CO2-1 Measurement of Gamma Ray and Neutron Spectrum of Curium Isotope (2)

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INTRODUCTION: The high energy (>3MeV) gamma rays (HEγs) would be an indicator of fuel materials since they are radiated either by the spontaneous fission, the neutron induced fission, or the radiative capture. When we apply the HEγ measurements to the fuel material accountancy, HEγ from the spontaneous fissions would be the significant background. For the subcriticality determination, the yield ratio of HEγ to neutron of the spontaneous fission is useful as well as that of the induced fissions. For the reason, measurements of HEγs and neutrons have been conducted for Cm isotopes [1].

CHARACTERIZATION OF SAMPLE: A disk type sample of Cm (CM246) sealed in an aluminum can was prepared. The isotopic composition was once measured in 2010 by thermal ionization mass spectrometry (TIMS) [2] and their activities are listed in Table 1. We measured a γ ray spectrum from CM246 and indentified γ rays from α-decay of ^{243,244,245}Cm (Fig.1). By using an ^{152}Eu calibration source, the absolute efficiency of γ ray detection was estimated. As for ^{244}Cm, the deduced activity is 1.43x10^{9}Bq(±11.9%) and agrees with one by TIMS.

NEUTRON MEASUREMENT: Neutrons from spontaneous fissions in CM246 and CM248 (Table 1, [1]) samples were measured by the H(n,γ) method [3]. As shown in Fig. 2, Polyethylene blocks of 35cm x 30cm x 30cm were piled as making a cavity of 10cm x 10cm x 5cm at the central part. We put the samples in the cavity and a Ge detector 10cm apart from the block and 2.223MeV γ rays were measured for each sample. The detection efficiencies for a neutron emission were calculated with the MCNP-5 code [4]. For the calculation, the neutron source spectra estimated by the SOUCES4C code [5] were used. In Table 1, the neutron yields by the H(n,γ) method are compared to those calculated by SOUCES4C based on the composition listed in Table 1. There is a discrepancy for CM246. The authors will review validity of H(n,γ) method, the isotopic composition and the database used in SOURCES4C. In addition, the authors shall measure radiations from another Cm sample of a different isotopic composition.

HIGH ENERGY GAMMA RAY MEASUREMENT: With a BGO detector of 3" in length and 3" in diameter, HEγ was measured for each sample. To reduce direct radiation of neutrons to the BGO, polyethylene blocks of 40cm thickness containing 10wt% boron are set between the sample and BGO although they were on a stage made of iron. The measured pulse height spectra of HEγ are compared in Fig. 3. The spectra consist of continuum regions (< 5MeV) and higher energy bumps. The former is the spontaneous fission γ rays and the latter is the capture ones by Fe(n,γ) reactions. It was found that fission γ ray spectra are almost similar for Cm isotopes and ^{252}Cf.

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Table1 Sample and neutron yield by H(n,γ) method.

Sample		CM246 (Bq)	CM248 (Bq)
Activity by	244Cm(Bq)	1.59E+09	1.90E+03
TIMS(CM246) or	246Cm(Bq)	1.48E+07	1.27E+05
α spect.(CM248)	248Cm(Bq)	3.13E+04	4.08E+04
neutron yield by SOURCES4C(/s)		2.62E+04	1.07E+04
Count rate of 2.2MeV γ ray (/s)		1.44E+00	1.07E+00
Efficiency by MCNP		2.50E−04	2.42E−04
n yield by H(n,γ) method (/s)		1.25E+04	9.54E+03

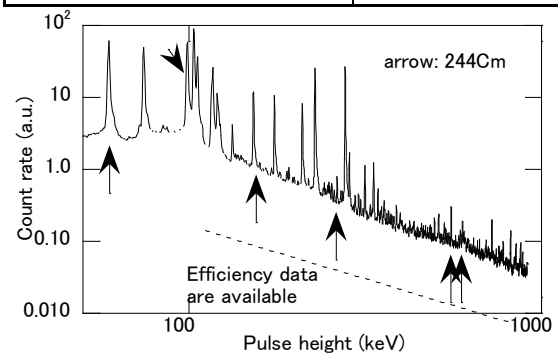

Fig.1 Low energy γ ray pulse height spectra from α decay of CM246 sample. ^{244}Cm components are pointed.

