

한국원자력학회
2021년 춘계 학술대회

A Study on the Uranium Enrichment Determination using Standard-less Gamma Spectrometry Technique

김 우 진

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- 3 분석 방법
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1. 개요

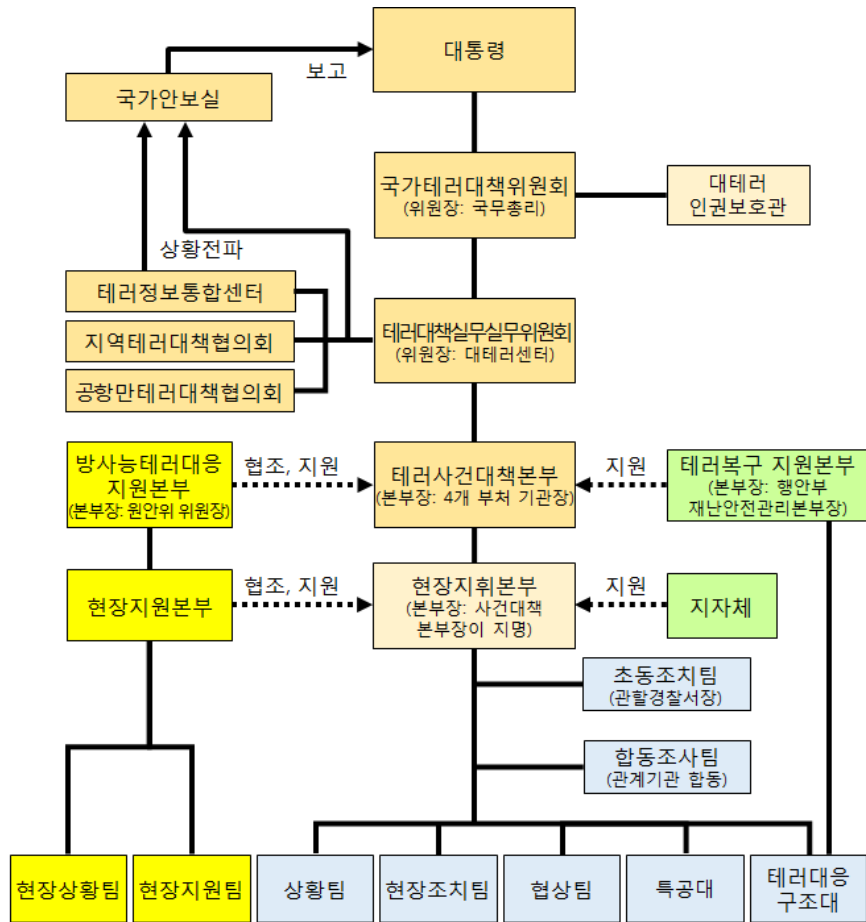
1. 개요 : 우라늄 농축도 측정 필요성

◆ 시나리오 : 핵물질 밀수/도난 등의 불법행위 의심 상황 발생

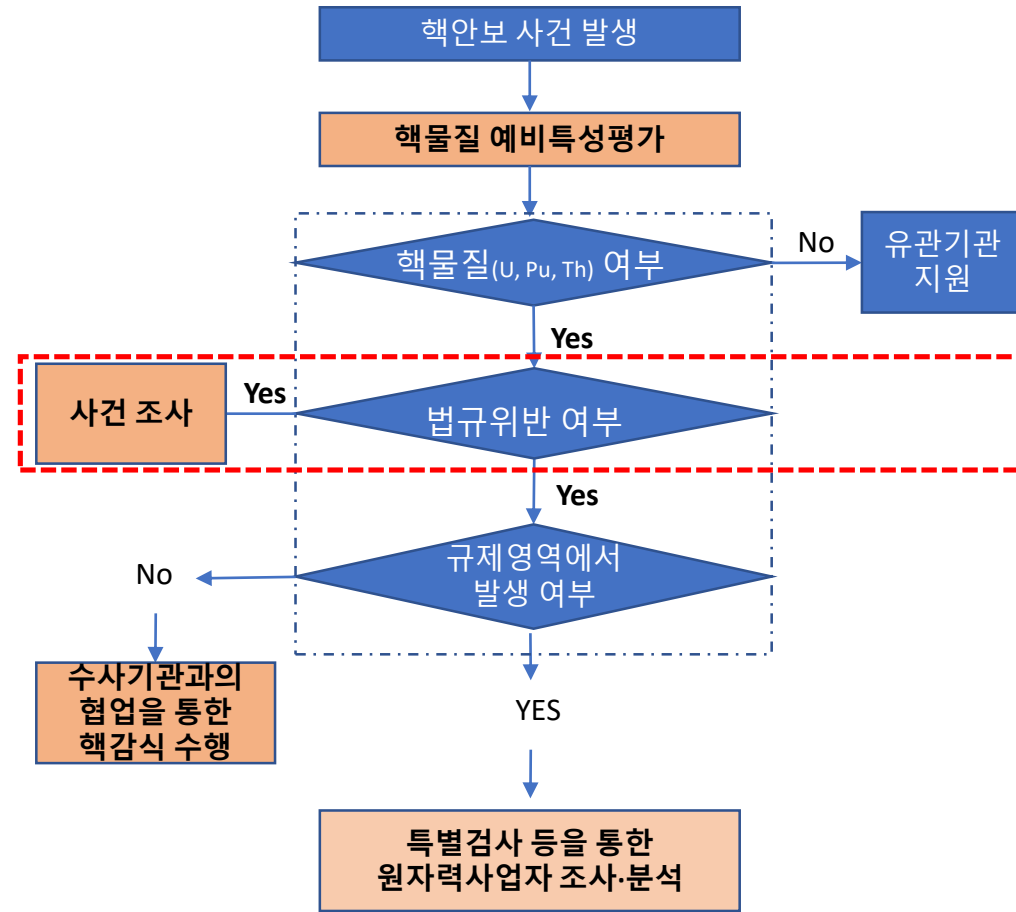


1. 개요 : 우라늄 농축도 측정 필요성

◆ 국가 방사능 대응 체계



국가 수준



전문기관(KINAC) 수준

1. 개요 : 우라늄 농축도 측정 필요성

◆ 미확인 핵물질 예비특성평가 개념

- 현장에서 신속하게 측정, 기준과의 비교 및 해석을 통해 법집행에 대한 의사결정에 도움을 주기 위한 과학적 기법

◆ 주요 예시



음주 단속 기준 : 도로교통법 제44조 제4항
- 운전자가 술에 취한 상태의 기준 :
혈중 알코올 농도가 0.03% 이상

휴대용 음주측정기와 경찰장비 비교

	휴대용 장비 측정값	경찰 장비 측정값
소주 4잔 먹고 난 뒤 	 0.03/0.06 등 수치가 오락가락했음. 0.06은 입건대상이지만, 0.03은 혼란조치에 해당하는 것	 .025 %BAC <5/01/15 14:20 0025>
소주 2병 먹고 난 뒤 		 .115 %BAC <5/01/15 14:20 0025>

