NKS RINFOR

NKS GammaRayX Seminar, 20-21 October 2021

Development of a Resource for the Improvement of National Nuclear Forensics Gamma Spectrometric Core Capabilities

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Core capabilities

Wallenius, M., Mayer, K. Nuclear Forensics Awareness and Understanding, Conference: *International Conference on Advances in Nuclear Forensics: Countering the Evolving Threat of Nuclear and Other Radioactive Material out of Regulatory Control*; Vienna, Austria: July 2014

"*Each State* should seek to acquire *nuclear forensic capabilities* enabling to provide *competent authorities* with relevant information ………… on the *main characteristics of the interdicted material*. Such capabilities are often referred to as ***nuclear forensics core capabilities***.

Besides conducting the *preliminary assessment* of the material ……………, the core capabilities help …… to strengthen overarching nuclear security controls, *enable rapid and appropriate response*, and in case advanced nuclear forensic analyses are desired, *enable States to request and receive international assistance*."

For the vast majority of organisations (including typical NKS participants) and the materials of most concern, core capabilities are conducted using gamma spectrometry based on the usual detector types – hence RINFOR. For many states, core capabilities should be present in safety authorities, regulatory bodies, etc.

Objective and Background

"….provide a means of enhancing (where they exist) or establishing (where they do not) core capabilities of institutes, agencies or entities with respect to gamma spectrometric responses to situations requiring, or potentially requiring, some measure of nuclear forensic analysis through the generation of a suite of *fit-for-purpose* materials….."

NKS project AFT/B(19)8, completed during 2019

Partners

[1]Norwegian Radiation and Nuclear Safety Authority
[2]Swedish Defence Research Agency
[3]Swedish Radiation Safety Authority
[4]Technical University of Denmark
[5]Radiation and Nuclear Safety Authority (STUK), Finland
[6]Icelandic Radiation Safety Authority

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RINFOR Objectives

Genenrate a set of synthetic spectra, typical of materials that:

- are outside of many practitioners typical experience
- are not that easy to obtain by other means
- may arise in nuclear security incidents
- facilitate appraisal of «*main characteristics*»
- come with ancilliary information such that conventional routines may be applied without too much trouble

Background

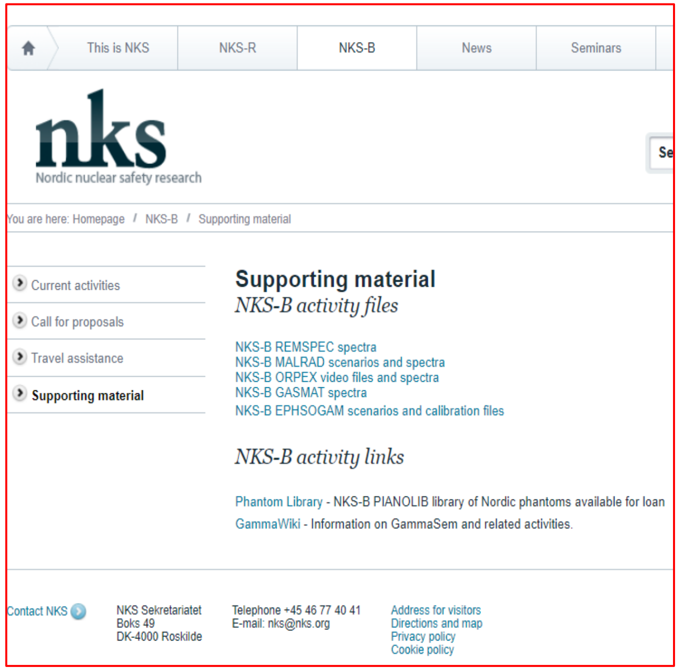
Supplementing the already existing suite of gamma spectra available on the NKS website.

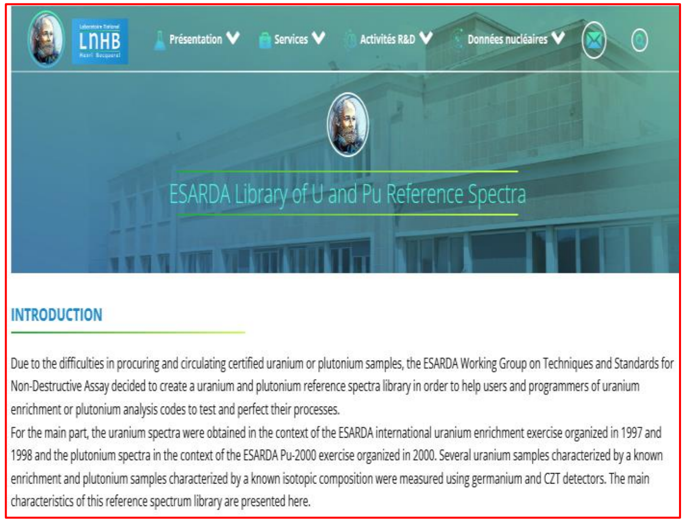
In the case of RINFOR – set of spectra of uranium and plutonium materials of different ages and compositions with associated calibration spectra for common general purpose detectors.

Target audience – those practitioners without easy access to such materials or spectra or analytical assets beyond those found in conventional spectrometry suites and who may at some point need to exercise "*core capabilities*".

