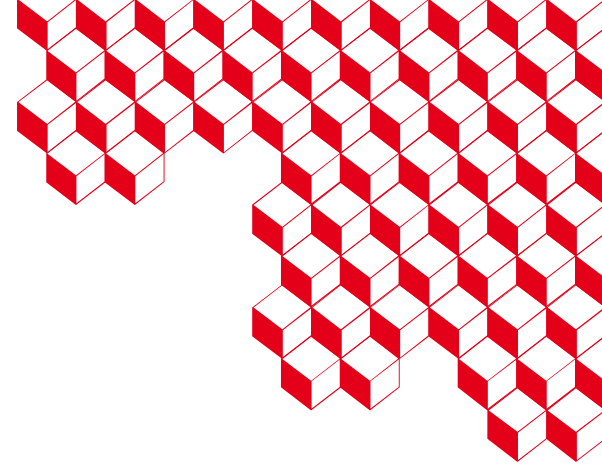




iresne



Scintillators for nondestructive assay in industrial and security applications

Bertrand PEROT, CEA Fellow – CEA DES IRESNE, DTN/SMTA – Nuclear Measurement Lab



Summary

- 1. NUCLEAR MEASUREMENT LAB**
- 2. X-RAY IMAGING**
- 3. GAMMA-RAY SPECTROSCOPY**
- 4. NEUTRON MEASUREMENTS**
- 5. NEUTRON & PHOTON ACTIVATION**
- 6. CONCLUSION & PROSPECTS**





1 ■ Introduction

The Nuclear Measurement Laboratory

- ❑ CEA, DES Energy Division, IRESNE institute at CEA Cadarache
- ❑ 25-30 permanent staff + 5-10 PhD, trainees, temporary contracts
- ❑ 50 - 60 projects or collaborations on R&D and applications



Cadarache



Non-destructive Nuclear Measurements

❑ Radiological characterization

- ✓ Gamma-ray spectroscopy
- ✓ Passive and active neutron measurements
- ✓ Photofission

❑ Physical characterization

- ✓ Photon radiography and tomography
- ✓ Neutron radiography

❑ Elemental characterization

- ✓ Thermal neutron activation analysis
- ✓ Fast neutron activation analysis

Applications

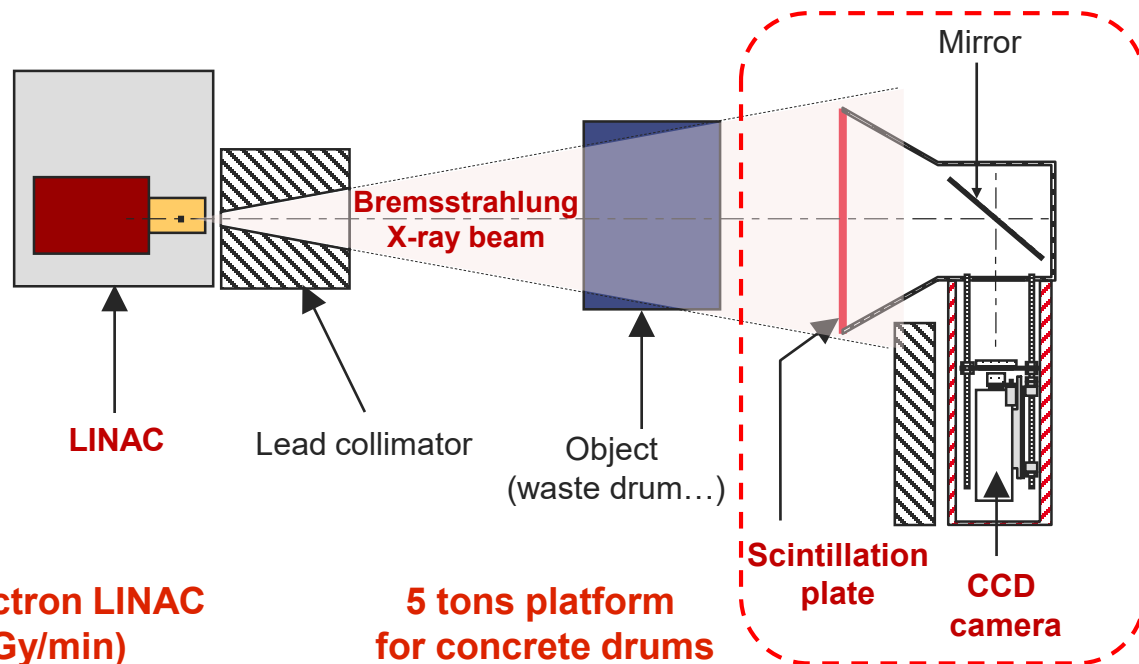


- ❑ **Nuclear fuel cycle**
 - ✓ Uranium mining, enrichment, fuel fabrication & reprocessing
 - ✓ Radioactive waste characterization
 - ✓ Dismantling and decommissioning
- ❑ **Reactors**
 - ✓ Non-destructive exams for Jules Horowitz Reactor
 - ✓ Nuclear accident studies and monitoring
- ❑ **Homeland Security and Safeguards**
 - ✓ CBRNE threats detection
 - ✓ Nuclear material controls
- ❑ **Waste recycling**
 - ✓ Characterization of valuable materials



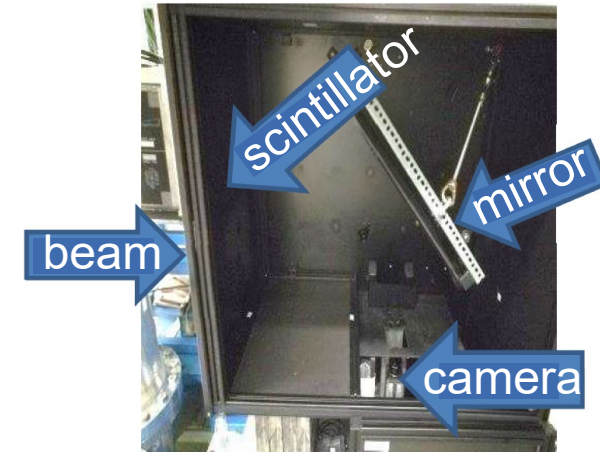
2 ■ **Scintillators for X-ray imaging**

High energy X-ray imaging in CINPHONIE casemate

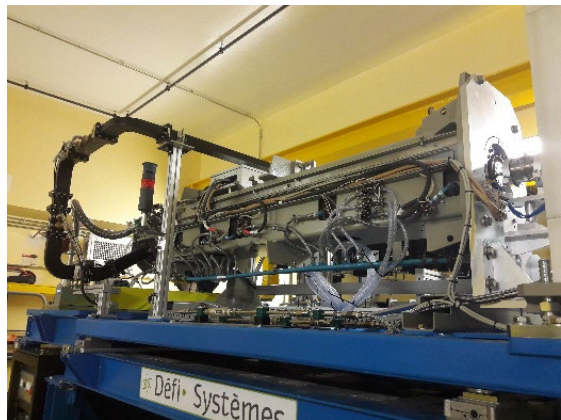


15-20 MeV electron LINAC
(up to 65 Gy/min)

5 tons platform
for concrete drums



Scintillation plate 800 x 600 mm²
Gadox Gd₂O₂S thickness 1.5 mm



High energy X-ray imaging of large concrete drums

Concrete waste package

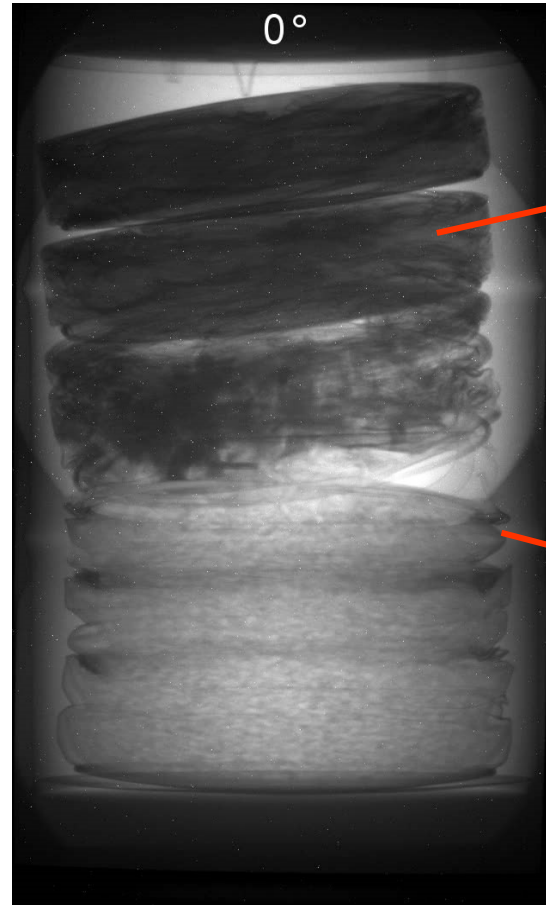
- Density $> 2 \text{ g/cm}^3$
- Mass $> 1,2 \text{ ton}$
- H = 120 cm
- $\varnothing = 84 \text{ cm}$

$\ll 1 \text{ s}$ per radiography

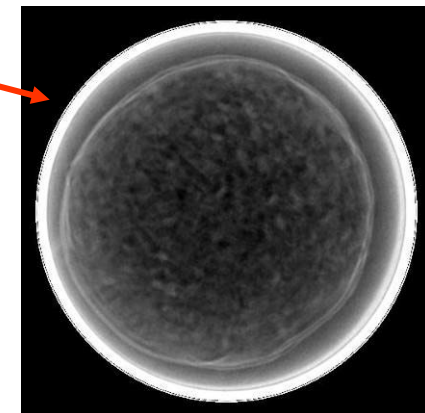
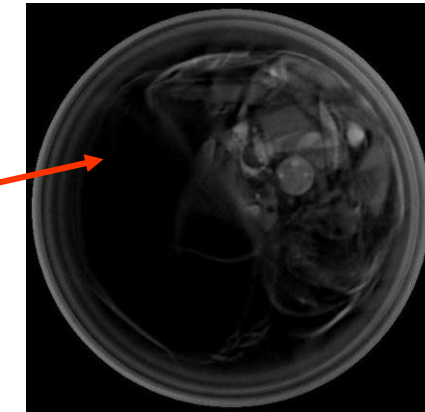
but due to mechanical movements

- Radiography $\approx 10 \text{ min}$
- Tomography $\approx 25 \text{ min}$

■ Radiography

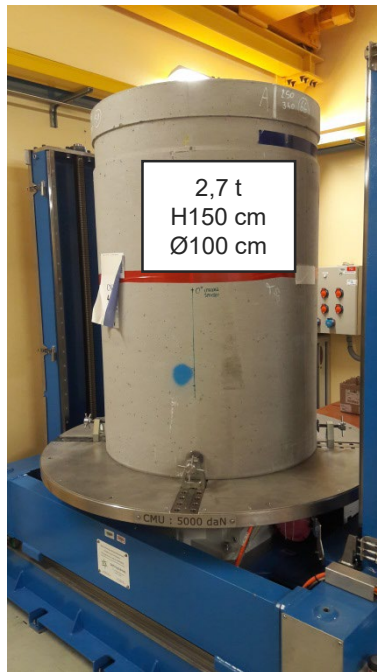


■ Tomography

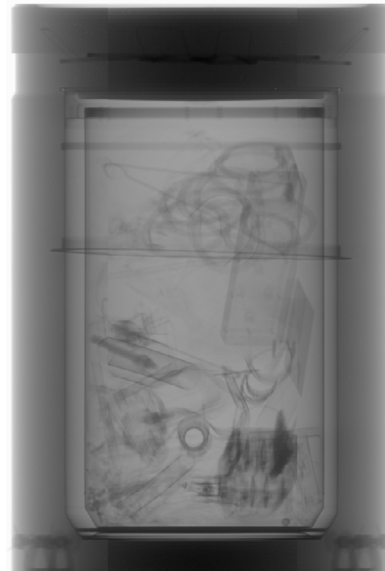


Detailed expertise of a large concrete drum

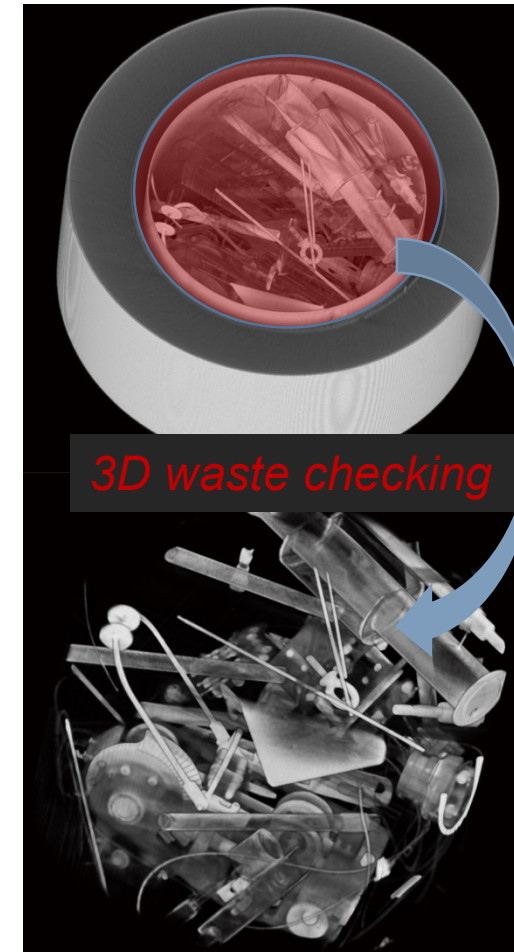
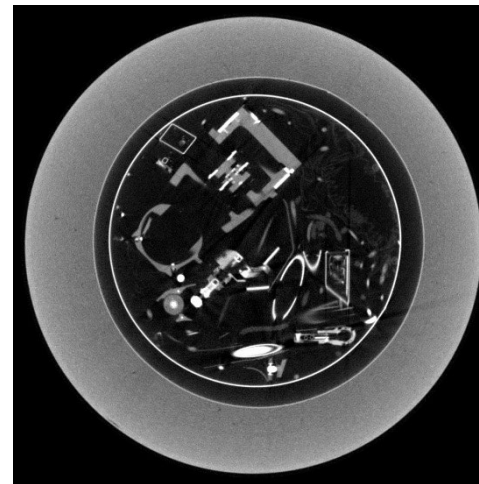
500 l waste blocked in a concrete matrix
(external volume 1.2 m³)



Radiographies
(duration 10 min)



2D/3D Tomographies
(duration 30 min)



High energy X-ray radioscopy

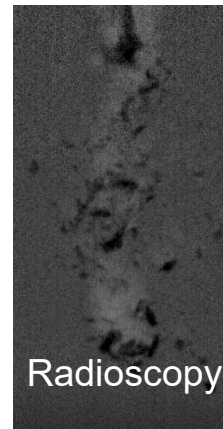
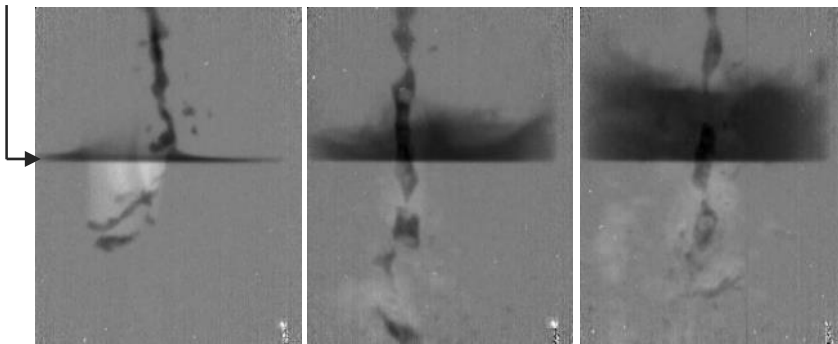
■ Dynamic radiography

- Original camera 50 pictures / sec – 230 × 240 pixels
- With 4 modern cameras ⇒ up to 200 pict/s – 2560 × 2160 pixels

■ Application to corium / water interaction

⇒ MC3D code validation (nuclear accident studies)

Water surface



KROTOS facility



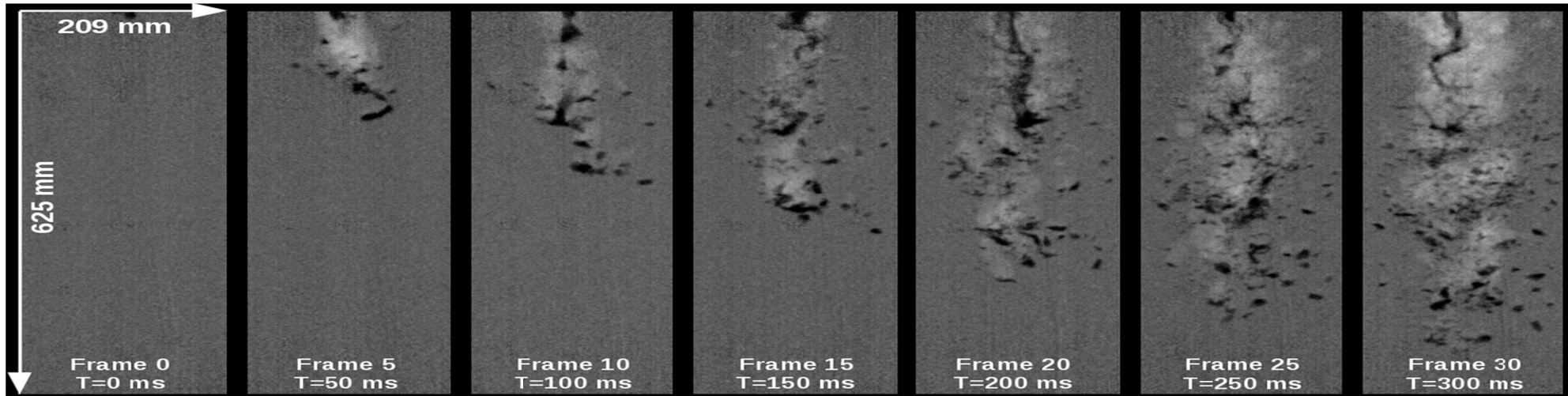
In yellow: 9 MeV LINAC
In black : imaging plate