휴대용 음주측정기와 경찰장비 비교/2015-01-18(한국일보)

1. 개요 : 우라늄 농축도 측정치 비교 기준

◆ 원자력안전법 제45조, 동법 시행령 71조 (핵물질 사용/소지를 위한 허가 요건)

제71조(사용허가가 필요하지 아니한 핵연료물질) 법 제45조제1항제3호에서 "대통령령으로 정하는 종류 및 수량의 핵연료물질"이란 다음 각 호의 어느 하나에 해당하는 핵연료물질을 말한다.

1. 우라늄 238에 대한 우라늄 235의 비율이 천연혼합물과 같은 우라늄 및 그 화합물의 경우에는 우라늄의 양이 300그램 이하인 것
2. 우라늄 238에 대한 우라늄 235의 비율이 천연혼합물에 미달하는 우라늄 및 그 화합물의 경우에는 우라늄의 양이 300그램 이하인 것
3. 제1호 또는 제2호의 물질이 하나 이상 함유된 물질로서 원자로의 연료로 사용되는 물질의 경우에는 우라늄의 양이 300그램 이하인 것
4. 토륨 및 그 화합물의 경우에는 토륨의 양이 900그램 이하인 것
5. 제4호의 물질이 하나 이상 함유된 물질로서 원자로의 연료로 사용되는 물질의 경우에는 토륨의 양이 900그램 이하인 것
6. 그 밖에 방사선장해 발생의 우려가 없다고 위원회가 정하여 고시하는 것

종류		수량	농도
감손우라늄		300 grams	N/A
천연우라늄		300 grams	N/A
그 외	U-235	1×10^4 Bq	1×10^1 Bq/g
	U-238	1×10^4 Bq	1×10^1 Bq/g

천연혼합물
(0.712 %)

1. 개요 : 우라늄 농축도 측정치 비교 기준

◆ 방사능방재법 제8조, 동법 시행령 15조 (물리적방호를 위한 핵물질 등급분류)

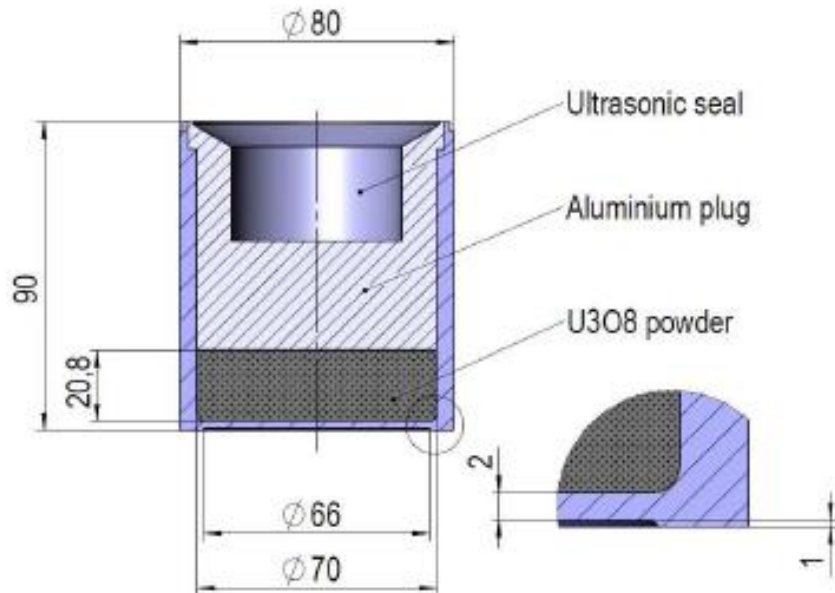
핵물질		등급		
물질	형태	등급 I	등급 II	등급 III
1. 플루토늄	미조사(未照射)	2킬로그램 이상	500그램 초과 2킬로그램 미만	15그램 초과 500그램 이하
2. 우라늄 235	우라늄 235의 농축도가 <u>20퍼센트 이상</u> 인 미조사 우라늄	5킬로그램 이상	1킬로그램 초과 5킬로그램 미만	15그램 초과 1킬로그램 이하
	우라늄 235의 농축도가 <u>10퍼센트 이상 20퍼센트 미만</u> 인 미조사 우라늄		10킬로그램 이상	1킬로그램 초과 10킬로그램 미만
	우라늄 235의 농축도가 <u>천연우라늄의 농축도 초과 10퍼센트 미만</u> 인 미조사 우라늄			10킬로그램 이상
3. 우라늄 233	미조사	2킬로그램 이상	500그램 초과 2킬로그램 미만	15그램 초과 500그램 이하
4. 조사(照射)된 연료			핵분열성물질 10퍼센트 미만의 감손우라늄, 천연우라늄, 토륨 또는 저농축연료	

2. 물질 및 장비

2. 물질 및 장비

◆ CBNM Uranium Set (“EC NRM 171” or “NBS SRM 969”)

Reference material	Mass %				
	²³⁵ U	²³² U [Ref. 5]	²³⁴ U	²³⁶ U	²³⁸ U
CBNM 031	0.3166(2)	$2.1(5) \times 10^{-10}$	0.0020(2)	0.0146(3)	99.6669(4)
CBNM 071	0.7119(5)	$< 1 \times 10^{-11}$	0.0052(2)	< 0.00002	99.2828(4)
CBNM 194	1.9420(14)	$3(3) \times 10^{-11}$	0.0171(2)	0.0003(1)	98.0406(18)
CBNM 295	2.9492(21)	$6(2) \times 10^{-11}$	0.0279(4)	0.0033(2)	97.0196(29)
CBNM 446	4.4623(32)	$3.7(3) \times 10^{-10}$	0.0359(3)	0.0068(2)	95.4950(32)