Existing resources – The Uranium and Plutonium Reference spectra at LNHB

http://www.lnhb.fr/esarda_wg/





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Fit for purpose

- analysis by software not specific for forensics and by practitioners not typically involved in such measurements
- parameters of most interest (age, enrichment, isotopes) can be estimated without dedicated software
- information provided should be sufficient to allow use of conventional software routines

The spectra were **NOT** expected to be fit for, nor intended, to be used for:

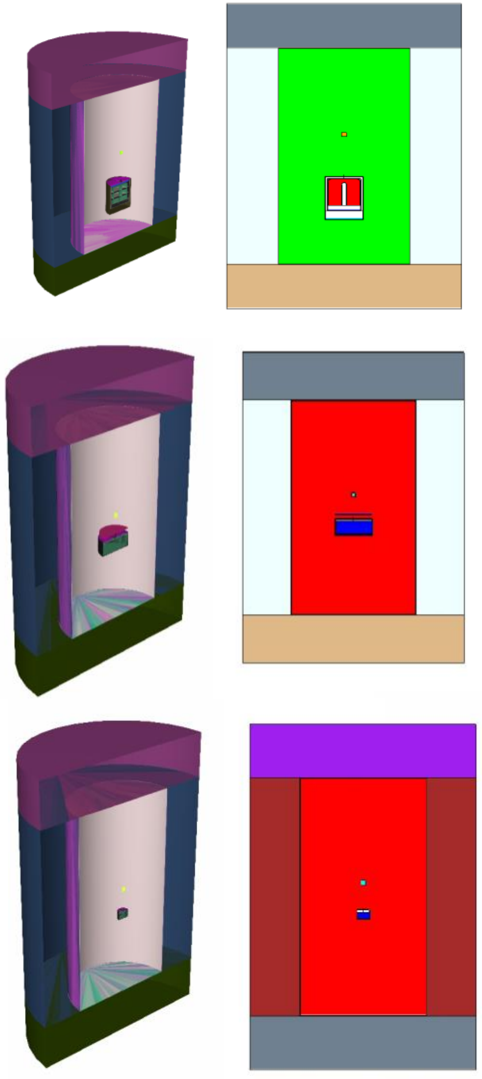
- Quality assurance/quality control, use as «reference» or «standard» spectra
- Method development or validation, proficiency testing

In general – if you have capabilities/software that go beyond «*core*» – these may not be the spectra for you !

Methodology - Detectors

5 detectors chosen – typical for those found in conventional gamma labs or deployed in the field

- coaxial p-type HPGe detector (46% relative efficiency), Al endcap (HPGe)
- planar detector, 0.6 mm carbon window (PLAN)
- low energy detector 0.20 mm beryllium window (LEGE)
- 4” x 4” x 16” NaI detector
- 5 cm x 5 cm LaBr detector



DSA Norwegian Radiation and Nuclear Safety Authority

Methodology - Sources

50 year old depleted uranium (metallic) - cylinder 1 cm tall and 1 cm in diameter, density 19.9512.

Isotope	Composition (w/w)
^{238}U	99.7495 %
^{235}U	0.25 %
^{234}U	0.0005 %

Enriched uranium (metallic) – cylinder 1 cm tall and 1 cm in diameter, density 18.94449. "*Commercial Grade*"

The composition of the material at the time of last separation was:

Isotope	Composition (w/w)
^{238}U	97.01 %
^{235}U	2.96 %
^{234}U	0.03 %

Compositions of the uranium were calculated for 1 year, 10 years and 50 years after last separation

Methodology - Sources

Enriched uranium (metallic) - cylinder 1 cm tall and 1 cm in diameter, density 18.7328. "*Highly Enriched Uranium I*"

The composition of the material at time of last separation was:

Isotope	Composition (w/w)
^{238}U	8.85 %
^{235}U	89.80 %
^{234}U	0.97 %
^{236}U	0.38 %

Compositions of the uranium were calculated for 1 year, 10 years and 50 years after last separation.

Enriched uranium (metallic) - cylinder of 1 cm tall and 1 cm in diameter, density 18.724. "*Highly Enriched Uranium II*"

The composition of the material at the time of last separation was:

Isotope	Composition (w/w)
^{238}U	5.42 %
^{235}U	93.16 %
^{234}U	0.98 %
^{236}U	0.45 %

Compositions of the uranium were calculated for 1 year, 10 years and 50 years after last separation.

Methodology - Sources

Plutonium, 10 mg - cylinder of 1 cm height and 1 cm diameter with an aqueous composition and density. “*Fuel Grade*”
Composition of the material at time of last separation.

Isotope	Composition (w/w)
^{238}Pu	0.10 %
^{239}Pu	86.10 %
^{240}Pu	12.00 %
^{241}Pu	1.60 %
^{242}Pu	0.20 %

Compositions of the plutonium were calculated for 1 year, 10 years and 50 years after last separation.

Plutonium, 10 mg, - cylinder of 1 cm height and 1 cm diameter with an aqueous composition and density. “*Reactor grade*”
The composition of the material at time of last separation was:

Isotope	Composition (w/w)
^{238}Pu	0.99 %
^{239}Pu	62.38 %
^{240}Pu	21.78 %
^{241}Pu	11.88 %
^{242}Pu	2.97 %

Compositions of the plutonium were calculated for 1 year, 10 years and 50 years after last separation.

Methodology - Sources

Plutonium (5.7 %), 10 mg - cylinder of 1 cm height and 1 cm diameter, aqueous composition and density. "*Weapons grade I*"
The composition (w/w) of the material at the time of last separation was:

Isotope	Composition (w/w)
^{238}Pu	0.03 %
^{239}Pu	93.92 %
^{240}Pu	5.70 %
^{241}Pu	0.32 %
^{242}Pu	0.03 %

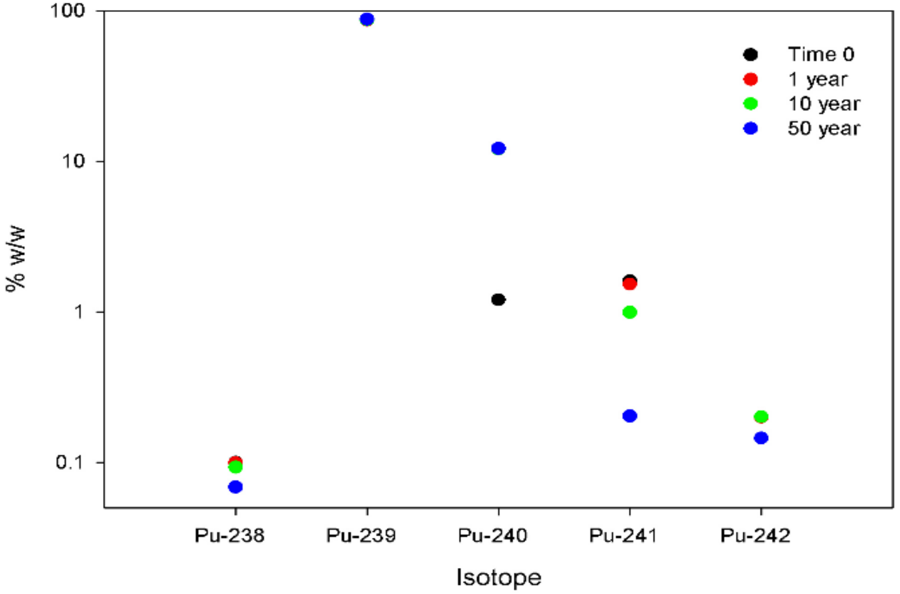
Compositions of the plutonium were calculated for 1 year, 10 years and 50 years after last separation.

