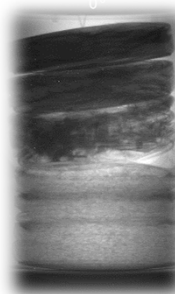


DE LA RECHERCHE À L'INDUSTRIE

cea

LES MESURES NUCLÉAIRES NON DESTRUCTIVES POUR LE CONTRÔLE DES DÉCHETS RADIOACTIFS, L'ASSAINISSEMENT ET LE DÉMANTÈLEMENT DES INSTALLATIONS

Bertrand PEROT, CEA DEN CADARACHE

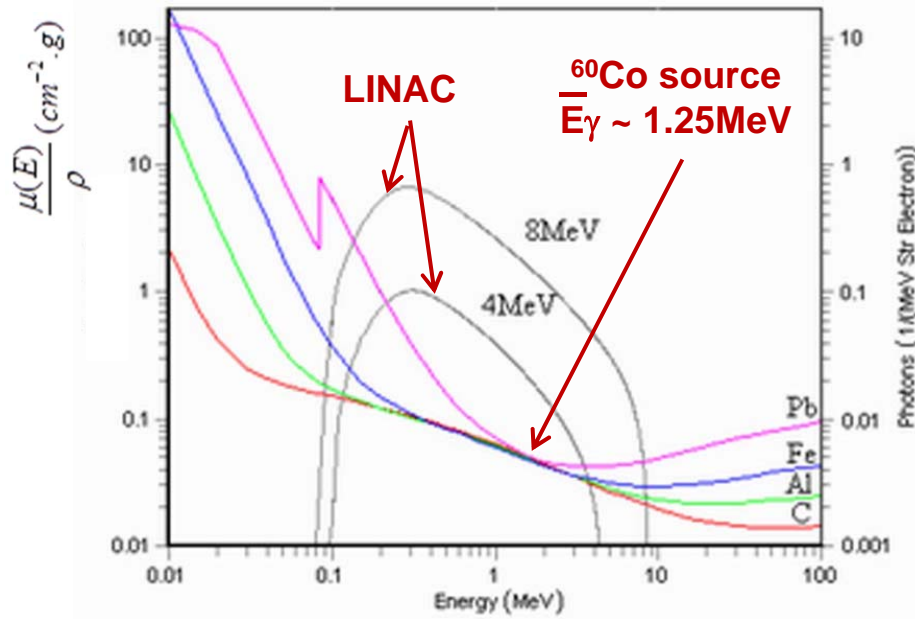


www.cea.fr

EFFMIN 4 | MARSEILLE, FRANCE | 18-21 JUILLET 2016

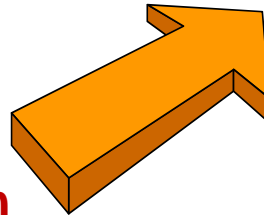
- ❑ Photon imaging
- ❑ Gamma-ray spectroscopy
- ❑ Passive and active neutron measurements
- ❑ Coupling measurement results
- ❑ R&D on new techniques

TRANSMISSION RADIOGRAPHY AND DENSITOMETRY

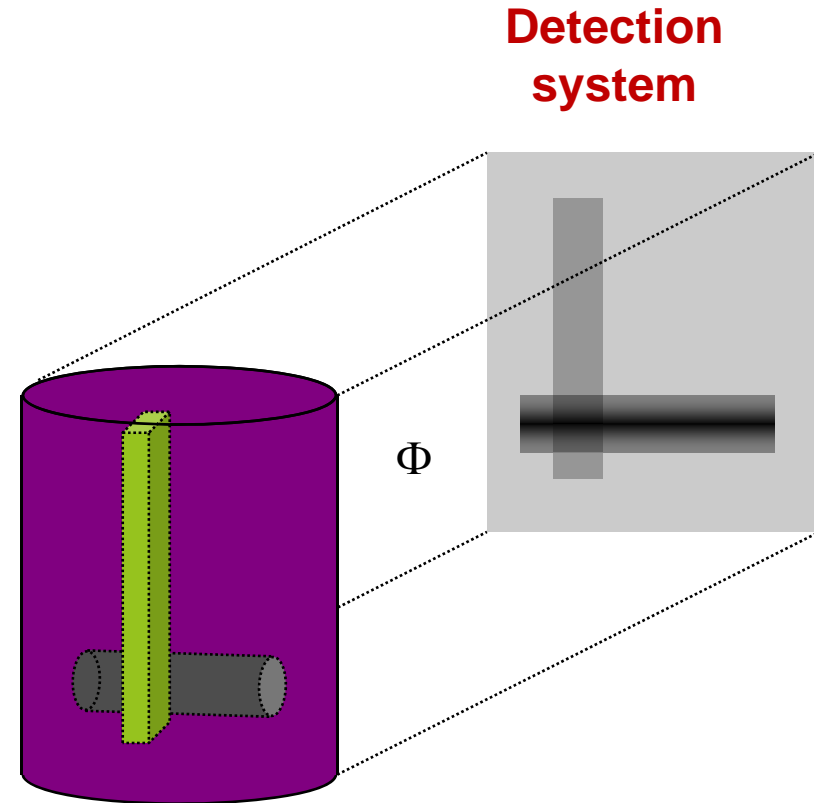


$$\rho = \frac{\ln(\Phi_0 / \Phi)}{\frac{\mu(E)}{\rho} \times L}$$

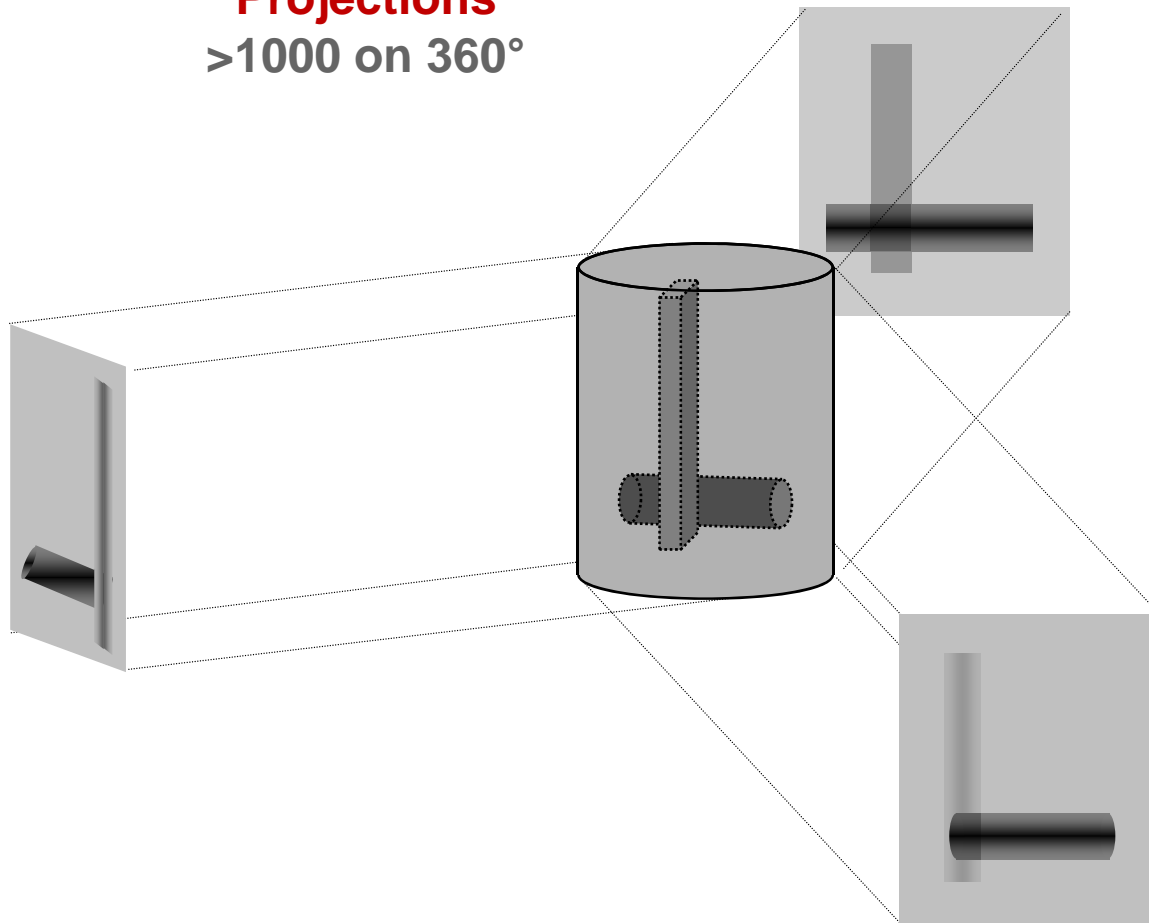
Photon source



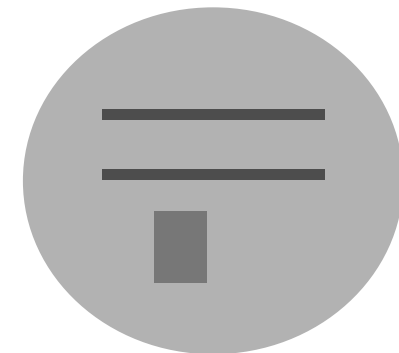
Φ_0



Projections
>1000 on 360°

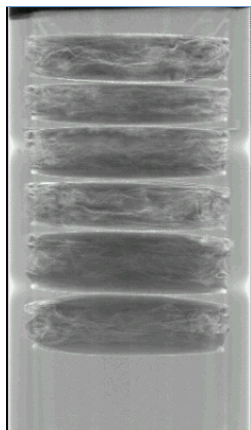


**Reconstructed
2D cross sections**

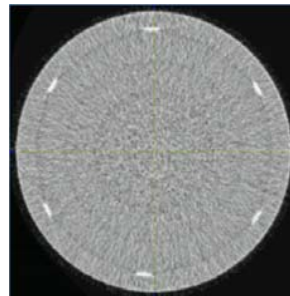


^{60}Co IMAGING: PRECISE DENSITY BUT LONG

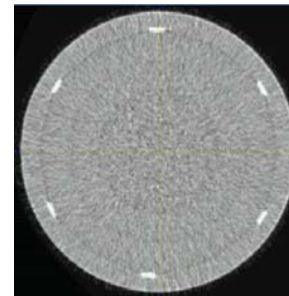
TRANSEC: historic ^{60}Co tomography system of LMN, CEA DEN CADARACHE



