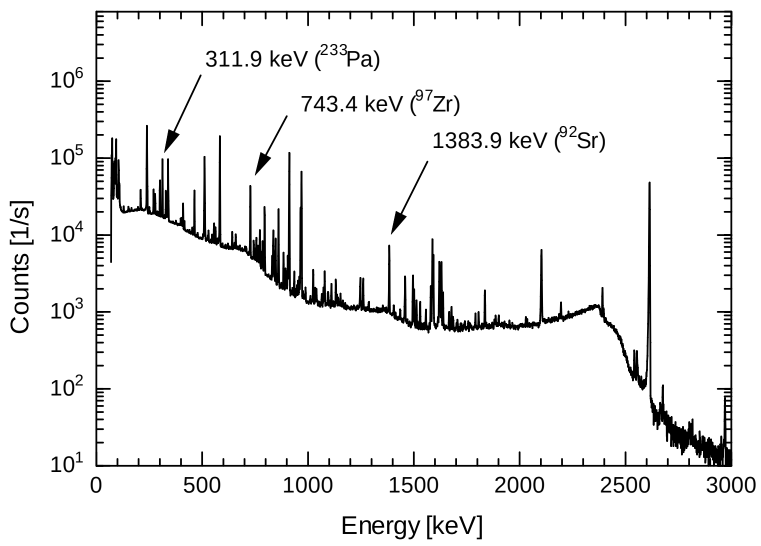

Fig. 3-1 Measured γ-ray spectrum of irradiated thorium plate in the critical state

3-2. Reaction rate distribution

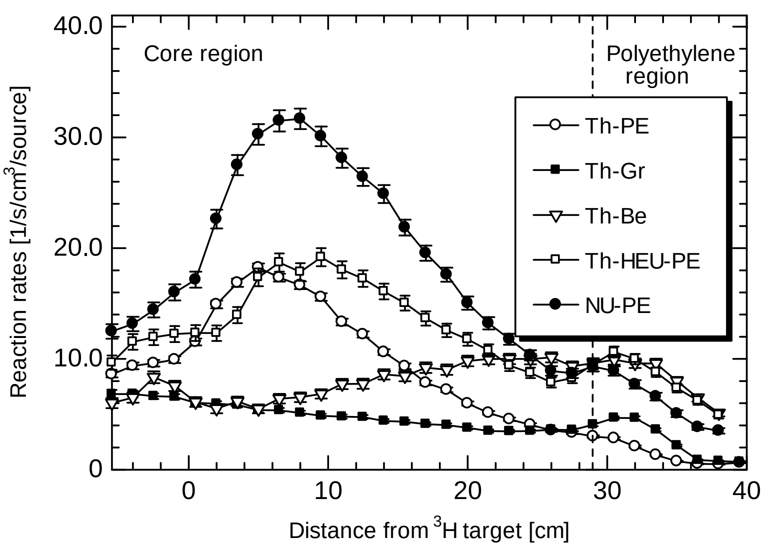

Fig. 3-2 Comparison between the measured reaction rates of indium wire obtained from the Th-loaded ADS experiment with 14 MeV neutrons for Th-PE, Th-Gr, Th-Be, Th-HEU-PE and NU-PE cores

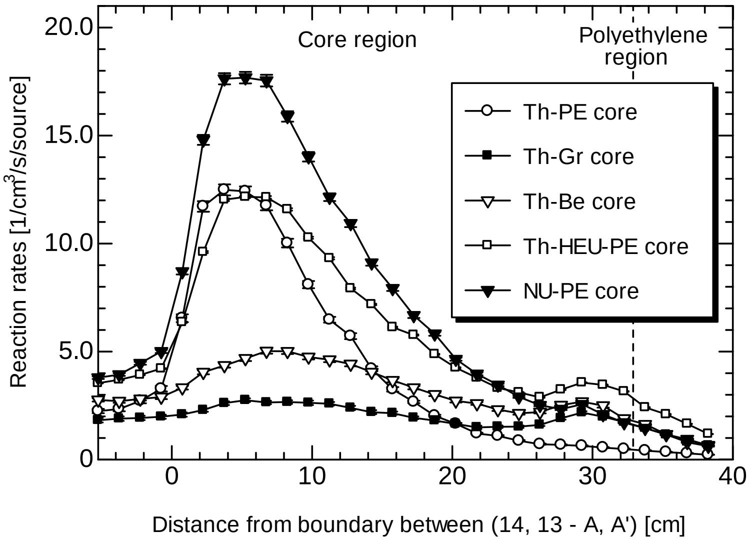

Fig. 3-3 Comparison between the measured reaction rates of indium wire obtained from the Th-loaded ADS experiment with 100 MeV protons for Th-PE, Th-Gr, Th-Be, Th-HEU-PE and NU-PE cores

