Isotopic Composition Analysis and Age Dating of Uranium Samples from Nuclear Fuel Cycle by High Resolution Gamma-ray Spectrometry

A. Apostol[1,2], A. Pantelica[1], O. Sima[2], V. Fugaru[1]

1. *Horia Hulubei National Institute for Physics and Nuclear Engineering (IFIN-HH), Bucharest-Magurele, Romania*
2. *University of Bucharest, Faculty of Physics, Bucharest-Magurele, Romania*

e-mail: andrei.apostol@nipne.ro

Content

1. Why?

2. What?

3. How?

Over 630 nuclear trafficking incidents were recorded in the Black Sea states between 1991 and 2012

Table 1. Highly credible incidents involving unauthorized possession of highly enriched uranium and plutonium-239, 1992–2013

Date	Location	Material	Amount (grams)	IAEA confirmed
6 Oct. 1992	Podolsk, Russia	HEU (90%)	1500	No
29 July 1993	Andreeva Guba, Russia	HEU (36%)	1800	No
28 Nov. 1993	Polyarny, Russia	HEU (20%)	4500	No
Mar. 1994	St Petersburg, Russia	HEU (90%)	2972	Yes
10 May 1994	Tengen-Wiechs, Germany	Pu	6.2	Yes
13 June 1994	Landshut, Germany	HEU (87.7%)	0.795	Yes
25 July 1994	Munich, Germany	Pu	0.24	Yes
8 Aug. 1994	Munich Airport, Germany	Pu	363.4	Yes
14 Dec .1994	Prague, Czech Republic	HEU (87.7%)	2730	Yes
June 1995	Moscow, Russia	HEU (21%)	1700	Yes
6 June 1995	Prague, Czech Republic	HEU (87.7%)	0.415	Yes
8 June 1995	Ceske Budejovice, Czech Republic	HEU (87.7%)	16.9	Yes
29 May 1999	Rousse, Bulgaria	HEU (72.65%)	10	Yes
2000	Elektrostal, Russia	HEU (21%)	3700	No
26 June 2003	Sadahlo, Georgia	HEU (89%)	-170	Yes
Jan. 2006	Tbilisi, Georgia	HEU (89%)	79.5	Yes
11 Mar. 2010	Tbilisi, Georgia	HEU (89%)	18	Yes
27 June 2011	Chisinau, Moldova	HEU (???)	4	Yes

HEU = highly enriched uranium; IAEA = International Atomic Energy Agency; Pu = plutonium.

FORENSICS IN NUCLEAR SECURITY

Sharing Knowledge

Nuclear Forensics

Age, Isotopic Composition, Physical Form, Chemical Composition, Microstructure, Impurities, Dimensions Surface roughness

National Nuclear Forensic Library

Point of Loss-of-Control

Why strengthen Nuclear Forensic capabilities in Romania?


Ukraine
Ukraine
21°
27°
Tisza River
Hungary
Satu Mare
Baia Mare
Suceava
Botosani
Somes R.
Bistrita R.
Dnestr R.
Zalau
Oradea
Bistrita
Iasi
Platra Neamt
Cluj-Napoca
Tirgu Mures
Bacau
Miercurea Ciuc
Arad
Alba Iulia
Mures R.
Romania
Siret R.
Olt R.
Tisa R.
Timisoara
Deva
Sibui
Sfintu Gheorghe
Brasov
Focsani
Danube R.
Arges R.
Galati
Resita
Braila
Rîmnicu Vîlcea
Tulcea
Buzau
45°
Tîrgu Jiu
Tirgoviste
Ploiesti
Danube R.
Drobeta-Turnu Severin
Pitesti
Slobozia
Slatina
Bucharest
Serbia
Craiova
Calarasi
Constanta
Black Sea
0
150 km
0
100 mi
Bulgaria
21°
27°

Example of Nuclear Forensics Library DATA

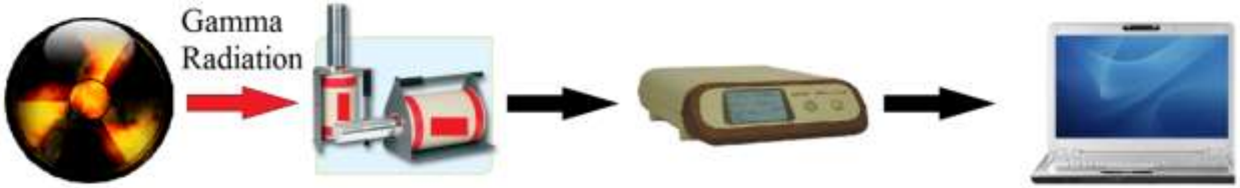
Uranium production facility of country R is producing UO_2 powder of ceramic type with enrichment lower then 20%. General characteristics of the produced powder of Uranium dioxide (UO_2):		
1	Uranium Enrichment (^{235}U%), % mass	From 0.7 to 20
2	Uranium Content, % mass	>87.42
3	Humidity concentration, % mass	<0.3
4	Oxigen coeficient (O/U)	2.077
5	Grain size, mm	<0.9

Mass concentration of metallic impurities		
No.	Element	ppm
1	Al	30
2	B	0.3
3	C	45
4	Ca	100
5	Cd	0.3
6	Cr	30
7	Cu	3
8	F	20
9	Fe	70
10	Mg	7
11	Mo	30
12	N	50
13	Ni	30
14	Si	30
15	Mn	3
16	Pb	30
17	V	30
18	W	19

Weapon-Usable

Uranium

Gamma Spectrometry System



Uranium Isotopic Composition Analysis and Age-Dating

Uranium Isotopic Composition Analysis of UMETA-UAERES samples using High Resolution Gamma Spectrometry and MGAU 4.2 code