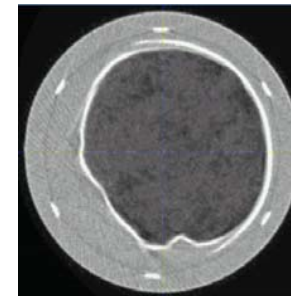
Density



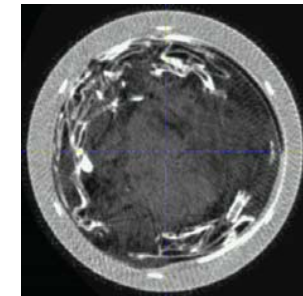
50 mm



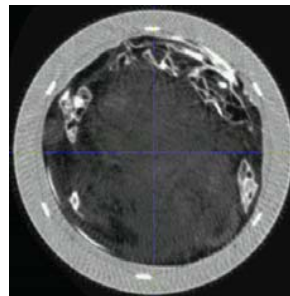
120 mm



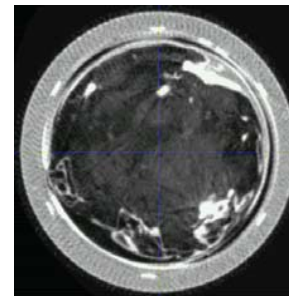
190 mm



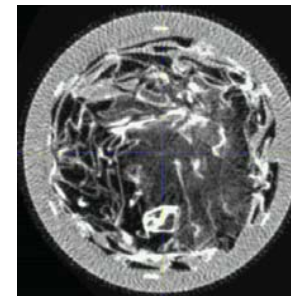
260 mm



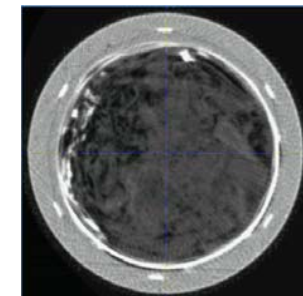
330 mm



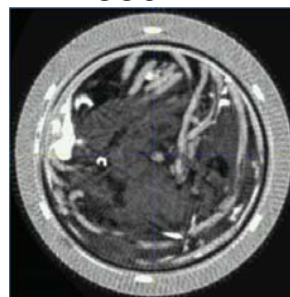
400 mm



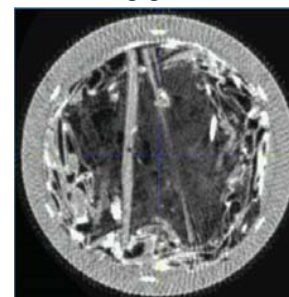
470 mm



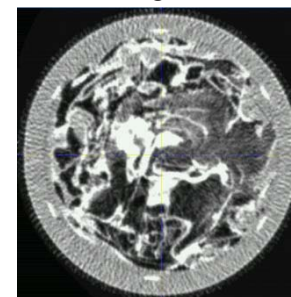
540 mm



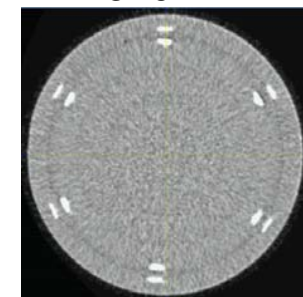
610 mm



680 mm

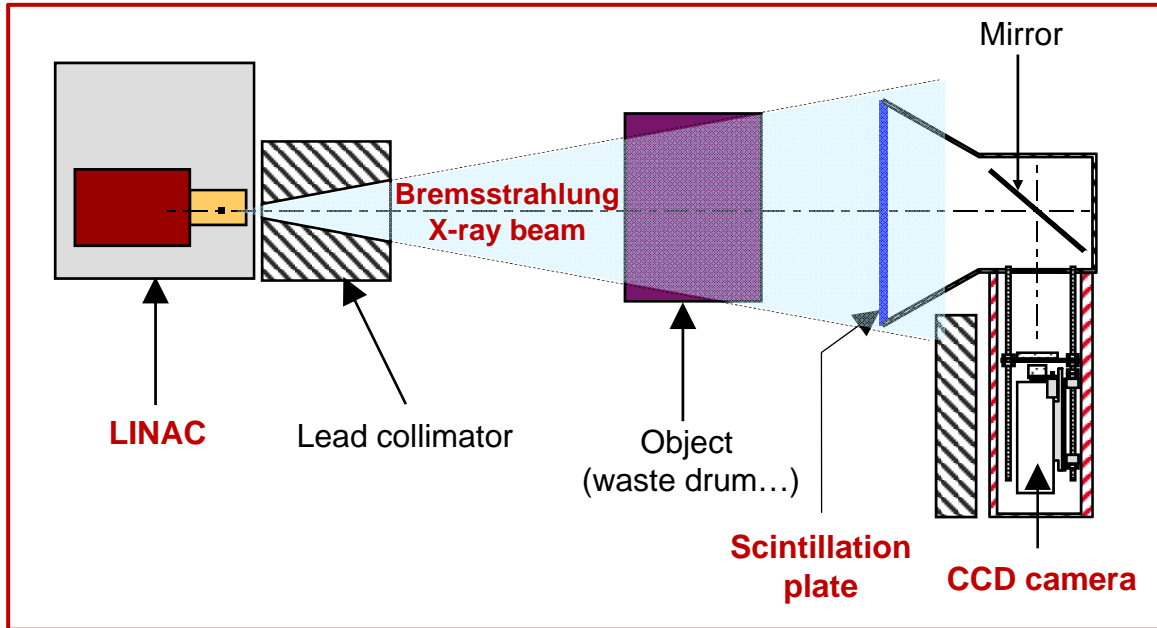


750 mm



820 mm

HIGH ENERGY X-RAY IMAGING WITH A LINAC



CINPHONIE: recent high energy imaging system of LMN, CEA DEN CAD

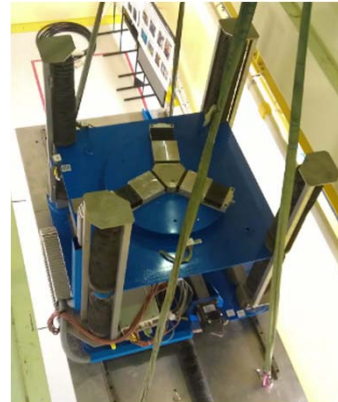
CINPHONIE Vs. TRANSEC:

- ❑ Radiography: 240 × faster (40 h → 10 min)
- ❑ Tomography: 36 × faster (15 h → 25 min)

9 MeV LINAC (15 Gy/min at 1m)



Moving platform (up to 2 tons)

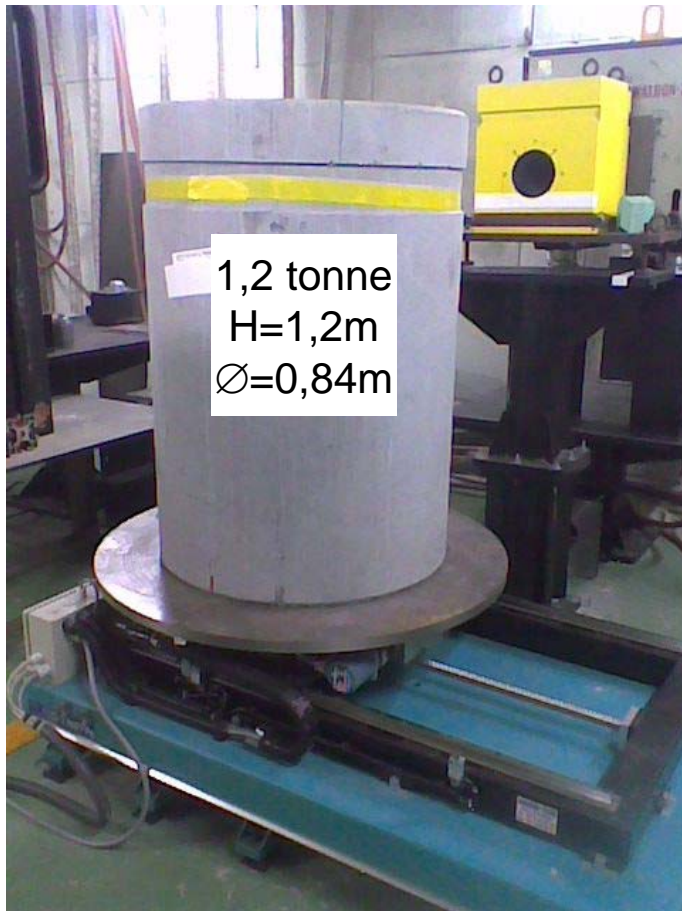


Scintillation plate (800 x 600 mm²)
Gadox Gd₂O₂S ; 1.5 mm

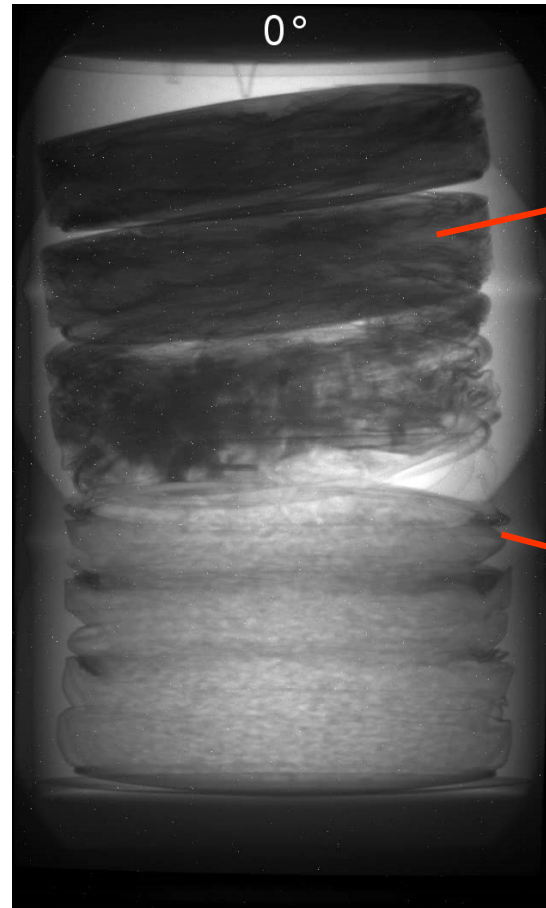


Concrete waste: density > 2

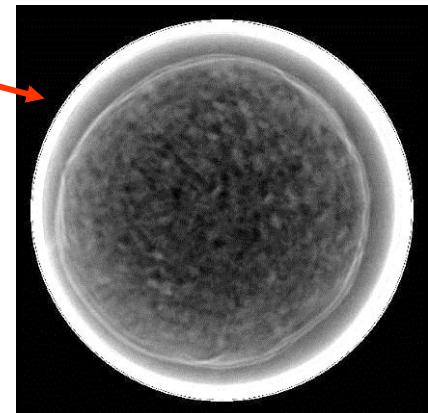
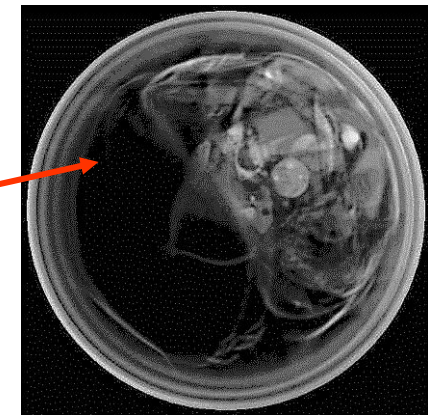
Less than 1 s per single radiography



■ Radiography

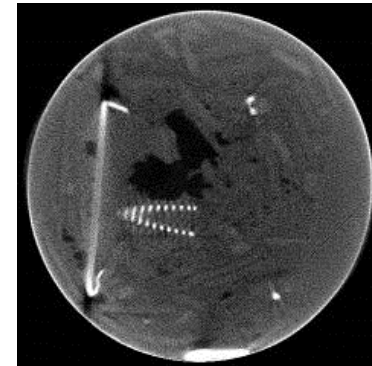
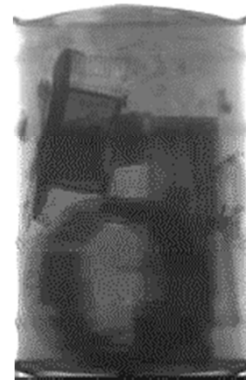


■ Tomography



IN SITU HIGH ENERGY X-RAY IMAGING

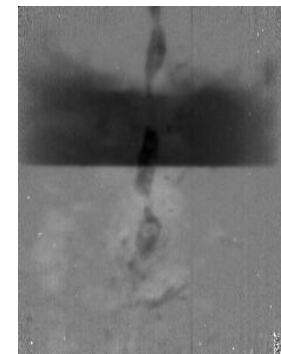
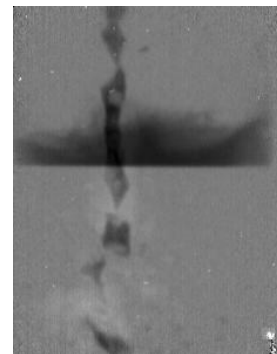
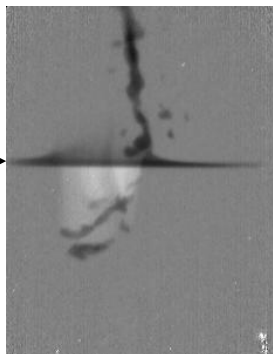
Bituminized waste drum characterization at BELGOPROCESS facility, Belgium