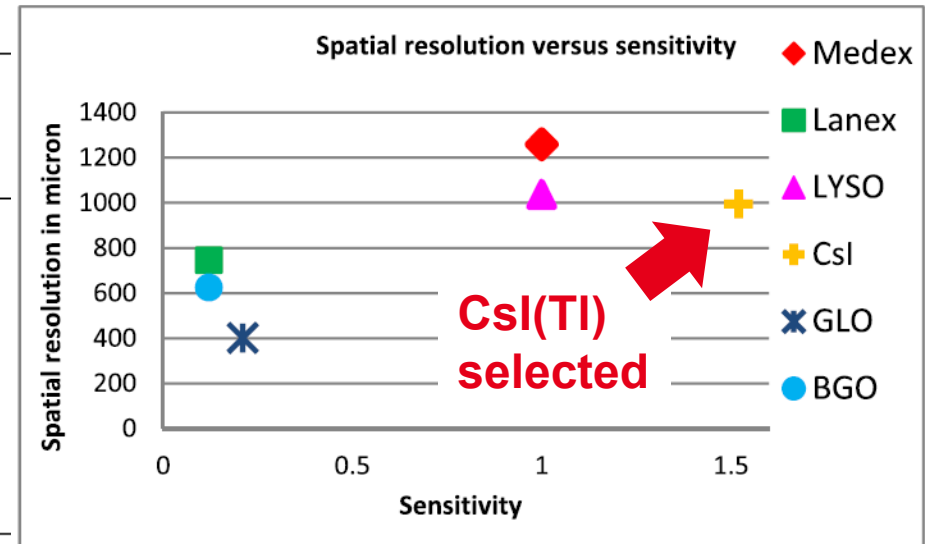
AGENCE NATIONALE DE LA RECHERCHE
ANR

« ICE » Project on Water-Corium Interaction

R&D on high resolution fast scintillation screens

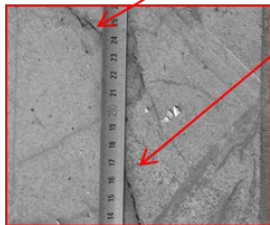
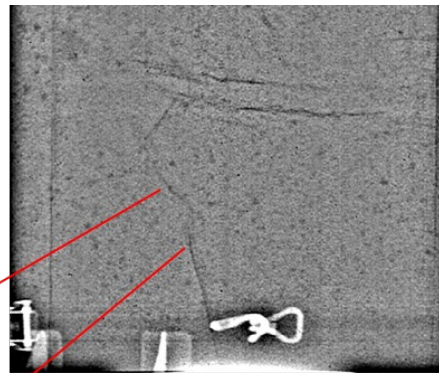
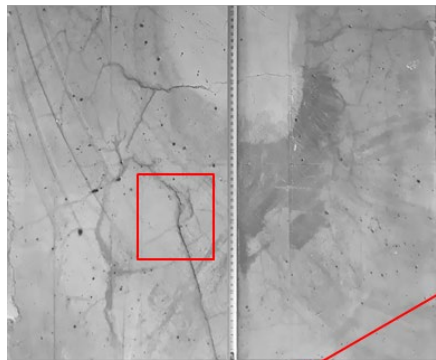
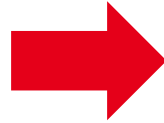


Scintillator	Density (g/cm ³)	Thickness (mm)	Surface density (mg/cm ²)	Average Light yield (photons/MeV)	Wavelength Peak (nm)
Medex (GOS)	4.45	0.780	347	65000	545
Lanex (GOS)	4.62	0.290	134	65000	545
CsI:Tl	4.51	2	902	54000	546
BGO	7.13	3	2139	9000	480
LYSO	7.25	20	14500	30000	420
GLO	9.10	1.58	1438	55000	589



R&D on high resolution linear detectors

Historical system = 25 CdTe collimated detectors



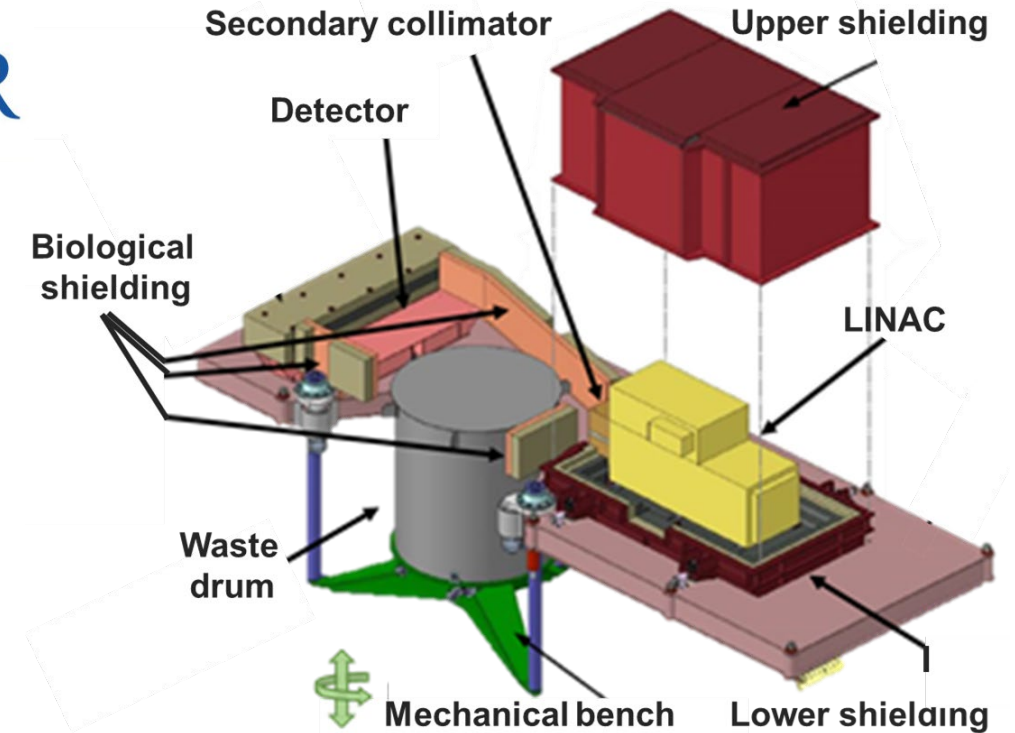
3 mm bubbles, 2 mm cracks

Scintillators typically used in imaging systems

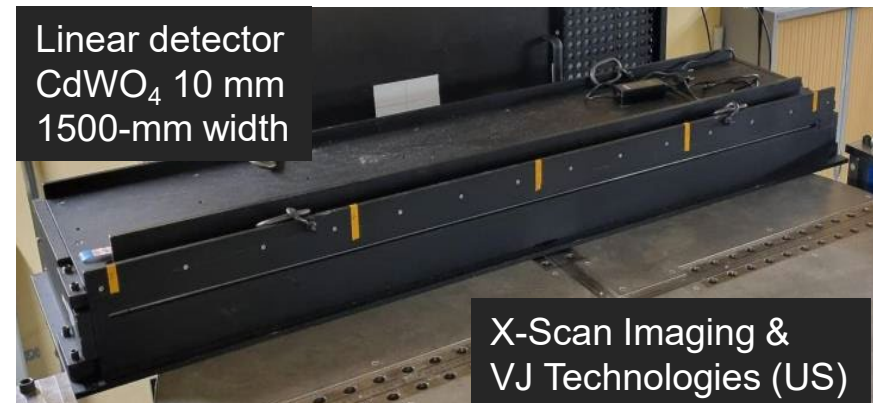
Scintillator	density ρ (g/cm ³)	Photon yield (ph/MeV)
GLO (GD _{0,3} Lu _{1,6} Eu _{0,1} O ₃)	9.1	55 000
CsI(Tl)	4.5	54 000
CdWO ₄	7.9	20 000
Gadox (GOS GD ₂ O ₂ S:Tb)	4.5	55 000

- ❑ Objective: high resolution imaging 200 μm
- ❑ Analysis of small objects and defects (bubbles, cracks...)
- ❑ Comparative study of linear detectors to replace the collimated CdTe detectors \Rightarrow reduced acquisition time
- \Rightarrow **GLO (if available), CdWO₄ or CsI(Tl) scintillators**

TOMIS mobile high-energy tomography



- ❑ 9 MeV LINAC, 30 Gy/min @ 1 m
- ❑ Integration in a 40 feet sea container
- ❑ **High performance CdWO₄ linear detector**
- ❑ 5.5 tons mechanical bench, up to 1.8 m³
- ❑ Self-shielded LINAC ⇒ small footprint (restricted area)
- ⇒ In situ imaging of legacy concrete waste packages



TOMIS in situ implementation



Commissioning and first measurements in 2024



3 ■ **Scintillators for gamma-ray spectroscopy**

But many other gamma detectors are available

Liquid nitrogen cooled HP Ge



Portable HP Ge
(electric cooling)



CdTe, CdZnTe



NaI(Tl)



BGO ($\text{Bi}_4\text{Ge}_3\text{O}_{12}$)



CeBr₃



LaBr₃(Ce)



Scintillators

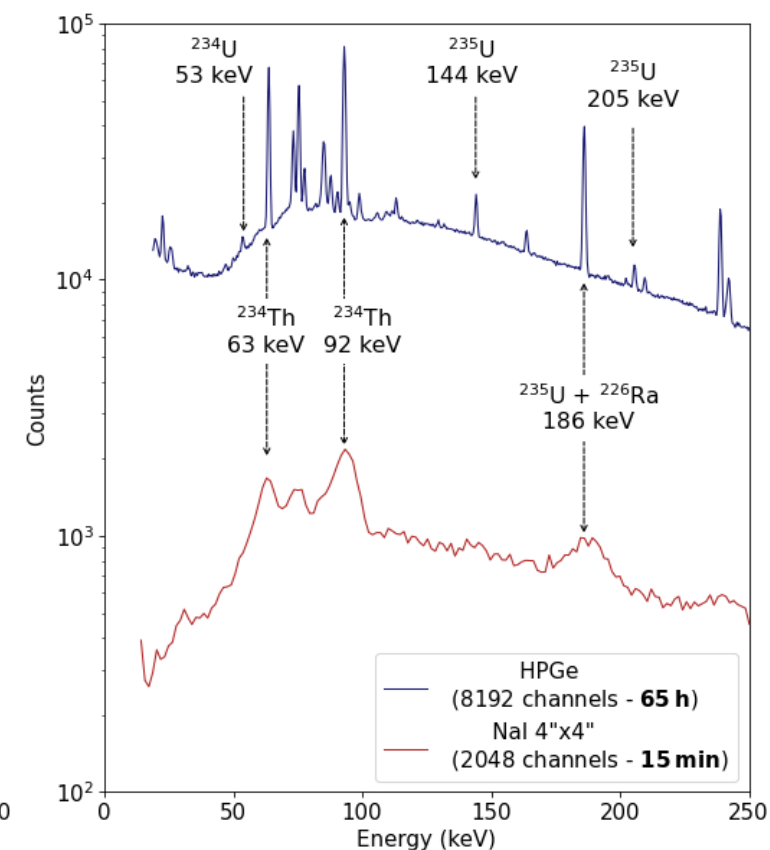
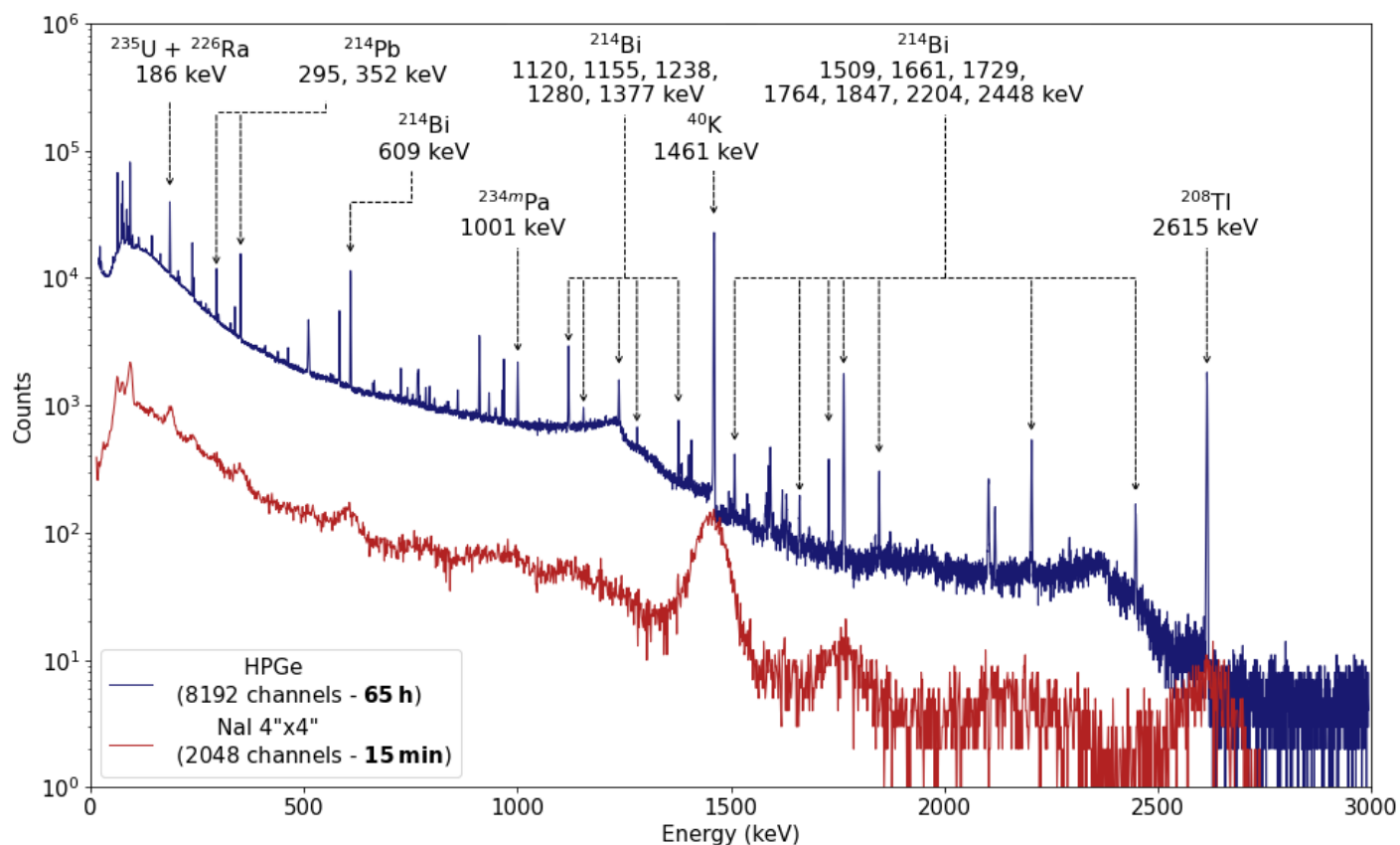
Energy resolution vs. efficiency

□ **HPGe is the standard of gamma spectroscopy**

⇒ Pu isotopic composition, spent fuel, nuclear accidents, etc. (many peaks)

□ **But scintillators are preferred in many instances**

for their robustness, efficiency/cost ratio, fast timing...
⇒ radioactive waste (few peaks), cleaning and dismantling, uranium mining, online analysis, security...