Detector Choice

energy-calibration gain set

Recommended Non-Destructive Assay (NDA) Systems *BEGe detector, LEGe detector*

0.075 keV/channel

307 keV on 4096 channels

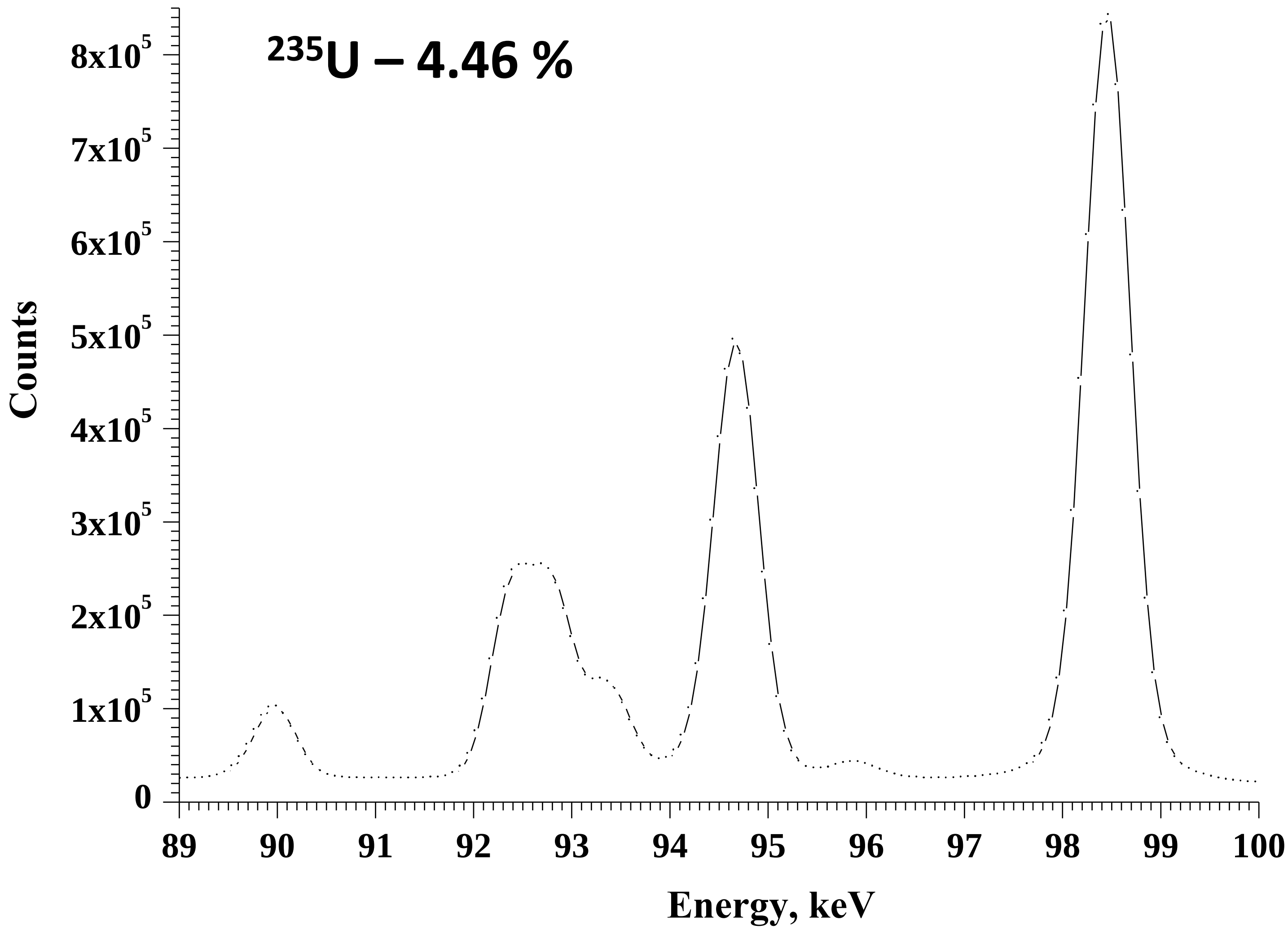
Large NDA Systems
Coaxial HPGe

0.095 keV/channel

1.5 MeV in a single 16k spectrum.

FWHM <750 eV at 122 keV

The main energy region analyzed by MGAU 4.2



[1] R. Gunnink, W.D. Ruhter, P. Miller, J. Goerten, M. Swisnhoe, H. Wagner, J. Verplancke, M. Bickel, S. Abousahl, MGAU: A new analysis code for measuring U-235 enrichments in arbitrary samples, IAEA-SM-333/88.

Detector Characteristics/Experimental Setup

1. GEM-30185, Relative efficiency: 30%, Resolution: 875 eV at 122 keV (Low background set-up, Lab. GammaSpec)

2. GL0105P, Resolution 495 eV at 122 keV (Remus), Active area: 100 mm^2

3. GL0055P, Resolution 495 eV at 122 keV (Romulus), Active area: 50 mm^2

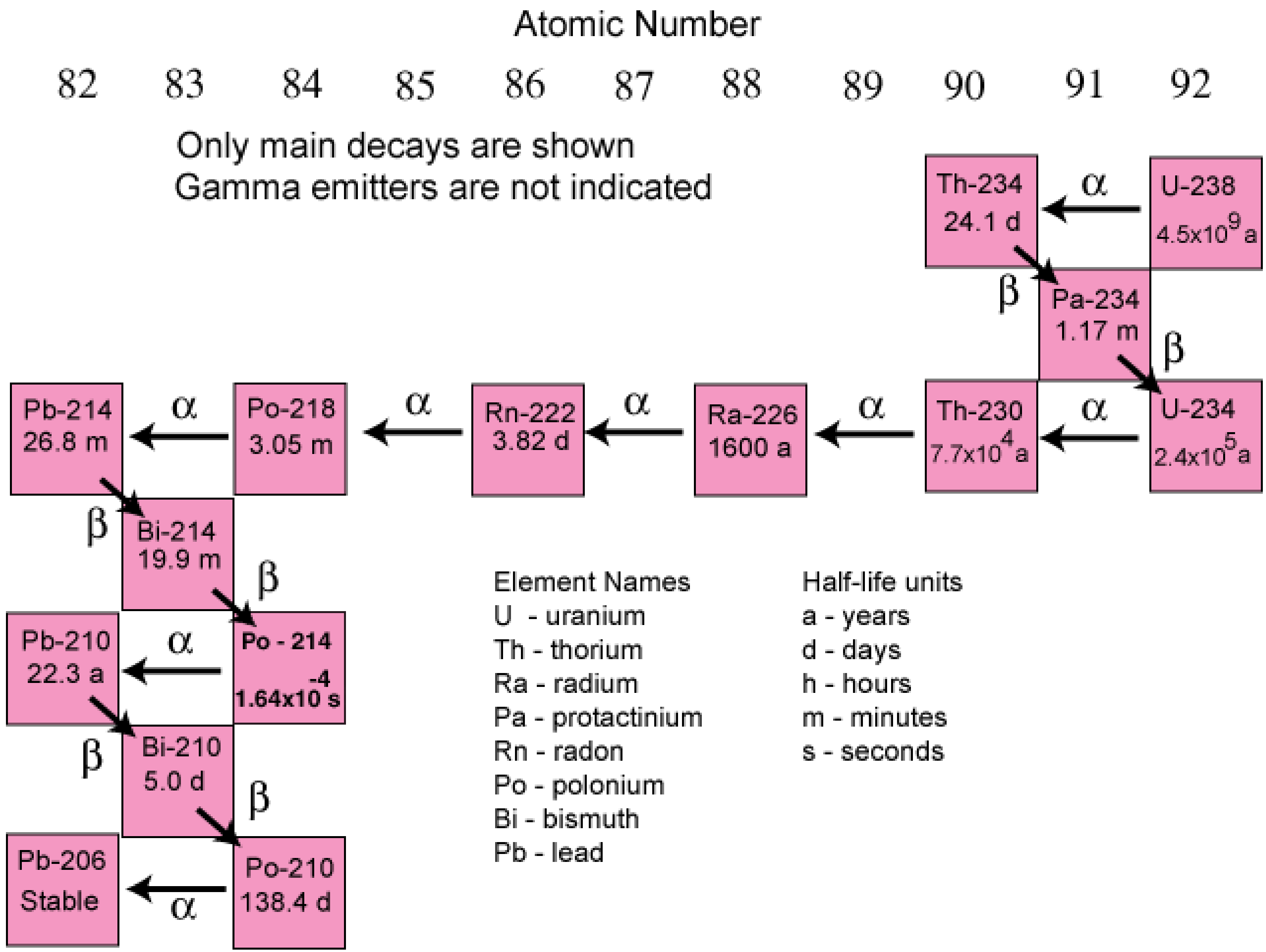
Nuclear Forensics Gamma Spectrometry Systems at IFIN-HH