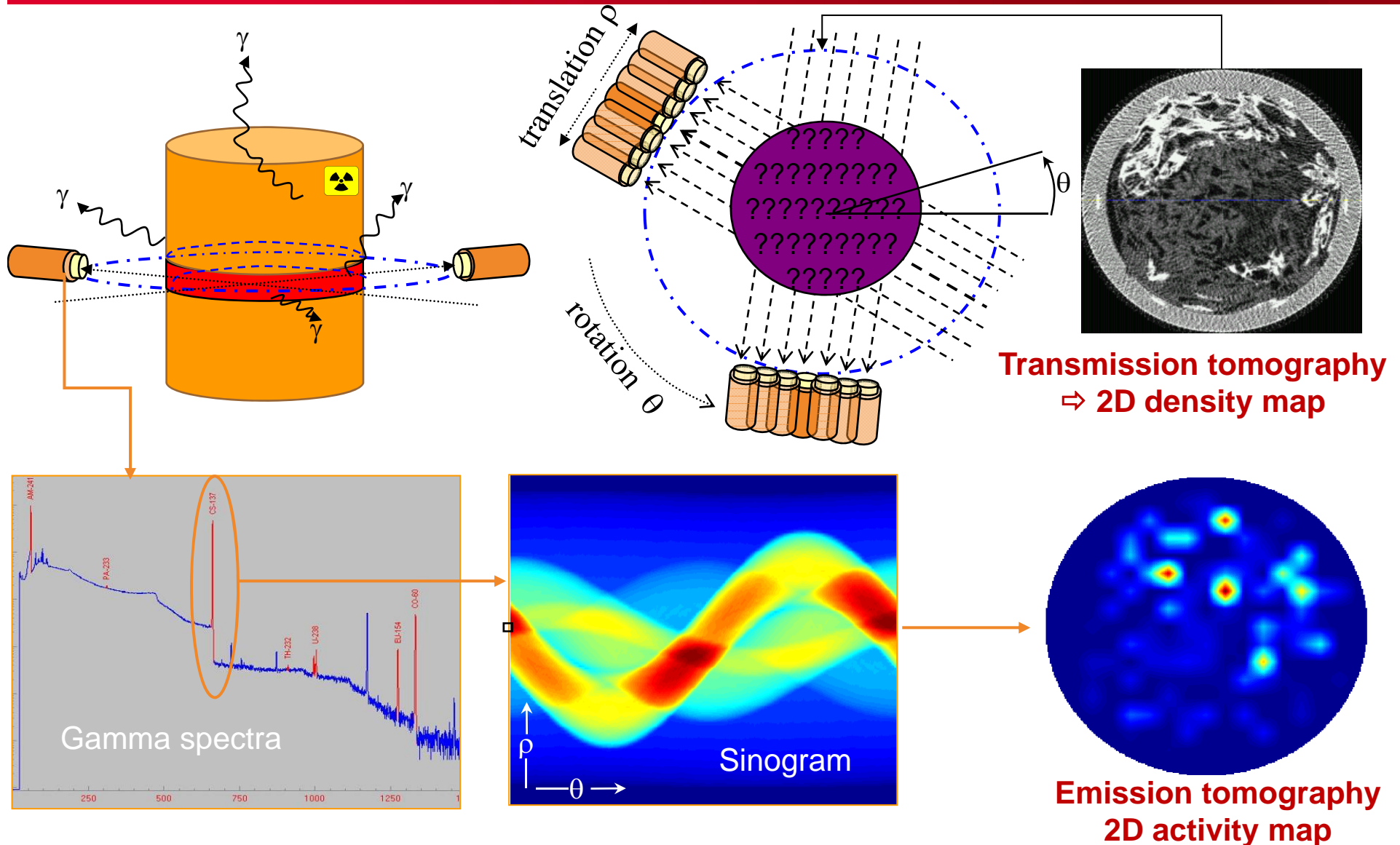
Radioscopy of the corium - water interaction Nuclear accident studies, KROTOS facility, CEA Cadarache



Water
surface



COMPUTED GAMMA EMISSION TOMOGRAPHY





PHOTON IMAGING

GAMMA SPECTROSCOPY

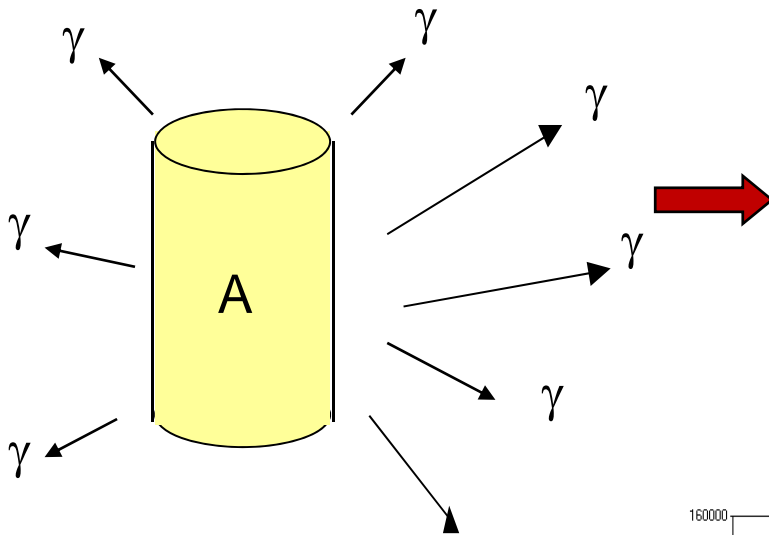
PASSIVE AND ACTIVE NEUTRON MEASUREMENTS

COMBINED MEASUREMENTS

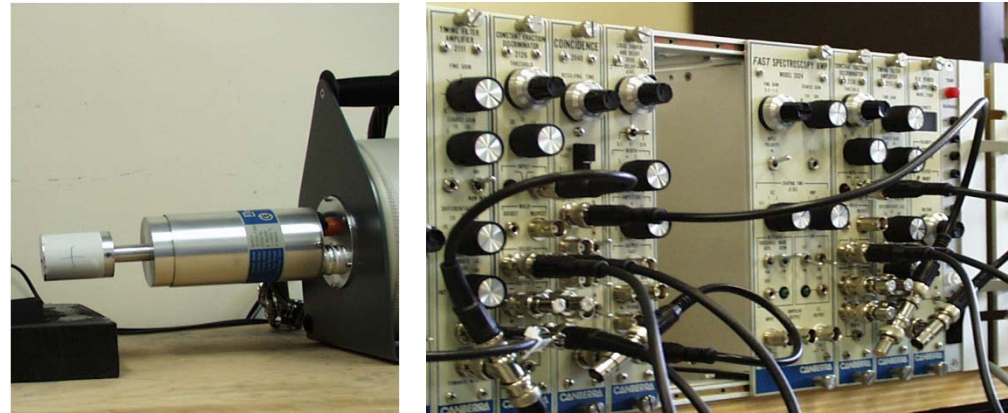
R&D

GAMMA-RAY SPECTROSCOPY

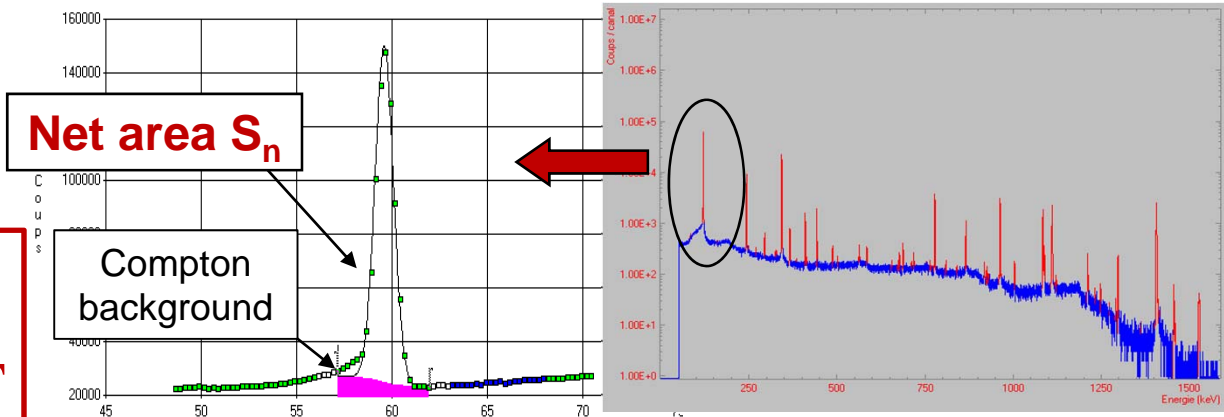
Radioactive object
(e.g. a waste package)



Detector and electronics



Gamma spectrum



Activity

$$A(E) = \frac{S_n(E)}{\epsilon(E) \times I(E) \times T}$$

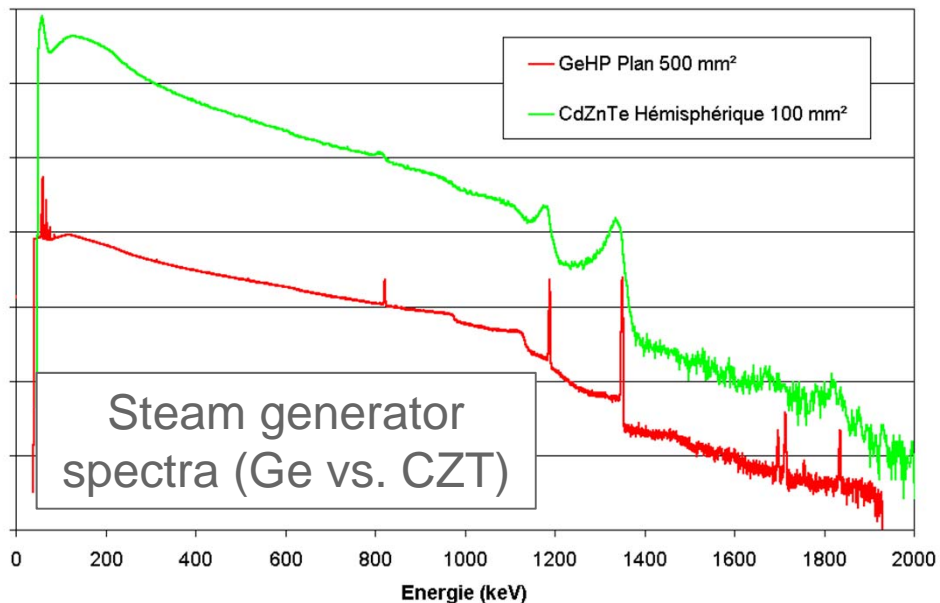
RADIOACTIVE WASTE CHARACTERIZATION

- **Specific gamma spectroscopy** stations the laboratory
- **Part of combined measurements** e.g. for ANDRA
“Super-Controls” with X-ray imaging and neutron assay



IN SITU γ SPECTROSCOPY IN NUCLEAR REACTORS

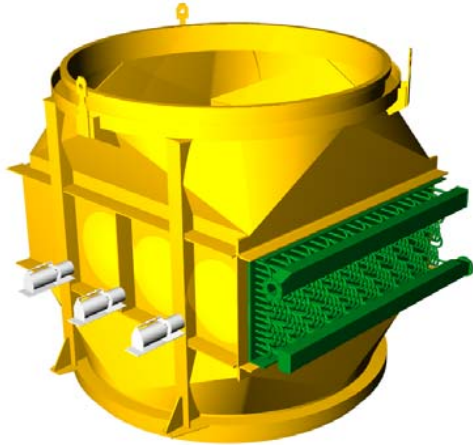
- **Historic activity** > 300 campaigns (end of 70's)
- **EDF** and foreign power plants
- **EMECC** device & software
 - Intermediate activities (1 MBq/m² → 100 GBq/m²)
 - Pipes, steam generators, heat exchangers...
 - Characterization of a whole plant (all circuits)
 - Deposited or volumetric activities (GBq / m² or m³)
 - Experimental qualification by EDF
 - Support to **OSCAR** contamination transfer code