Plutonium (10–13 %), 10 mg - cylinder of 1 cm height and 1 cm diameter, aqueous composition and density. "*Weapons grade II*"
The composition (w/w) of the material at the time of last separation was:

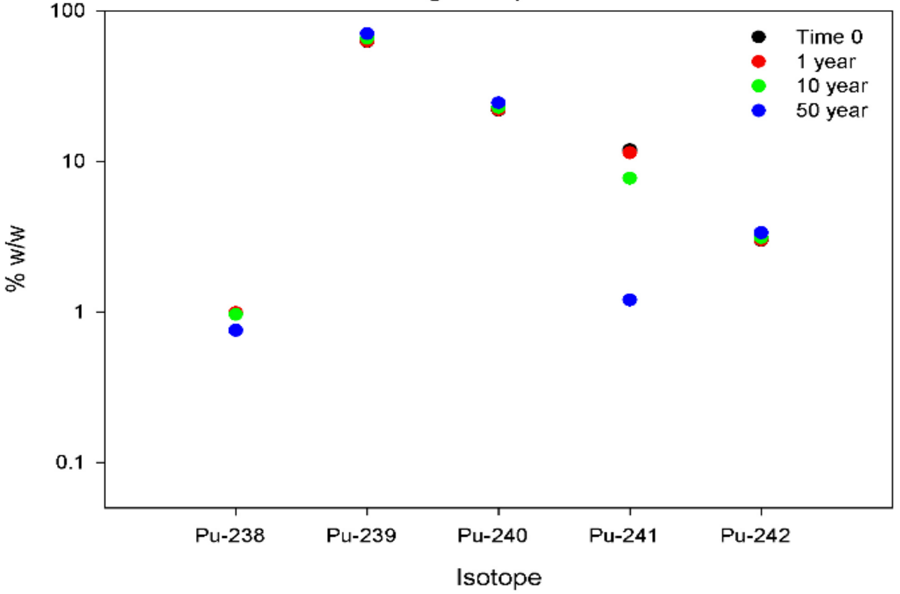
Isotope	Composition (w/w)
^{238}Pu	0.0892 %
^{239}Pu	86.1901 %
^{240}Pu	11.7081 %
^{241}Pu	1.844 %
^{242}Pu	0.1686 %

Compositions of the plutonium were calculated for 1 year, 10 years and 50 years after last separation.

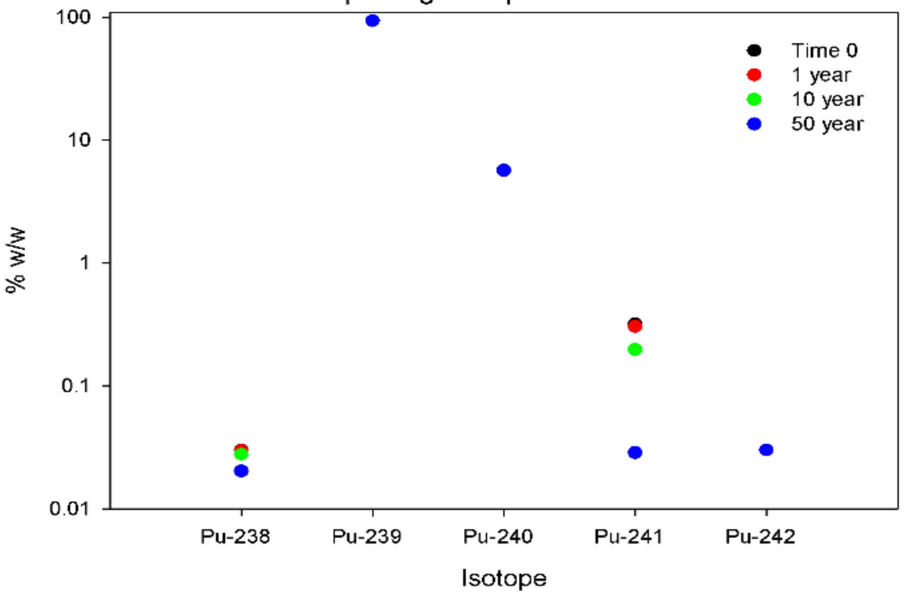
Fuel grade plutonium



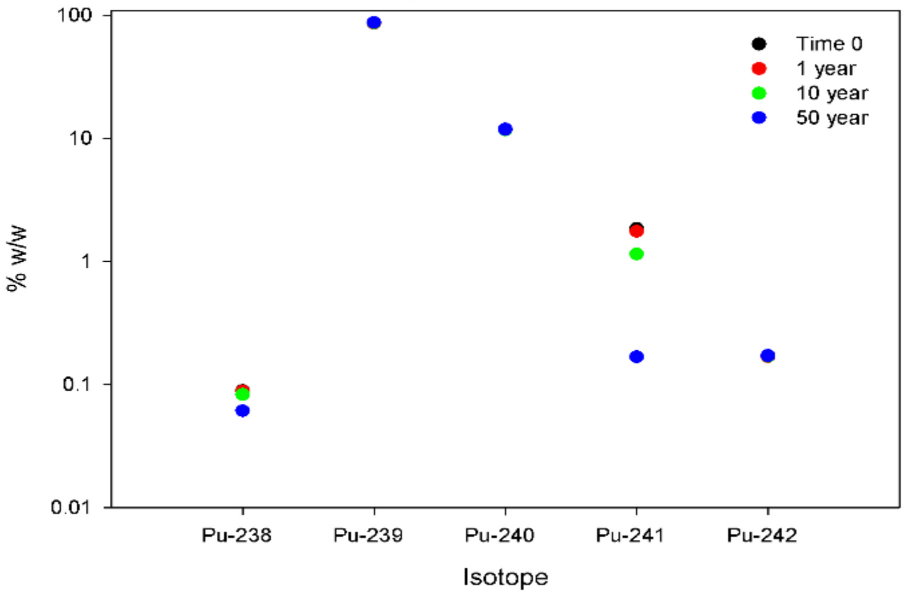
Reactor grade plutonium



Weapons grade plutonium 5.7 %



Weapons grade plutonium 11.7 %



Methodology - Sources

For relevant materials, daughters were calculated for the three time periods after last separation.