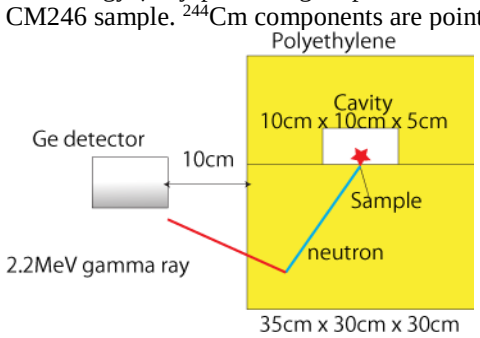

Fig. 2 Geometry of H(n,γ) measurements.

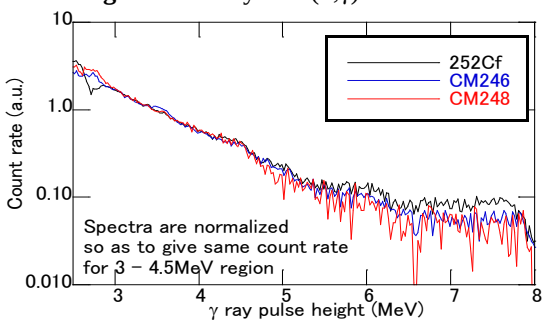

Fig. 3 γ ray pulse height spectra from Cm and Cf sam-ples.

CO2-2

Development of Absolute Measurement Method for Epi-Thermal Neutrons Using a LGB Scintillator

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INTRODUCTION: Evaluation of neutron dose equivalent for the epi-thermal neutron region is very important in work places with neutron sources or nuclear fuels as well as irradiation fields in a boron neutron capture therapy. A new calibration method for the response of neutron dosimeters has been developed using a pulsed white neutron beam produced by the KURRI-LINAC and the 4 MV Pelletron accelerator of AIST [1]. On the other hand, it is not easy to measure the neutron fluence with small uncertainty for epi-thermal neutrons in an irradiation field, although the fluences for thermal and fast neutrons are precisely determined using the gold activation reaction and the elastic neutron scattering reaction with hydrogen atoms. In the present study, we have developed an absolute measurement method for epi-thermal neutrons using a $^{6}Li_{6}{}^{nat}Gd^{10}B_3O_9$:Ce+ (LGB) and an NaI(Tl) scintillators. In future, the absolute measurement method developed in the KURRI experiments will be used for determination of neutron fluence in the 24-keV mono-energetic neutron standard field at AIST.

EXPERIMENTS: A neutron beam through collimators was obtained by the photo-neutron reaction using a water-cooled tantalum target at the KURRI Linac [2]. The 50-mm diameter and 5-mm thick LGB scintillator was set at the center of the neutron beam. The 5.08 cm-diameter and 5.08 cm thick NaI(Tl) was set out of neutron beam as shown in Fig.1. When the LGB scintillator detects neutrons by the $^{10}B(n,\alpha\gamma)$ reaction, 478 keV monoenergetic gamma rays are produced and subsequently detected with the NaI(Tl) scintillator as shown in Fig.2. In the coincidence measurements, the neutron capture reaction rate in the LGB scintillator is obtained without detection efficiencies of the LGB and the NaI(Tl) scintillators. Moreover, gamma rays from the $^{10}B(n,\alpha\gamma)$ reaction are measured with the NaI(Tl) scintillator in setting a thick ^{10}B total absorption sample in front of the LGB scintillator. The absolute neutron fluence is determined by comparison between results obtained from the measurements with and without the thick ^{10}B sample. On the other hand, although neutrons are detected by the $^{10}B(n,\alpha\gamma)$, the $^{6}Li(n,\alpha)T$ and the $Gd(n,\gamma)$ reactions by the LGB scintillator, only a peak due to the $^{10}B(n,\alpha\gamma)$ reaction is appeared by the coincidence measurements. Reliability of this absolute measurement method will be investigated using Monte-Carlo simulation and measurements under other beam conditions in future.