Uranium Isotopic Composition Analysis
UMETA-AAERES (MGAU 4.2)

Material ID	Declared Enrichment	m (g)	T (h)	Measured Isotopic Composition					
				Uranium 234		Uranium 235		Uranium 238	
				Weight (%)	σ (%)	Weight (%)	σ (%)	Weight (%)	σ (%)
UMETA-0	**0.5**	2.11	19.9	-	-	**0.49**	7.53	**99.50**	0.04
UMETA-01	**1**	2.27	48.1	**0.011**	18.46	**1.01**	1.75	**98.98**	0.02
UMETA-03	**3**	2.06	5.4	**0.028**	31.63	**2.99**	2.49	**96.98**	0.08
UMETA-06	**6**	1.96	15.6	**0.046**	9.33	**5.99**	0.98	**93.97**	0.06
UMETA-1	**10**	2.03	16.7	**0.080**	6.15	**9.97**	0.85	**89.95**	0.1
UMETA-2	**20**	2.36	8.6	**0.164**	3.8	**19.81**	0.77	**80.02**	0.19
UMETA-3	**30**	2.37	15.5	**0.247**	2.92	**29.80**	0.67	**69.95**	0.29
UMETA-6	**60**	1.84	7.9	**0.504**	2.40	**59.02**	0.72	**40.47**	1.07
UAERES	**93**	1.87	23.2	**0.988**	1.25	**92.18**	0.79	**6.83**	10.75

Uranium age-dating of UMETA-UAERES samples using High Resolution Gamma Spectrometry

The Uranium-238 Decay Chain



Uranium Age Dating

^{214}Bi/^{234}U Chronometer

$$\frac{A_{\mathrm{Bi214}}(T)}{A_{\mathrm{U234}}(T)} = \frac{A_{\mathrm{Bi214}}(T)}{A_{\mathrm{U234}}(0)} = \frac{A_{\mathrm{Ra226}}(T)}{A_{\mathrm{U234}}(0)} = \lambda_2\lambda_3\left[\frac{\mathrm{e}^{-\lambda_1 T}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)} + \frac{\mathrm{e}^{-\lambda_2 T}}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)} + \frac{\mathrm{e}^{-\lambda_3 T}}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)}\right]$$

	Nuclide	$T_{1/2}$ (y)	λ (y^{-1})
1	^{234}U	2.46E+05	2.823E-06
2	^{230}Th	7.538E+04	9.195E-06
3	^{226}Ra	1600	4.332E-04

Taylor Series around T=0

$$\frac{A_{\mathrm{Bi214}}(T)}{A_{\mathrm{U234}}(T)} = \frac{1}{2}\lambda_2\lambda_3 T^2$$

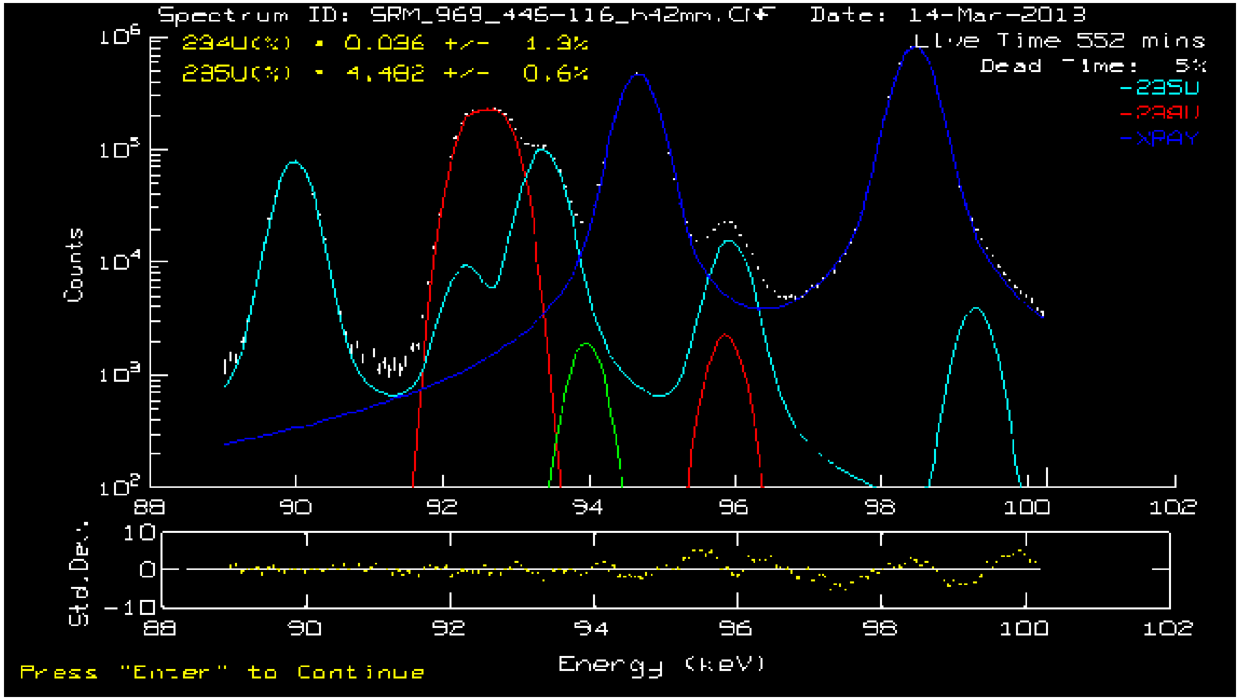
[2] Nguyen, C. T., J. Zsigrai, Gamma-spectrometric uranium age-dating using intrinsic efficiency calibration, Nuclear Instruments and Methods in Physics Research B 243 (2006) 187–192.

$$\frac{A(^{214}Bi)}{A(^{234}U)} = \frac{A(^{214}Bi)}{A(^{238}U)} \left(\frac{A(^{235}U)}{A(^{238}U)} \frac{A(^{234}U)}{A(^{235}U)} \right)^{-1}$$

$$\frac{A(^{214}Bi)}{A(^{238}U)} = \frac{cps_{609.3}/I_{609.3}}{\mathbf{F(609.3)}}$$

$$\frac{A(^{234}U)}{A(^{235}U)} = \frac{cps_{120.9}/I_{120.9}}{\mathbf{f(120.9)}}$$

$$\frac{A(^{235}U)}{A(^{238}U)}$$



$$\frac{A(^{214}Bi)}{A(^{234}U)} = \frac{A(^{214}Bi)}{A(^{238}U)}\left(\frac{A(^{235}U)}{A(^{238}U)}\frac{A(^{234}U)}{A(^{235}U)}\right)^{-1}$$

$$\frac{A(^{214}Bi)}{A(^{238}U)} = \frac{cps_{609.3}/I_{609.3}}{\mathbf{F(609.3)}}$$