«Fuel grade» plutonium shown as example.

<u>Isotope activities (Bq) of the “fuel grade” plutonium materials.</u>

Isotope	*1 year old*	*10 years old*	*50 years old*
^{241}Pu	5.829612E+08	3.774441E+08	5.466790E+07
^{239}Pu	1.975995E+07	1.975402E+07	1.973229E+07
^{235m}U	1.974809E+07	1.974217E+07	1.972043E+07
^{240}Pu	1.007419E+07	1.006412E+07	1.002180E+07
^{238}Pu	6.286938E+06	5.855388E+06	4.268603E+06
^{241}Am	9.569178E+05	7.715302E+06	1.751698E+07
^{237}U	1.429870E+04	9.257139E+03	1.341153E+03
^{242}Pu	2.914000E+03	2.914000E+03	2.913709E+03
^{234}U	1.781964E+01	1.720496E+02	7.388942E+02
^{236}U	2.981252E-01	2.980244E+00	1.487100E+01
^{237}Np	1.606080E-01	1.355226E+01	1.984197E+02
^{233}Pa	1.306278E-01	1.328916E+01	1.978079E+02
^{235}U	1.945367E-02	1.945169E-01	9.719919E-01
^{231}Th	1.937265E-02	1.944379E-01	9.719919E-01

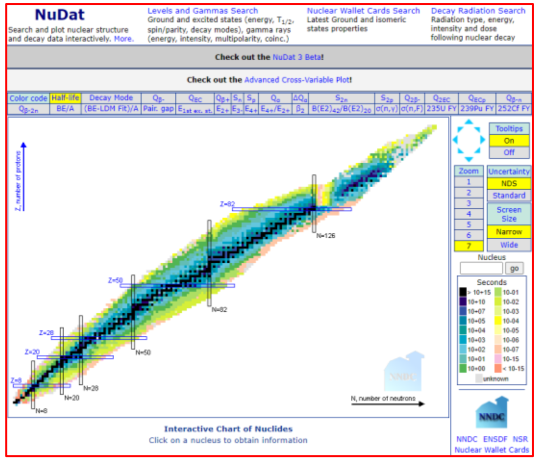
Methodology - Simulations

All simulations conducted using using MCNP-6.

All material compositional data taken from:

McConn, R.J., Gesh, C.J., Pagh, R.T., Rucker, R.A. And Williams III, R.G. 2011. *Compendium of Material Composition Data for Radiation Transport Modeling*. Pacific Northwest National Laboratory (PNNL), PIET-43741-TM-963, PNNL-15870 Rev 1.

Nuclear data (energies, emission probabilities etc) were drawn from NUDAT 2.6 (www.nndc.bnl.gov) as of 2015.



Methodology - Simulations

No background included in any spectrum from any source.

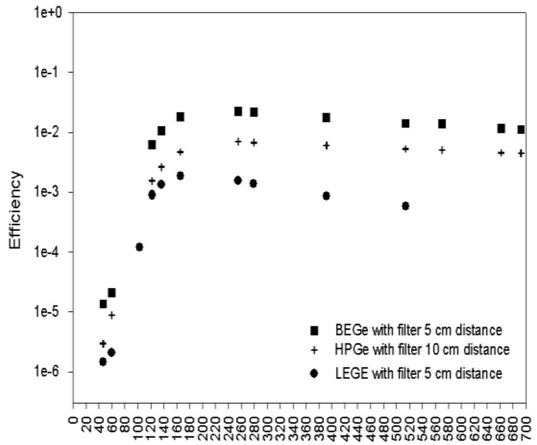
For the plutonium spectra, all spectra generated using both a filter (2mm Cd/1 mm Sn) and no filter present.

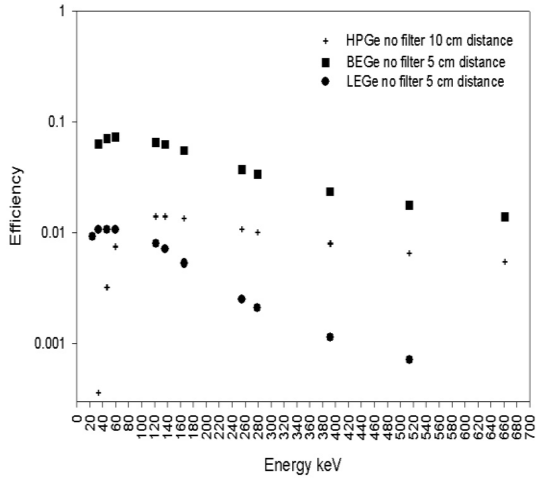
For each simulation, samples were presented at a distance of 10 or 5 cm from the detector face.

True coincidence summation effects were not included. Sample holders were not included in the simulations.