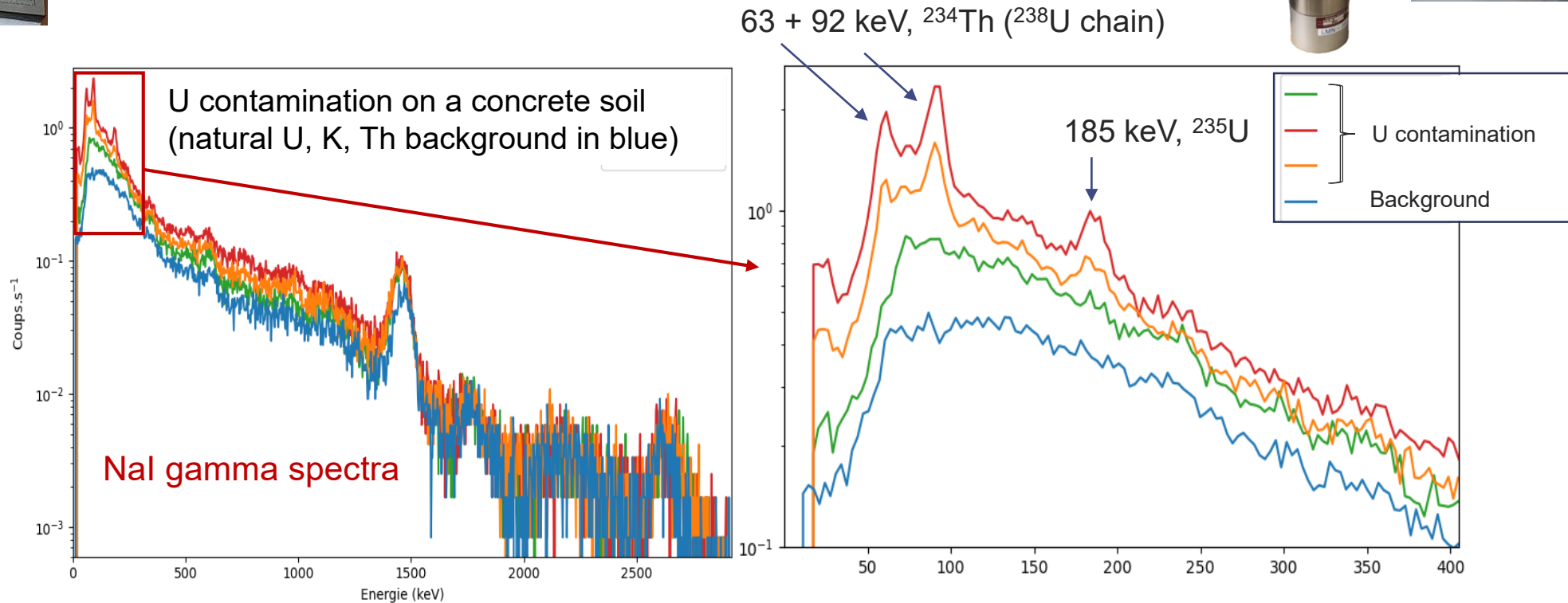


Alpha-beta-gamma scintillators for D&D

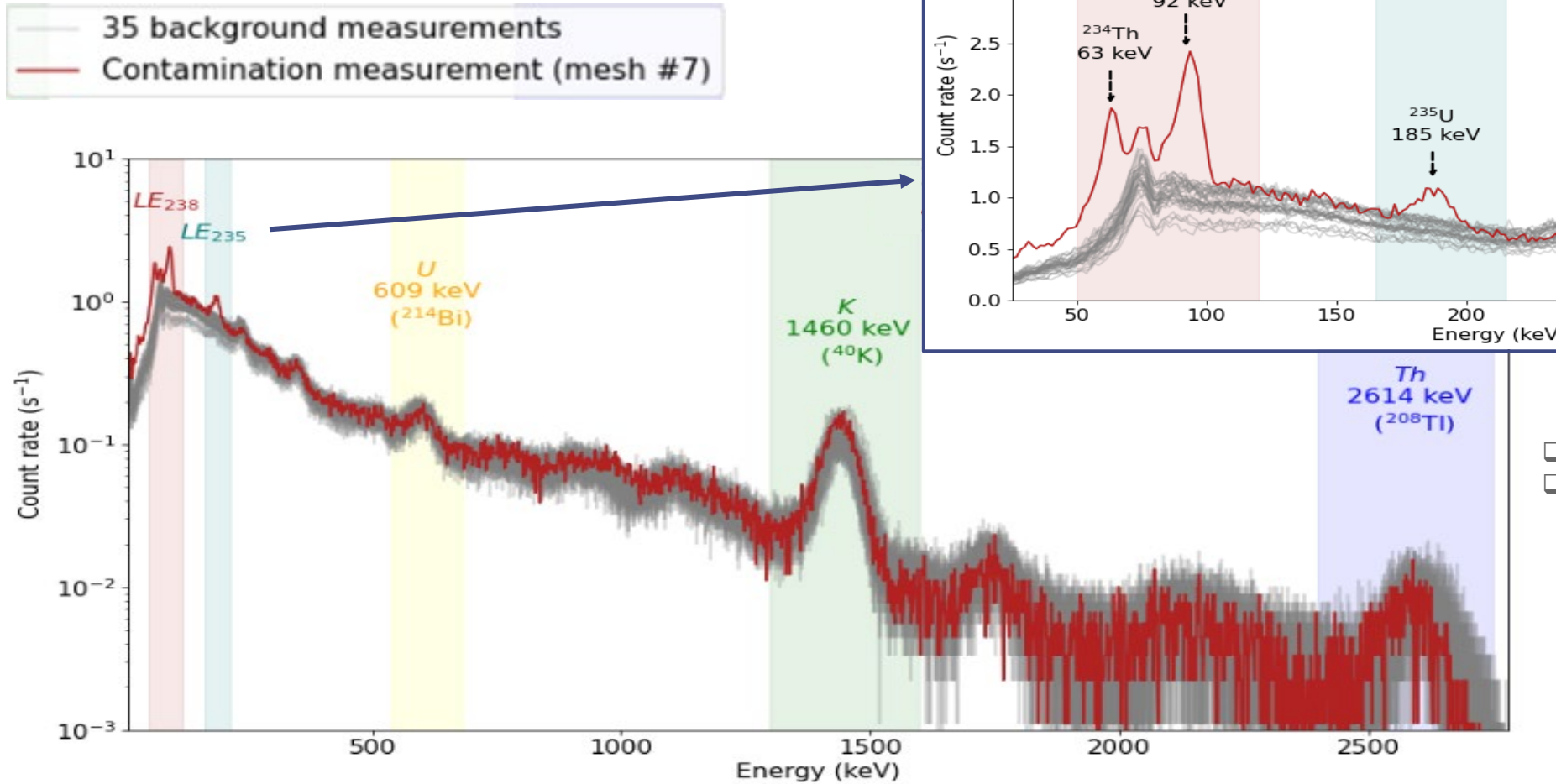
ZnS(Ag)
scintillator



- ❑ Fast alpha and beta detection (a few min) of uranium contamination deposited on concrete soil with **ZnS(Ag) and plastic PVT scintillators**
- ❑ Shielded **Nal(Tl) scintillator** close to the ground to confirm the activity within about 15 min



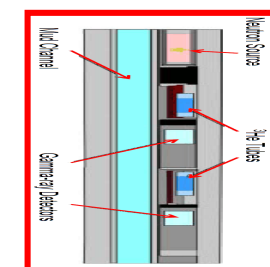
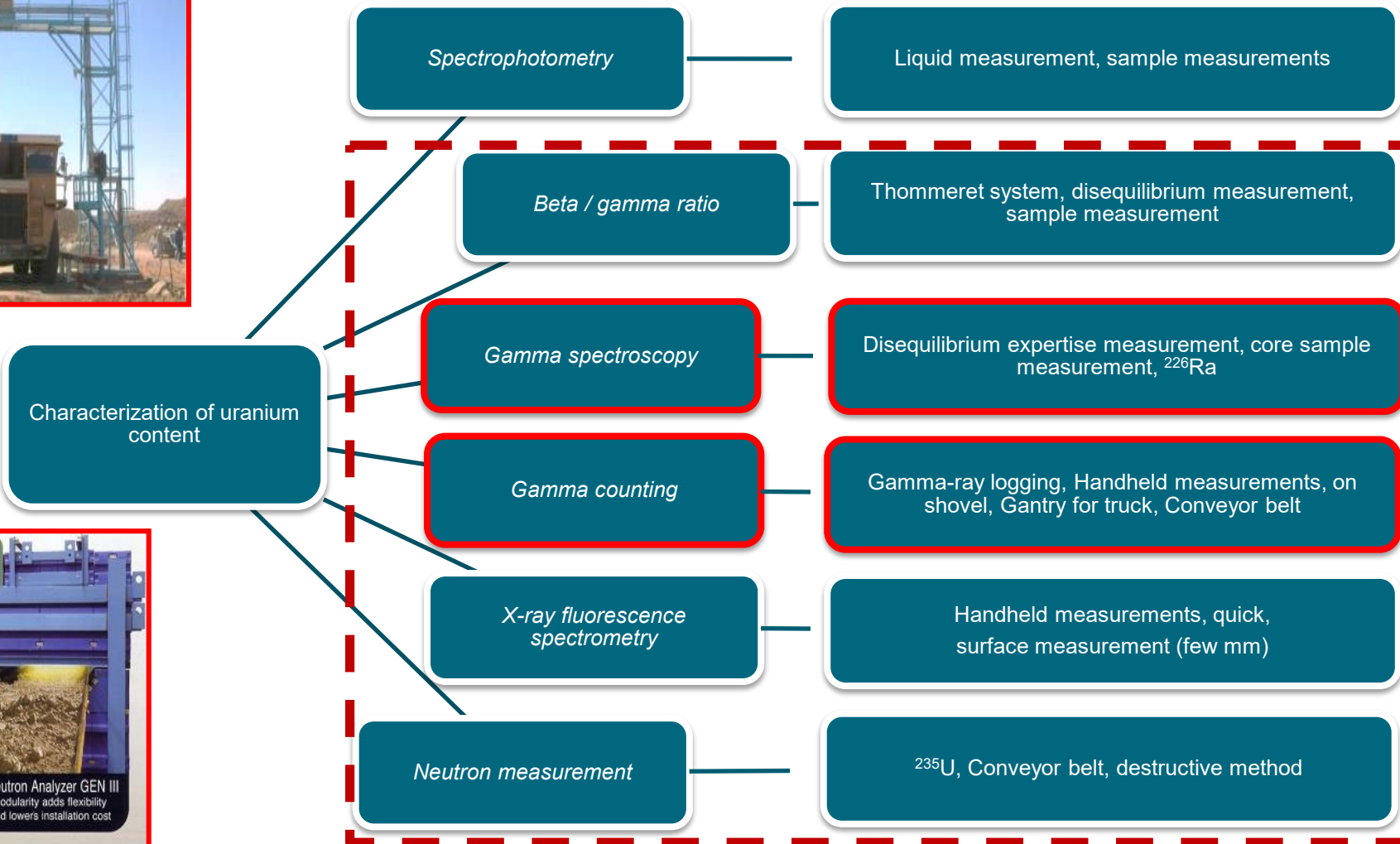
Nal(Tl) spectrum analysis with energy bands



□ Patent application FR2210553
 □ Salvador et al., Low and high resolution gamma-ray spectroscopy for the characterization of uranium contamination, submitted to NIM A, NIMA-D-24-00254

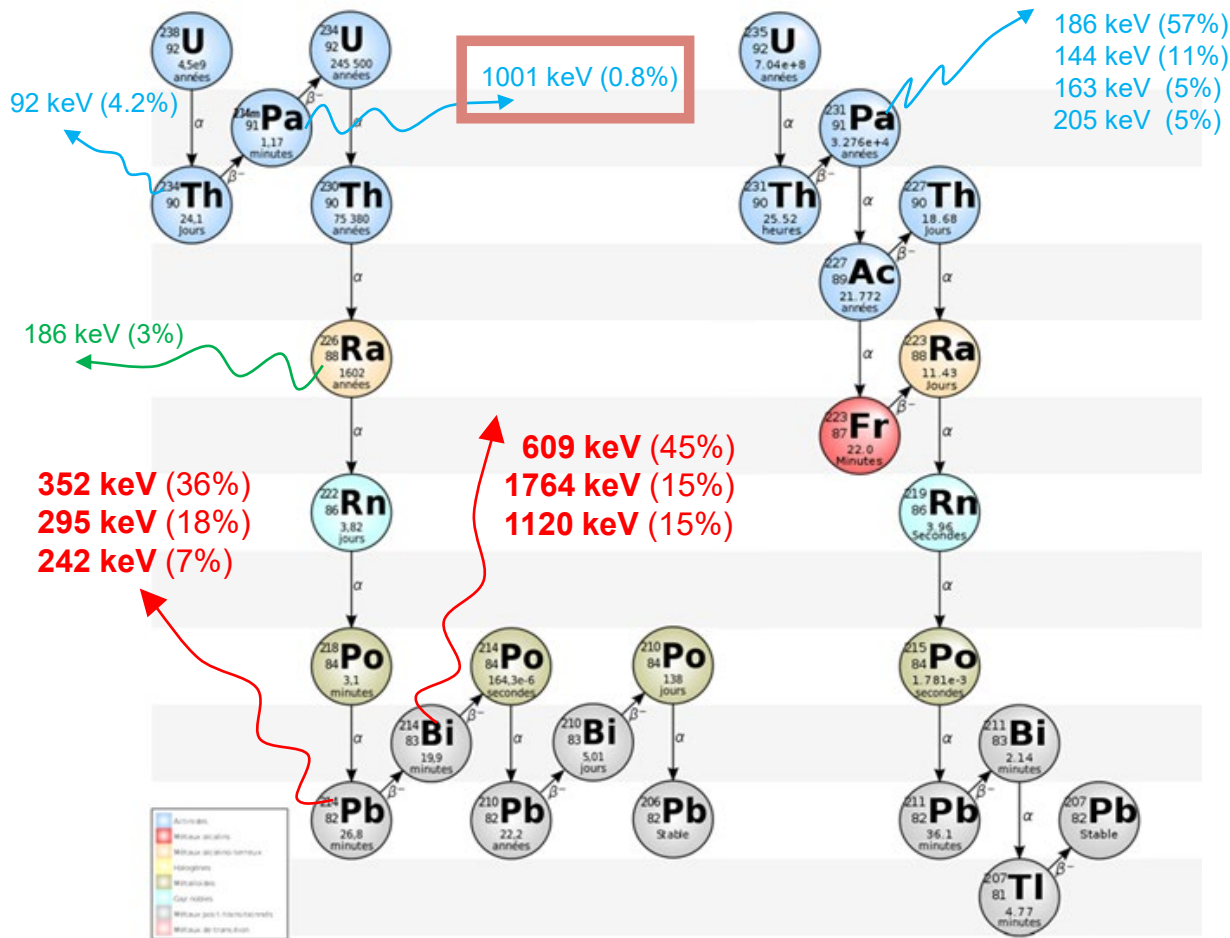
	HPGe (reference)	Nal(Tl)
Acquisition time	64 h	15 min
U surface activity	$(7.47 \pm 0.75) \text{ Bq.cm}^{-2}$	$(9.8 \pm 1.4) \text{ Bq.cm}^{-2}$
U enrichment	$(0.83 \pm 0.07) \%$	$(0.73 \pm 0.15) \%$

Uranium mining ⇒ gamma scintillators for samples, logging probes, truck portal monitors, conveyor belts

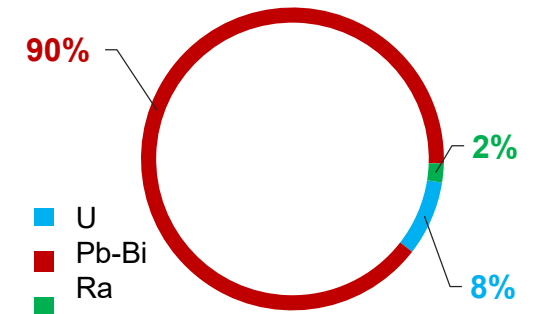


Non-destructive Measurements

Uranium chain gamma emissions



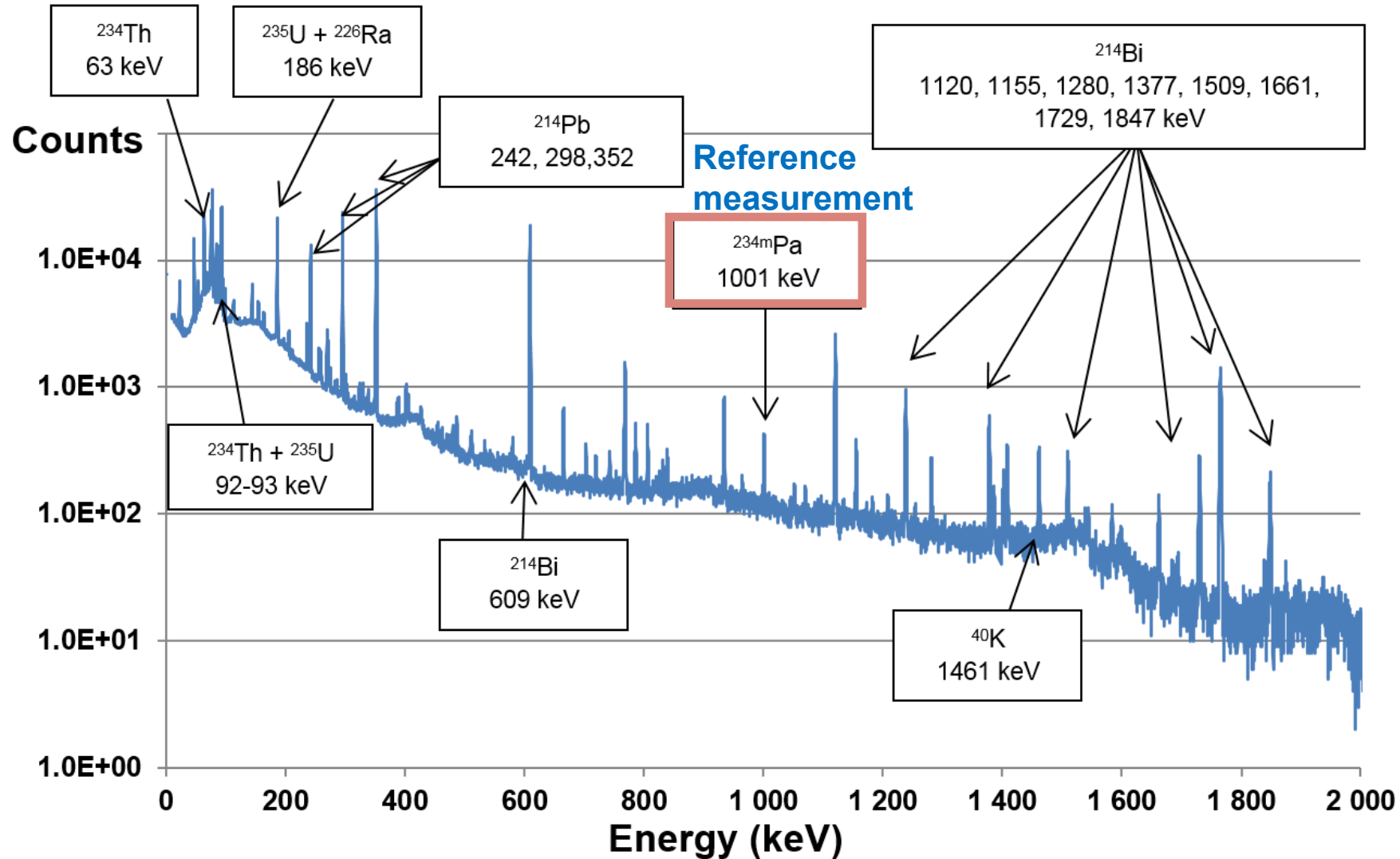
Origin of gamma rays



Disequilibria in U chain (U vs. Ra differential leaching in roll fronts)