IN SITU γ SPECTROSCOPY OF LARGE COMPONENTS

Uranium hold-up measurement in a heat exchanger

Segmented gamma spectrometry



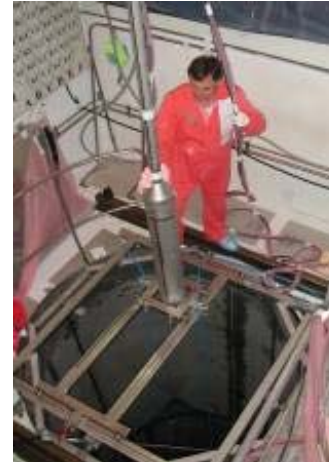
SFR cold trap characterization

HP Ge + CZT in inspection hall



Vessel contamination and activation

Underwater HP Ge gamma-ray spectroscopy



Characterization of activated components

TORE SUPRA wastes – HP Ge



■ Dismantling operations

Fuel fabrication and reprocessing units

■ HP Ge detector

⇒ isotopic composition, activity or mass

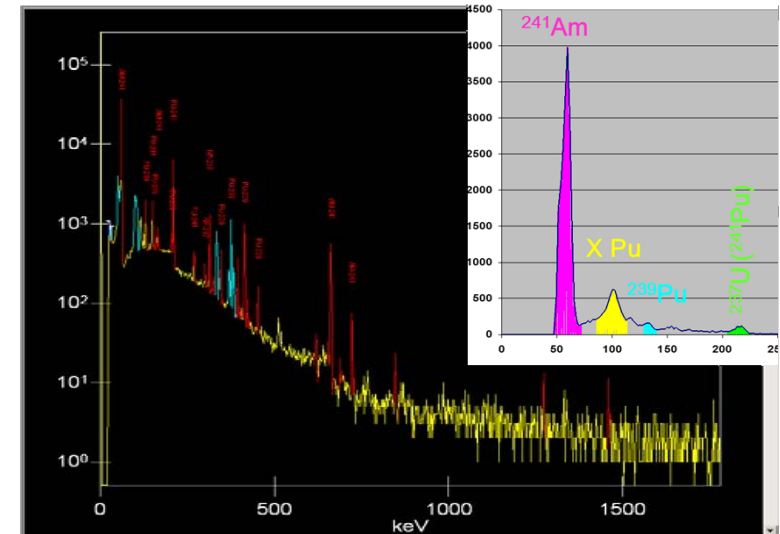
■ CZT probe introduced in glove boxes

⇒ localization

Characterization of Pu in glove boxes
of a Pu fuel fabrication facility



Fissile mass estimation in a
reprocessing unit



HP Ge detector



CZT probe



Inside

After removing all components



Outside

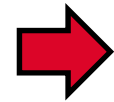
Walls control



High efficiency HP Ge with tungsten shield

PHOTON IMAGING

GAMMA SPECTROSCOPY

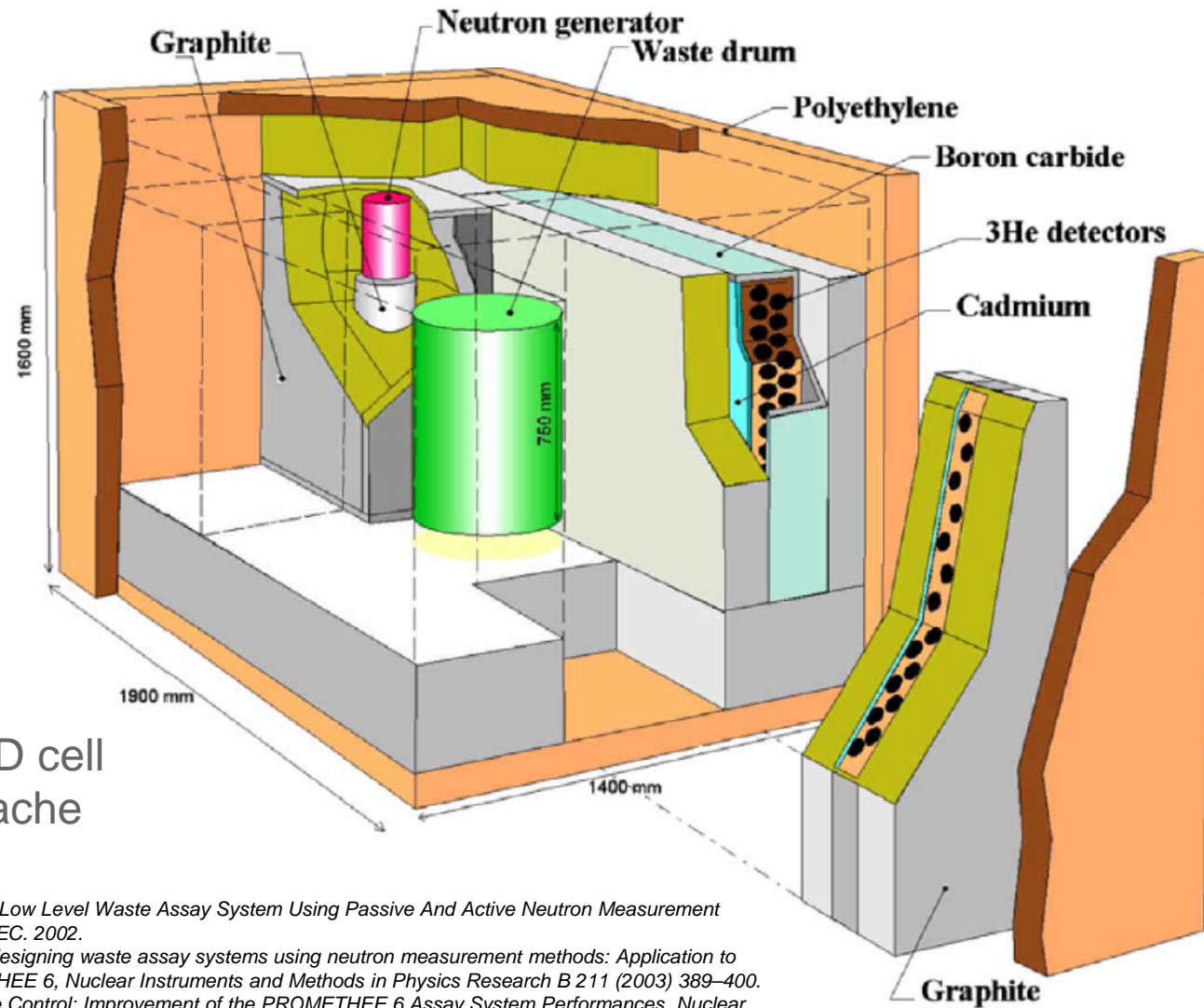


PASSIVE AND ACTIVE NEUTRON MEASUREMENTS

COMBINED MEASUREMENTS

R&D

MIXED PASSIVE & ACTIVE NEUTRON CELL (R&D)



“PROMETHEE VI” R&D cell LMN, CEA DEN Cadarache

- ❑ Christian Passard et al., *PROMETHEE: An Alpha Low Level Waste Assay System Using Passive And Active Neutron Measurement Methods*, NUCLEAR TECHNOLOGY VOL. 140 DEC. 2002.
- ❑ A. Mariani et al., *The help of simulation codes in designing waste assay systems using neutron measurement methods: Application to the alpha low level waste assay system PROMETHEE 6*, Nuclear Instruments and Methods in Physics Research B 211 (2003) 389–400.
- ❑ Fanny Jallu et al., *Alpha-Particle Low-Level Waste Control: Improvement of the PROMETHEE 6 Assay System Performances*, Nuclear Technology Vol. 153 Jan. 2006.