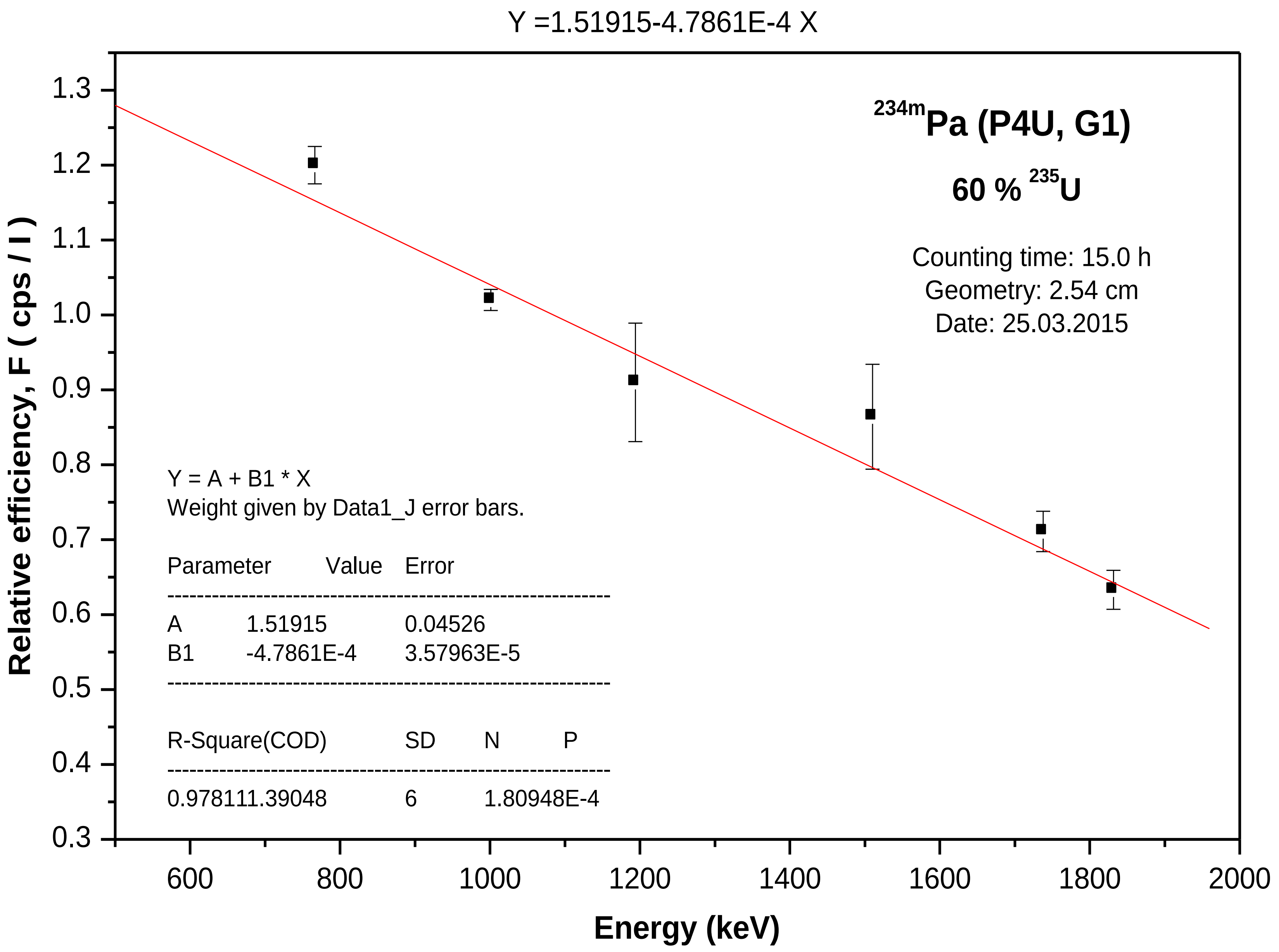
$$\frac{A(^{234}U)}{A(^{235}U)} = \frac{cps_{120.9}/I_{120.9}}{\mathbf{f(120.9)}}$$

<u>Activity ratios determination / Relative efficiency curves</u>

A = R / (ε · I) => **A · ε = R / I**

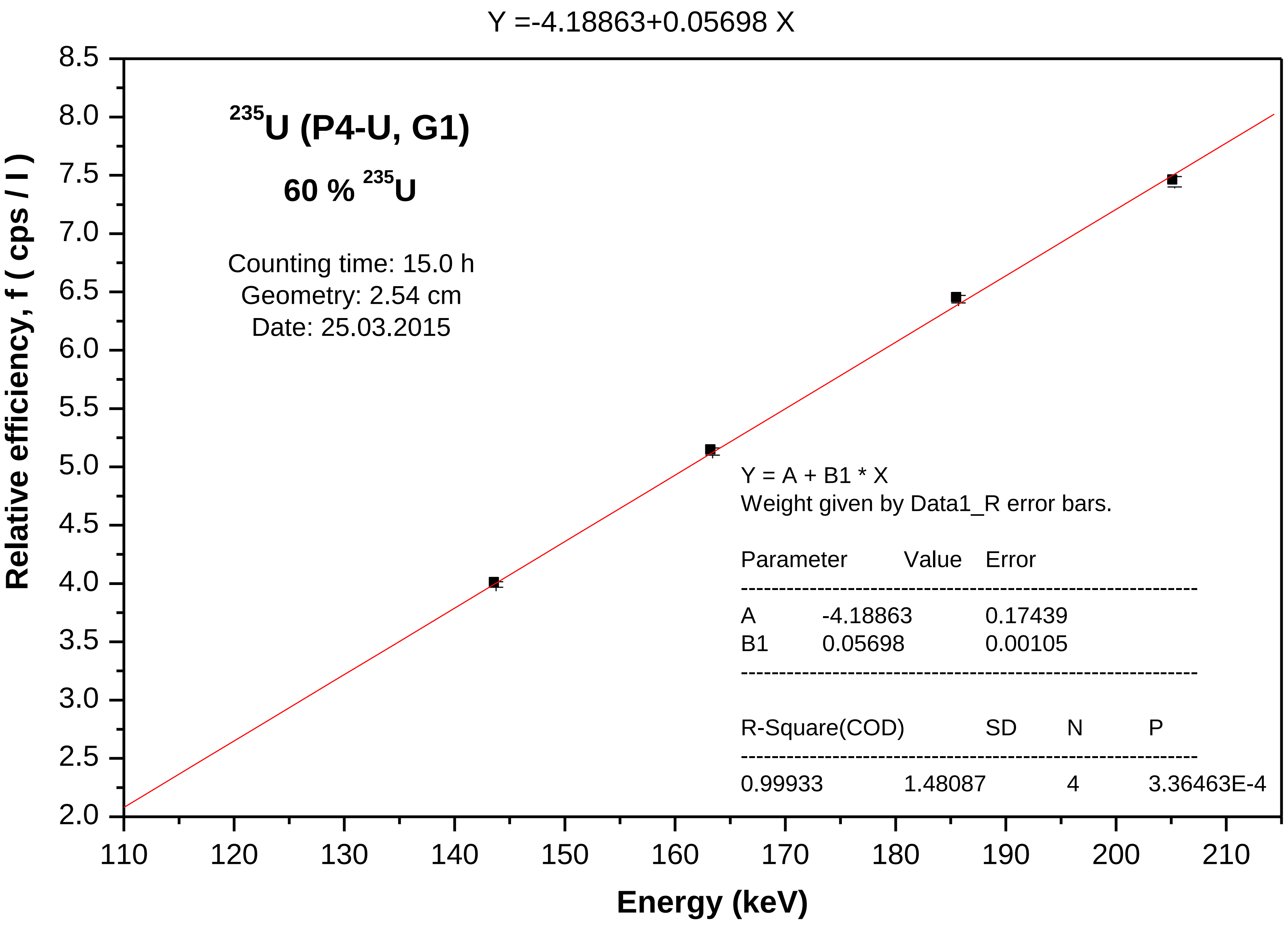
A – activity
R – photon detection rate [counts per second, cps]
ε – Full Energy Peak Efficiency
I – Emission probability