For all simulation setups, the calibration sources, presented in an aqueous matrix (density 1) of the same physical dimensions as the sample materials, was as follows (values in kBq):

Isotope	HPGe,LEGe and PLAN, with filter	HPGe, LEGe and PLAN, no filter	NaI and LaBr,
^{210}Pb	21000	110.1	11.1
^{241}Am	2000	100.11	1.11
^{57}Co	590	59.2	0.592
^{139}Ce	74	74	0.74
^{203}Hg	22.2	22.2	2.22
^{113}Sn	28.1	28.1	2.81
^{85}Sr	35.5	35.5	3.55
^{137}Cs	25.9	25.9	2.59
^{60}Co			3.03
^{88}Y			5.92

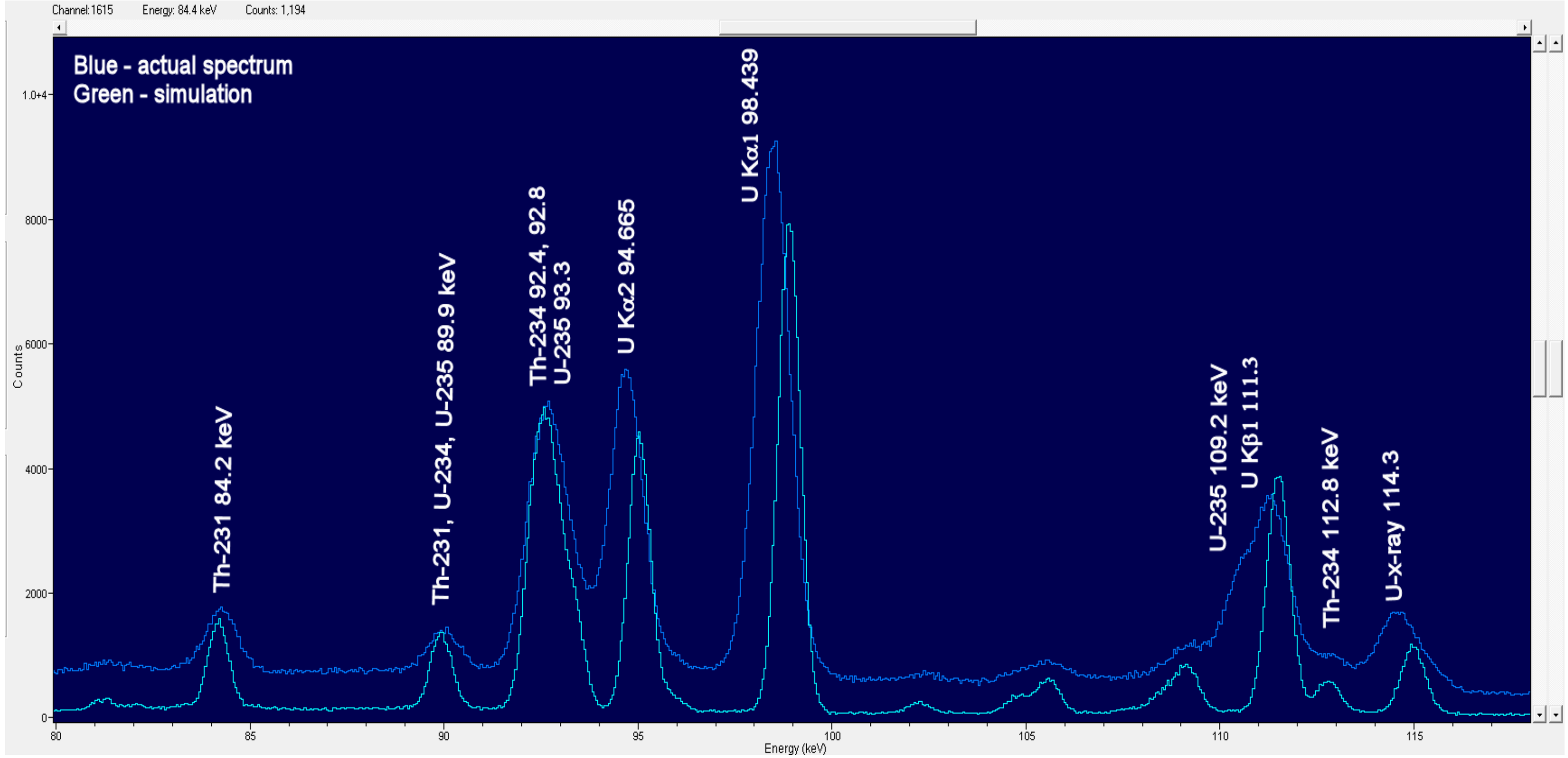




Methodology - Issues

MCNP – (un)documented feature. Energy shift in flourescence X-ray energies for high z-values.

G-1542d Coax spectrum (U, 0-214 keV)