⇒ **U grade over/under estimated by total gamma counting**

Disequilibrium \Rightarrow HPGe spectroscopy in the lab

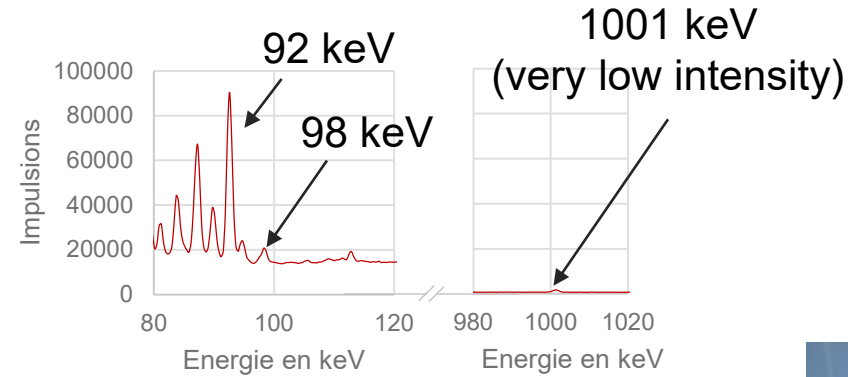


Low intensity of the 1001 keV ray \Rightarrow long measurement (hours)
Low energy of ^{234}Th gamma rays \Rightarrow large attenuation effects

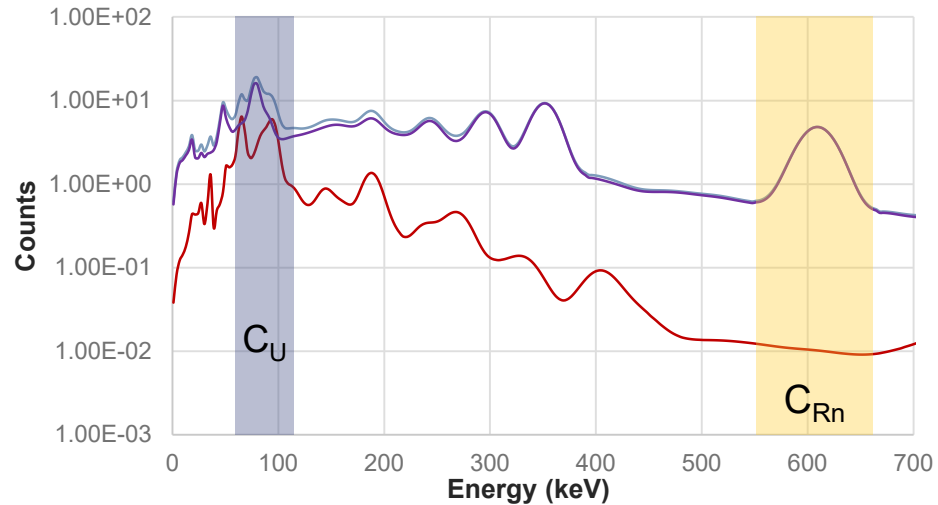
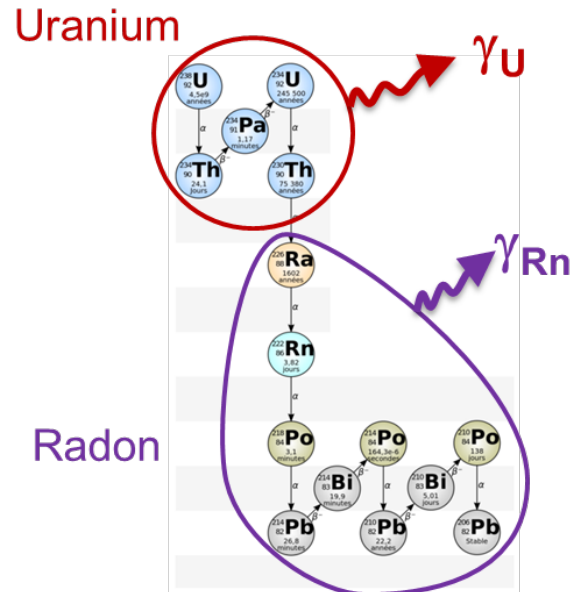
Gamma spectroscopy on samples: from Ge to NaI

New U-grade measurements (faster than with 1001 keV line and HPGe)

1. HPGe high-resolution spectroscopy
 - 92 keV γ -ray of ^{234}Th
 - 98 keV self-induced fluorescence X-ray



2. NaI(Tl) low-resolution spectroscopy : count rates in energy bands (C_U and C_{Rn})



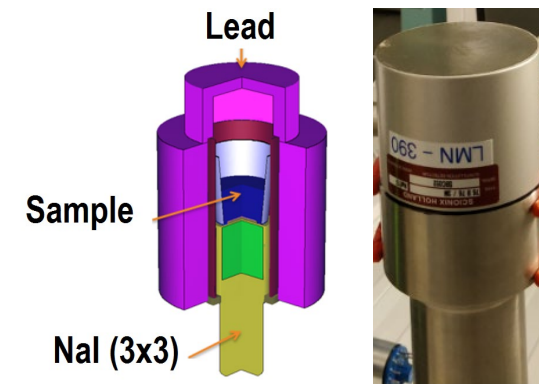
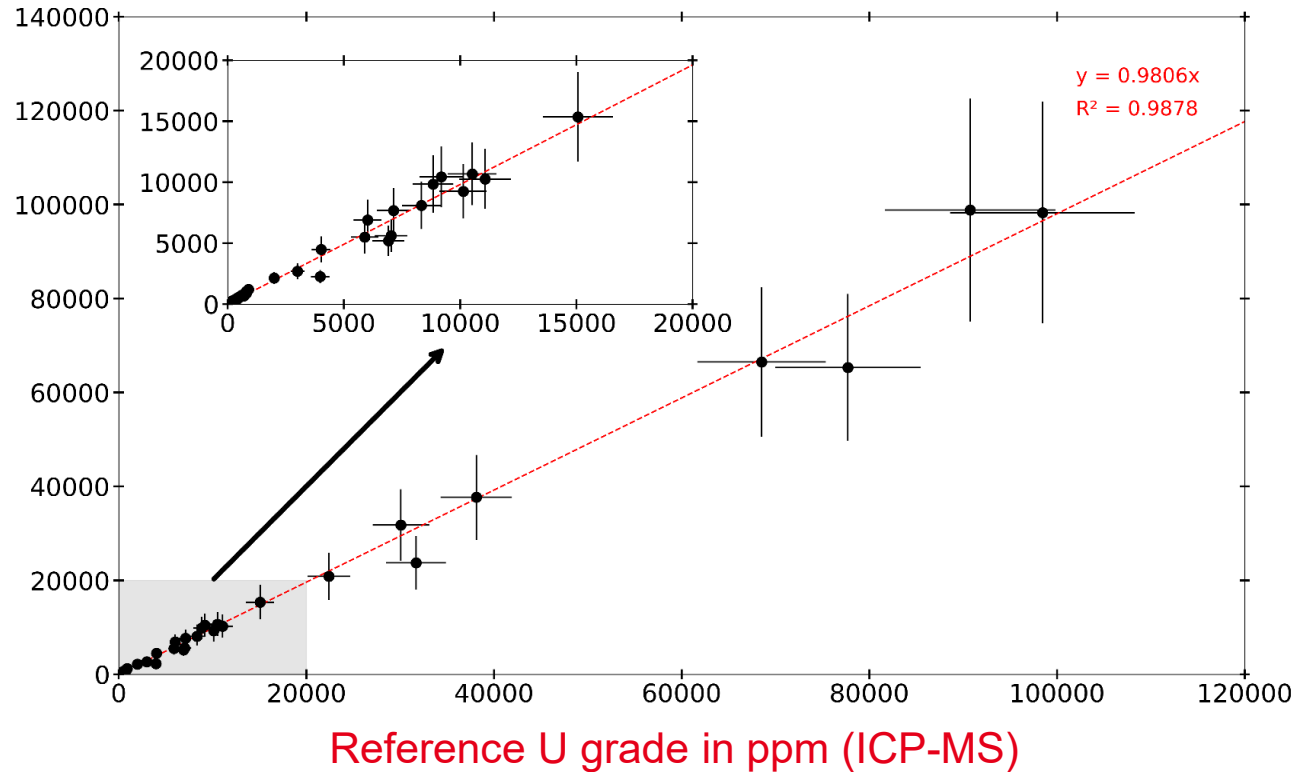
Energy-band approach with NaI(Tl)

$$\frac{C_U}{C_{Rn}} = \alpha \frac{A(U)}{A(Rn)} + \beta$$

PATENT
FR3088445

- ✓ Method validated with 38 samples
- ✓ Measurement time < 3 min

U grade
in ppm
with
NaI(Tl)



Logging probes with gamma scintillators



GEOVISTA NGRS Probe
Total counting

KOBRA KSP Probe
Total counting

GEOVISTA LaBr₃(Ce) Probe
Spectrometric probe

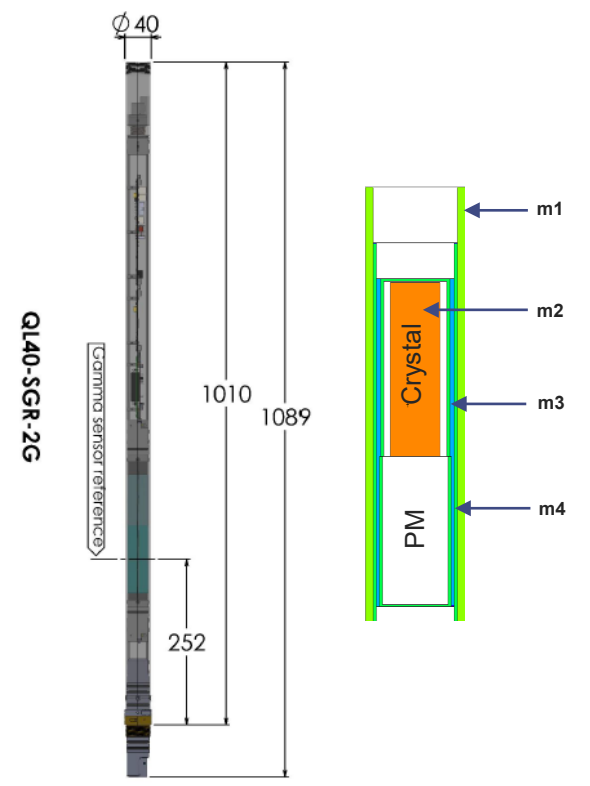
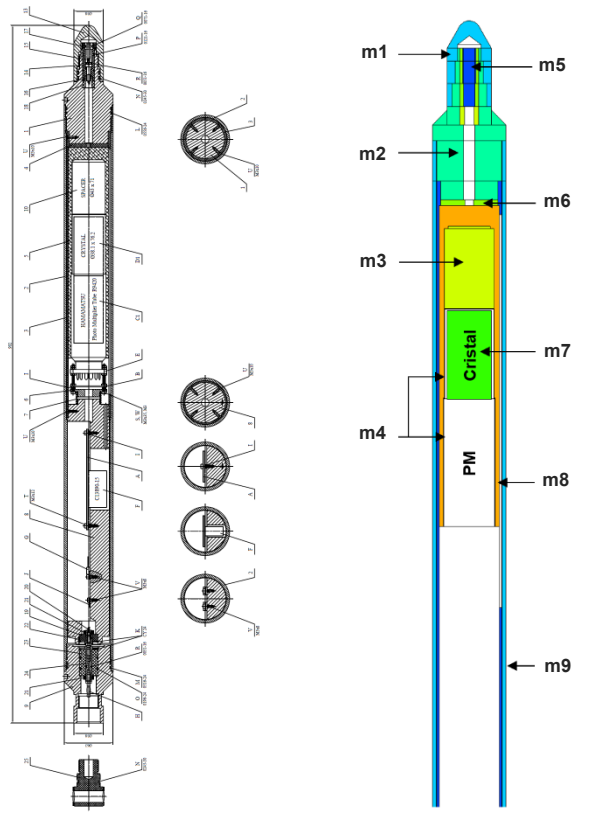
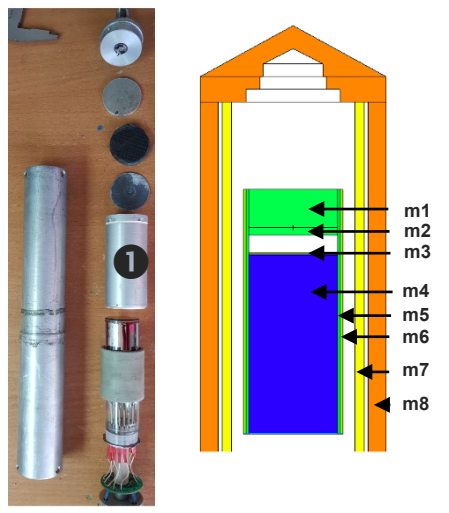
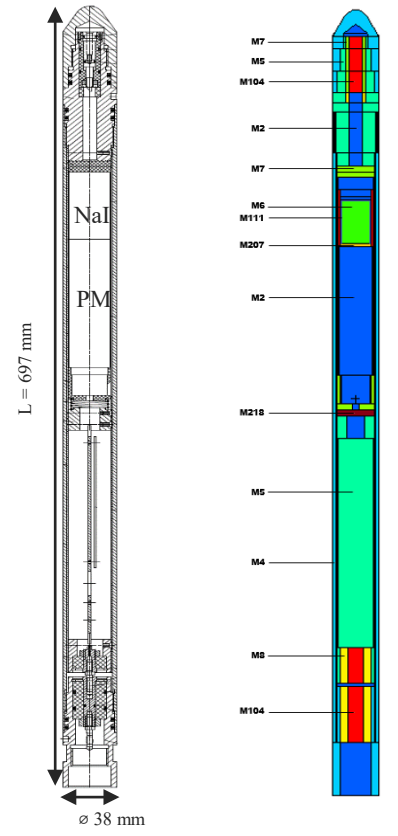
ALT CeBr₃ Probe
Spectrometric probe
Advanced Logic Technology

Nal(Tl) 2.4 cm x 3.9 cm

Nal(Tl) 3.0 cm x 7.0 cm

LaBr₃(Ce) 3.81 cm x 7.62 cm

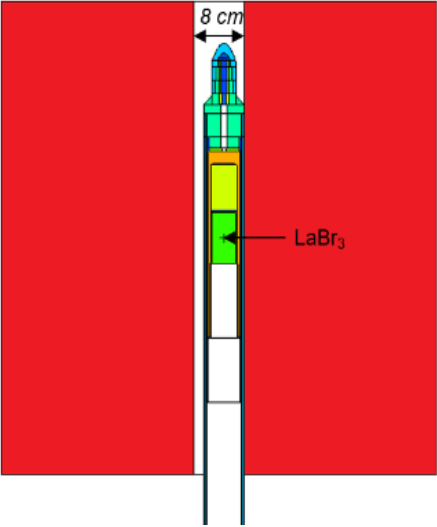
CeBr₃ 2.0 cm x 9.6 cm



Development of new spectroscopic methods

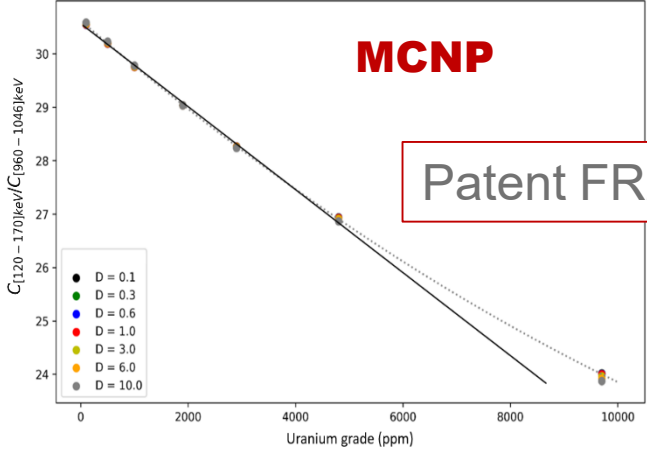
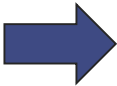
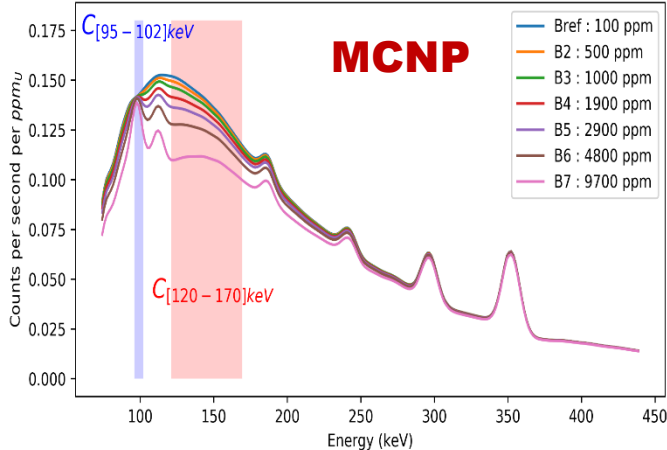
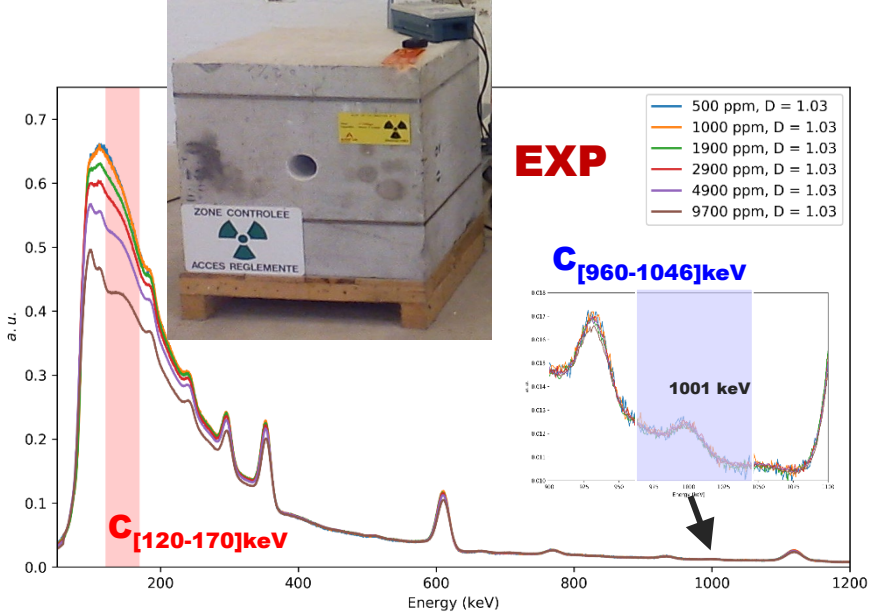