Can	Height	90 ± 0.2
	Outer diameter	80 ± (no data)
	Edge below window	1 ± 0.05
Can window	Thickness	2 ± 0.02
	Deviation from flatness	± 0.1
	Diameter	66 ± 0.05
U ₃ O ₈ filling	Height for material "446"	15.8 ± 0.5
	Height for other materials	20.8 ± 0.5
	Diameter	70 + 0.05 - 0

2. 물질 및 장비

◆ ORTEC Micro Detective, Electrically Cooled, HPGe Detector



Crystal : 50mm X 30mm

Resolution : 2.0 keV at 1332 keV, 1.0 keV at 122 keV

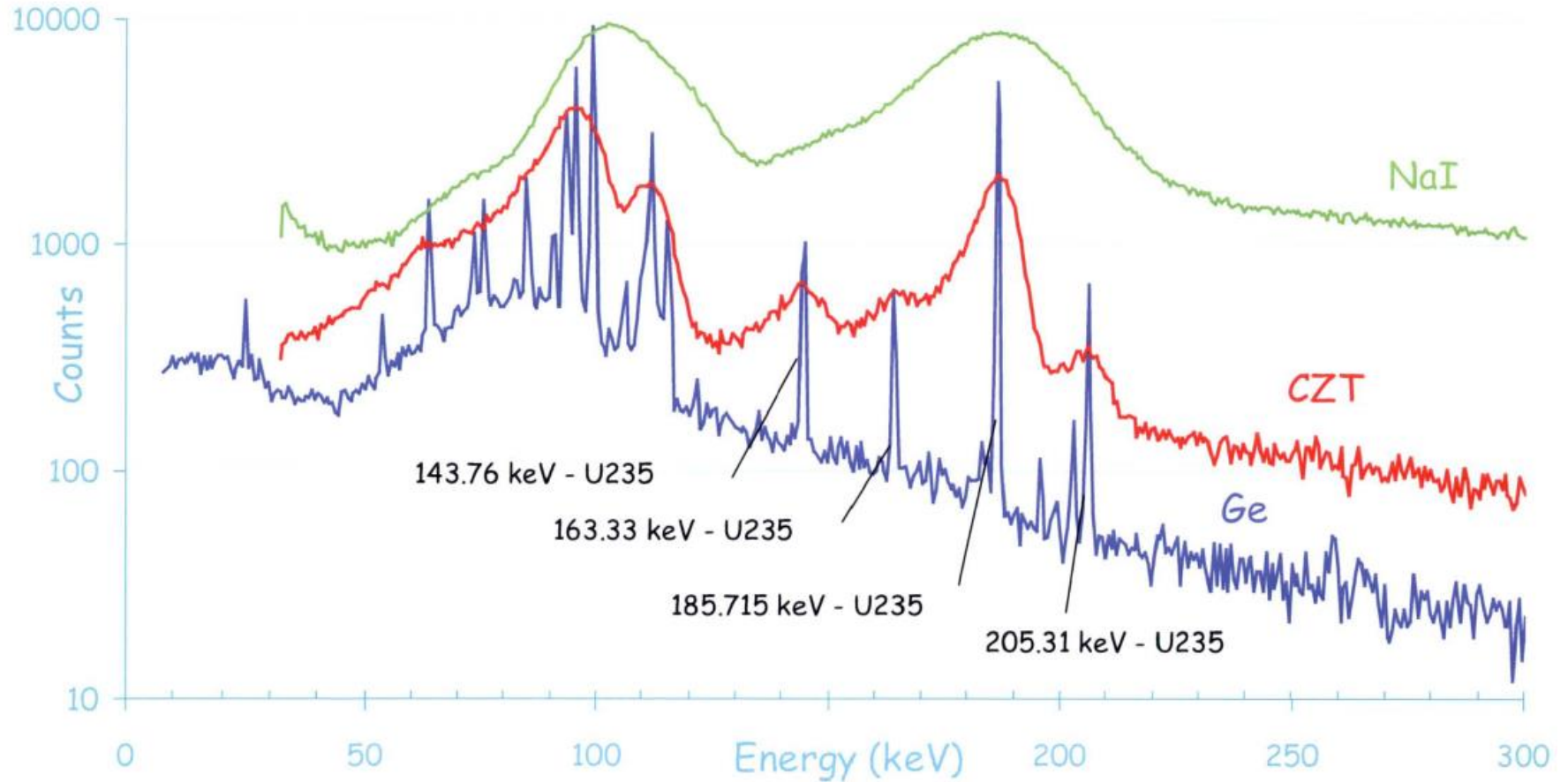
Total MCA Channels : 8192 ch

Conversion gain : 0.36621 keV/ch

3. 분석 방법

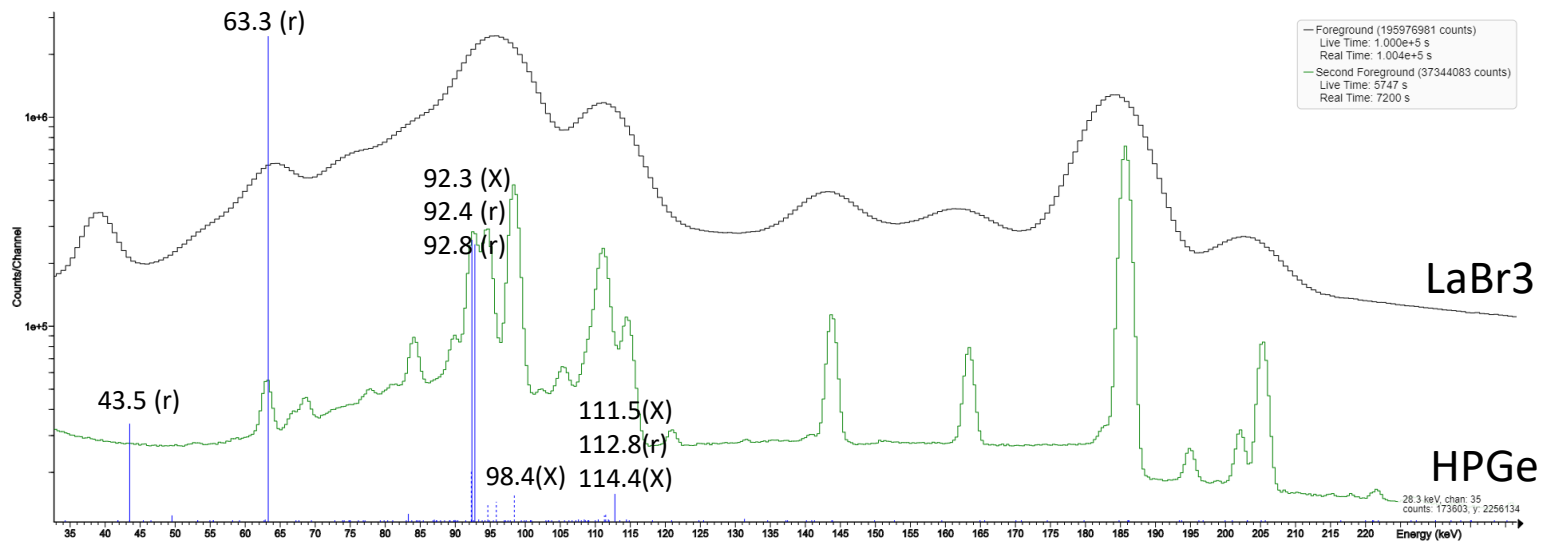
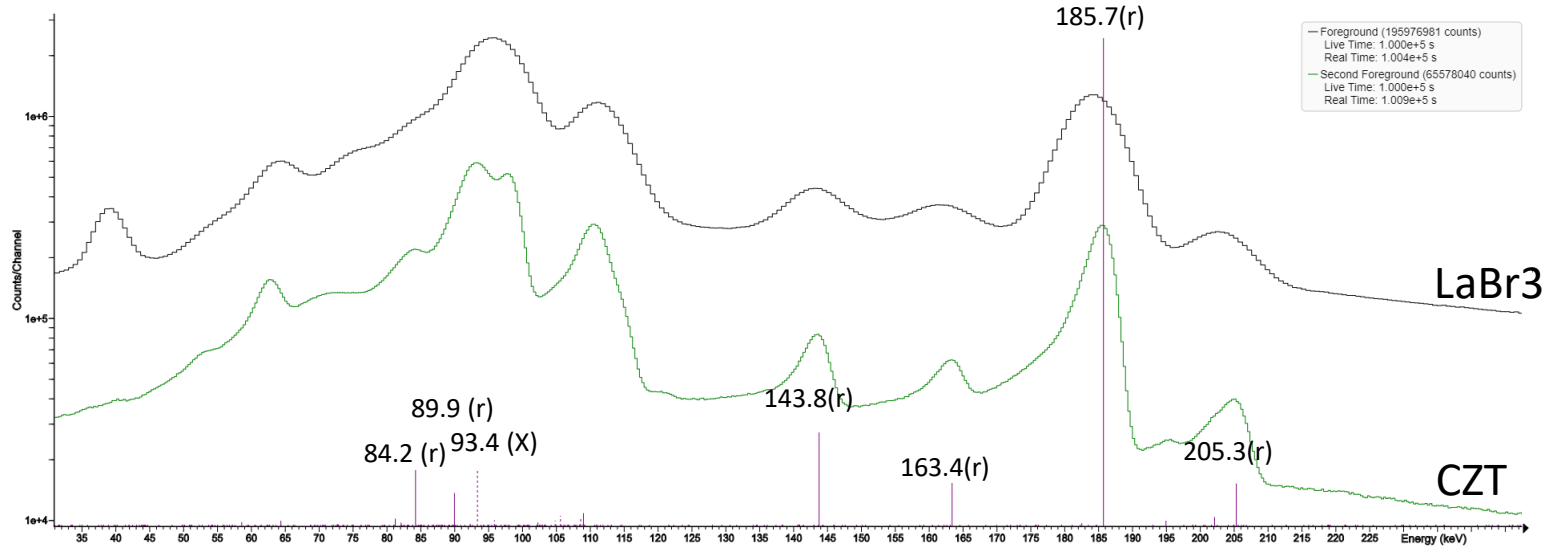
3. 분석 방법

◆ Uranium Spectra (Uranium Only)



3. 분석 방법

◆ Uranium Spectra (2.95 w/o)



3. 분석 방법

◆ Peak Ratio Method