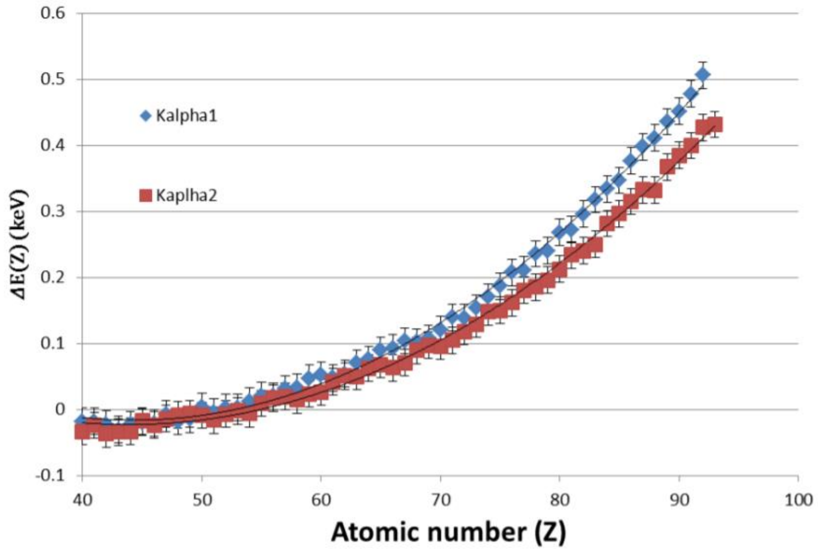


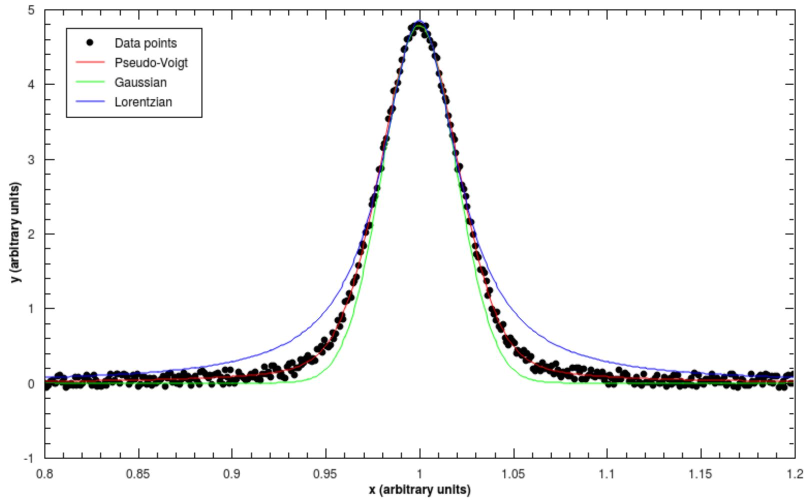
Fig. 9. Energy shift between the energy of fluorescence X-rays calculated with MCNP and the theoretical values taken from [4].

T. Marchais, B. Pérot, C. Carasco, P.-G. Allinei, P. Chaussonnet, et al.. Detailed MCNP Simulations of Gamma-Ray Spectroscopy Measurements With Calibration Blocks for Uranium Mining Applications. IEEE Transactions on Nuclear Science, Institute of Electrical and Electronics Engineers, 2018, 65 (9), pp.2533-2538. ff10.1109/TNS.2018.2797312ff. ffhal-01990707f

Methodology - Issues

Peak shapes – X-rays

X-ray peaks have different shapes to gamma peaks (Lorentzian/Gaussian Voight)



Not accounted for in MCNP (in so far as I can use it or understand it!)

May cause problems for dedicated codes (PCFRAM, MGA)

Not so simple to correct for within the context of the project - fixable with some kind of post-processing (probably)

Isotope	Composition (w/w)
^{238}U	5.42 %
^{235}U	93.16 %
^{234}U	0.98 %
^{236}U	0.45 %

Isotope	Age y	Composition (w/w ± 1σ) as determined by PCFRAM - HPGe	Composition (w/w ± 1σ) as determined by PCFRAM - PLAN	Composition (w/w ± 1σ) as determined by PCFRAM - LEGe
^{238}U	1	3.9 ± 0.87 %	3.78 ± 1.52 %	4.54 ± 7.59 %
	10	5.48 ± 1.3 %	5.76 ± 0.84 %	4.63 ± 7.8 %
	50	3.91 ± 1.7 %	8.8 ± 10.3 %	4.65 ± 7.87 %
^{235}U	1	94.63 ± 1.08 %	94.73 ± 2.08 %	94.35 ± 7.51 %
	10	93.12 ± 1.51 %	92.802 ± 0.95 %	94.27 ± 7.8 %
	50	94.69 ± 2.19 %	89.7 ± 12.76	94.23 ± 7.8 %
^{236}U	1	0.23 ± 0.05 %	0.23 ± 0.11 %	0.24 ± 0.12 %
	10	0.251 ± 0.07 %	0.254 ± 0.04 %	0.85 ± 0.07 %
	50	0.2334 ± 0.121 %	0.275 ± 0.46 %	0.24 ± 0.12 %
^{234}U	1	1.19 ± 0.41 %	1.24 ± 1.02 %	0.85 ± 0.07 %
	10	1.14 ± 0.46 %	1.18 ± 0.3 %	0.855 ± 0.07 %
	50	1.156 ± 0.97 %	1.12 ± 6.05 %	0.86 ± 0.07 %

Isotope	Composition (w/w)
^{238}Pu	0.10 %
^{239}Pu	86.10 %
^{240}Pu	12.00 %
^{241}Pu	1.60 %
^{242}Pu	0.20 %

Isotope	Age (y)	Composition (w/w ± 1σ) and age (y) as determined by PCFRAM – HPGe, with filter		Composition (w/w ± 1σ) and age (y) as determined by PCFRAM – PLAN, with filter		Composition (w/w ± 1σ) and age (y) as determined by PCFRAM – LEGe, with filter	
^{238}Pu	1	0.092 ± 0.003%	0.91 ± 0.14	0.1 ± 0.003 %	1.01 ± 0.06	0.094 ± 0.01 %	0.97 ± 0.17
	10	0.075 ± 0.009 %	9.38 ± 0.40	0.091 ± 0.004 %	10.09 ± 0.17	0.089 ± 0.9 %	10.3 ± 0.32
	50	0.062 ± 0.03 %	52.68 ± 3.1	0.07 ± 0.004 %	50.28 ± 0.54	0.076 ± 0.014 %	51.58 ± 2.8
^{239}Pu	1	86.43 ± 0.43 %		86.40 ± 0.41 %		86.46 ± 1.2 %	
	10	87.52 ± 1.4 %		86.70 ± 0.51 %		88.34 ± 0.9 %	
	50	88.66 ± 4.21 %		88.36 ± 0.58 %		89.497 ± 0.84 %	
^{240}Pu	1	11.74 ± 0.44 %		11.71 ± 0.41 %		11.76 ± 1.2 %	
	10	11.16 ± 1.41 %		11.95 ± 0.52 %		10.37 ± 0.90 %	
	50	10.03 ± 4.3 %		11.20 ± 0.57 %		10.11 ± 0.82 %	
^{241}Pu	1	1.45 ± 0.013 %		1.49 ± 0.015 %		1.401 ± 0.06 %	
	10	1.00 ± 0.03 %		0.97 ± 0.01 %		0.98 ± 0.03 %	
	50	0.134 ± 0.02 %		0.1445 ± 0.004 %		0.12 ± 0.01 %	
^{242}Pu	1	0.27 ± 0.13 %		0.29 ± 0.13 %		0.28 ± 0.04 %	
	10	0.24 ± 0.04 %		0.28 ± 0.015 %		0.23 ± 0.034 %	
	50	0.21 ± 0.104 %		0.224 ± 0.15 %		0.20 ± 0.03 %	