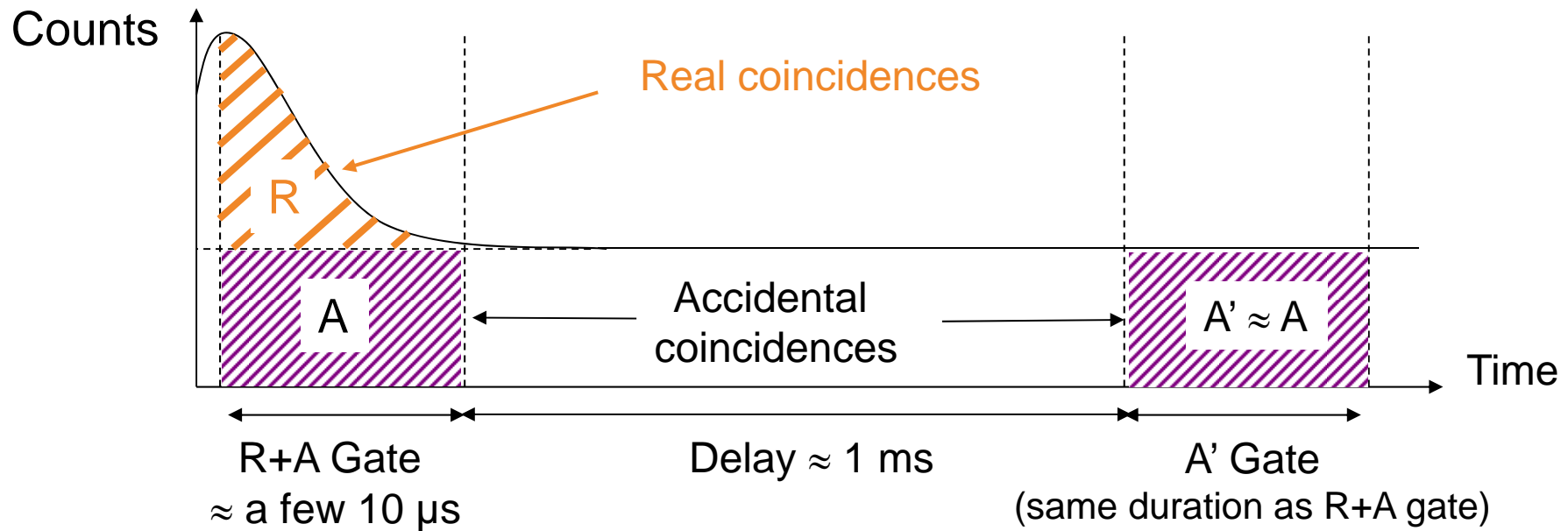
PASSIVE NEUTRON MEASUREMENT

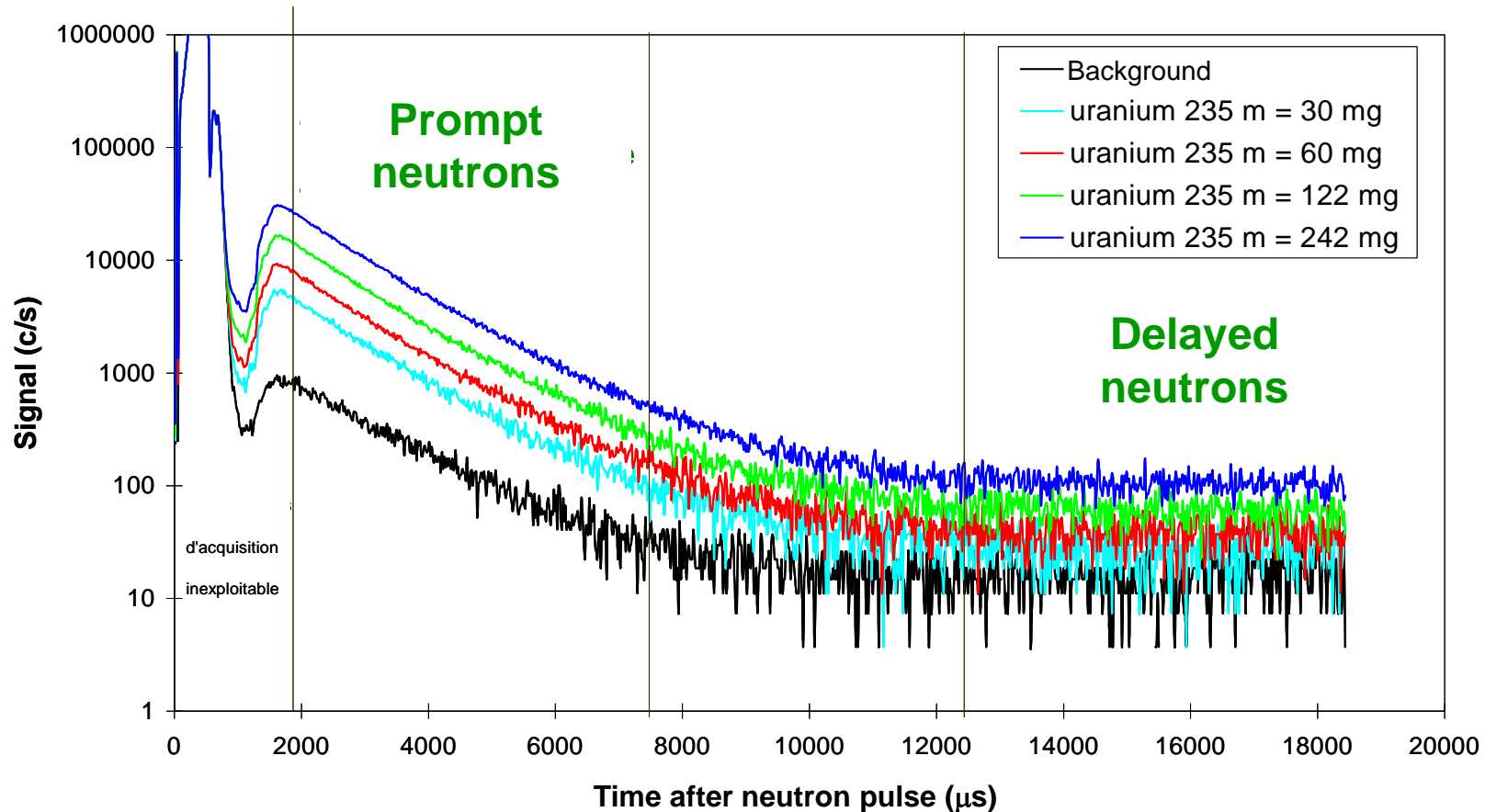
❑ Total counting

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

❑ Coincidence counting

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

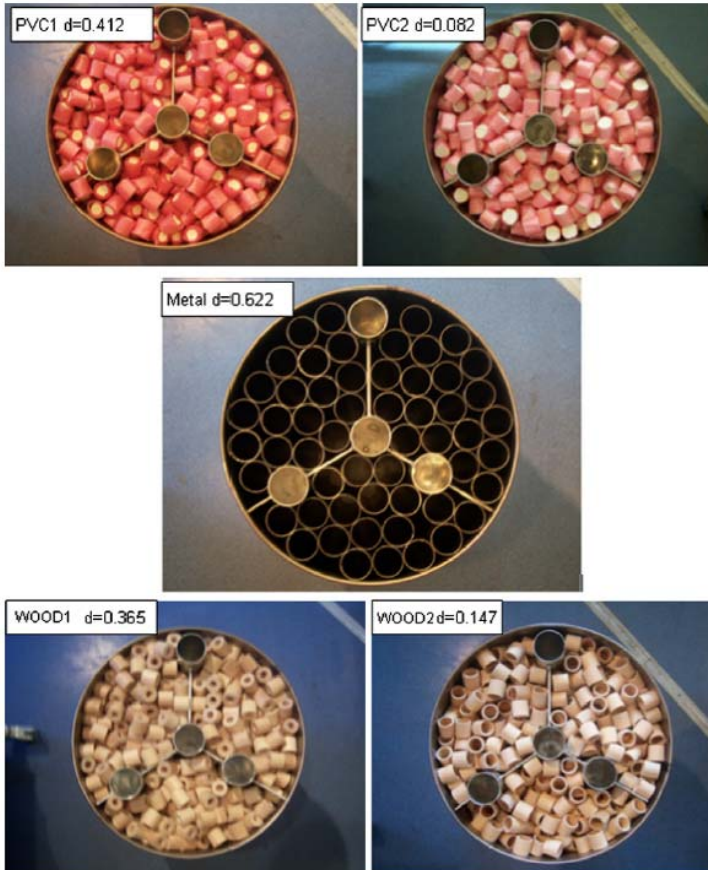




Prompt and delayed neutron signals \Rightarrow ^{235}U and ^{239}Pu discrimination

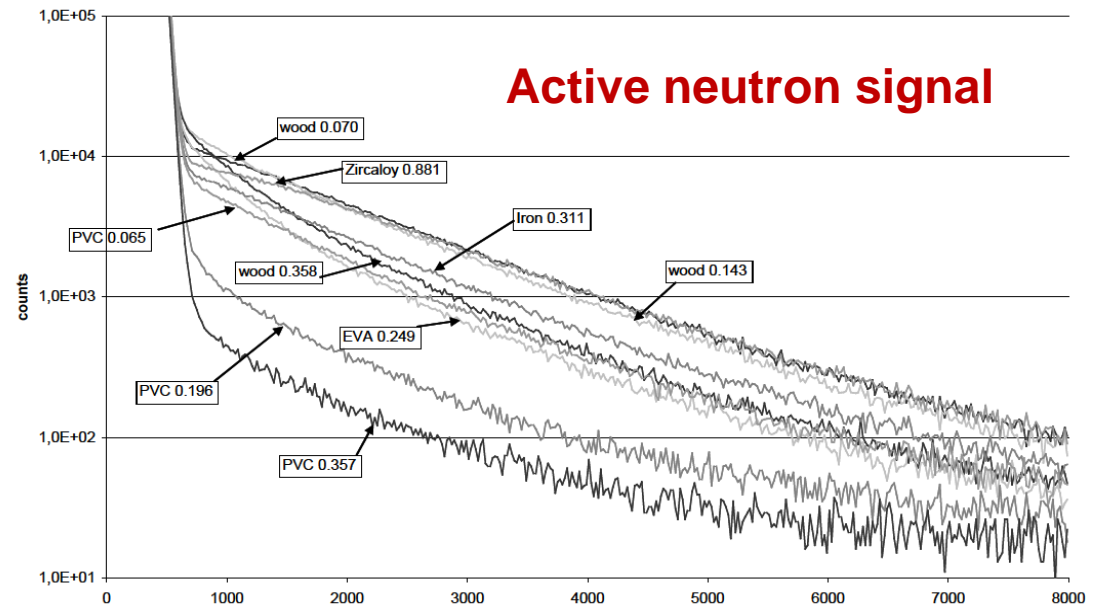
- **Prompts neutrons** \Rightarrow ^{235}U , ^{239}Pu and ^{241}Pu
- **Delayed neutrons** \Rightarrow ^{235}U , ^{239}Pu and ^{241}Pu + ^{238}U and ^{232}Th

CALIBRATION WITH DIFFERENT MATRIX MATERIALS



Passive detection

Matrix	Detection efficiency (%)	
	Measured	Calculated
Empty cavity	10.13 ± 0.22	10.27 ± 0.01
PVC1 (d = 0.412)	9.05 ± 0.20	9.19 ± 0.01
PVC2 (d = 0.082)	10.37 ± 0.23	10.54 ± 0.01
WOOD1 (d = 0.365)	8.92 ± 0.20	8.88 ± 0.01
WOOD2 (d = 0.147)	10.44 ± 0.23	10.65 ± 0.01
Metal (d = 0.622)	10.49 ± 0.23	10.67 ± 0.05



- ❑ F. Jallu et al. / Nuclear Instruments and Methods in Physics Research B 271 (2012) 48-54
- ❑ A.-C. Raoux et al. / Nuclear Instruments and Methods in Physics Research B 266 (2008) 4837-4844
- ❑ R. Antoni et al. Matrix effect correction with internal flux monitor in radiation waste characterization with the Differential Die-away Technique, IEEE Transactions on Nuclear Science, Vol. 61, No. 4 (2014) 2155-2160

PASSIVE & ACTIVE NEUTRON CELL

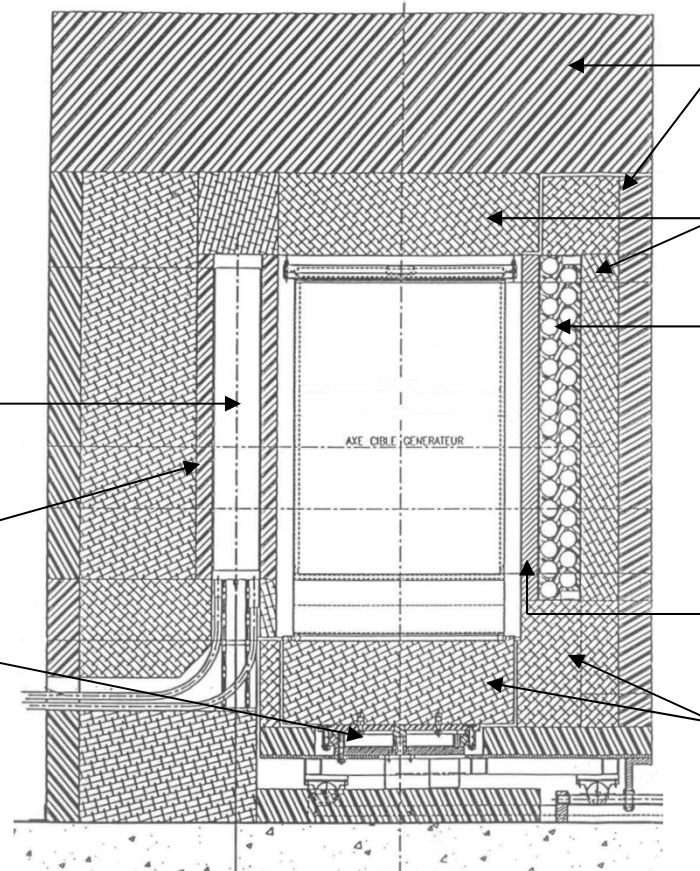


Pulsed DT neutron generator
GENIE 36 SODERN, 2.10^9 n/s

Neutron generator

Lead ring around
the generator

Turntable



Polyethylene

Graphite

99 ^3He counters
in 3 detection
blocks ($\text{CH}_2 + \text{Cd}$)

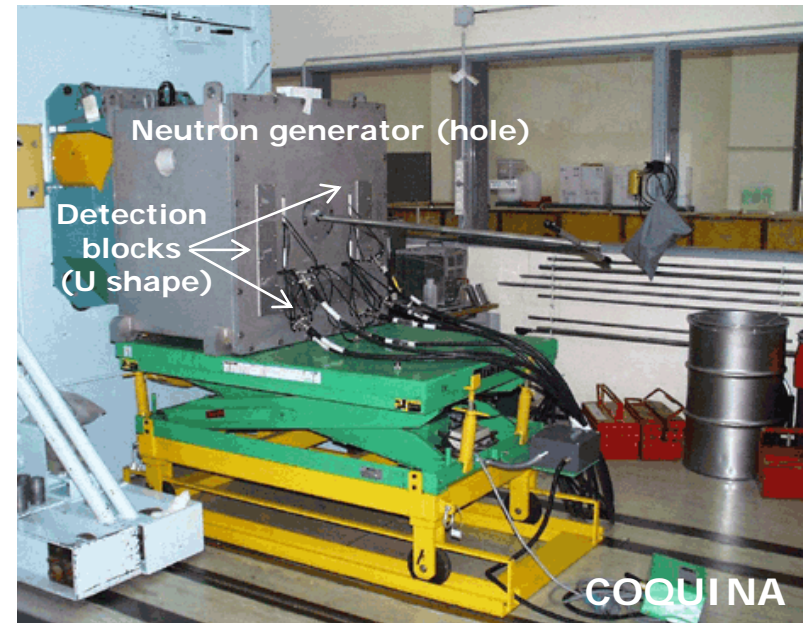
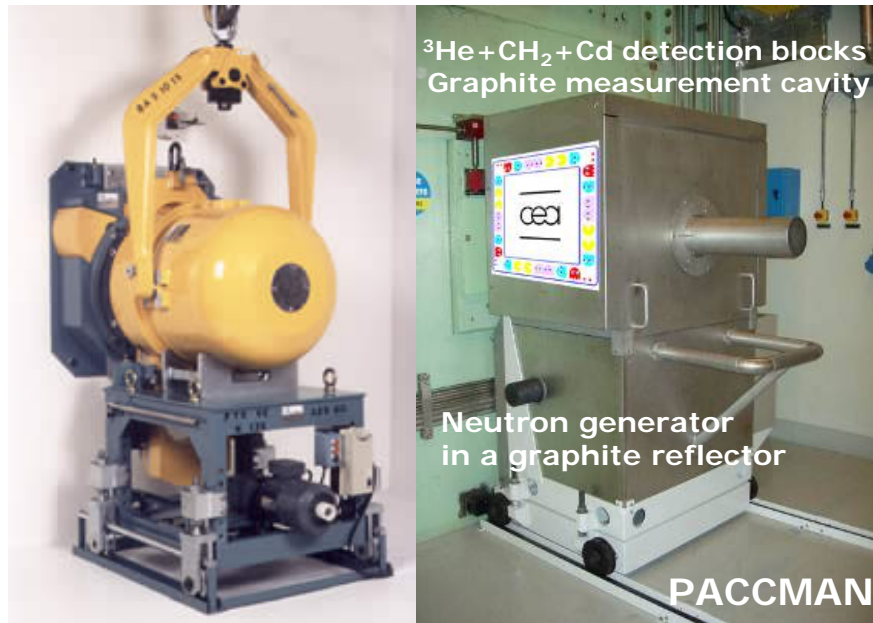
ϵ empty cell ~ 23%