$$C_i(E_n) = \lambda_i \overset{\text{Yield}}{N_i} \overset{\text{Efficiency}}{\xi_{i,n}} \varepsilon(E_n)$$

$$C_{U^{235}}(E_{186keV}) = \lambda_{U^{235}} N_{U^{235}} \xi_{U^{235},186keV} \varepsilon(E_{186keV})$$

$$C_{U^{238}}(E_{766keV}) = \lambda_{U^{238}} N_{U^{238}} \xi_{U^{238},766keV} \varepsilon(E_{766keV})$$

Absolute Eff.



Relative Eff. (or Intrinsic Eff.)

$$f_{238}^{235} = \frac{N_{235}}{N_{238}} = \frac{C_{235}(E_{186})}{C_{238}(E_{766})} \cdot \frac{\lambda_{238}}{\lambda_{235}} \cdot \frac{\xi_{238,766}}{\xi_{235,186}} \cdot \frac{\eta(E_{766})}{\eta(E_{186})}$$

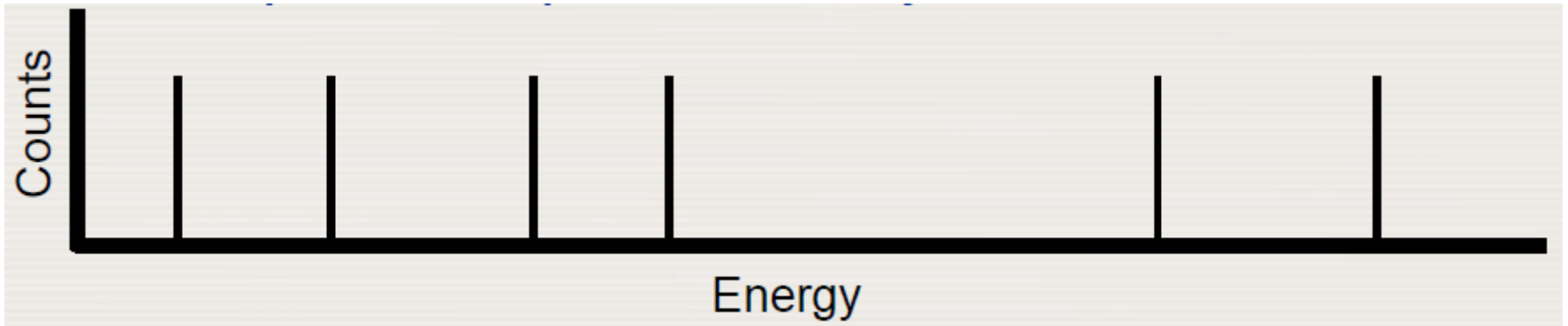
$$f_k^i = \frac{N_i}{N_k} = \frac{C(E_i^n)}{C(E_k^m)} \cdot \frac{\lambda_k}{\lambda_i} \cdot \frac{\xi(E_k^m)}{\xi(E_i^n)} \cdot \frac{\eta(E_k^m)}{\eta(E_i^n)}$$