F(609.3)



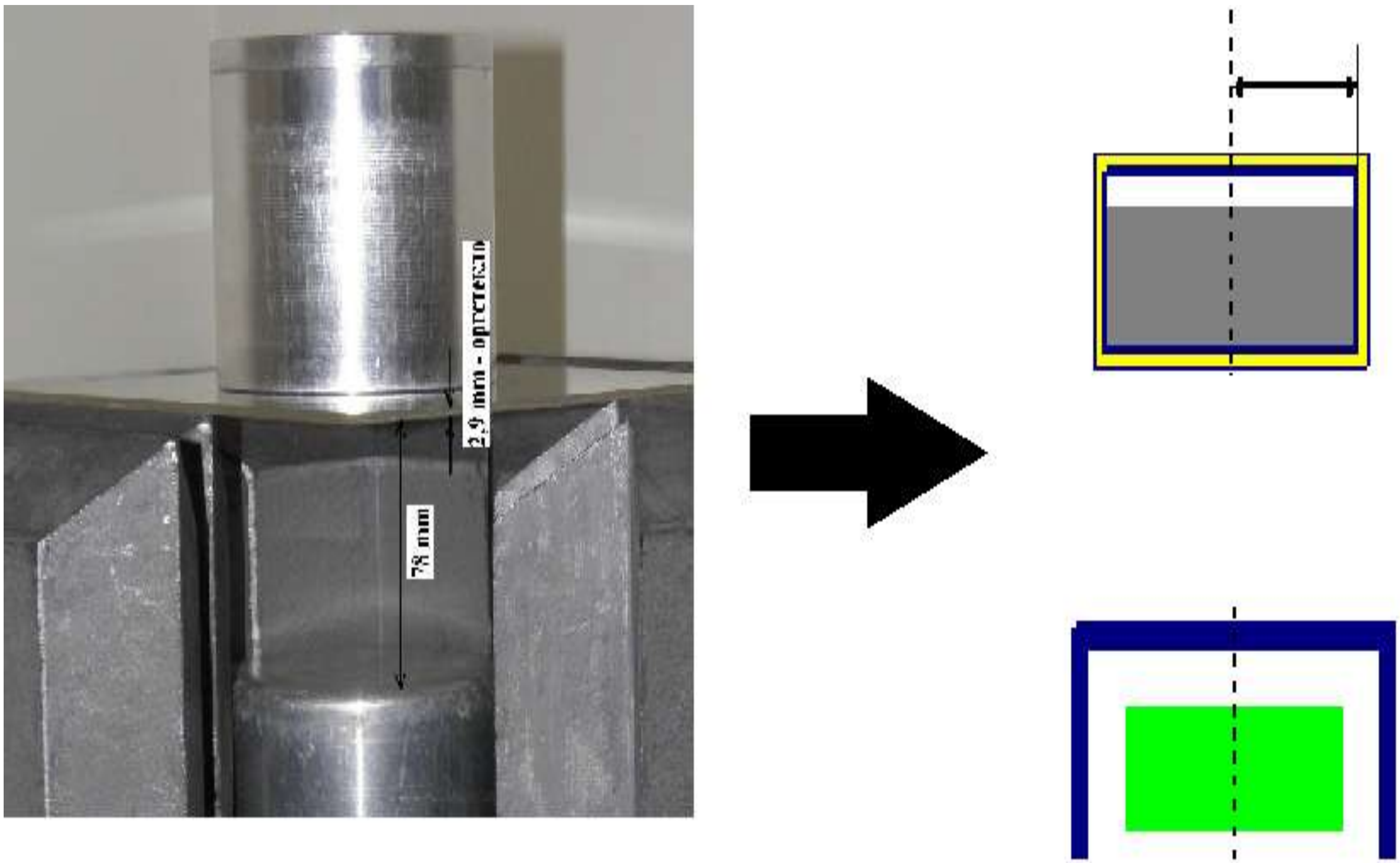
Y =1.51915-4.7861E-4 X
234mPa (P4U, G1)
60 % 235U
Counting time: 15.0 h
Geometry: 2.54 cm
Date: 25.03.2015
Y = A + B1 * X
Weight given by Data1_J error bars.
Parameter Value Error
A 1.51915 0.04526
B1 -4.7861E-4 3.57963E-5
R-Square(COD) SD N P
0.97811 1.39048 6 1.80948E-4
Relative efficiency, F (cps / I)
Energy (keV)
1.3
1.2
1.1
1.0
0.9
0.8
0.7
0.6
0.5
0.4
0.3
600
800
1000
1200
1400
1600
1800
2000

f(120.9)