Isotope	Age (y)	Composition (w/w ± 1σ) and age (y) as determined by PCFRAM – HPGe, no filter		Composition (w/w ± 1σ) and age (y) as determined by PCFRAM – PLAN, no filter		Composition (w/w ± 1σ) and age (y) as determined by PCFRAM – LEGe, no filter	
^{238}Pu	1	0.092 ± 0.005 %	1.12 ± 0.10	0.097 ± 0.004 %	0.91 ± 0.10	0.099 ± 0.003 %	0.98 ± 0.06
	10	0.093 ± 0.04 %	9.93 ± 0.20	0.0896 ± 0.005 %	10.06 ± 1.09	0.093 ± 0.01 %	9.86 ± 0.41
	50	0.064 ± 0.106 %	52.16 ± 6.8	0.07 ± 0.004 %	49.55 ± 0.55	0.076 ± 0.005 %	49.14 ± 0.7
^{239}Pu	1	88.07 ± 1.1 %		86.5 ± 0.89 %		86.23 ± 0.63 %	
	10	86.96 ± 0.73 %		86.79 ± 0.74 %		86.20 ± 1.25 %	
	50	88.29 ± 1.8 %		87.78 ± 0.6 %		87.56 ± 0.83 %	
^{240}Pu	1	10.14 ± 1.1 %		11.62 ± 0.91 %		11.85 ± 0.64 %	
	10	11.7 ± 0.74 %		11.87 ± 0.76 %		12.38 ± 1.3 %	
	50	11.29 ± 1.87 %		11.76 ± 0.58 %		11.96 ± 0.84 %	
^{241}Pu	1	1.47 ± 0.03 %		1.5 ± 0.024 %		1.51 ± 0.02 %	
	10	0.976 ± 0.015 %		0.967 ± 0.016 %		1.02 ± 0.03 %	
	50	0.13 ± 0.008 %		0.15 ± 0.0038 %		0.148 ± 0.005 %	
^{242}Pu	1	0.23 ± 0.04 %		0.284 ± 0.026 %		0.297 ± 0.02 %	
	10	0.2779 ± 0.021 %		0.2777 ± 0.035 %		0.301 ± 0.05 %	
	50	0.216 ± 0.045 %		0.236 ± 0.03 %		0.253 ± 0.022 %	

Challenges

What sort of challenges will the non-expert analyst be faced with?

- The spectra are complex and include isotopes not encountered daily
- The spectra regions of interest may not be typical of what many analysts deal with routinely
- Extreme density/matrix correction for the uranium samples
- Parent daughter relationships to account for in the Pu spectra
- Activities are very high relative to more routine measurements
- Nuclear libraries as provided by manufacturers may be out of date or not comprehensive enough (especially for the isotopes of interest in this instance)
- What units are such materials best reported in?

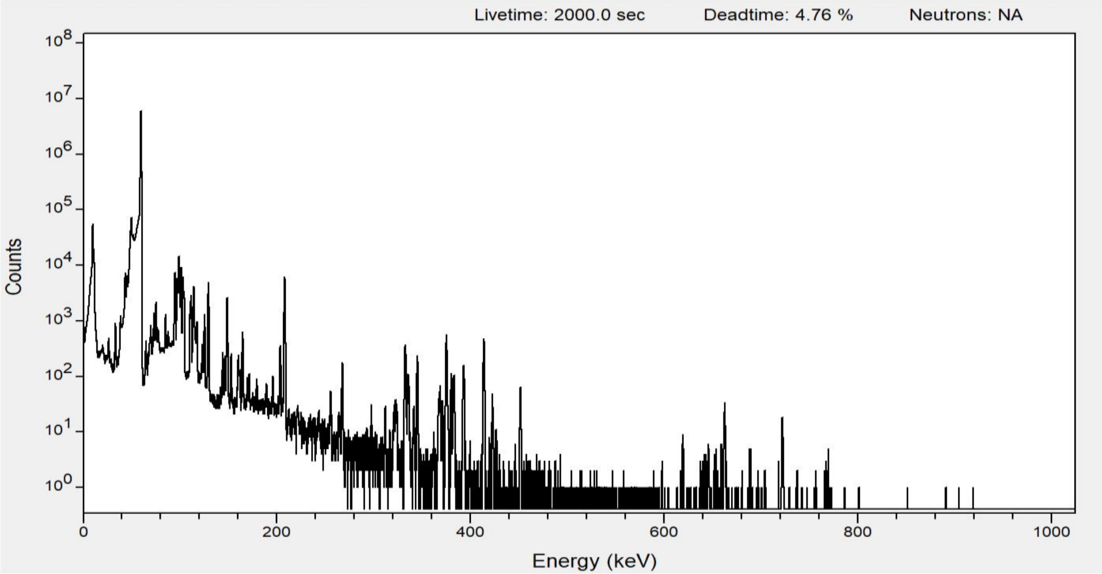
Distributed spectra

Spectrum 1

«*Fuel grade*» Pu (12% ^{240}Pu)