Generalization

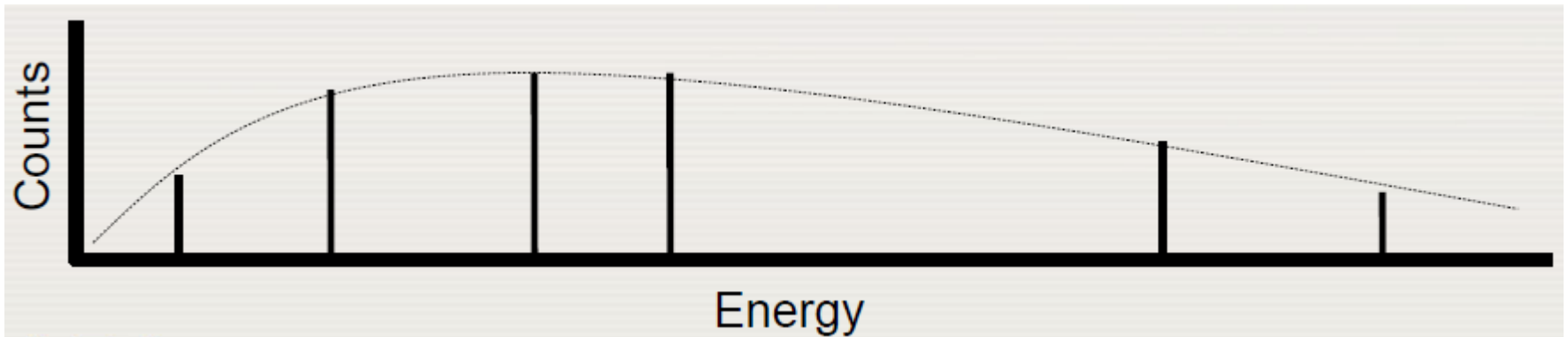
3. 분석 방법

◆ Relative Efficiency Curve

- Assume : Same branching ratio, 100% efficient detector



- Real : different branching ratio by isotopes, different efficient detector by energy



3. 분석 방법

◆ Comparisons of the commercially utilizing analysis software

구분	MGA	FRAM
전체명칭	<u>Multi Group Analysis</u>	Fixed energy Response functions Analysis with Multiple efficiency
개발기관	Lawrence Livermore National Laboratory	Los Alamos National Laboratory
판매사	ORTEC MGA++ V1.06 (MGA ~ 100 keV, U235 < 300 keV, MGAHI : 200-1,000 keV) CANBERRA MGA v4.2	ORTEC FRAM v5.2 CANBERRA PC/FRAM v4.4

ORTEC® MGA++ Safeguards Software MGA-B32 V1.0

Advanced Gamma-Ray Isotopic-Ratio Actinide Analysis from Ge Detector Spectra

- Analyzes both Pu and Pu₂ isotopes with MGA software. Only with U235 software
- The latest LLN algorithms for actinide analysis
- No calibration standards necessary to correct for matrix or container effects
- Rapid operation with display of spectra, results, and peak-fit residuals
- Easily modified parameter sets for different results
- Flexible reporting: instant results and archive copy to Access® database
- Integrated data collection and analysis
- Easy-to-use Windows® Graphical User Interface
- ORTEC ConnectPro-32 compliant
- Operates with all ORTEC and many non-ORTEC MCAs
- Developer's locked options aid system development

Introduction

MGA++ is a suite of two software programs (MGA and U235) for analysis of Actinide spectra acquired by germanium detectors. MGA++ is the result of years of continuing development at Lawrence Livermore National Laboratory.

The MGA++ suite represents the latest versions of the LLN-developed Multi-Group Analytic Codes, which have been in use worldwide since 1970 for a range of measurements related to nuclear materials control and accountability. MGA was originally developed at LLN, to determine relative abundances of plutonium isotopes in samples to an accuracy ~ 1%. U235 has been subsequently developed to determine weight percentage results for U235.

The programs, requiring no special calibration sources or calculations, use only information obtained from the sample spectra to determine the isotopic ratios.

A full member of the CONNECTPRO-32 software family, MGA++ can display the data being acquired, then rapidly analyze and report results—all from a single, easy-to-use program.

User Interface

For each operational mode, separate "viewer" programs (MGAViewer and U235Viewer) and analysis modules (MGAEXE and U235EXE) require the integrity of the analytical methods. The Viewer program provides the user interface and the hardware control function. In MGA++, it is possible to access all spectra displays.

MGAViewer and U235Viewer present the same friendly user interface. The MCA mode operation interface is shown in Fig. 1. The current status of the detector is shown on the right. A special count-rate meter mode shows the instantaneous count rate of a selected region. The spectrum may be viewed during acquisition.

MIRION TECHNOLOGIES

MGAIU™

Multi-Group Analysis for Uranium

KEY FEATURES

- Corrects system isotopic inhomogeneity accurately in low detector live time applications
- Sub-optimal analysis using multiple detector channels allows the need for efficiency calibration based on matrix density, matrix type, or container characteristics
- In the environment entry mode, only one calibration measurement is required
- Integrates into Mirion suite and safeguards instruments to perform analysis with minimal operator intervention (auto mode for repeated measurements)
- Standard analysis mode available to report results in terms of spectra, % of residual, and more
- Developed in collaboration with top experts in safeguards NDA applications

KEY BENEFITS

- Fast and accurate uranium isotopic measurements
- No calibration standards or operators required
- Easy to use for regular operations, limited operator training is required

APPLICATIONS

- Nuclear Safeguards
- Inventories control and verification facilities
- Work measurements

INTRODUCTION

The unique non-destructive range of uranium bearing materials to determine uranium content by what is known as safeguards, waste management, and fuel cycle control control measurements.