GEOVISTA

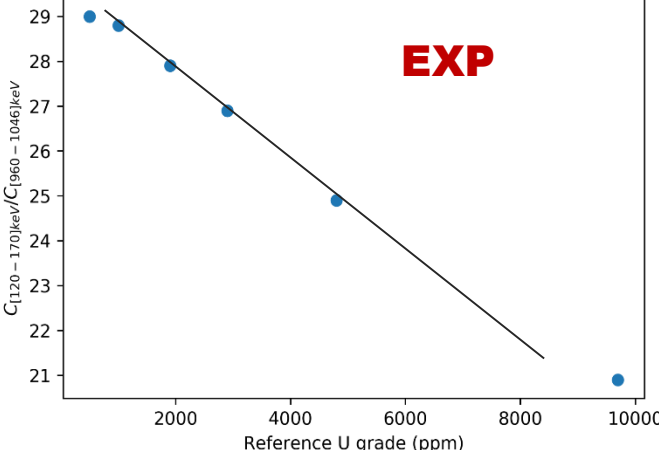


- ❑ **MCNP simulations**
 - ✓ Compton counts vs. uranium grade (calibration)
 - ✓ Uncertainties (disequilibrium, density, mineralogy...)
- ❑ **EXP: ORANO CIME calibration station**
 - ✓ Concrete blocks with U grades up to 10 000 ppm

$$U_{ppm} = \frac{\beta_{Compton} \cdot \frac{C_{[120-170]keV}}{C_{[960-1046]keV}}}{\alpha_{Compton}}$$



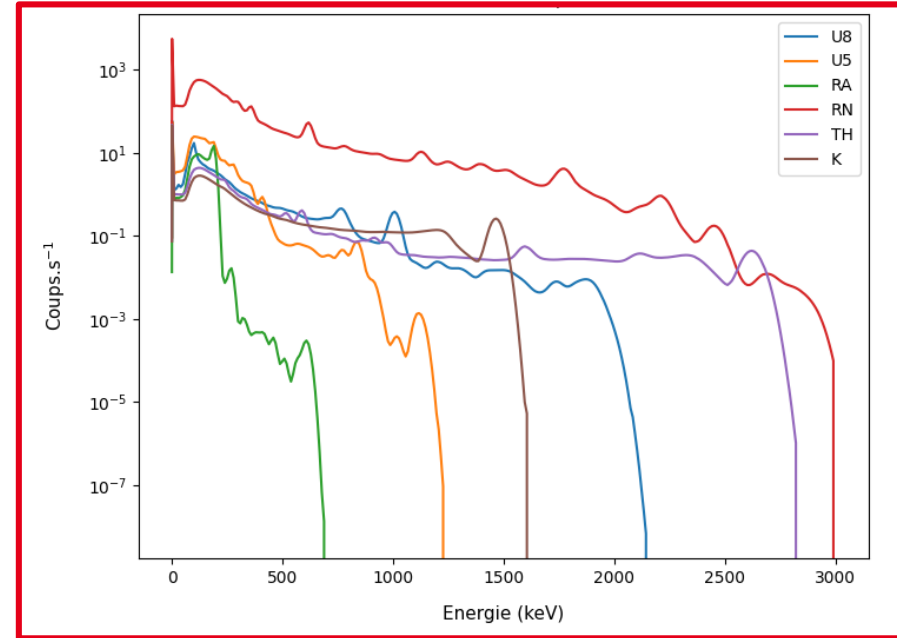
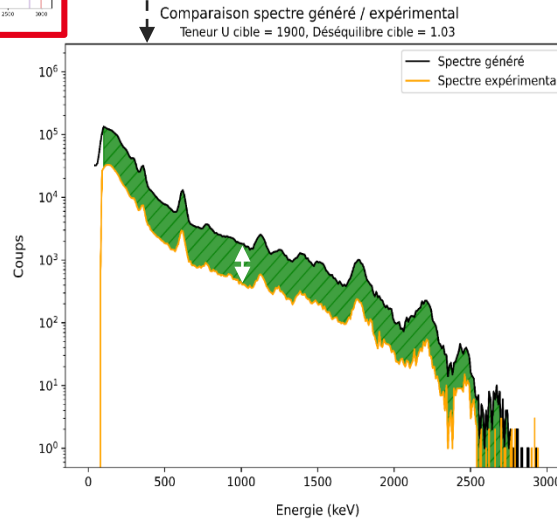
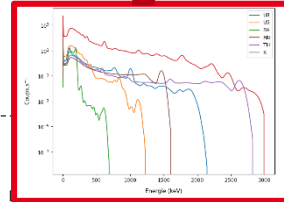
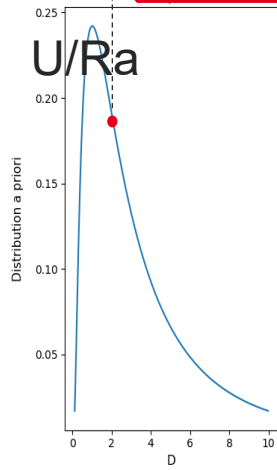
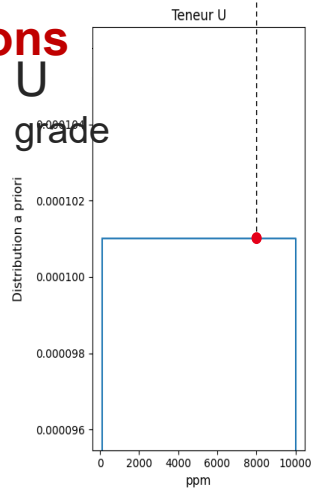
Patent FR2109325



Approaches using the entire spectrum

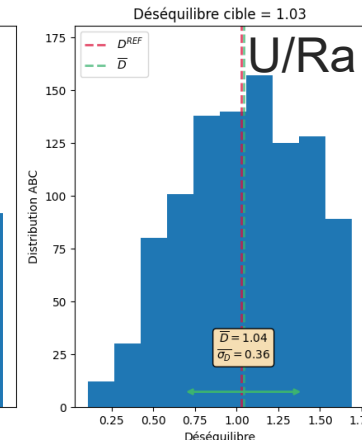
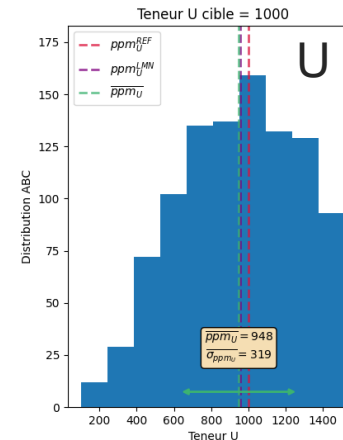
- Unfolding (see further in slide 42)
- Machine Learning, neural networks
- Bayesian inference (here)

Prior distributions



$$distance = \sum_i (canal_i^{EXP} - canal_i^{GEN})^2$$

if $distance < \epsilon$



Posterior distributions



4. Scintillators for passive neutron coincidence counting

Main neutron detectors



³He proportional counter is the gold standard in radioactive waste and nuclear material characterization, nuclear process monitoring, homeland security, etc.

Detector Type	Size	Neutron Active Material	Incident Neutron Energy	Neutron Detection Efficiency ^a (%)	Gamma-Ray Sensitivity (R/h) ^b
Plastic scintillator	5 cm thick	¹ H	1 MeV	78	0.01
Liquid scintillator	5 cm thick	¹ H	1 MeV	78	0.1
Loaded scintillator	1 mm thick	⁶ Li	thermal	50	1
Hornyak button	1 mm thick	¹ H	1 MeV	1	1
Methane (7 atm)	5 cm diam	¹ H	1 MeV	1	1
⁴ He (18 atm)	5 cm diam	⁴ He	1 MeV	1	1
³ He (4 atm), Ar (2 atm)	2.5 cm diam	³ He	thermal	77	1
³ He (4 atm), CO ₂ (5%)	2.5 cm diam	³ He	thermal	77	10
BF ₃ (0.66 atm)	5 cm diam	¹⁰ B	thermal	29	10
BF ₃ (1.18 atm)	5 cm diam	¹⁰ B	thermal	46	10
¹⁰ B-lined chamber	0.2 mg/cm ²	¹⁰ B	thermal	10	10 ³
Fission chamber	2.0 mg/cm ²	²³⁵ U	thermal	0.5	10 ⁶ – 10 ⁷

Passive Non Destructive Assay of Nuclear Materials, Los Alamos National Laboratory, NUREG/CR-5550, 1991

Passive neutron measurements with ^3He counters

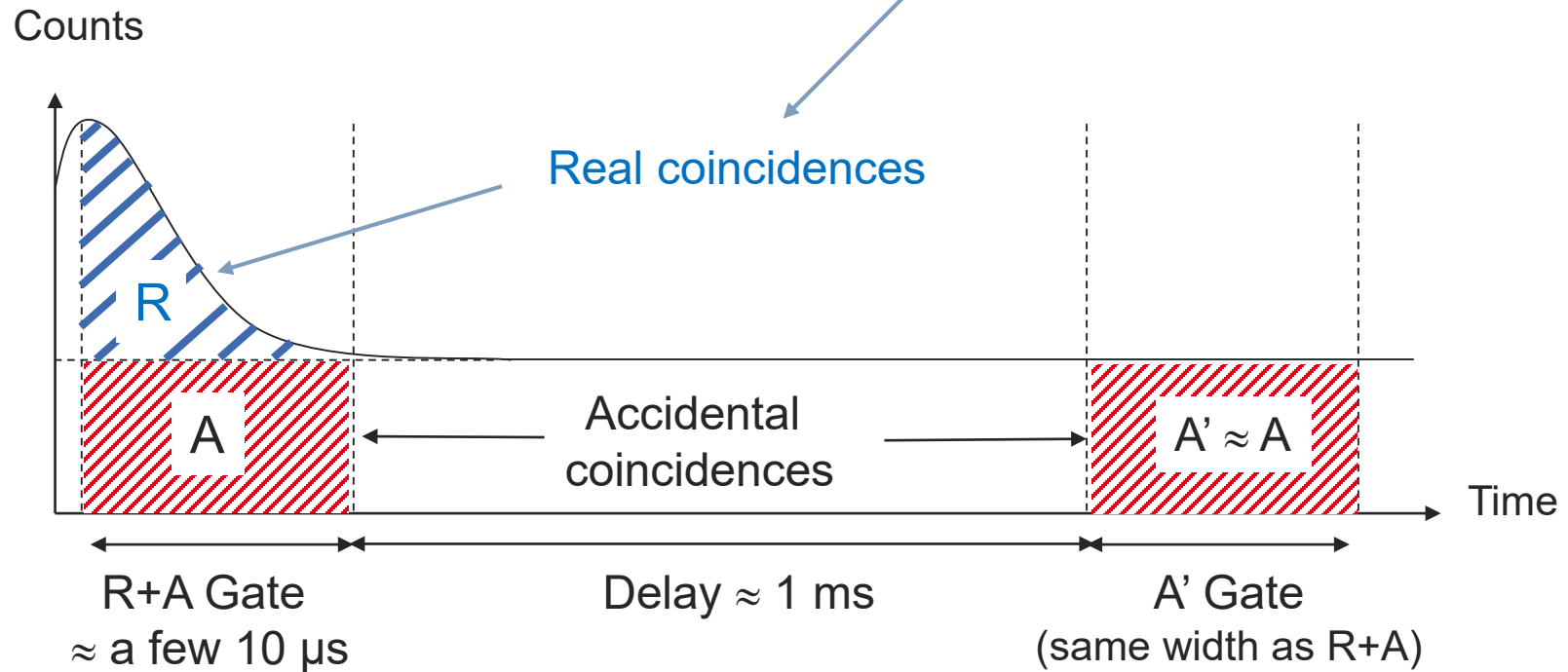
❑ Total counting

Spontaneous fission + (α, n) neutrons \Rightarrow ^{242}Cm and ^{244}Cm , ^{238}Pu , ^{240}Pu and ^{242}Pu , ^{241}Am
(+ ^{238}U if mass > kg)







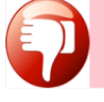



❑ Coincidences counting

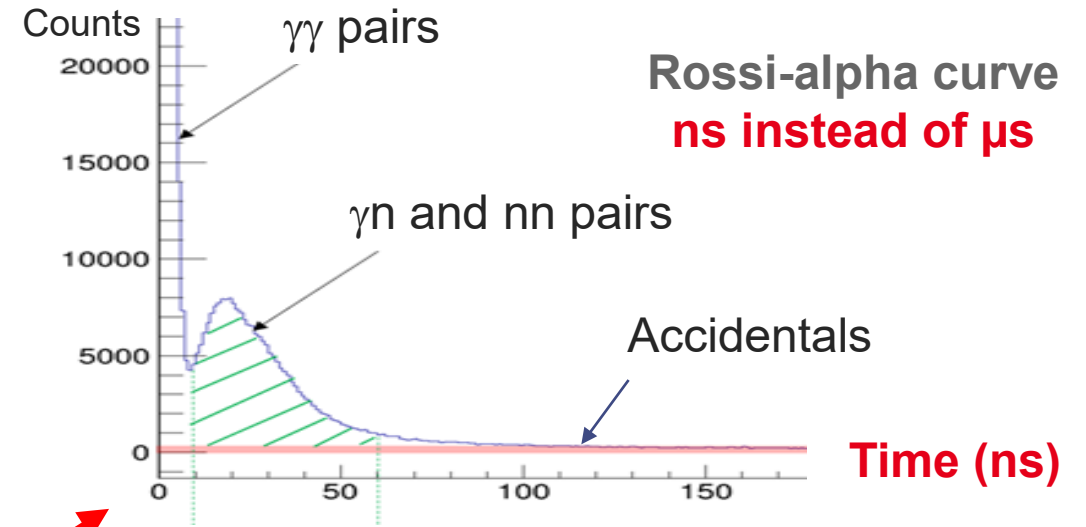
Spontaneous fission neutrons only \Rightarrow ^{242}Cm and ^{244}Cm , ^{238}Pu , ^{240}Pu and ^{242}Pu (+ ^{238}U)

“Rossi-alpha”
curve



Alternatives to ^3He counters \Rightarrow PVT plastic scintillators

^3He proportional counters	Plastic Scintillators
	
 GOOD EFFICIENCY	 GOOD EFFICIENCY
 EXPENSIVE (since 09/11/2001)	 CHEAP (~ factor 5 wrt. ^3He)
 SLOW RESPONSE (~ μs) (thermalization needed)	 FAST RESPONSE (~ ns) (recoil proton)
 UNSENSITIVE TO γ RAYS AND CROSS TALK	 VERY SENSITIVE TO γ RAYS AND CROSS-TALK



- 118 L drums with ~ 0.5 g $^{240}\text{Pu}_{\text{eq}}$
- PVT scintillators + 5 cm Pb shield
- 1500 s acquisition time

MCNPX PoliMi
Feasibility study



0.5 g/cm³ metallic matrix

Multiplicity	Pu only	Am only	Mix (Am, Pu)
0	232,383 ± 482	450,562 ± 671	684,157 ± 827
1	32,356 ± 180	3,675 ± 61	36,287 ± 190
2	2,400 ± 49	9 ± 3	2,365 ± 49
3	66 ± 8	0	66 ± 8

0.2 g/cm³ organic matrix

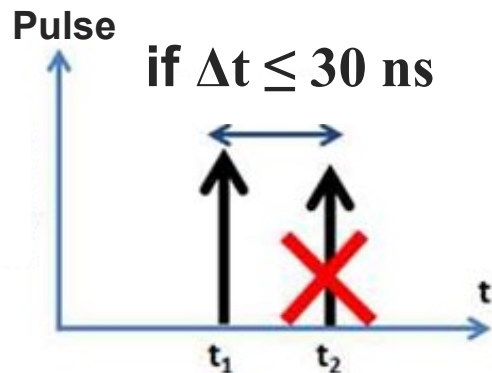
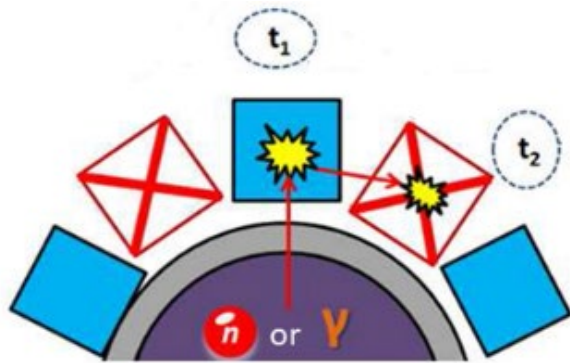
Multiplicity	Pu only	Am only	Mix (Am, Pu)
0	178,516 ± 423	299,817 ± 548	478,372 ± 692
1	11,744 ± 108	2,795 ± 53	14,777 ± 122
2	372 ± 19	12 ± 3	365 ± 19