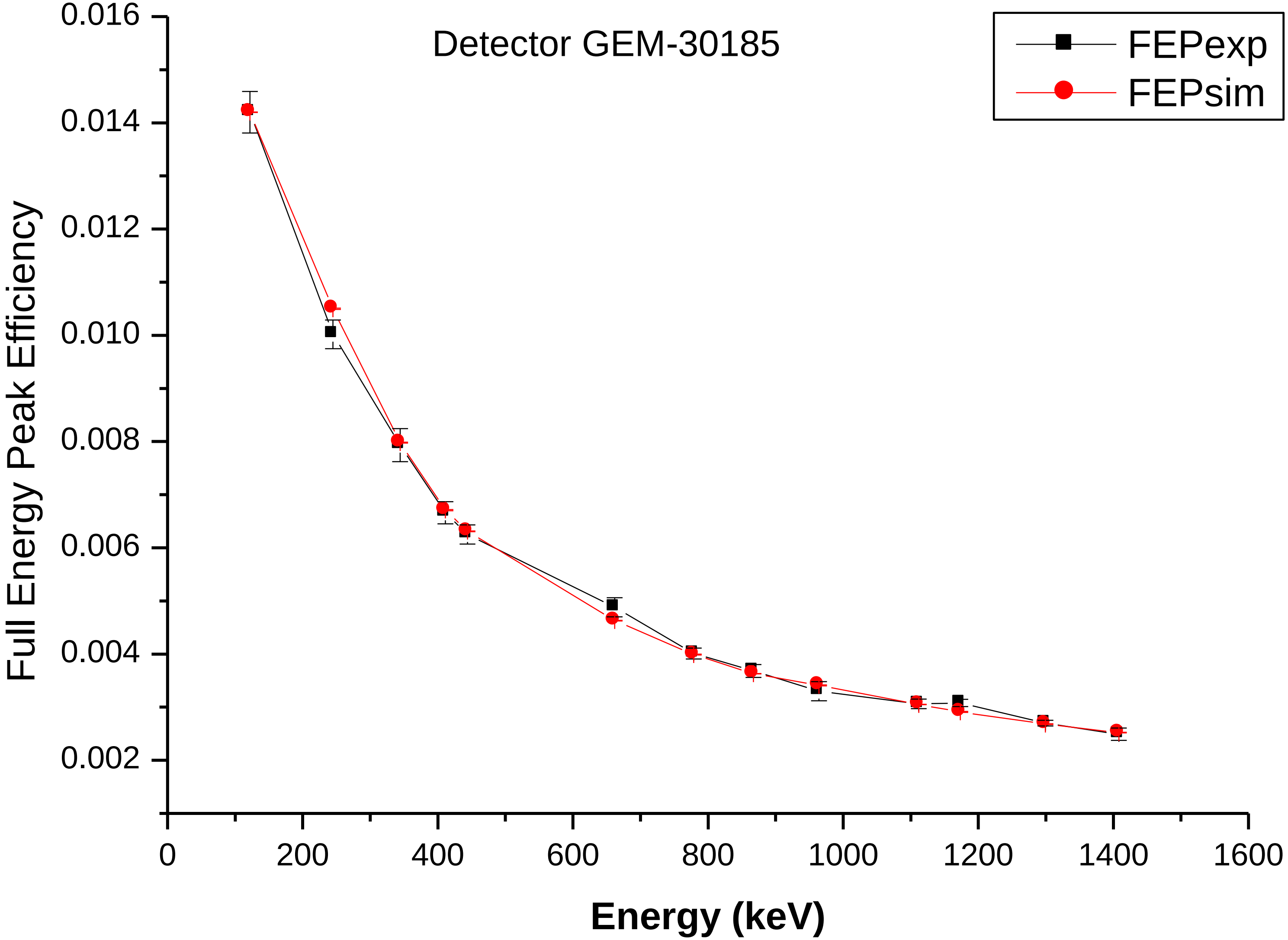

Y =-4.18863+0.05698 X
235U (P4-U, G1)
60 % 235U
Counting time: 15.0 h
Geometry: 2.54 cm
Date: 25.03.2015
Y = A + B1 * X
Weight given by Data1_R error bars.
Parameter Value Error
A -4.18863 0.17439
B1 0.05698 0.00105
R-Square(COD) SD N P
0.99933 1.48087 4 3.36463E-4
Relative efficiency, f (cps / I)
Energy (keV)
8.5
8.0
7.5
7.0
6.5
6.0
5.5
5.0
4.5
4.0
3.5
3.0
2.5
2.0
110
120
130
140
150
160
170
180
190
200
210

		GEM (fit 609 keV)		GEM (fit 121 keV)		Remus						
		R1		R2		R3		R4= R1/(R2*R3)				
CPR Code	**235U(%) (aprox)**	**A(214Bi)/ A(238U)**	**σ (%)**	**A(234U)/ A(235U)**	**σ (%)**	**A(235U)/ A(238U)**	**σ (%)**	**A(214Bi)/ A(234U)**	**σ (%)**	**ΔT (y)**	**σ (y)**	Date (zero)
U-META-0	0.5	**3.365E-05**	100	**17.863**	36.4	**0.0319**	7.5	**5.90E-05**	106.7	**172**	184	**1843.3???**
U-META-01	1	**1.673E-05**	100	**23.971**	8.8	**0.0656**	1.8	**1.06E-05**	100.4	**73.1**	73.4	**1942.2???**
U-META-03	3	**3.924E-05**	17.9	**20.215**	8.48	**0.1984**	2.5	**9.78E-06**	20.0	**70.1**	14.0	**1945.3(14.0)**
U-META-03	3	**3.924E-05**	17.9	**26.951**	31.7	**0.1984**	2.5	**7.34E-06**	36.5	**60.7**	22.2	**1954.6(22.2)**
U-META-06	6	**4.827E-05**	16.6	**20.561**	10.4	**0.4101**	1.0	**5.73E-06**	19.6	**53.6**	10.5	**1961.7(10.5)**
U-META-06	6	**4.827E-05**	16.6	**22.459**	9.4	**0.4101**	1.0	**5.24E-06**	19.1	**51.3**	9.8	**1964.0(9.8)**
U-META-1	10	**1.380E-04**	12.6	**21.466**	9.10	**0.713**	0.9	**9.02E-06**	15.5	**67.3**	10.5	**1948.0(10.5)**
U-META-1	10	**1.380E-04**	12.6	**23.370**	6.21	**0.713**	0.9	**8.28E-06**	14.0	**64.5**	9.0	**1950.8(9.0)**
U-META-2	20	**1.184E-04**	8.3	**21.581**	9.71	**1.594**	1.4	**3.44E-06**	12.8	**41.6**	5.3	**1973.8(5.3)**
U-META-2	20	**1.184E-04**	8.3	**24.720**	7.6	**1.594**	1.4	**3.01E-06**	11.3	**38.8**	4.4	**1976.5(4.4)**
U-META-2	20	**1.364E-04**	13.6	**21.796**	5.7	**1.594**	1.4	**3.93E-06**	14.8	**44.4**	6.6	**1971.0(6.6)**
U-META-2	20	**1.364E-04**	13.6	**24.720**	7.6	**1.594**	1.4	**3.46E-06**	15.6	**41.7**	6.5	**1973.7(6.5)**
U-META-3	30	**2.783E-04**	8.1	**20.886**	9.3	**2.740**	0.7	**4.86E-06**	12.3	**49.4**	6.1	**1966.0(6.1)**
U-META-3	30	**2.783E-04**	8.1	**24.049**	3.0	**2.740**	0.7	**4.22E-06**	8.6	**46.1**	4.0	**1969.4(4.0)**
U-META-6	60	**7.312E-04**	6.6	**21.443**	8.1	**9.378**	1.3	**3.64E-06**	10.5	**42.7**	4.5	**1972.6(4.5)**
U-META-6	60	**7.312E-04**	6.6	**24.721**	2.5	**9.378**	1.3	**3.15E-06**	7.2	**39.8**	2.9	**1975.5(2.9)**
U-AE-RES	93.3	**1.377E-02**	10.4	**26.456**	10.4	**86.729**	10.8	**6.00E-06**	18.2	**54.9**	10.0	**1960.4(10)**