In a range of applications for the possible detection of special nuclear materials, there is a need for the accurate and precise measurement of uranium content and enrichment of uranium content.

Uranium measurements are typically affected because of varying container characteristics, container wall thickness, geometry, and other factors.

The MGAIU software can be configured to improve the accuracy of these measurements over traditional methods, while simplifying or automating the setup and operation of the system.

Other methods require MGA software with a range of options for both uranium and plutonium isotopic analysis, including background subtraction, and a ready application interface for integration into existing measurement systems.

In the environment entry mode, MGAIU software addresses the "Matrix Gamma Bias" (MGB) effect and reports just one calibration measurement with a relative standard deviation of about 0.5% (from 1000 counts) with this mode. The results of this calibration measurement are automatically stored for future use on any samples with known composition and thickness. The standard water mode is useful for very thick samples with high uranium content. The results from this mode are stored in the results from the standard mode, and also can be used for analyzing samples where the uranium isotopes are not in equilibrium with their daughter products, thereby separating uranium.

ORTEC® PC/FRAM-B32 V3.3 Safeguards Software

Advanced Isotopic Ratio Analysis Software from Ge Detector Gamma-Ray Spectra

- Analyzes Pu, and a wide variety of heterogeneous samples containing Pu, Am, U, and other isotopes including Pu₂
- Operates with a single HPGe detector
- No calibration standards necessary
- Works with shielded samples
- Select from a large number of supplied sample/geometry conditions or add more types
- Dynamic selection of English or Russian Graphical-User Interface
- ORTEC ConnectPro-32 compliant
- Operates on a variety of hardware platforms and is a 32-bit Windows® 95/98/NT application
- Decay Correction for isotopic fractions

Introduction

The PC/FRAM code has been under evolutionary development at Los Alamos National Laboratory since the mid 1980s. PC/FRAM-B32 Version 3.3 is a 32-bit version for the Windows 95/98/NT operating platforms. It operates within the ORTEC ConnectPro network spectroscopy architecture, giving greater flexibility in choice of MCA hardware.

PC/FRAM-B32 analyzes the gamma-ray spectrum taken with a germanium detector, of plutonium-bearing, uranium-bearing, or mixed items and quantifies the distribution of plutonium or uranium isotopes, ²³⁹Am/uranium and other transuranic isotopes (including uranium in trace amounts) relative to plutonium. PC/FRAM-B32 analyzes spectra from items containing only uranium and quantifies the uranium isotopic distribution. These measurements are performed on samples of arbitrary size, geometry, and physical and chemical composition.

MIRION TECHNOLOGIES

FRAM

Fixed-Energy, Response Function Analysis with Multiple Efficiency

KEY FEATURES

- Calibrates relative plutonium isotopic abundances or uranium isotopic abundances or uranium isotopic abundance to known
- Uses accurate calibration with HPGe detectors
- Incorporates a sophisticated peak fitting and multiple deconvolution algorithm to analyze complex peak regions
- Requires no efficiency calibration for matrix density, thickness or container characteristics
- Developed by Los Alamos and licensed to CANBERRA

DESCRIPTION

FRAM (Fixed-Energy, Response Function Analysis with Multiple Efficiency) is a code developed by Los Alamos to analyze gamma-ray spectra generated by high-resolution germanium detectors to obtain relative isotopic abundances of plutonium or uranium isotopes. Its design and flexibility allow it to easily measure ratios and distributions of isotopes other than plutonium isotopic ratios.

In nuclear safeguards, FRAM complements calorimetry and neutron coincidence counting by allowing measurements to be interpreted in terms of total plutonium mass.

Table 1 - Isotopic Types Measured with FRAM

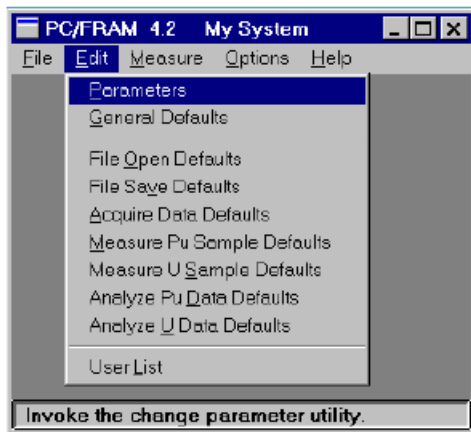
²³⁹ Pu	²⁴⁰ Pu (total plutonium)	²⁴¹ Pu
²³⁸ Pu	²⁴² Pu (total plutonium)	²⁴³ Pu
²⁴⁴ Pu (total plutonium)	²⁴⁵ Pu (total plutonium)	²⁴⁶ Pu
²⁴⁷ Pu (total plutonium)	²⁴⁸ Pu (total plutonium)	²⁴⁹ Pu
²⁵⁰ Pu (total plutonium)	²⁵¹ Pu (total plutonium)	²⁵² Pu
²⁵³ Pu (total plutonium)	²⁵⁴ Pu (total plutonium)	²⁵⁵ Pu
²⁵⁶ Pu (total plutonium)	²⁵⁷ Pu (total plutonium)	²⁵⁸ Pu
²⁵⁹ Pu (total plutonium)	²⁶⁰ Pu (total plutonium)	²⁶¹ Pu
²⁶² Pu (total plutonium)	²⁶³ Pu (total plutonium)	²⁶⁴ Pu
²⁶⁵ Pu (total plutonium)	²⁶⁶ Pu (total plutonium)	²⁶⁷ Pu
²⁶⁸ Pu (total plutonium)	²⁶⁹ Pu (total plutonium)	²⁷⁰ Pu
²⁷¹ Pu (total plutonium)	²⁷² Pu (total plutonium)	²⁷³ Pu
²⁷⁴ Pu (total plutonium)	²⁷⁵ Pu (total plutonium)	²⁷⁶ Pu
²⁷⁷ Pu (total plutonium)	²⁷⁸ Pu (total plutonium)	²⁷⁹ Pu
²⁸⁰ Pu (total plutonium)	²⁸¹ Pu (total plutonium)	²⁸² Pu
²⁸³ Pu (total plutonium)	²⁸⁴ Pu (total plutonium)	²⁸⁵ Pu
²⁸⁶ Pu (total plutonium)	²⁸⁷ Pu (total plutonium)	²⁸⁸ Pu
²⁸⁹ Pu (total plutonium)	²⁹⁰ Pu (total plutonium)	²⁹¹ Pu
²⁹² Pu (total plutonium)	²⁹³ Pu (total plutonium)	²⁹⁴ Pu
²⁹⁵ Pu (total plutonium)	²⁹⁶ Pu (total plutonium)	²⁹⁷ Pu
²⁹⁸ Pu (total plutonium)	²⁹⁹ Pu (total plutonium)	³⁰⁰ Pu