In the present study, we also try to measure responses of manganese activation detectors. The responses are obtained by measuring transmitted neutrons with and without the manganese activation detector. Neutron resonances in total cross sections of ^{55}Mn at neutron energies of 341 eV, 1.10 keV and 1.66 keV are used in the measurements. The results obtained from the measurements will be used in a manganese bath detector for determination of neutron emission rate of a radioactive neutron source.

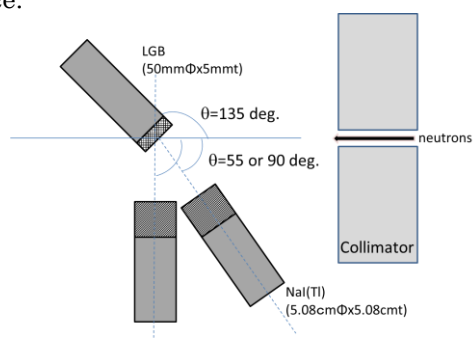

Fig. 1. Typical experimental setup

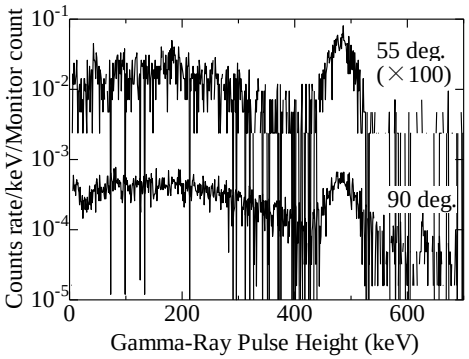

Fig.2. Pulse height spectra of the NaI(Tl) scintillator obtained from the coincidence measurements with the LGB and NaI detector.

ACKNOWLEDGMENT
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採択課題番号 25050 熱中性子フルエンス率の測定の高度化とその国際標準化に関する研究 共同通常
（産総研）原野英樹、松本哲郎、増田明彦（名大）井口哲夫、瓜谷章、河原林順、渡辺賢一、
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CO2-3 Experimental Study on Non-Destructive Assay with a Pulsed Neutron Source

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INTRODUCTION: Non-destructive assay of nuclear fuel materials in spent nuclear fuel is a key technology for nuclear material accountancy and critical safety. Neutron Resonance Densitometry (NRD) with a pulsed neutron source has been developed for a non-destructive assay of material [1]. In this method, we can identify and quantify the target nuclide by measuring the neutron resonance absorption with the Time-of-Flight (TOF) technique. There are two ways to measure the neutron resonance absorption. One is Neutron Resonance Transmission Analysis (NRTA) [2] and another is Neutron Resonance Capture Analysis (NRCA) [3, 4].

However, there are difficulties to apply an assay for spent fuel. In the NRTA, many resonances of other nuclides which are contained in the fuel may make it difficult to identify and quantify the target nuclide. In the NRCA, it is expected that the intense decayed gamma rays from the fuel result in high background and large dead time of gamma-ray detector.

In this work, we have proposed a new concept of "self-indication method" as a complementary assay to overcome those difficulties. In the self-indication method, we set an indicator consisted of target nuclide with a high purity at the beam-downstream from a sample. By detecting the reaction products such as neutron capture gamma rays or fission products from the indicator with the TOF method, the transmission neutron can be measured indirectly. The self-indicator is a transmission neutron detector which has a high efficiency around the objective neutron resonance energies of target nuclide, so that it enables us to quantify effectively the amount of resonance absorption of the target nuclide. Moreover, it is hard to be affected by the decayed gamma rays from the fuel. We carried out the experimental validation for application of the self-indication method. The details of experiment were reported in Ref. [5].

EXPERIMENTS: In order to verify the self-indication method, we performed the experiments using a 46-MeV electron linear accelerator at the Research Reactor Institute (KURRI-LINAC). A sample and an indicator were located at a distance of 11.0 m and 12.7 m from the photo-neutron source, respectively. The capture gamma rays from the indicator were measured by a $Bi_4Ge_3O_{12}$ (BGO) assembly, which consists of 12 scintillation detectors [6]. At first, the area-densities of gold foils with different thickness were estimated by area analysis for the 4.9-eV resonance of ^{197}Au. At the next step, a 50-μm thick silver foil was added to a 10-μm thick gold foil to form a sample, and the area-density of gold foil was measured. It is worth noting that silver has a large resonance at 5.2 eV close to 4.9 eV resonance of gold. Finally we demonstrated a non-destructive assay to nuclear materials using a mixture composed of a natural uranium plate and sealed minor actinide samples of ^{237}Np and ^{243}Am.