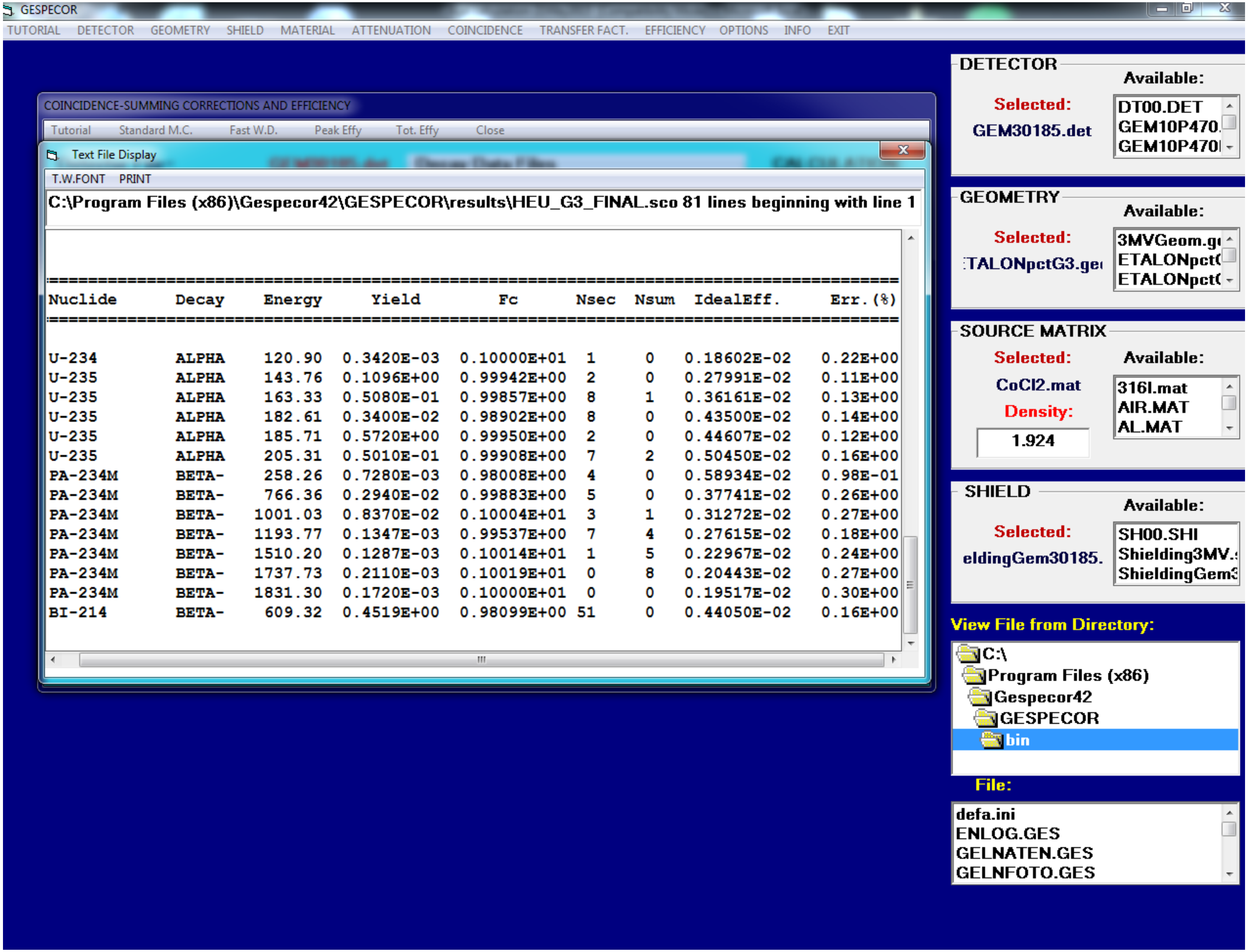
Age dating of 93% enriched Uranium sample U-AE-RES using Monte Carlo simulations software GESPECOR and high resolution low background gamma spectrometry



The geometry transfer to GESPECOR software



Detector GEM-30185
FEPexp
FEPsim
Full Energy Peak Efficiency
0.016
0.014
0.012
0.010
0.008
0.006
0.004
0.002
0
200
400
600
800
1000
1200
1400
1600
Energy (keV)

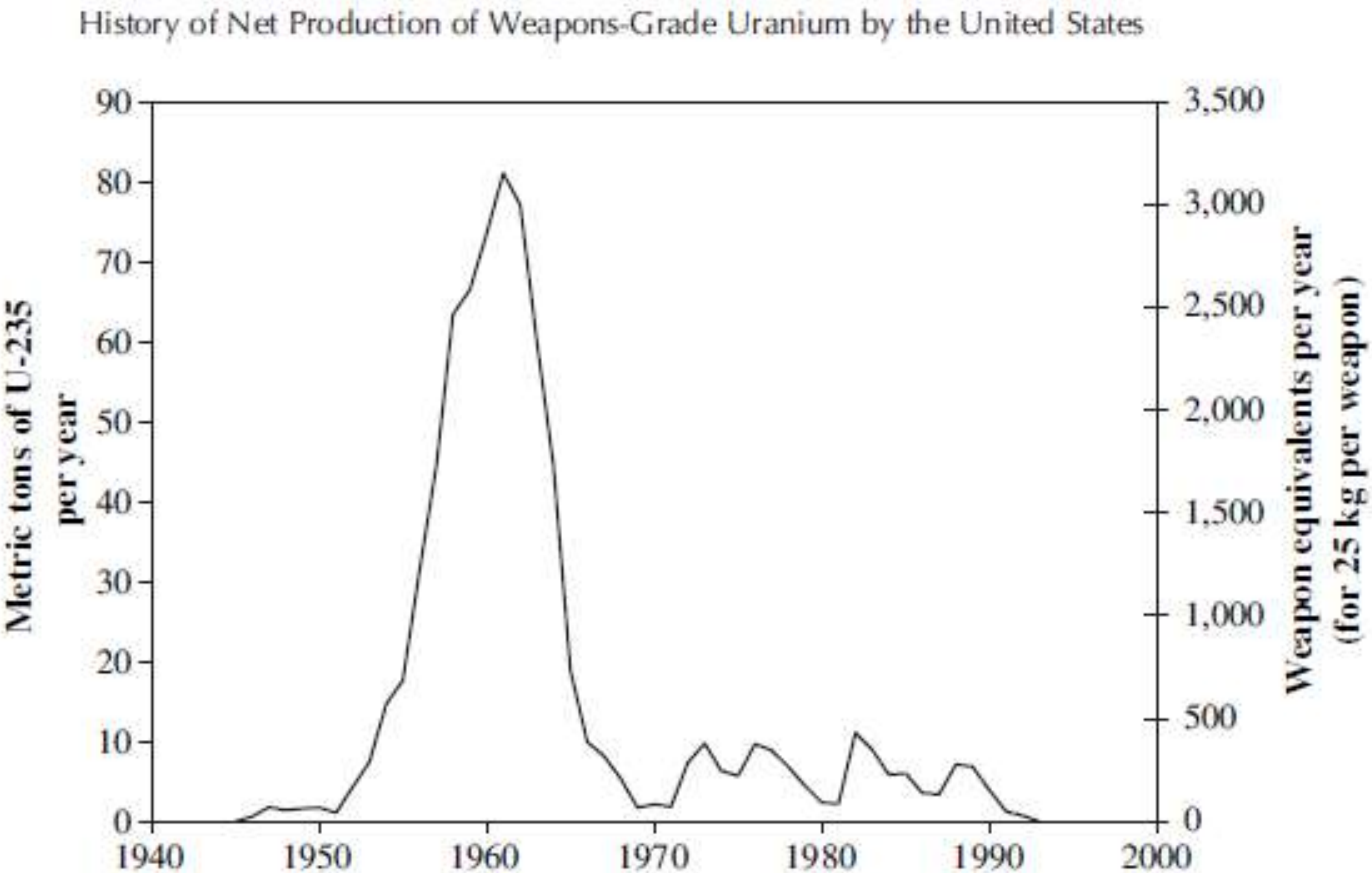


[3] O. Sima, D. Arnold, C. Dovlete, GESPECOR: A versatile tool in gamma-ray spectrometry, Journal of Radioanalytical and Nuclear Chemistry, Vol. 248, No. 2 (2001) 359-364