10 years old

Normal Coax HPGE with no filter present

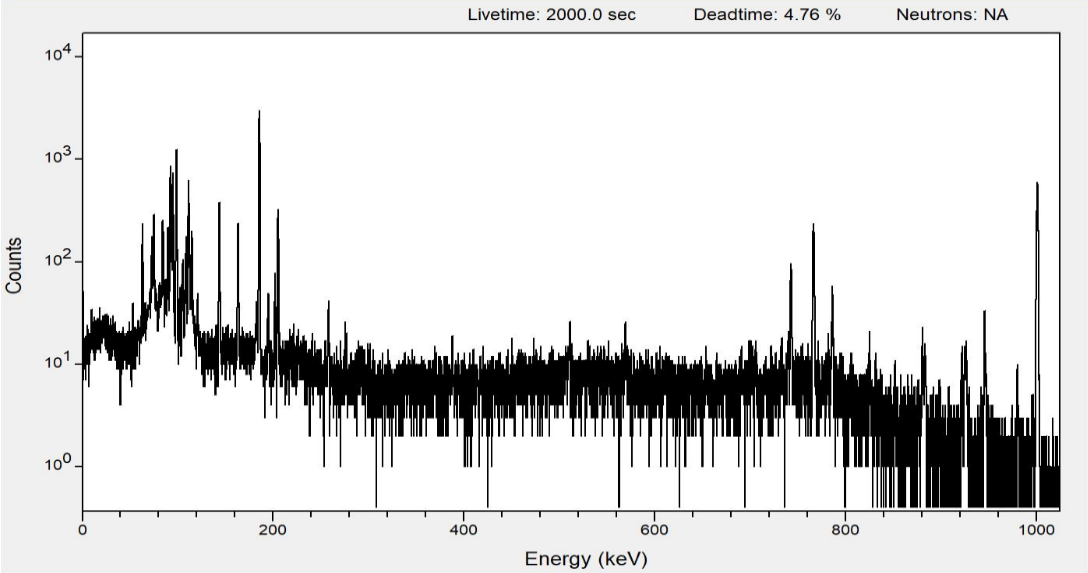


Spectrum 2

«Commercial grade» enriched uranium – (2.96% ^{235}U)

50 years old

Normal Coax HPGe with no filter present



What was hoped for..........

Generation of «*core capability*» information.

→ An identification of what the materials were (plutonium or uranium or whatever) ?

→ An estimate of the enrichment level where applicable

→ A possible positing of the type of material ("*fuel grade*" or "*weapons grade*" etc) ?

→ A possible estimate of the age of the material

→ An estimate of the amount of various isotopes present (preferably in mass quantities)

Example participant: 1

Approach: complex spectral analysis with post-processing and determination of relative efficiencies by Monte Carlo. No attempt at age determination for Pu or mass ratios.

Spectrum 1

Isotope	Reported result Bq	True value Bq	Mass derived from reported activity mg	True mass value mg
^{241}Am	8.53E+06 ± 1.1%	7.72E+06		
^{241}Pu	3.64E+08 ± 0.8%	3.77E+08	0.09503	0.09843
^{237}U	9.00E+03 ± 0.8%	9.26E+03		
^{239}Pu	1.93E+07 ± 0.6%	1.98E+07	8.404	8.61

Spectrum 2 Reported enrichment grade: 2.8 % (true grade: 2.9 %)

Isotope	Reported Activity Bq	True Activity Bq	Reported mass g	True mass g
^{234}U	1.15E+06 ± 11%	1.045E+06	0.005	0.0045
^{235}U	3.32E+04 ± 0.9%	3.52E+04	0.415	0.440
^{238}U	1.81E+05 ± 1.3%	1.7956E+05	14.554	14.434

Example participant: 2

Approach: complex spectral analysis with post-processing. No attempt at age determination for Pu.

Spectrum 1

Isotope	Reported mass fraction at time of counting %	True mass fraction at time of counting %
^{238}Pu	0.1%	0.1%
^{239}Pu	88.82%	86.3%
^{241}Am	0.53%	0.61%
^{240}Pu	9.62%	12.0%
^{241}Pu	0.93%	0.985%
^{237}U	0.001%	0.005%

Spectrum 2

Isotope	Reported mass g	True mass g
^{235}U	0.3	0.440
^{238}U	11.29	14.43

Example participant: 3

Approach: conventional gamma spectrometry and post processing. *This is a Pu sample that was created/purified (Pu extracted) roughly 10 years ago….*

Spectrum 1

Isotope	Reported result MBq/mg	True value MBq/mg
^{239}Pu	1.97 ± 2.3 %	1.98
^{240}Pu	0.750 ± 4.7 %	1.006
^{241}Pu	37.3 ± 2.4 %	37.7
^{241}Am	0.71 ±12.5 %	0.77
^{237}U	0.000946 ± 2.8 %	0.000926

Spectrum 2   Reported enrichment grade: 3.51%

Isotope	Reported activity kBq	True activity kBq
^{238}U	20.9 ± 15%	12.06
^{235}U	4.181 ± 8%	2.36
^{234}U	139.2 ± 27.8%	70.2

Example participant: 4

Approach: conventional gamma spectrometry

Spectrum 1: *Pu from the gamma measurement could arise from nuclear fuel*. No age estimate.

Isotope	Reported activity kBq/mg	True value kBq/mg
^{238}Pu	5.2E+05	5.86E+05
^{239}Pu	2.0E+06	1.98E+06
^{240}Pu	1.2E+06	1.01E+06
^{241}Pu	3.9E+07	3.77E+07
^{241}Am	8.5E+05	7.72E+05

Spectrum 2 Reported enrichment grade: 2.6 ± 0.7% (True value 2.9%)

Isotope	Reported Activity kBq/g	True Activity kBq/g
^{235}U	0.3	2.366
^{238}U (^{234}Th/^{234m}Pa)	14	12.07

What was learned……..

Speaking personally…….

1. The non-expert analyst with routine software is probably in a position to fulfill most of the «*core capabilities*»
2. Some of characteristics of the materials are possible to determine using routine analysis but it is possible that non-expert analysts are unaware they can/should attempt it ?
3. Training in what to report or how to report is probably of more benefit to the routine analyst than how to determine it ?
4. In some cases generating «*core capability*» data that would stand up in court is probably best left to either regional experts or international entities ? Perhaps………..

What could be worth doing…………….

Accepting that the chance of interception of such materials is small (although the consequences could be significant!)…. such interception will more often than not involve communication of data with another country/organisation/entity. On the regional level it would be nice if (perhaps):

1. There was a regional agreement as to how data pertaining to such materials is reported
2. There was a common understanding of how characteristics of these materials are determined
3. There was agreement as to what units are employed

On a wider level…. «*core capability*» information generated for an interdicted material is subject to pressures or demands or will be utilised in ways that would not be typical for any other gamma measurement.

How to address the above?

Availability

http://www.nks.org/en/nksb/supporting_material/nks-b-rinfor-report-and-spectra.htm

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