RESULTS: The TOF spectra obtained by both methods of the NRTA (dot lines) and the self-indication (solid lines) around the 4.9-eV resonance of ^{197}Au are shown in Fig. 1. The resonance absorption due to the 4.9-eV resonance was observed as a dip and a lack of peak for the NRTA and the self-indication, respectively. It was confirmed that the area-densities of the target nuclide can be determined by the self-indication method within 3 % accuracy. Moreover, it was shown that the contribution from the other nuclide can be remarkably suppressed by applying the self-indication method. It will be useful tool for non-destructive assaying the distribution of nuclear material in the spent fuel or the melted fuel debris.

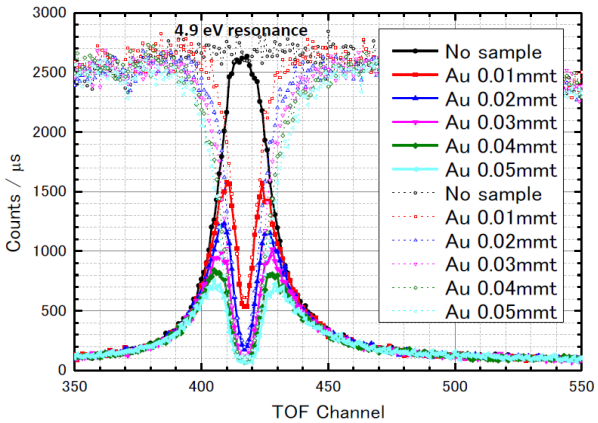

Fig. 1. Comparison of TOF spectra obtained by NRTA and self-indication method for 4.9 eV resonance of ^{197}Au

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CO2-4 Measurement of Prompt Gamma-rays for Therman-Neutron Induced Fission of ^{235}U

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INTRODUCTION: The Great East Japan Earthquake has damaged the reactors of Fukushima I Nuclear Power Plants, which consequently caused the nuclear fuel to melt. The Japanese government has a plan to recover the land by the full deconstruction of the power plants, for which the first action should be a removal the melted fuel (debris). This operation must be carried out without reaching the criticality of the melted core. This requires the use of an adopted surveillance system. This system necessitates an idea to detect increased fission rate correlated with reaching the criticality. The detection system should also work in a strong γ-ray field arising from the fission products contained in the debris.

We are proposing to develop a system based on detecting high-energy prompt γ-rays originating from fission events to monitor the increased fission rate. The measured γ-ray energy spectrum for the spontaneous fission of ^{252}Cf shows a high energy γ-ray component around E_γ = 14 MeV associated with giant dipole resonance (GDR) [1]. The energy is significantly larger than that for the background γ-rays from the fission products in debris. The high energy γ-rays can penetrate the material shielding the surveillance detector, necessary to reduce the low energy but intense background γ-rays.

To design such a surveillance detector and its sensitivity to criticality, it is essential to know the spectrum of prompt fission γ-rays up to more than 8 MeV for thermal neutron-induced fission of ^{235}U, ^{235}U(n_{th},f). No data are, however, available in the energy range larger than 7 MeV [2]. The purpose of this experimental campaign is to measure the energy spectrum up to around 16 MeV.

EXPERIMENTS: First, the measuring conditions, such as background γ-ray yield and spectrum, were experimentally measured. A test experiment to detect prompt γ-rays from fission fragments in ^{235}U(n_{th},f) was then carried out.

The experiment was performed at the B4 super-mirror neutron guide tube of the Research Reactor Institute, Kyoto University. The enriched uranium target (enrichment 99.1%) was made by a filtration method [3] with a thickness of about 1 mg/cm^2, where the uranium material was deposited on a circular area with diameter 18 mm. Fission fragments were detected by two multi-wire proportional counters (MWPC) with an active area of 80 × 80 mm^2, located on both sides of the targets at a distance of 50 mm. Due to the target-backing thickness, only single fission fragments can be emitted from the target layer. Two targets were mounted back to back such that fission fragments are detected by each MWPC. The MWPC was operated with isobutene gas.