At least 3 coincident pulses within 100 ns

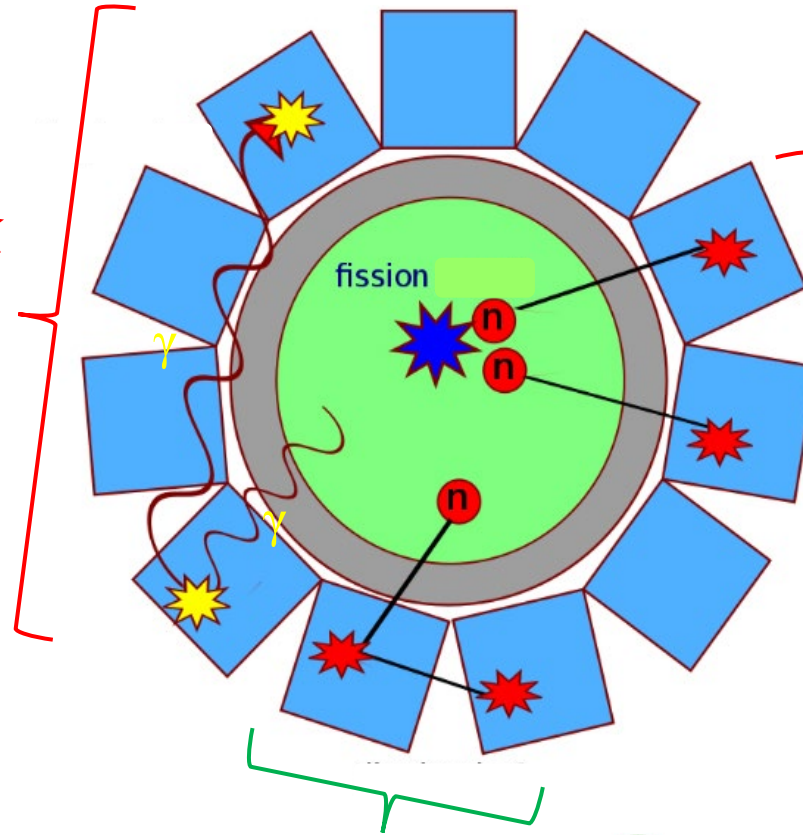
Scattering cross talk mitigation

Principle

suppression of the 2nd pulse
in double detections
in adjacent detectors within Δt



Cross talk
not
rejected



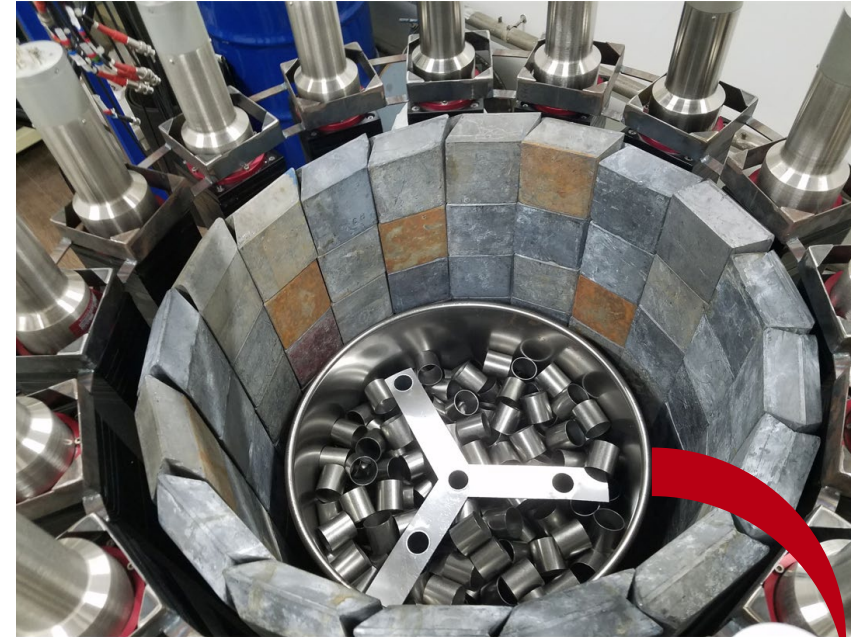
Useful signal
(fission
coincidence)
rejected



Cross talk
Rejected



Validation in the lab with 118 L mock-up drums



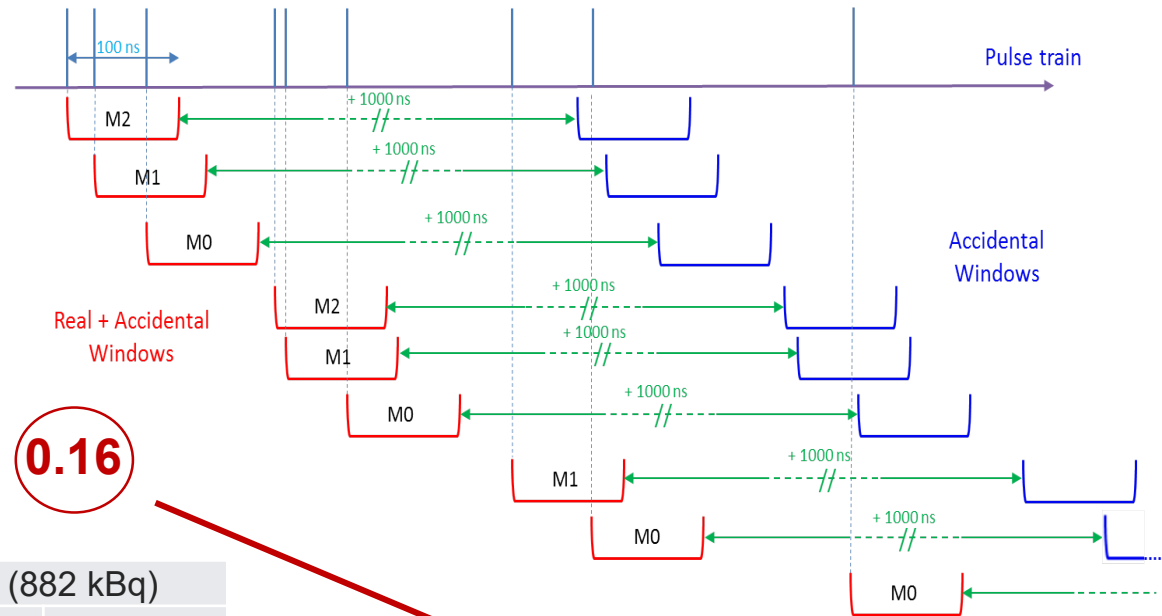
- 16 PVT scintillators 10 cm × 10 cm × 100 cm
- ^{137}Cs , ^{60}Co , ^{252}Cf , AmBe sources, Pu samples
- 5 cm lead shield
- FASTER electronics (CNRS)



Mock-up drums
filled with iron or
wood matrixes

Coincidence Multiplicity analysis

- ❑ **Shift register method** to unfold real from accidental coincidences
- ❑ **At least 3 pulses in coincidence** to separate spontaneous fissions (Pu or ^{252}Cf) from parasitic coincidences of (α, n) reactions, gamma cascades...



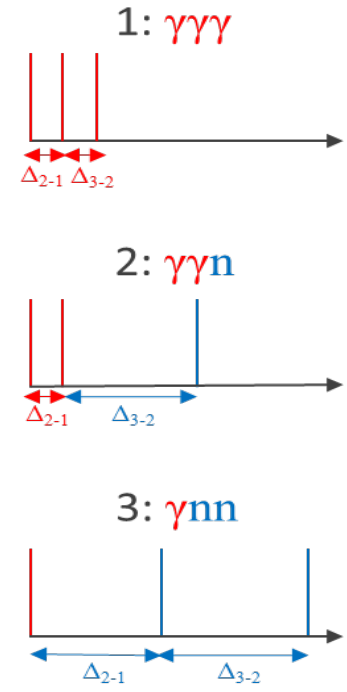
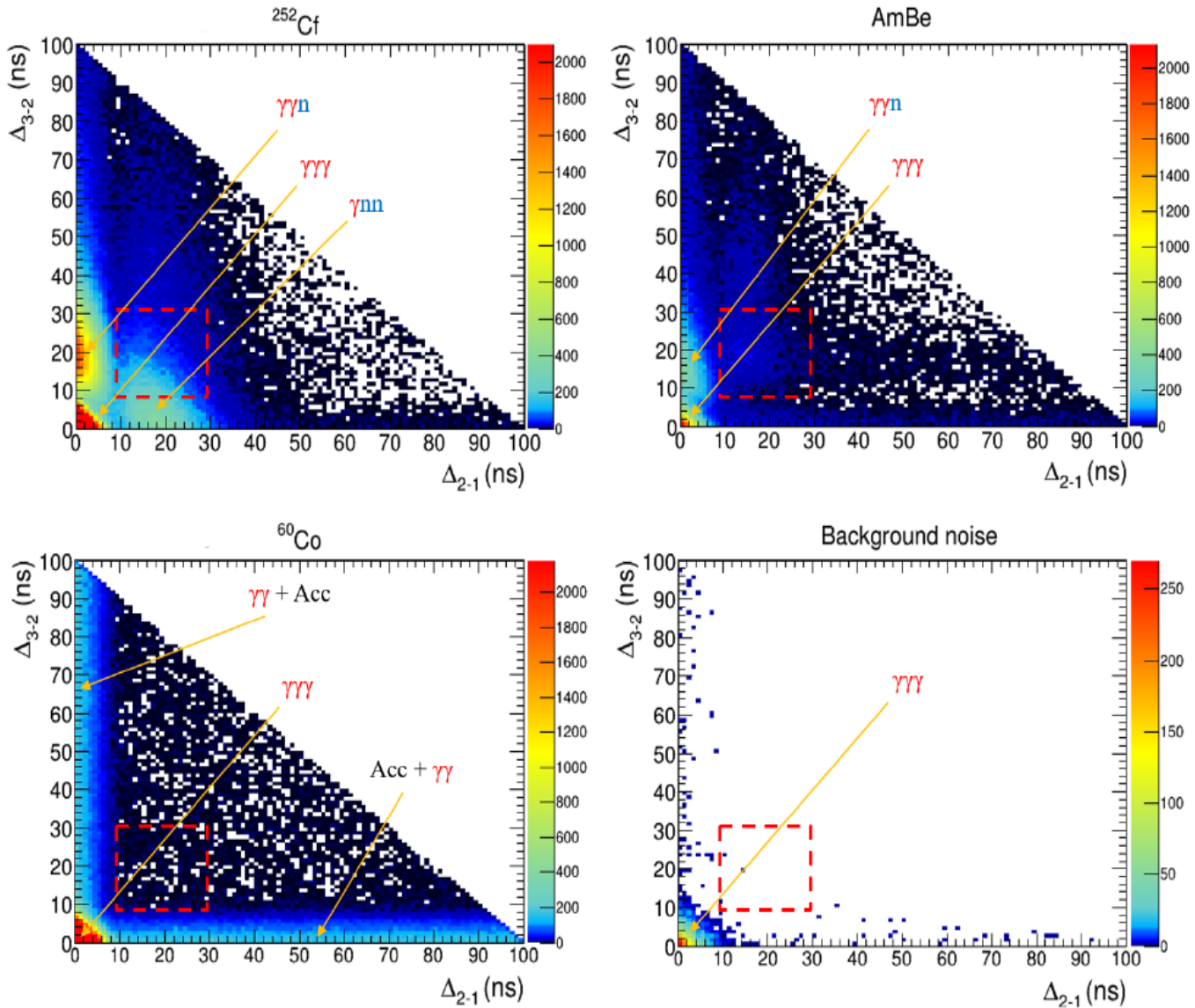
Spontaneous fission neutrons of ^{252}Cf
(α, n) neutrons of AmBe ≈ 0.16

	^{252}Cf (3.6×10^4 n/s)		AmBe (2.3×10^5 n/s)		^{60}Co (882 kBq)	
	R+A	A	R+A	A	R+A	A
M0	3379137	4479459	15577017	16671802	53638186	57771864
M1	857974	8310	1141198	102194	4947762	989025
M2	209237	1296	59474	5790	251249	81991
M3	38098	257	2312	253	9079	3516
M4	4806	42	67	14	239	97
M5	401	5	-9	0	-11	2
M6	18	1	-3	0	-7	0
M7	-1	0	-2	1	-2	0
S	14 965.6 s $^{-1}$		55 933.5 s $^{-1}$		196 155.0 s $^{-1}$	
D	4 667.6 s $^{-1}$		3 842.5 s $^{-1}$		14 381.3 s $^{-1}$	
T	1 174.8 s $^{-1}$		174.9 s $^{-1}$		342.2 s $^{-1}$	

Triples $\frac{T(^{252}\text{Cf})}{T(\text{AmBe})} \approx 6.7$

S, D, T = singles, doubles, triples after real vs. accidental coincidence unfolding

SNR improvement using 2D TOF maps (patented)



(γnn)
region of
interest

Radioactive source	Counts in ROI
^{252}Cf ($3.6 \times 10^4 \text{ n.s}^{-1}$)	10894
AmBe ($2.3 \times 10^5 \text{ n.s}^{-1}$)	761
^{60}Co (882 kBq)	522
SNR ($^{252}\text{Cf}/\text{AmBe}$)	14.3*

* After neutron and gamma scattering cross talk rejection

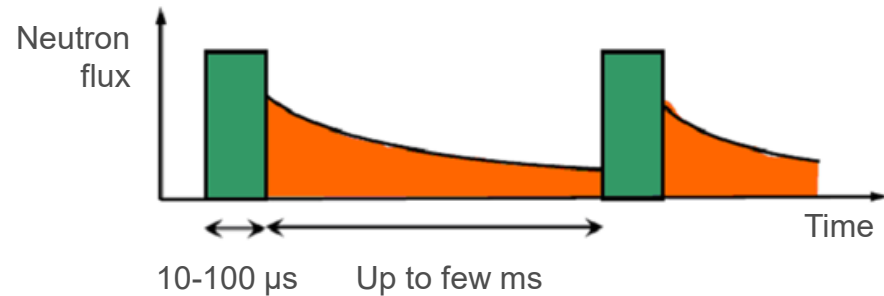




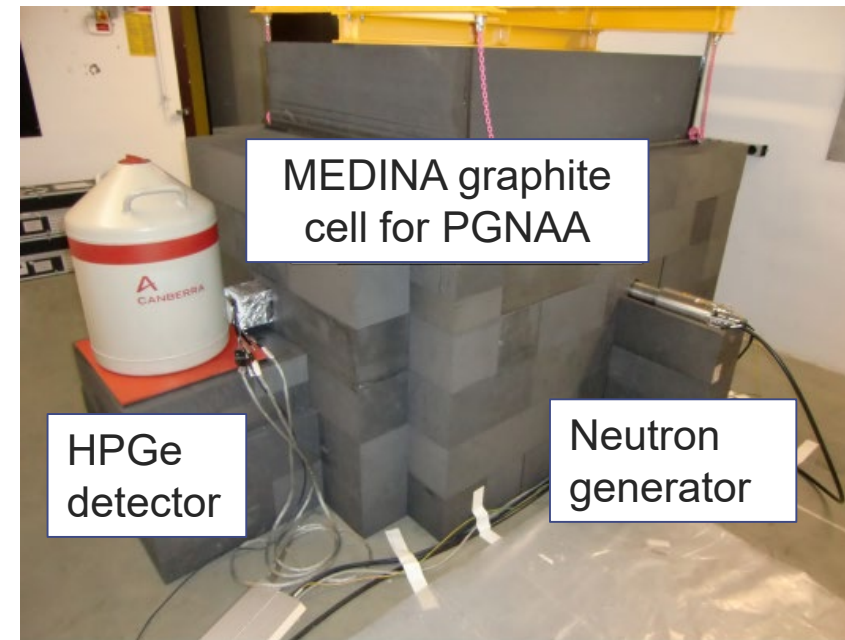
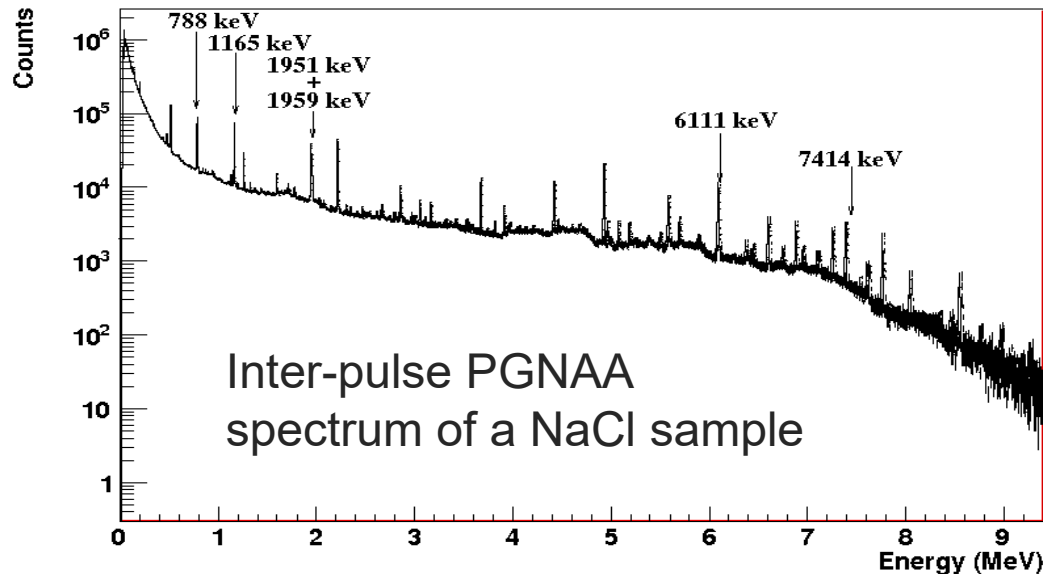
5 ■ **Scintillators for neutron & photon activation analysis**

Traditional HPGe neutron activation analysis)