Lead shield

Graphite

SYMETRIC measurement cell
LMN, CEA DEN Cadarache
ANDRA -CEA "Super-controls"

PACCMAN, COQUINA: active neutron interrogation systems plugged to hot cells to assess Pu and U in high level wastes

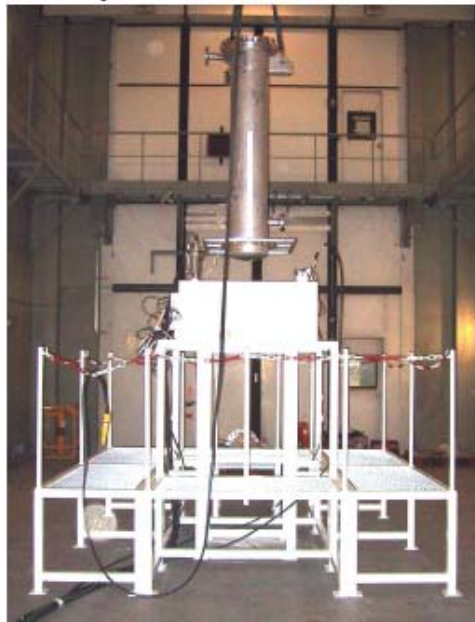


LMN, CEA DEN CADARACHE
ATALANTE, CEA DEN MARCOULE

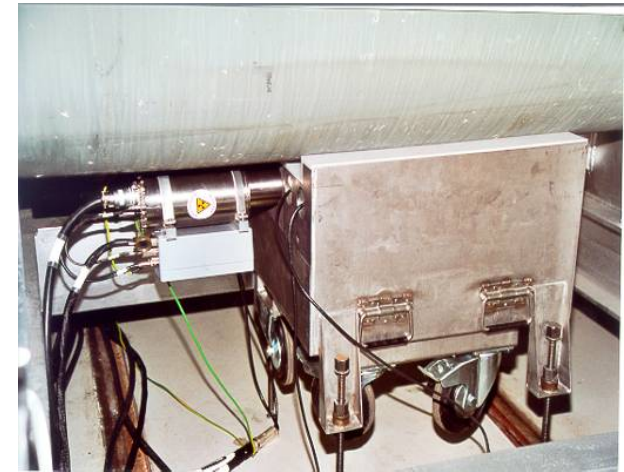
Characterization of large components in uranium enrichment facilities (UF₆ gas diffusion technology)



Passive neutron measurement of the uranium hold-up in UF₆ compressors



Passive (and active) neutron measurements of uranium hold-up in an UF₆ crystallizer



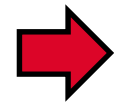
Measurement of the uranium enrichment in an UF₆ container by active neutron interrogation

F. Jallu et al., Dismantling and decommissioning: The interest of passive neutron measurement to control and characterise radioactive wastes containing uranium, Nuclear Instruments and Methods in Physics Research B 271 (2012) 48–54

PHOTON IMAGING

GAMMA SPECTROSCOPY

PASSIVE AND ACTIVE NEUTRON MEASUREMENTS



COMBINED MEASUREMENTS

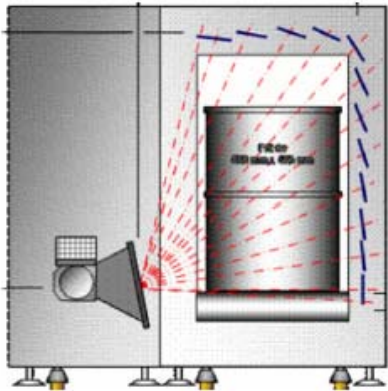
R&D

RADIOACTIVE WASTE CHARACTERISATION

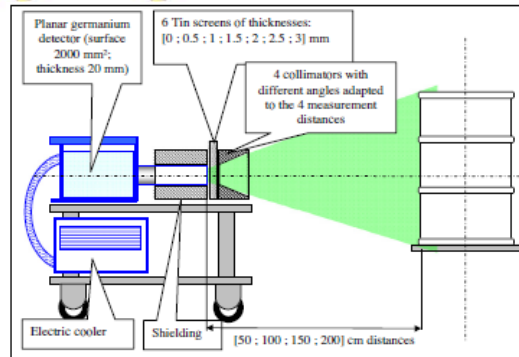
X-RAY RADIOGRAPHY + γ SPECTROSCOPY + PASSIVE NEUTRON COUNTING

X-ray radiography

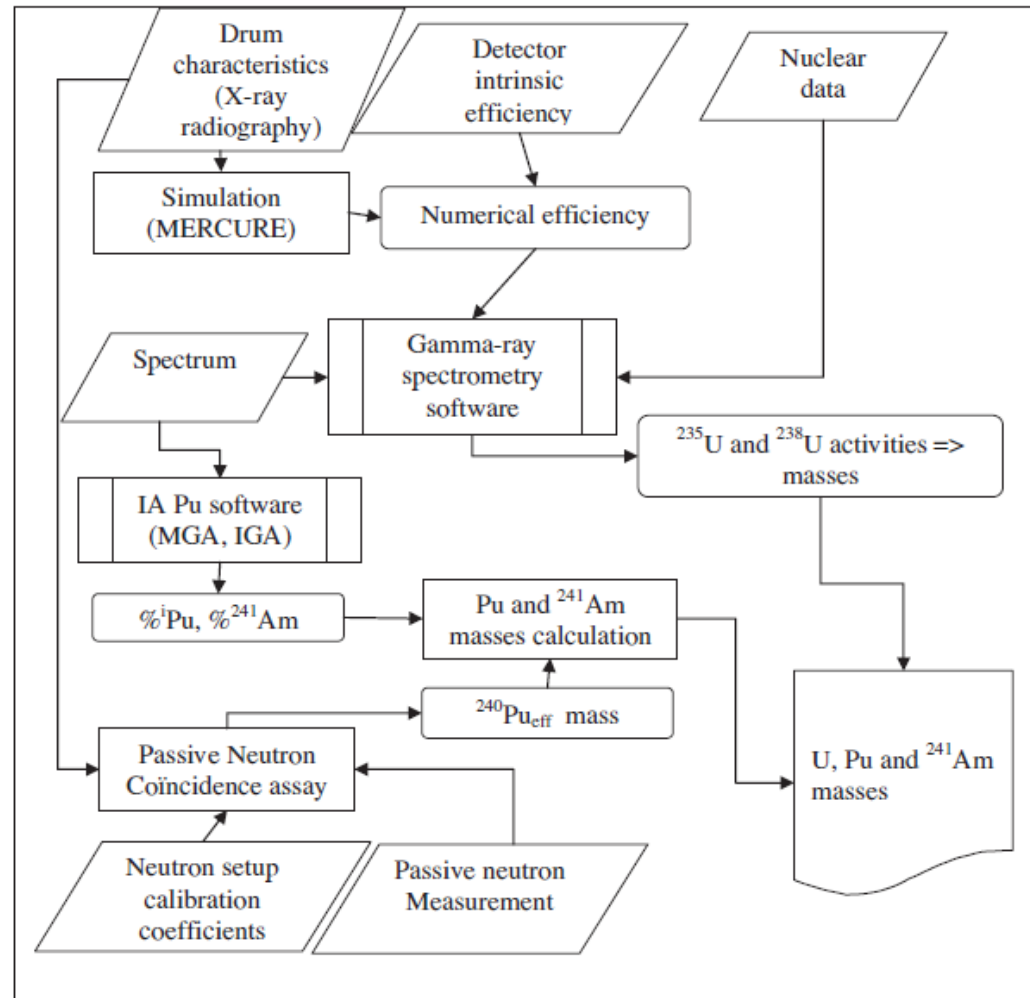
PEGAZE 100 L historic waste drums, CEA DEN CADARACHE



γ -ray Spectro.



Passive Neutron Coinc. Counting



F. Jallu et al. / Nuclear Instruments and Methods in Physics Research B 271 (2012) 48–54

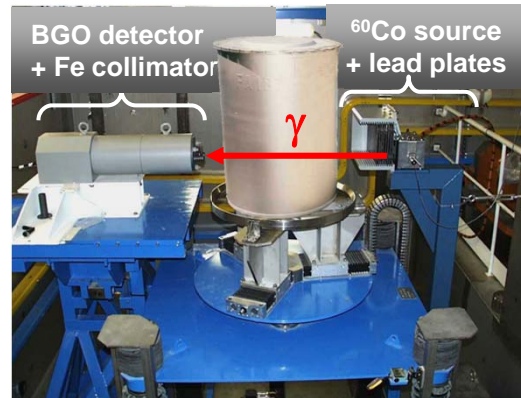
COMBINED MEASUREMENT SYSTEM

γ RADIOGRAPHY + γ SPECTROSCOPY + ACTIVE & PASSIVE NEUTRON CELL

> 60,000 historic 225 L bituminized waste drums (Marcoule, France) \rightarrow **Non Destructive Assay** performance study



Low resolution gamma radiography (^{60}Co)

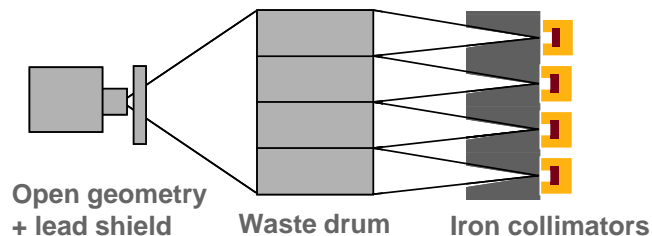


Gamma spectroscopy

$\Rightarrow \beta, \gamma$ radionuclides $\Rightarrow ^{241}\text{Am}$ α activity

Coaxial HP Ge
(100% rel. eff.)

Planar HP Ge + BGO
Compton suppression

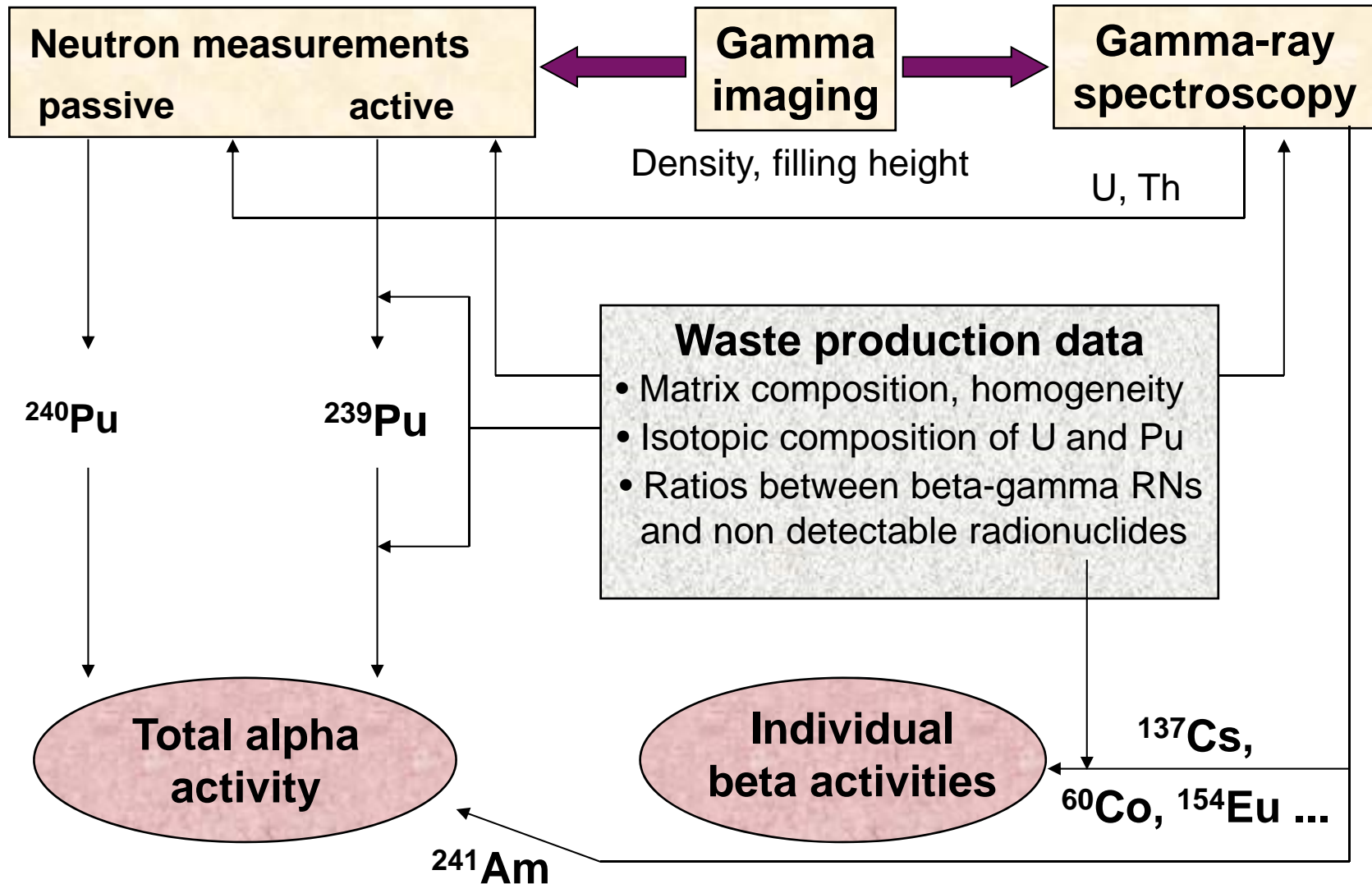


Passive & active neutron cell \Rightarrow Pu, U



- Graphite and polyethylene cell
- 99 ^3He counters + Pb and Cd shields
- Pulsed 2.10^9 n/s neutron generator