Prompt γ-rays accompanied by fission were detected by a $LaBr_3$(Ce) detector located 200 mm from the uranium target. The detector has a cylindrical shape with a length of 127 mm and a diameter of 102 mm. The γ-ray detector was surrounded by lead blocks with a thickness of 100 mm except the entrance to the γ-ray detector, which was furthermore surrounded polyethylene blocks at a thickness of 50 mm. The entrance of the γ-ray detector was shielded by a ^{nat}LiF plate to prevent the scattered neutron entering in the γ-ray detector. Pulse heights from the $LaBr_3$ detector and cathode signals from two MWPCs as well as time differences between the $LaBr_3$ and MWPCs were recorded with a VME based data acquisition system. The experiment was carried out at a 1 MW operation of the KUR (expected neutron flux is 1.0×10^7 /s/cm^2). Typical counting rates for fission fragments and γ-rays were 4.6×10^3 cps and 4.6×10^3 cps, respectively.

RESULTS: Figure 1 shows the γ-ray spectrum for ^{235}U(n_{th},f) obtained in the 70 min run. The peaks at 7.7 MeV originate from the γ-rays in the neutron capture of aluminum, which is not fully subtracted in the analysis. To obtain a spectrum at higher energy region with reasonable beam time, we plan to employ a larger amount ^{235}U material.

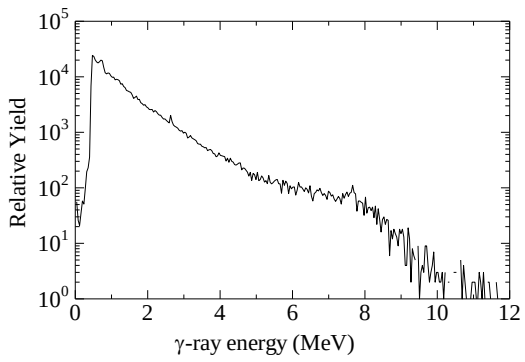

Fig. 1 Spectrum of prompt fission γ-ray for ^{235}U(n_{th},f)

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採択課題番号 25120　新たな未臨界監視検出器をめざした核分裂高エネルギーガンマ線の測定　共同即時
（原子力機構・先端基礎センター）西尾勝久、牧井宏之、廣瀬健太郎、西中一朗、Riccardo Orlando、Romain Léguillon、James Smallcombe（京大・原子炉）関本　俊、高宮幸一、大槻　勤

CO2-5 Neutronic Characteristics of Lead-Bismuth in KUCA A Core for Accelerator-Driven System

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INTRODUCTION: The Japan Atomic Energy Agency (JAEA) has investigated the accelerator-driven system (ADS) to transmute minor actinides discharged from nuclear power plants. The ADS investigated by JAEA is a lead bismuth eutectic (LBE) cooled-tank-type ADS. It has been known that there was a major upgrade for the cross section data of lead isotopes from JENDL-3.3 to JENDL-4.0. Due to this upgrade, the value of k_{eff} of the core was significantly changed, from 0.97 calculated by JENDL-3.3 to 1.00 calculated by JENDL-4.0 [1]. The difference was mainly caused by the cross section data of the lead isotopes [1] from JENDL-3.3 to JENDL-4.0.

This study aims to measure sample worth reactivity from aluminum plates to lead or LBE ones to validate the nuclear data of lead and bismuth isotopes.

EXPERIMENTS: The A-core (EE1) of KUCA was employed for the experiment. Figure 1 shows the A-core of KUCA and Fig. 2 presents the composition of fuel assembly and special assemblies. The special assemblies were composed of the Al or Pb plates instead of polyethylene, and were placed at the center of the core. The core with five Al special assemblies was the reference case in this study. Then, the Al special assembly was replaced to the Pb special assembly and the difference of excess reactivities were calculated as the sample worth reactivity.

Four experiment cases (Pb-3X, 3Y, 4, 5) as shown in Fig. 3 were carried out. Three to five Al special assemblies were substituted for the Pb ones to measure the sample worth reactivity.