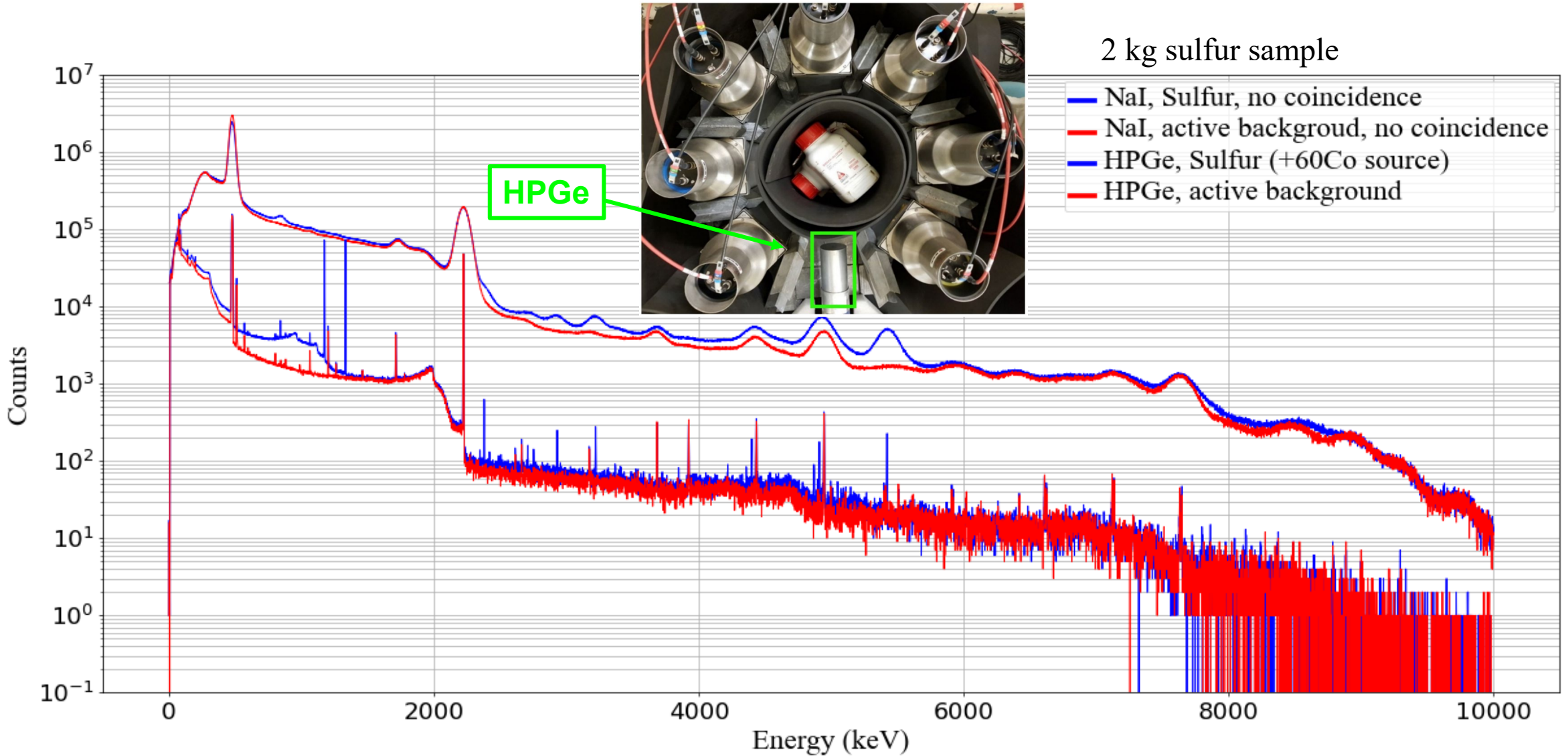
- ❑ Pulsed neutron generator + HPGe \Rightarrow Prompt Gamma Neutron Activation Analysis (PGNAA)
- ❑ Toxic chemicals (water, soils, rad-waste...), recycling (batteries, permanent coils, e-waste...)



- Fast neutrons during the pulses $\Rightarrow (n, n'\gamma)$
 - Thermal neutrons between pulses $\Rightarrow (n, \gamma)$
- (+ delayed gamma rays of activation or fission products after irradiation)

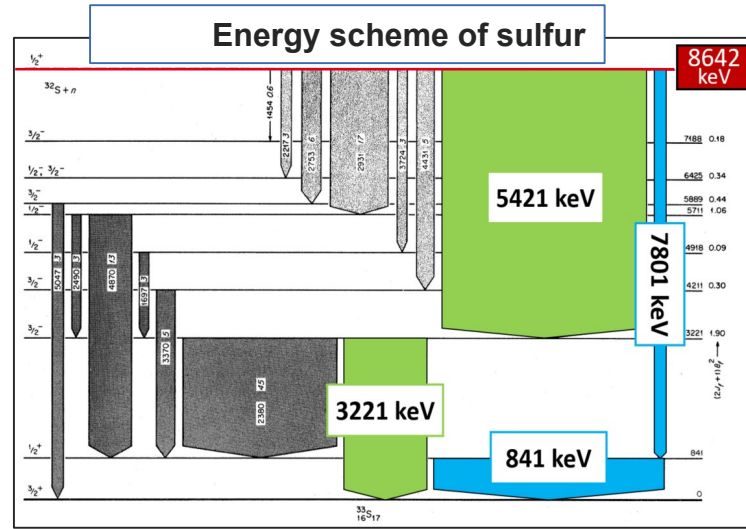


Large NaI scintillators (5"x5"x10") vs. HPGe (30 %)



Pulsed NAA with scintillators

Gamma coincidences with large **Nal(Tl) scintillators**



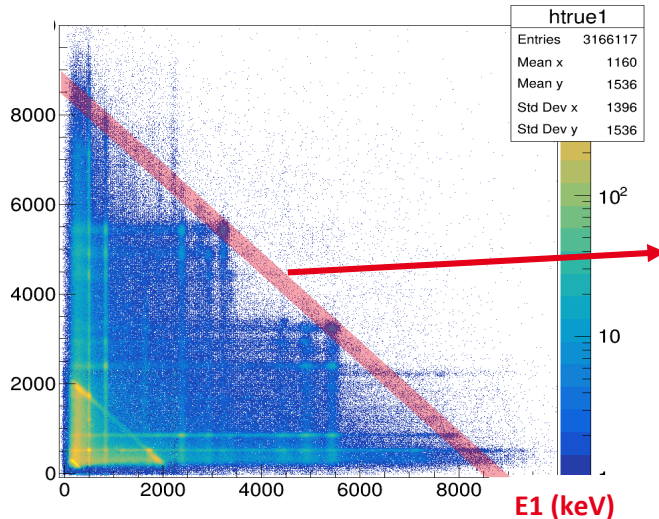
SODERN's online elemental analyzer for cement, coal and mining industries

with BGO scintillators

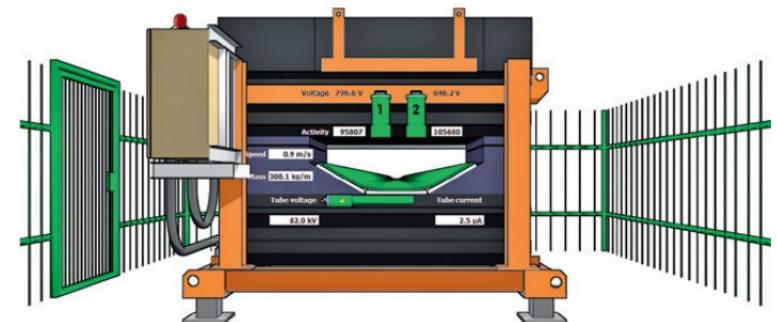
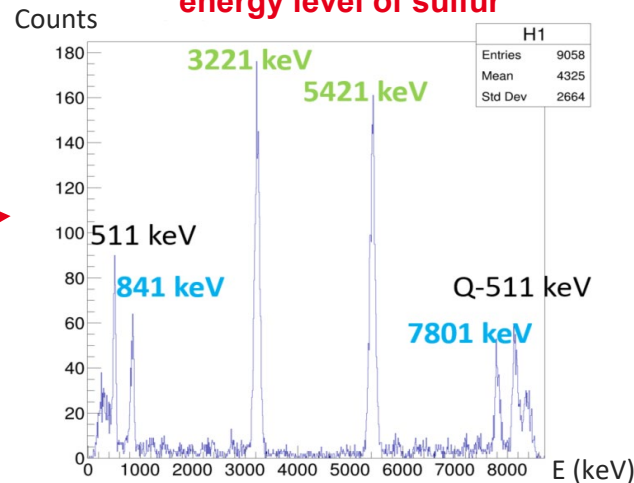
(spectrum unfolding \Rightarrow see next slide)



E2 (keV)

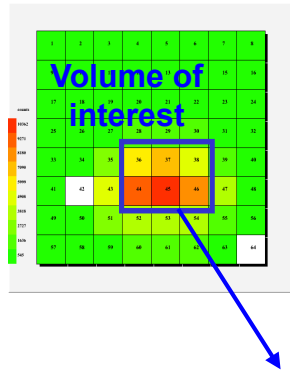


2D projection on the 8642 keV energy level of sulfur

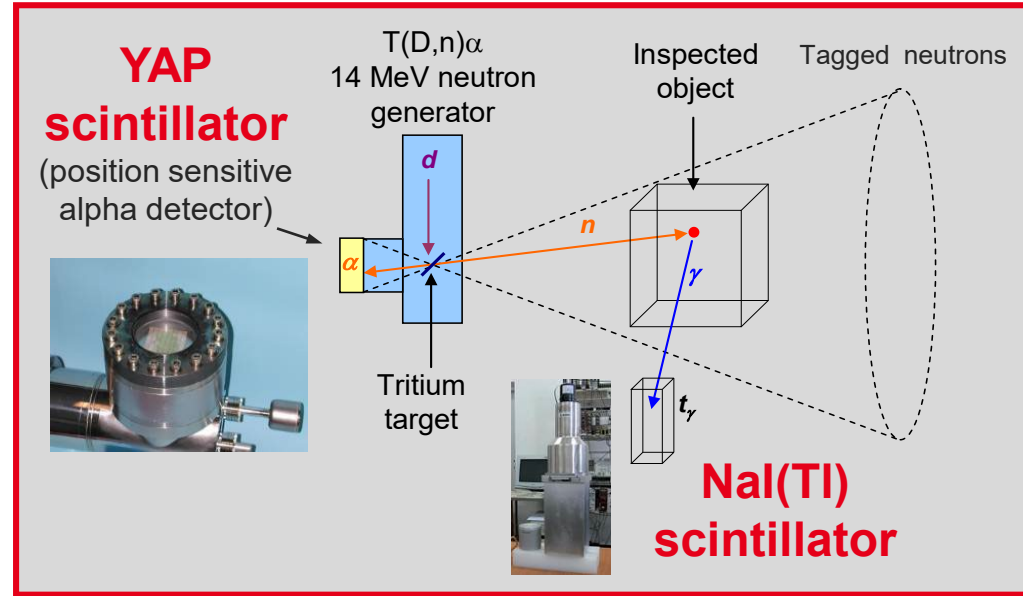
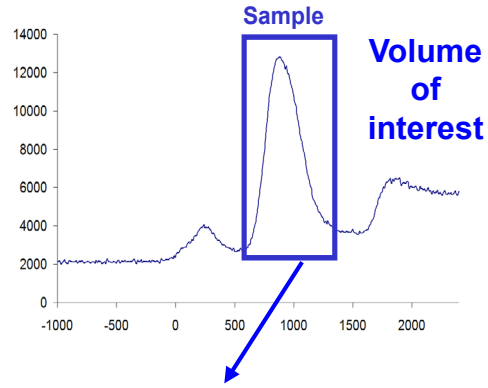


NAA with the Associated Particle Technique (APT)

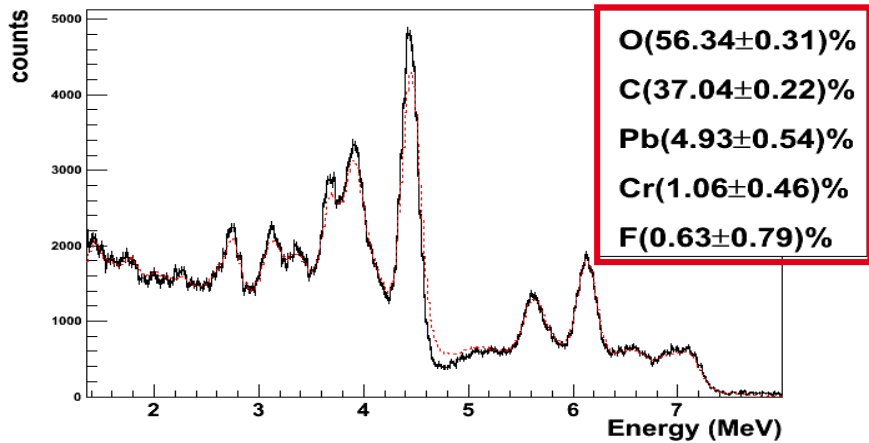
YAP position sensitive alpha detector



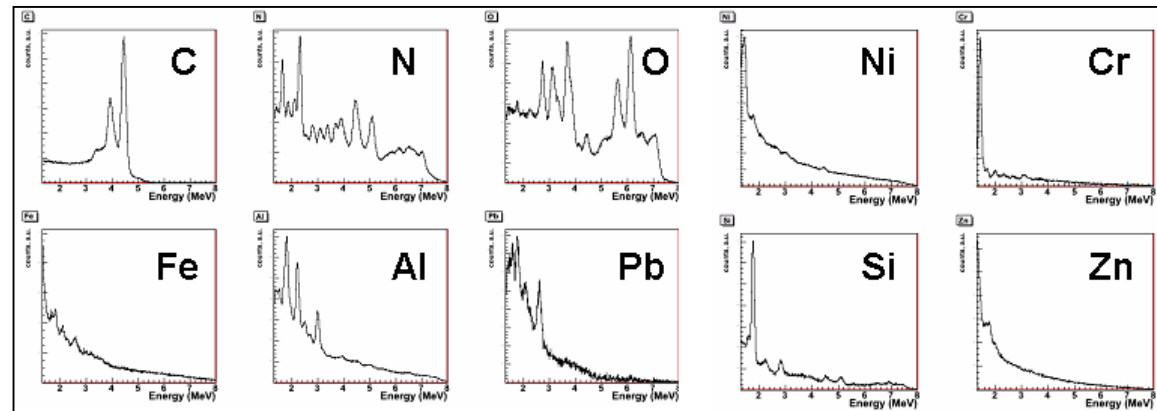
Neutron TOF (alpha-gamma coincidence)



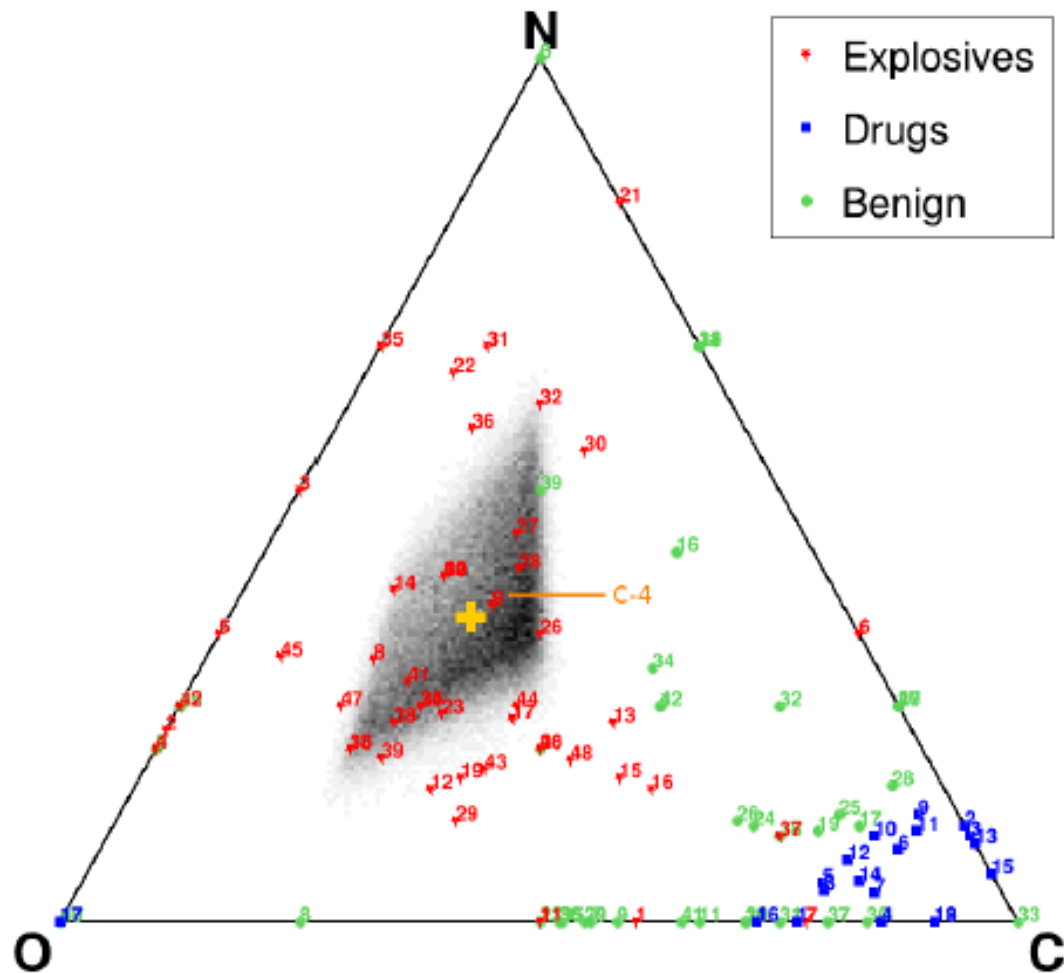
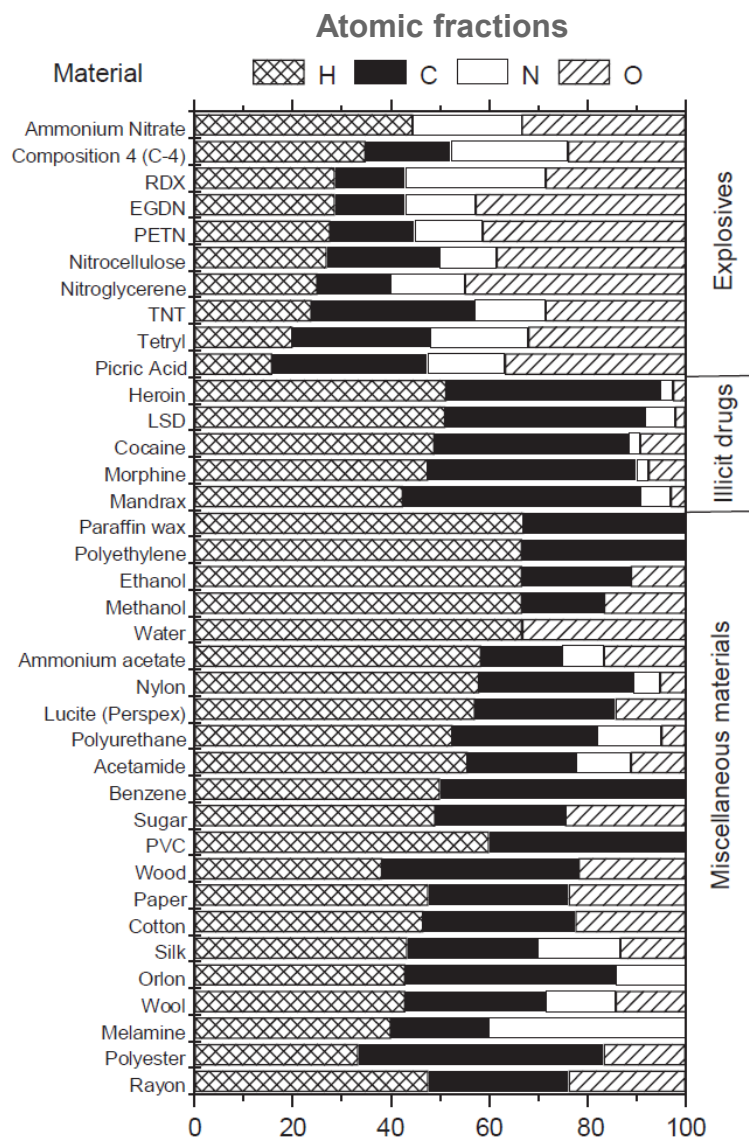
NaI(Tl) gamma spectrum (selected voxel)