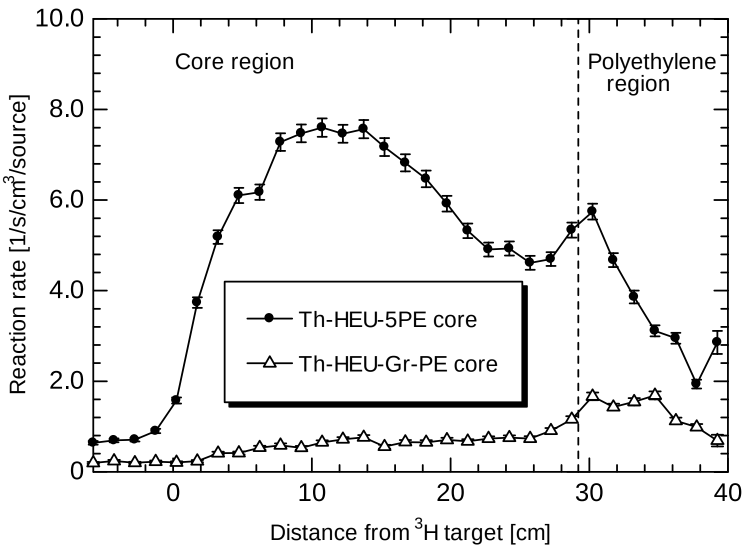

Fig. 3-4 Comparison between the measured reaction rates of indium wire obtained from the Th-loaded ADS experiment with 14 MeV neutrons for Th-HEU-5PE and Th-PE-Gr-PE cores

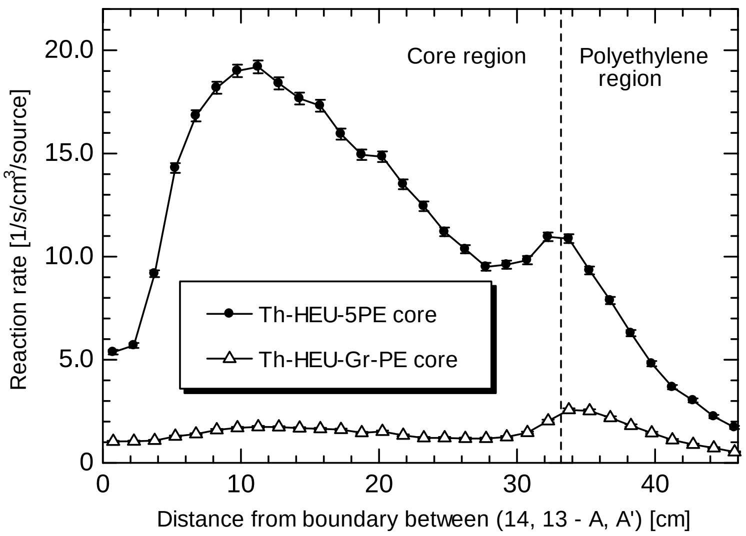

Fig. 3-5 Comparison between the measured reaction rates of indium wire obtained from the Th-loaded ADS experiment with 100 MeV protons for Th-HEU-5PE and Th-PE-Gr-PE cores

3-3. Time evolution data of PNS and Noise methods

Table 3-5 List of subcriticality data in all the cores shown in Table 2-1

Core	14 MeV neutrons		100 MeV protons	
	PNS method	Noise method	PNS method	Noise method
Th-PE	Available	Available	-	-
Th-Gr	Available	Available	-	-
Th-Be	Available	-	-	-
Th-HEU-PE	Available	Available	Available	Available
NU-PE	Available	-	-	-
Th-HEU-5PE	Available	Available	Available	Available
Th-HEU-Gr-PE	Available	Available	Available	Available

PNS method: Pulsed neutron source method; Noise method: Feynman-α method

Table 3-6 Time evolution data of thorium-loaded cores on the PNS and Noise methods

Core	14 MeV neutrons		100 MeV protons	
	Repetition [Hz]	Width [μs]	Repetition [Hz]	Width [ns]
Th-PE	100	10	-	-
Th-Gr	100	10	-	-
Th-Be	100	10	-	-
Th-HEU-PE	100	10	20	60
NU-PE	100	10	-	-
Th-HEU-5PE	10	10	20	100
Th-HEU-Gr-PE	10	10	20	100

Table 3-7 Measured results in neutron decay constants deduced by least-squared fitting in PNS method

	Neutron decay constant α [1/s]					
	14 MeV neutrons			100 MeV protons		
Core	^{3}He #1	^{3}He #2	^{3}He #3	^{3}He #1	^{3}He #2	^{3}He #3
Th-PE	6642±11	6224±27	5751±25	-	-	-
Th-Gr	6451±12	5945±15	5701±17	-	-	-
Th-Be	6515± 8	6111±17	5746±20	-	-	-
Th-HEU-PE	5692±11	5275± 7	5231± 9	5777±11	5527±35	5236±37
NU-PE	5748±11	6592±15	5010±11	-	-	-
Th-HEU-5PE	3110±11	3104±10	-	2776±17	2917± 8	-
Th-HEU-Gr-PE	4980±40	4939±50	-	3186±72	4656±12	-

Table 3-8 Measured results in subcriticality in dollar unit deduced by extrapolated area ratio method

	Subcriticality ρ [$] (by Area ratio method)					
	14 MeV neutrons			100 MeV protons		
Core	^{3}He #1	^{3}He #2	^{3}He #3	^{3}He #1	^{3}He #2	^{3}He #3
Th-PE	144.77±1.56	1051.97±28.70	2778.37±135.26	-	-	-
Th-Gr	108.47±18.06	400.34±5.92	880.49±20.41	-	-	-
Th-Be	33.42±0.17	37.04±0.16	105.35±0.85	-	-	-
Th-HEU-PE	12.05±0.03	29.75±0.09	63.53±0.35	32.30±0.45	26.52±0.29	44.27±0.74
NU-PE	13.40±0.04	26.64±0.12	82.76±0.57	-	-	-
Th-HEU-5PE	16.61±.0.21	11.35±0.09	-	3.77±0.01	10.35±0.05	-
Th-HEU-Gr-PE	63.76±1.42	42.92±0.66	-	3.83±0.30	64.54±11.13	-