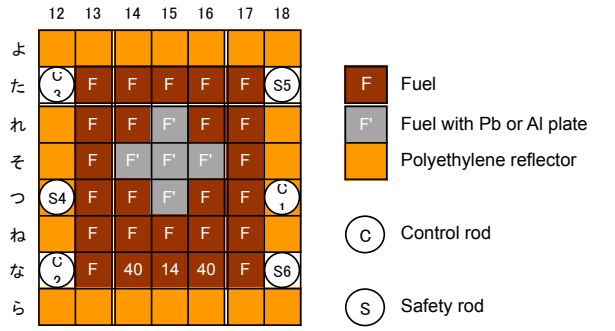

Fig. 1 Core configuration of A-core (EE1) of KUCA

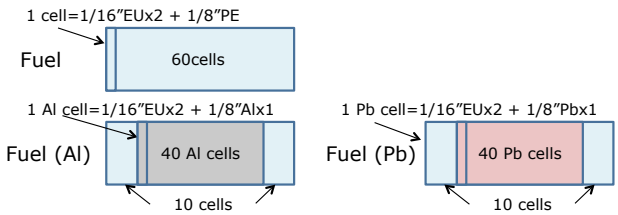

Fig. 2 Composition of fuel and special assemblies

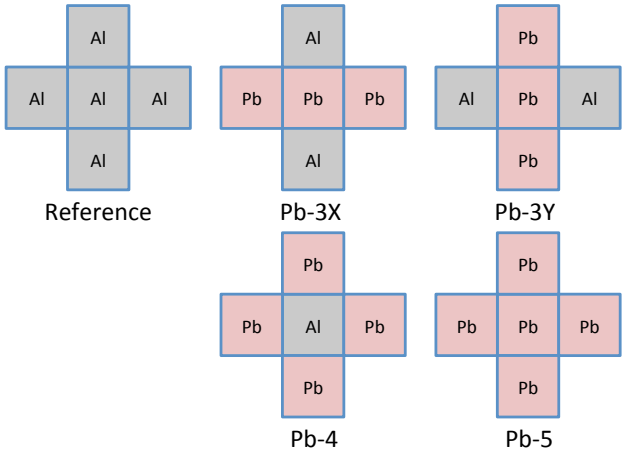

Fig. 3 Experiment cases to measure sample worth reactivity

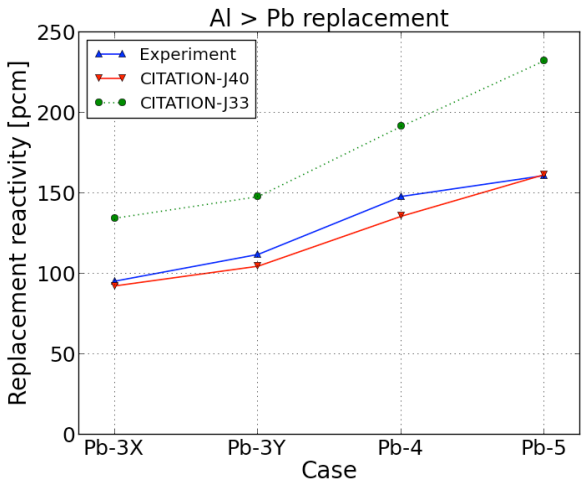

Fig. 4 Comparison between measured and calculated results

RESULTS: Figure 4 shows the experimental and calculation results. SLAROM-CITATION codes on the MARBLE system [2] were employed for the calculation with JENDL-4.0 and JENDL-3.3 libraries. As the number of the Pb plates was increased, the sample worth reactivity from Al to Pb was also large. The sample worth reactivity of 162 pcm was measured in the Pb-5 case. The calculated results with JENDL-4.0 revealed good accuracy with the comparison with the measured ones. The C/E values based on JENDL-4.0 were significantly ranging between 0.92 and 1.01, whereas, those based on JENDL-3.3 were less accurate between 1.30 and 1.45. From these results, the nuclear data of the lead isotopes in JENDL-4.0 were considered more reasonable than those in JENDL-3.3.

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採択課題番号 CA2501 KUCA-A 架台を用いた加速器駆動炉における鉛ビスマスの核特性評価 共同通常（原子力機構）菅原隆徳、大泉昭人、北村康則、岩元大樹（京大・原子炉）卞哲浩、八木貴宏