COUPLING MEASUREMENT RESULTS



PHOTON IMAGING

GAMMA SPECTROSCOPY

PASSIVE AND ACTIVE NEUTRON MEASUREMENTS

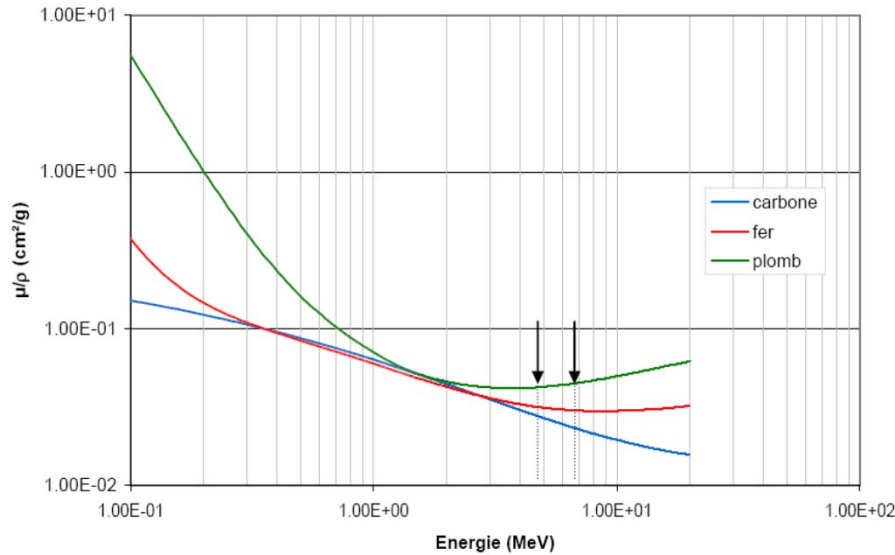
COMBINED MEASUREMENTS



A FEW R&D TOPICS

DUAL ENERGY IMAGING (SATURNE LINAC PROJECT)

Mass attenuation coefficient μ/ρ

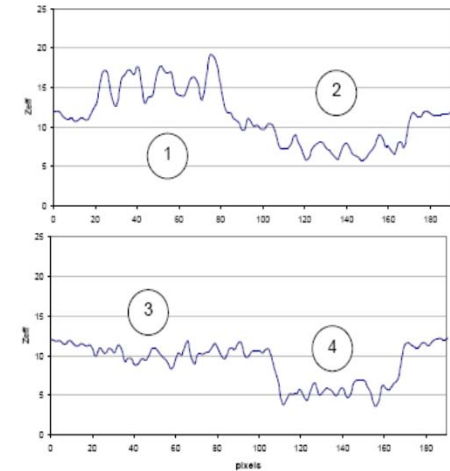
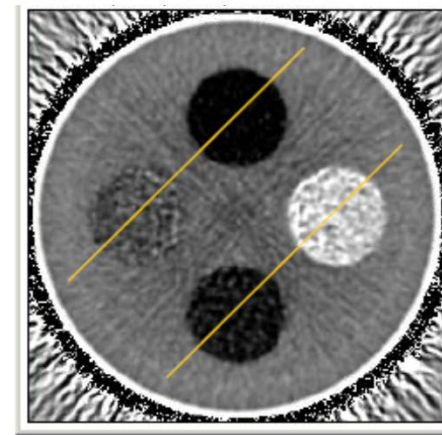


LINAC acquisitions at 15 MeV and 9 MeV

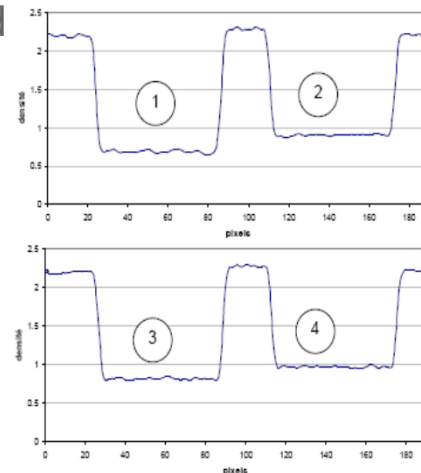
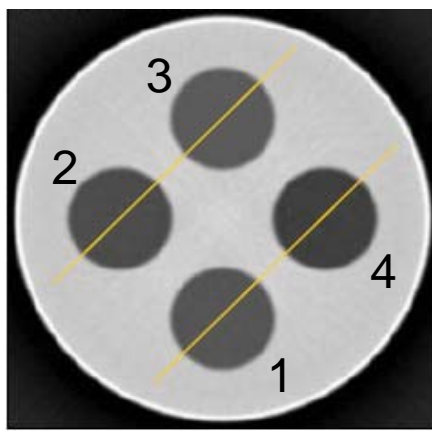
$$\mu_{15\text{MeV}} (\bar{E} \sim 6 \text{ MeV}) / \mu_{9\text{MeV}} (\bar{E} \sim 4 \text{ MeV})$$



Atomic number Z (simulation of a 870 L drum)



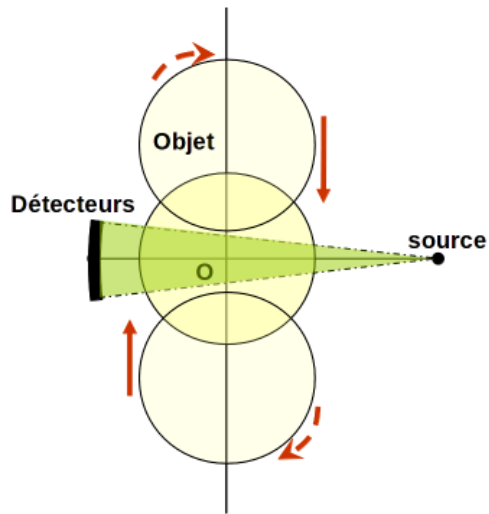
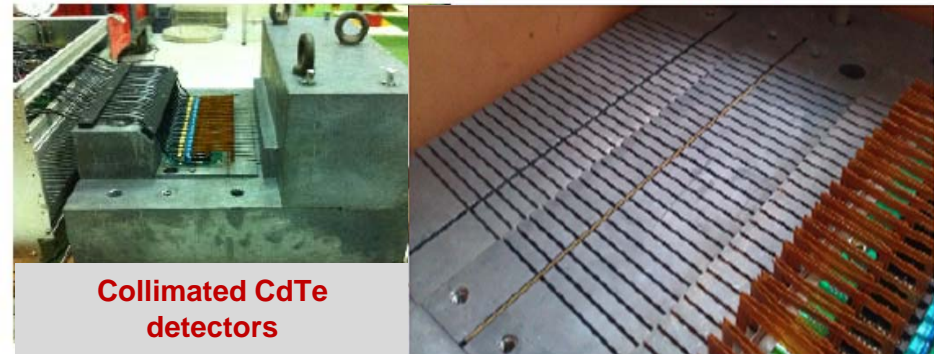
Density (simulation of a 870 L)



N° insert	matériau	Z_{eff} théorique	Z_{eff} calculé
1	PVC	12	$15.90 \pm 11.3\%$
2	Cellulose	6.68	$7.05 \pm 15\%$
3	plastique	9.3	$9.96 \pm 11.1\%$
4	CH ₂	5.28	$5.46 \pm 15.5\%$

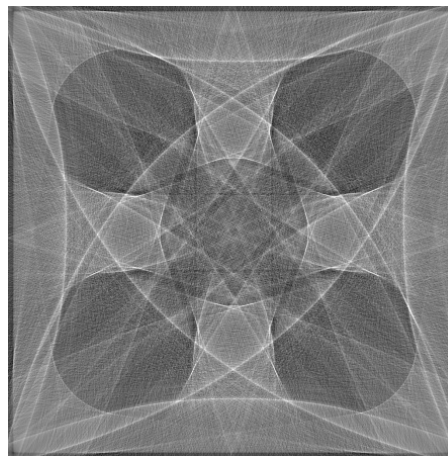
HIGH ENERGY X-RAY TOMODENSITOMETRY

- **Attenuation up to 5 decades (40 cm steel)**
- Radiography and 2D plane tomography
- 3D helicoidally tomography
- **0.5 to 2 mm spatial resolution**
- 1 h (2D) to several days (3D) acquisitions

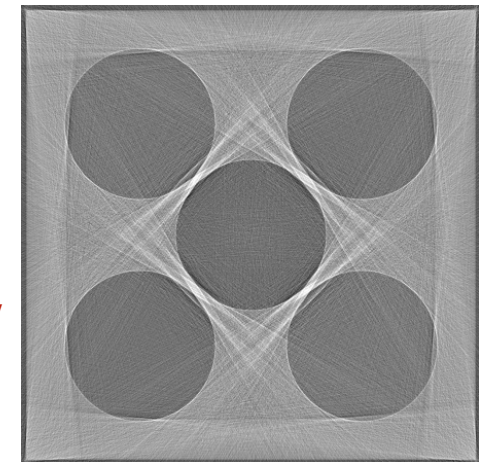


Simulations of a 5 m³ concrete waste package

**9 MeV
LINAC**



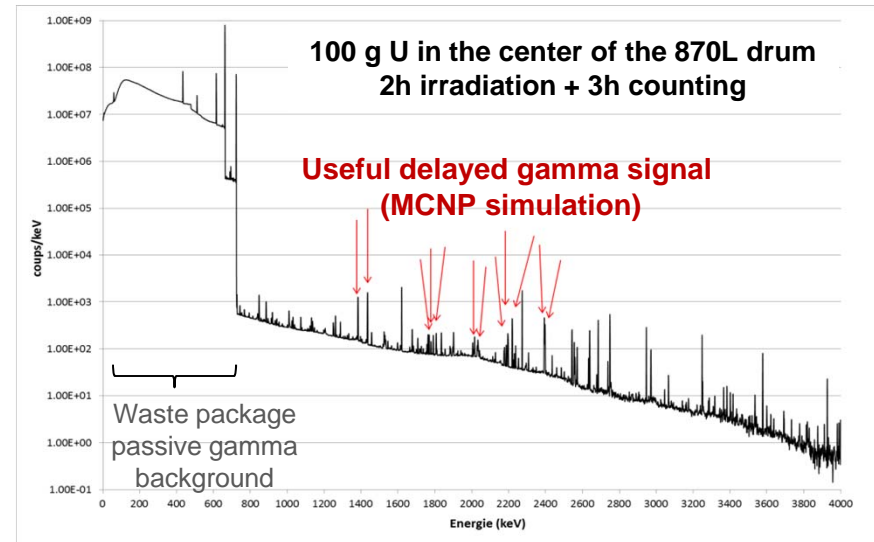
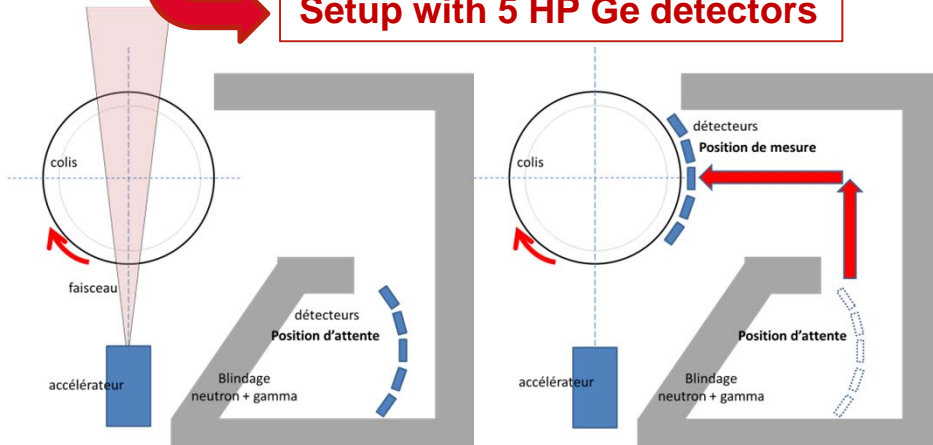
**15 MeV
LINAC**