SDD = 7.62 cm

t_m (h)= 15.92

		GEM (fit 609 keV)		GEM (fit 121 keV)		Remus				
		R1		R2		R3		R4= R1/(R2*R3)		
Sample Code	**235U(%) (aprox)**	**A(214Bi)/ A(238U)**	**σ (%)**	**A(234U)/ A(235U)**	**σ (%)**	**A(235U)/ A(238U)**	**σ (%)**	**A(214Bi)/ A(234U)**	**σ (%)**	Date (zero)
U-AE-RES	93.3	**1.377E-02**	10.4	**26.456**	10.4	**86.729**	10.8	**6.00E-06**	18.2	1960.4(10)
		(Gespecor, 609/1001)		(MGAU)		(MGAU)				
U-AE-RES	93.3	**1.4769E-02**	7.0	**31.047**	1.5	**86.729**	10.8	**5.49E-06**	12.9	1962.8(6.8)
		(Gesp., 609/1001)		(Gesp., 121/186)		(MGAU)				
U-AE-RES	93.3	**1.4769E-02**	7.0	**25.399**	3.3	**86.729**	10.8	**6.70E-06**	13.3	1957.3(7.7)
		(Gesp., 609/1001)		(Gesp., 121/186)		(Gesp.,186 /1001)				
U-AE-RES	93.3	**1.4769E-02**	7.0	**25.399**	3.3	**80.077**	4.9	**7.26E-06**	9.2	1954.9(5.5)
Gespecor								(Gespecor, 609/121 keV)		
U-AE-RES	93.3							**7.26E-06**	5.94	1954.9(3.6)



[4] Steven Aftergood and Frank N. von Hippel, REPORT THE U.S., HIGHLY ENRICHED URANIUM DECLARATION: TRANSPARENCY DEFERRED BUT NOT DENIED

Limitations

- Characteristics determination (Chemistry)
- Software limitations (Geometry description)

Future Work

Nuclear Forensics

Age, Isotopic Composition, Physical Form, Chemical Composition, Microstructure, **Impurities**, Dimensions Surface roughness

National Nuclear Forensic Library

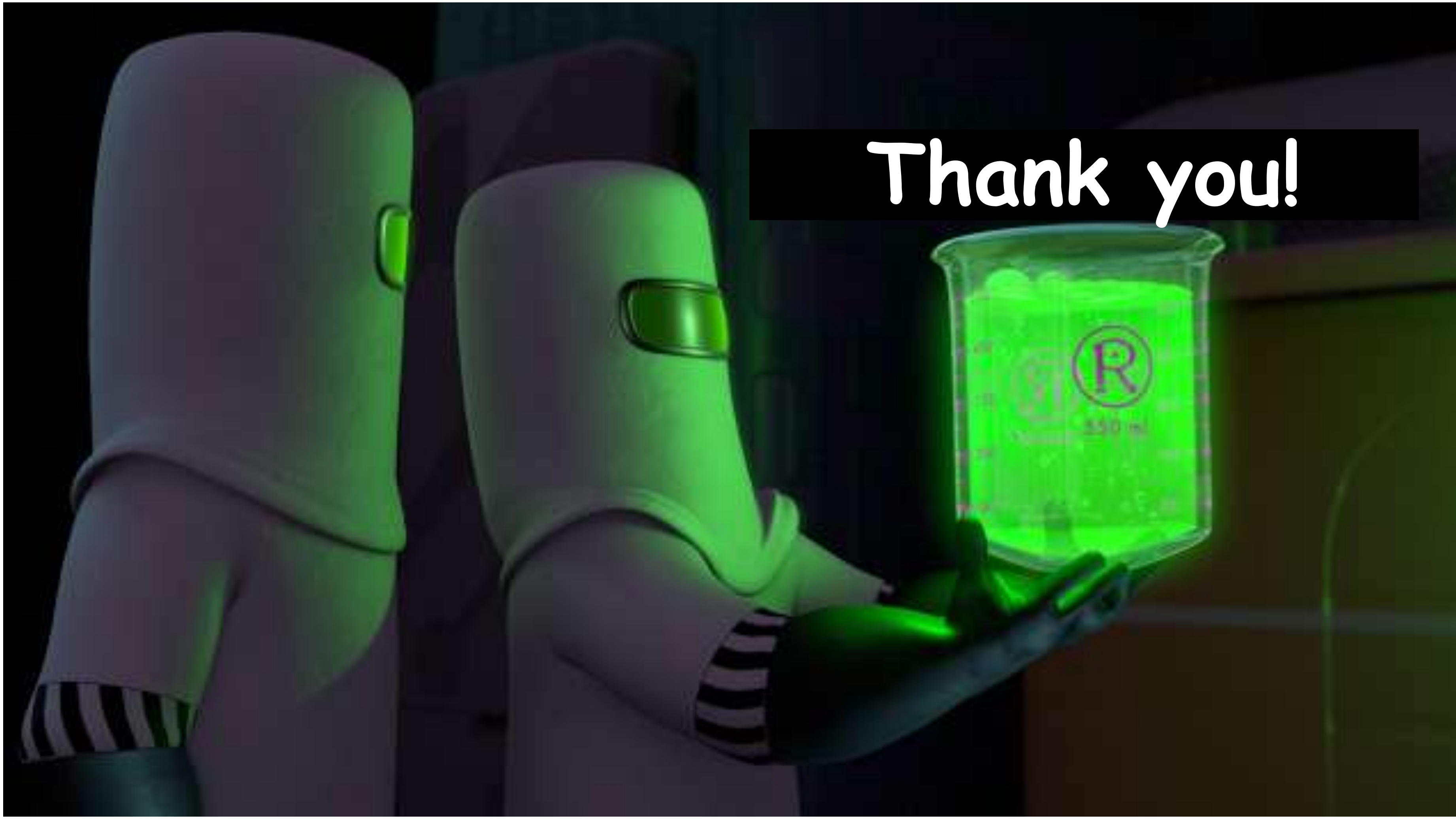
Point of Loss-of-Control

3 MV TANDETRON ACCELERATOR

1 MV TANDETRON ACCELERATOR (AMS)

Forensics can help reproduce the radiological incidents or even better –

Prevent Them



"In a strange turn of history, the threat of global nuclear war has gone down, but the risk of a nuclear attack has gone up." ©