MEASUREMENT PRINCIPLES

In its principal application, FRAM analyzes photomasks in the spectrum of plutonium gamma rays obtained by a high-purity germanium detector. The spectrum contains peaks from the plutonium isotopes ²³⁹Pu, ²⁴⁰Pu, and other isotopes such as ²⁴¹Pu or ²⁴²Pu. FRAM combines this information to obtain isotopic ratios independent of sample size, shape, physical and chemical composition, measurement geometry, and container characteristics. The results are compared using only the known data and known fundamental nuclear branching fractions and half lives and do not require calibration with standards.

3. 분석 방법

◆ Parameter Set (Analysis Method)



Parameter List

Name	Descriptions
U100keVLEU	Planar, E<=10%, 100keV
U100keVHEU	Planar, E>=2%, 100keV
ULow121_1001Cx	Coaxial, E<70%, 121-1001keV
UHi121_1001Cx	Coaxial, E>10%, 121-1001keV
ULowENR	Coaxial, E<=50%, 1-1024keV
UHiENR	Coaxial, E>20%, 1-1024keV

	isotope	peak energy	line width	branching ratio	fix area to	sum area with	used for eff	used for act	used for ecal	used for fcal	used for scal	
<input type="checkbox"/>	1	U234	120.905	0.00	3.33000e-004	0	0	<input type="checkbox"/>	<input checked="" type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	2	U235	140.760	0.00	2.62000e-003	0	0	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	3	U235	143.760	0.00	1.09500e-001	0	0	<input checked="" type="checkbox"/>	<input checked="" type="checkbox"/>	<input checked="" type="checkbox"/>	<input checked="" type="checkbox"/>	<input checked="" type="checkbox"/>
<input type="checkbox"/>	4		145.940	0.00	0.00000e+000	0	0	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	5	U235	163.330	0.00	5.08900e-002	0	0	<input checked="" type="checkbox"/>	<input checked="" type="checkbox"/>	<input checked="" type="checkbox"/>	<input checked="" type="checkbox"/>	<input checked="" type="checkbox"/>
<input type="checkbox"/>	6	U235	182.610	0.00	4.02400e-003	0	0	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>
<input type="checkbox"/>	7	U235	185.715	0.00	5.73900e-001	0	0	<input checked="" type="checkbox"/>	<input checked="" type="checkbox"/>	<input checked="" type="checkbox"/>	<input checked="" type="checkbox"/>	<input checked="" type="checkbox"/>
<input type="checkbox"/>	8	U235	202.110	0.00	1.10000e-002	0	0	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>	<input type="checkbox"/>

Peaks to be used in an analysis

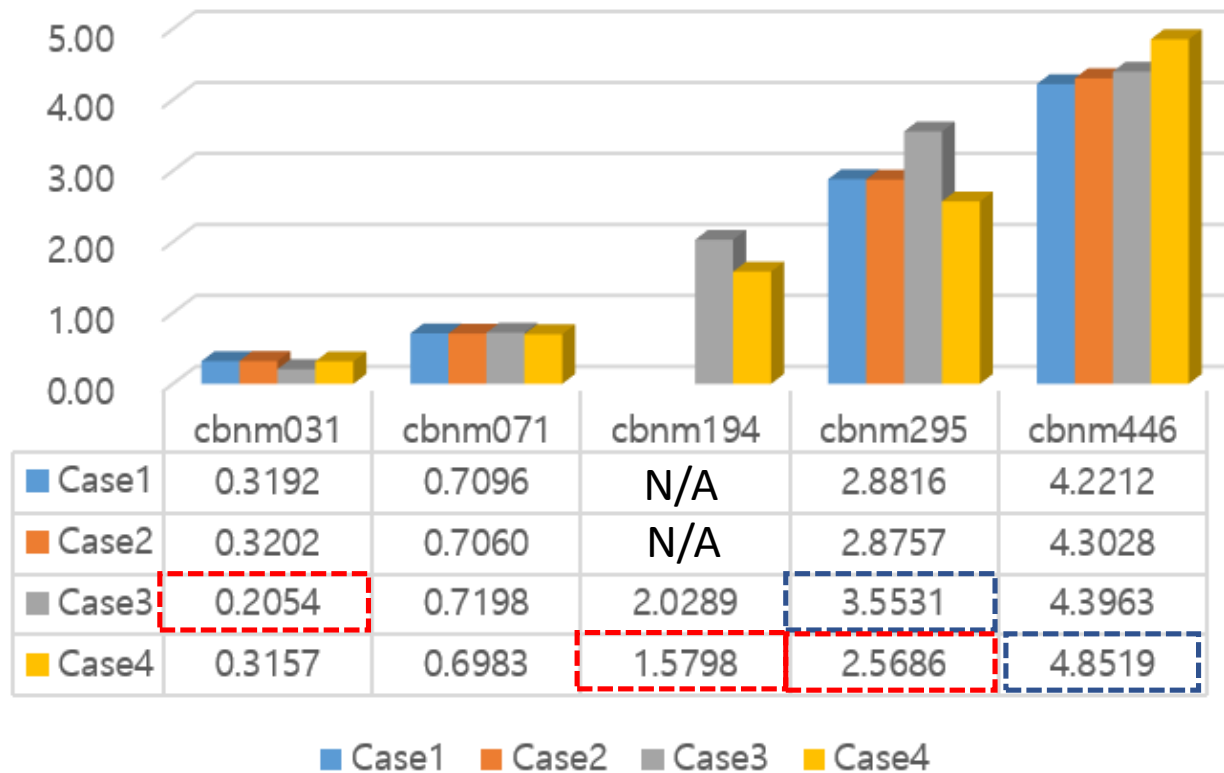
	region start	region end	BKG #1 start	BKG #1 end	BKG #2 start	BKG #2 end	BKG #3 start	BKG #3 end	BKG #4 start	BKG #4 end	background type
<input type="checkbox"/>	1	958	977	950	956	979	987	0	0	0	linear
<input type="checkbox"/>	2	1114	1176	1094	1102	1103	1112	1177	1186	1188	linear step
<input type="checkbox"/>	3	1286	1320	1267	1278	1278	1288	1330	1339	1342	linear step
<input type="checkbox"/>	4	1450	1500	1416	1428	1440	1448	1513	1524	1526	bilinear step
<input type="checkbox"/>	5	1585	1656	1580	1584	1600	1604	1656	1664	1688	linear step
<input type="checkbox"/>	6	2052	2080	2032	2048	2084	2100	0	0	0	linear
<input type="checkbox"/>	7	4534	4571	4496	4504	4575	4583	0	0	0	linear
<input type="checkbox"/>	8	5888	5968	5776	5792	5992	6008	0	0	0	linear