Unfolding (with a calibration database)



Organic Materials identification with APT

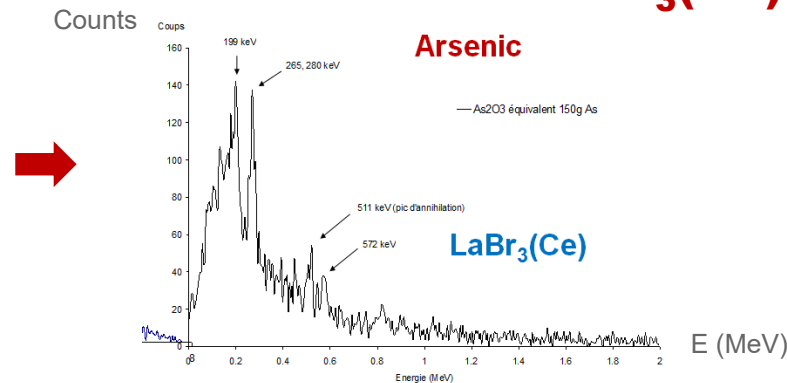


Different scintillators used with APT

Cargo containers ⇒ large 5"x5"x10" NaI(Tl)

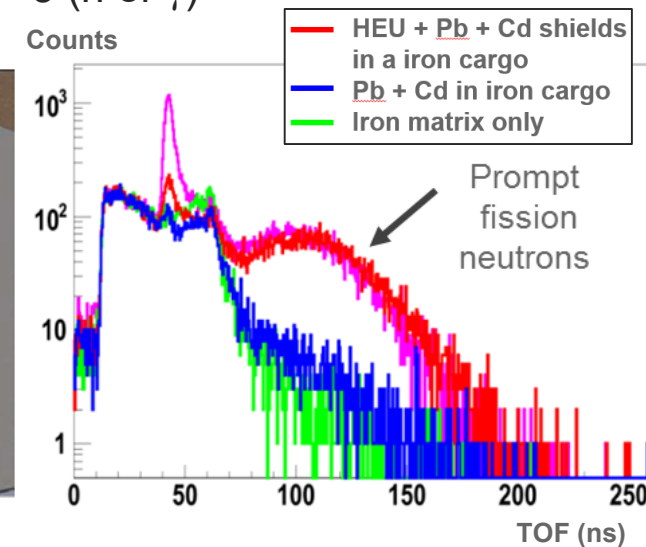
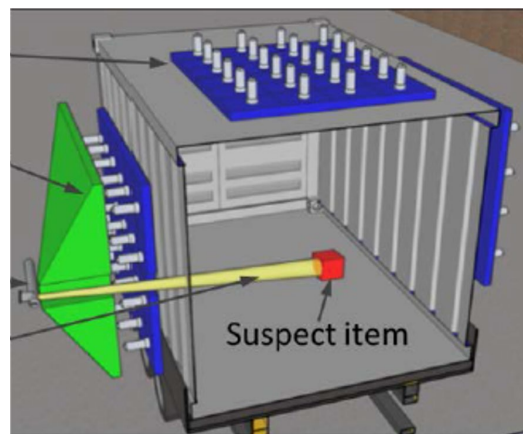


Chemical warfare detection ⇒ 3"x3" LaBr₃(Ce)

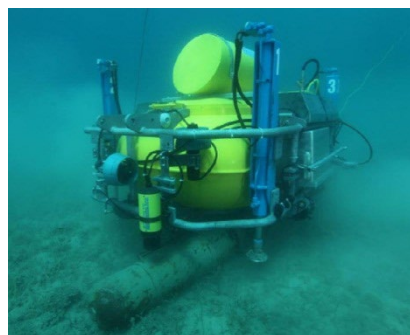
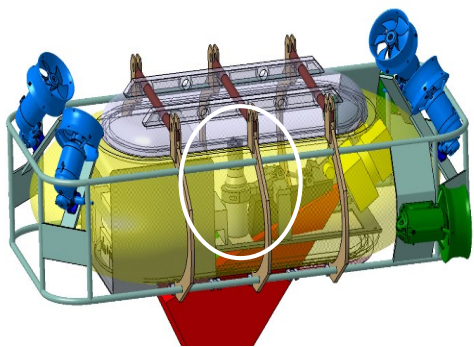


Nuclear material detection ⇒ large PVT panels

Coincidences between 1 α + 3 (n or γ)



Submarine explosive detection ⇒ 3"x3" LaBr₃(Ce)





Photofission delayed gamma rays \Rightarrow U, Pu characterization

15-20 MeV LINAC with its shielded head



Irradiation 2h



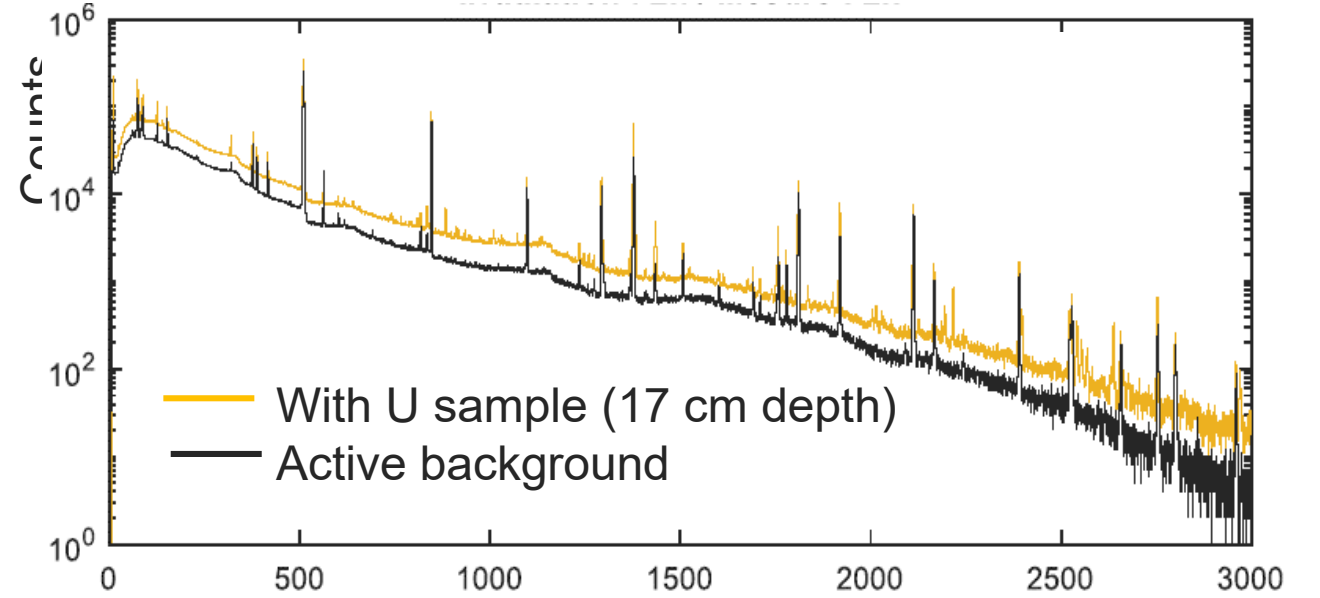
Transfer time to HPGe + cooling delay of a few min



HPGe acquisition 2h



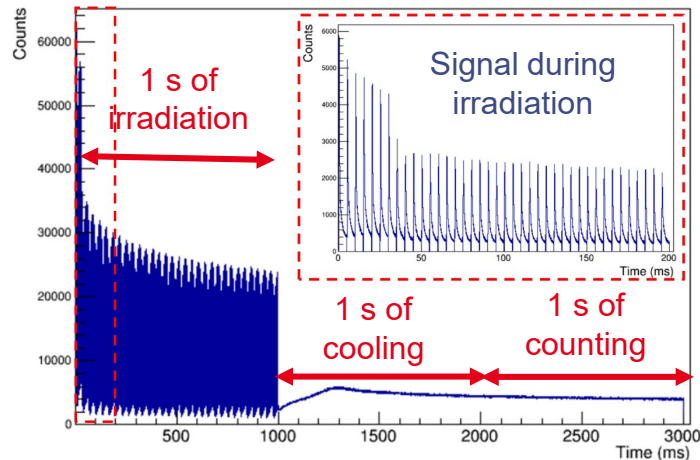
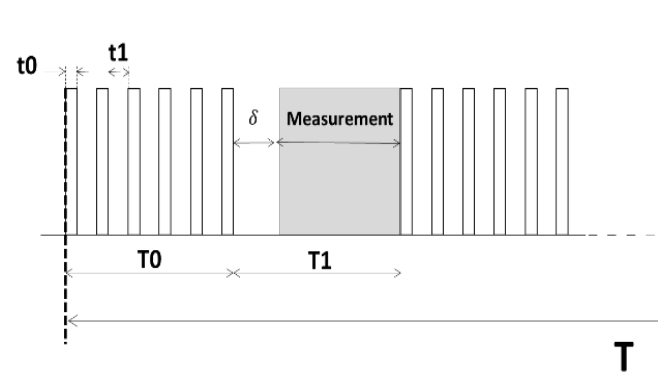
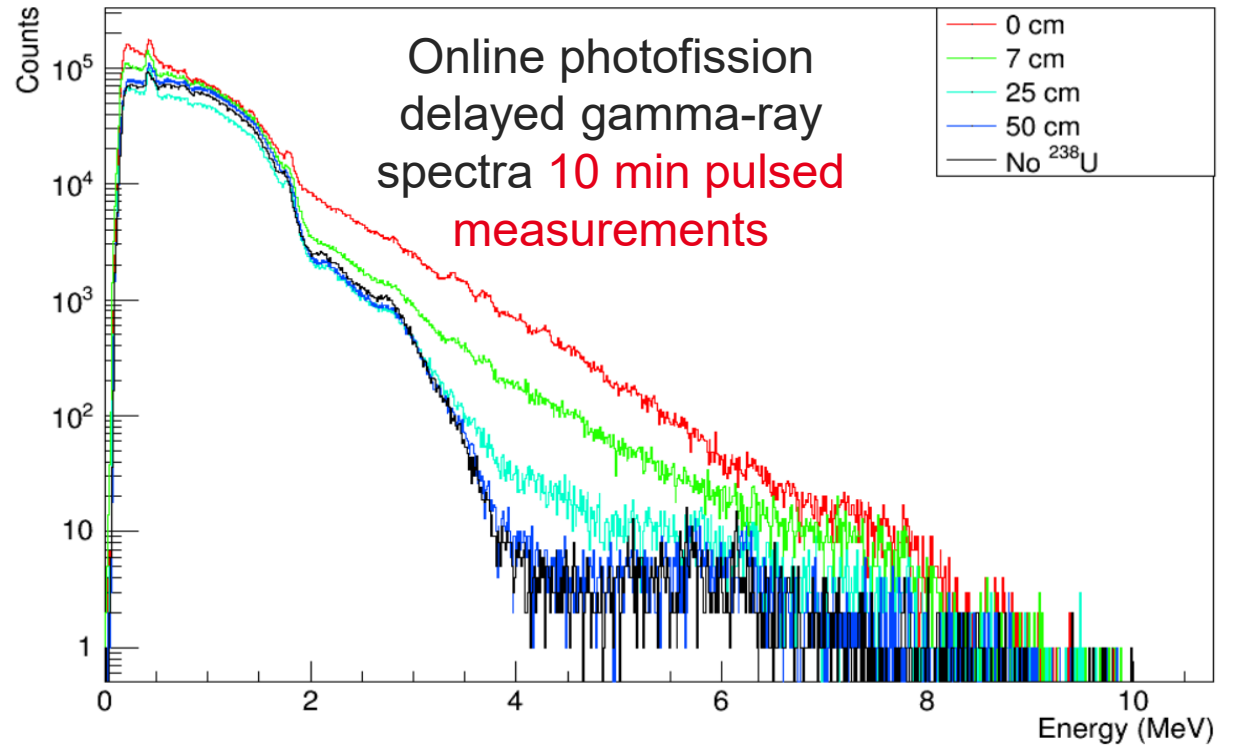
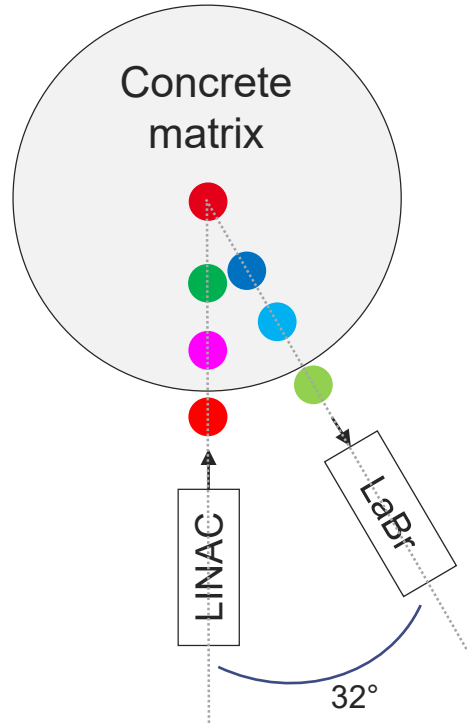
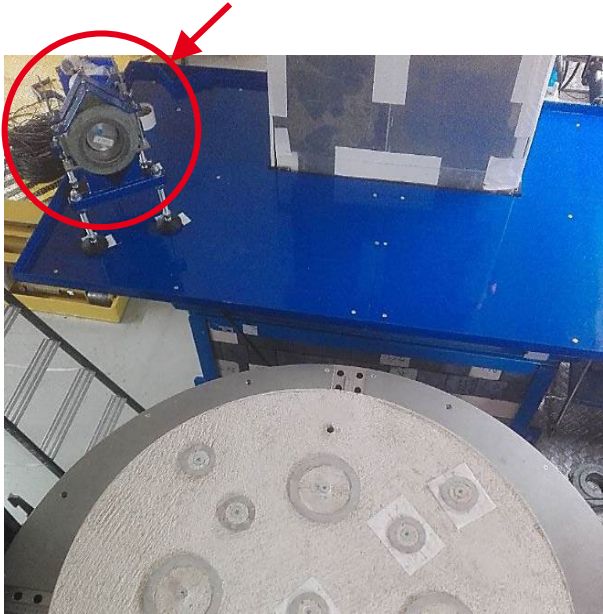
Highly shielded HPGe



Using a $\text{LaBr}_3(\text{Ce})$ scintillator in pulsed irradiation



$\text{LaBr}_3(\text{Ce})$ scintillator



Energy threshold	Net counts from ^{238}U	Background counts	Net counts / background
2	544017 ± 826	69371	8
2.5	308041 ± 599	25434	12
3	171349 ± 429	6496	26
3.5	92585 ± 310	1824	51
4	48954 ± 226	1184	41
4.5	24960 ± 164	1004	25
5	12357 ± 118	866	14





6 ■ **Conclusion and prospects**



Conclusion and prospects

□ Many advantages for scintillators

- Fast timing, high count rates, high efficiency, low cost, rugged, no cooling, many shapes...
- ⇒ Appropriate for a wide range of in situ and online applications

□ Constant innovations

- Developments of scintillation materials, setups (mixtures of scintillation materials, light guides, optical fibers, segmented detectors, multi-anodes PM, SiPM, ...), fast numeric electronics, onboard signal processing (PSD, CFD, coincidences, ToF...), etc.
- Improvements in time and energy resolutions, detection efficiency, throughput (small count losses at high count rates), specificity (e.g. by loading organic scintillators with B or Li, or coating with Cd or Gd to detect thermal neutrons, n/γ discrimination using pulse shape or detection times, ...), spatial selectivity with segmented light readout, segmented detectors, large scintillator arrays at reasonable cost...
- Machine learning, ANN and other mathematical approaches to counterbalance some drawbacks such as the poor spatial and energy resolutions, cross talk, γ sensitivity, etc.

⇒ **More and more applications open up for scintillators**



iresne



THANK YOU

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