Regions by Channel

4. Results

4. Results

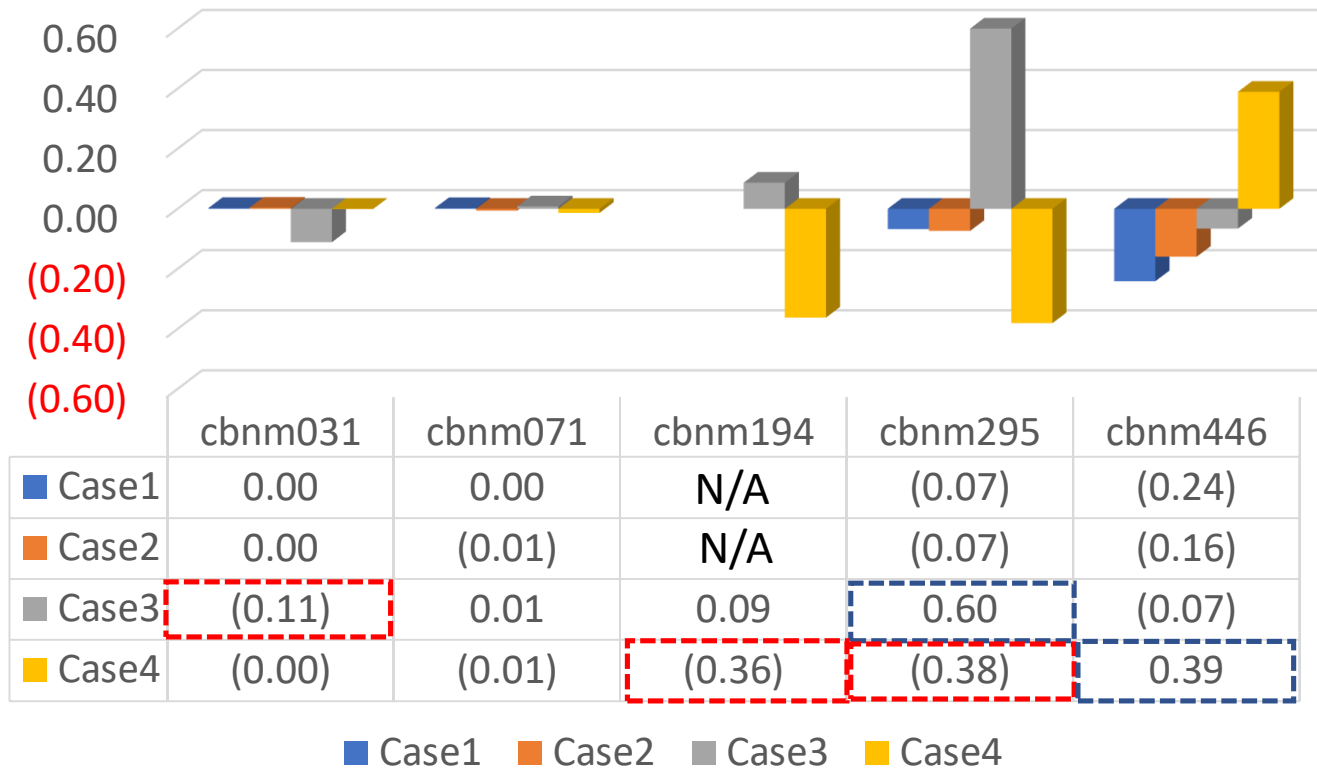
◆ Calculated U-235 Values



	Source to Detector Distance	Measuring time
Case 1	10 cm	16 hours
Case 2	2 cm	16 hours
Case 3	10 cm	5 minutes
Case 4	2cm	5 minutes

4. Results

◆ Relative Deviations (Mea - Ref)



	Source to Detector Distance	Measuring time
Case 1	10 cm	16 hours
Case 2	2 cm	16 hours
Case 3	10 cm	5 minutes
Case 4	2cm	5 minutes

5. Conclusions

5. Conclusion

◆ 천연 우라늄 이하의 농축도에서는 측정조건에 관계없이 농축도 예측 정확도가 높음

◆ Case 3와 4는 계측시간이 충분하지 않은 것이 예측 오차를 크게 만든 이유로 판단됨

<참조값과의 차이>

- Case3의 CBNM031에서 약 - 0.11 w/o,
CBNM295에서 약 + 0.60 w/o
- Case4의 CBNM194에서 약 - 0.36 w/o
CBNM446에서 약 - 0.38 w/o
CBNM446에서 약 + 0.39 w/o

◆ 특히, 가장 불리한 계측조건(Case3, 10 cm, 5 minutes)에서도

Ref 2.95w/o에서 0.6w/o 차이 존재.

- 현장에서 신속하게 불법여부를 판정하는데 미치는 영향은 없다고 볼 수 있음.
다만, 천연우라늄 부근(0.712 ± 0.6)에서는 동 방법이 판정에 어려움이 있을 수 있음.
- 충분한 계측시간과 가까운 거리를 확보할 수록 예측 정확도를 높일 수 있음.

감사합니다

Q&A