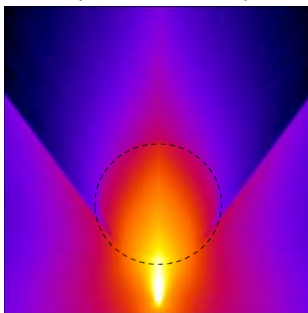
ACTIVE PHOTON INTERROGATION: PHOTOFISSION

- ❑ U and Pu measurement in **large concrete waste packages**
- ❑ **High energy X rays** produced by an electron LINAC (photofission threshold ~ 6 MeV)
- ❑ **Delayed neutron counting** between the pulses
- ❑ **Delayed gamma-ray spectroscopy** after stopping irradiation

Setup with 5 HP Ge detectors



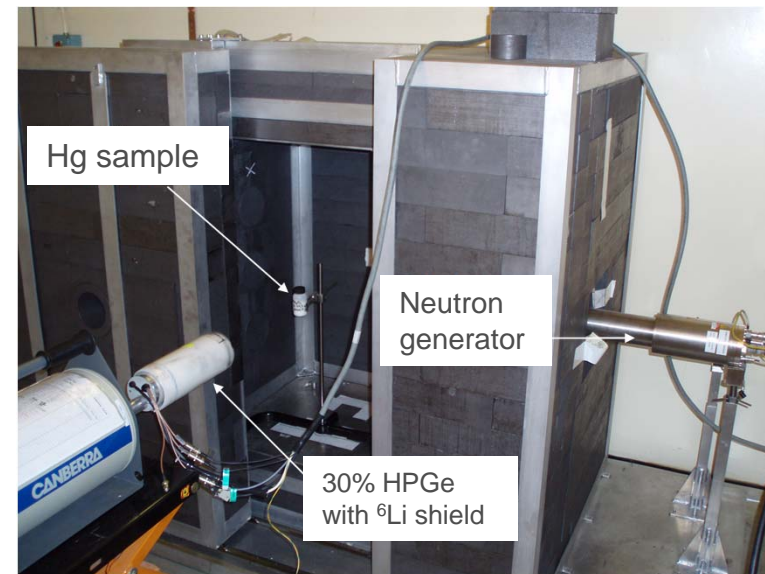
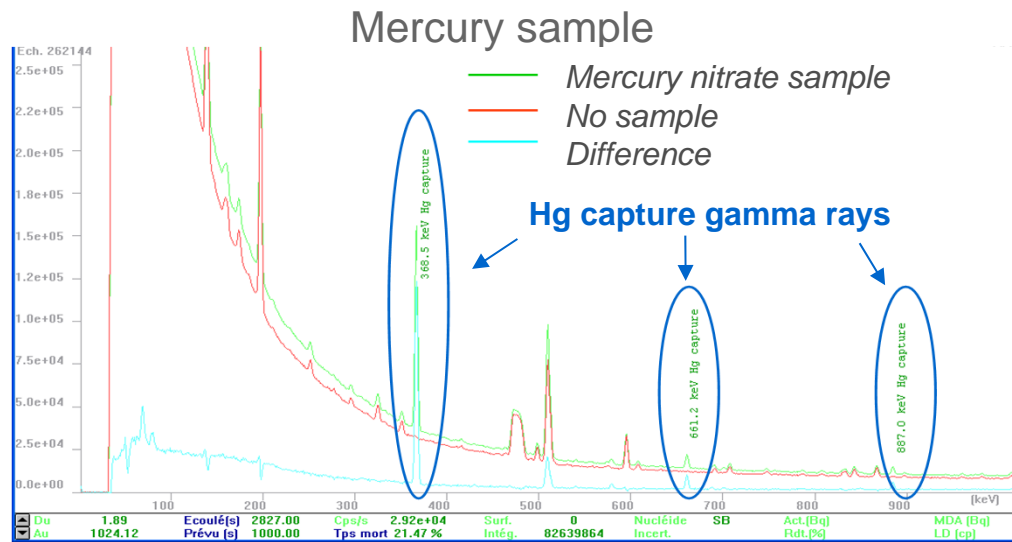
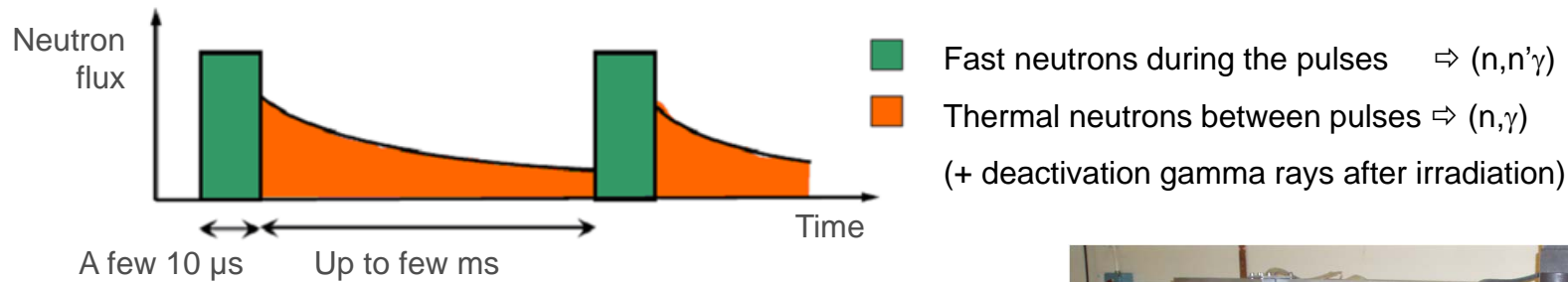
15 MV Linac beam
(870 L drum)



γ ray (keV)	U in the center			Background & detection limit - 1 detector			Backgr. & detection limit - 5 detector cluster		
	B (counts)	L_D (counts)	L_D (g ^{235}U)	B (counts)	L_D (counts)	L_D (g ^{235}U)	B (counts)	L_D (counts)	L_D (g ^{235}U)
1384	80	44	0,92	240	75	0,312	240	75	0,312
1436	116	53	0,84	580	115	0,36	580	115	0,36
1768	64	40	7,98	320	86	3,43	320	86	3,43
1791	64	40	7,98	320	86	3,43	320	86	3,43
1807	60	39	6,45	300	83	2,78	300	83	2,78
2016	96	48	9,66	480	105	4,18	480	105	4,18
2032	100	49	8,21	500	107	3,56	500	107	3,56
2176	34	30	9,94	170	63	4,22	170	63	4,22
2196	32	29	2,89	158	61	1,22	158	61	1,22
2218	34	30	1,74	168	63	0,74	168	63	0,74
2392	36	31	1,70	180	65	0,72	180	65	0,72
2398	36	31	2,35	180	65	1,00	180	65	1,00

NEUTRON ACTIVATION ANALYSIS

- ❑ **Toxic chemical or matrix material** characterization in radioactive wastes
- ❑ **PGNAA** Prompt Gamma Neutron Activation Analysis
- ❑ **Pulsed neutron generator + HP Ge gamma-ray detector**



REGAIN cell, CEA DEN CAD, LMN

J.-L. Ma et al., Prompt Gamma Neutron Activation Analysis of toxic elements in radioactive waste packages, Applied Radiation and Isotopes 70 (2012) 1261

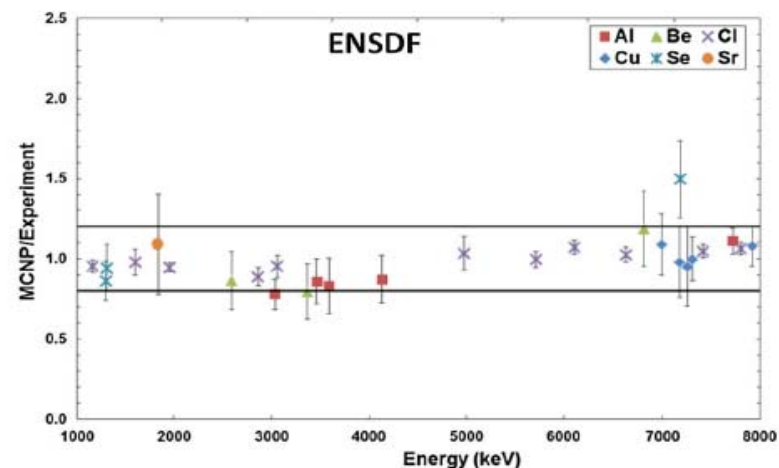
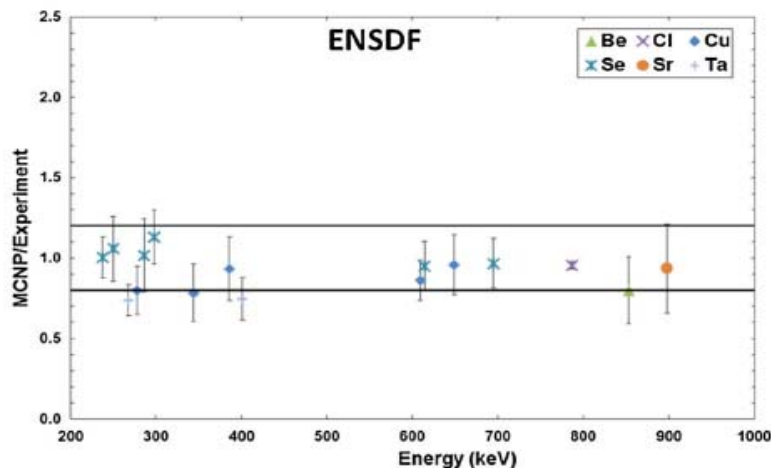
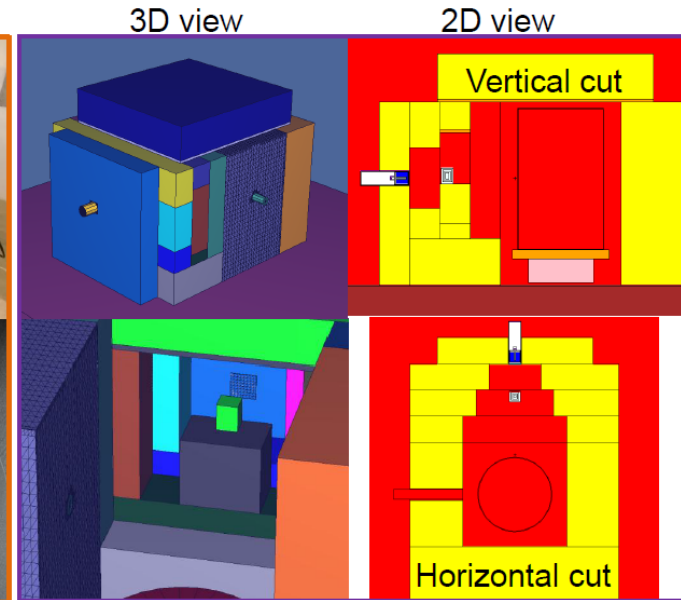
- ❑ **MEDINA PGNAA cell**
FZJ, Germany, GENIE 16 neutron generator and ~ 100% HP Ge
- ❑ **EXP / MCNP** comparisons
- ❑ **MCNP performance** optimization
- ❑ **Fission delayed γ** (see next slide)

POSTER Tangi NICOL – EFFMIN 4
tangi.nicol@cea.fr

Exp. setup

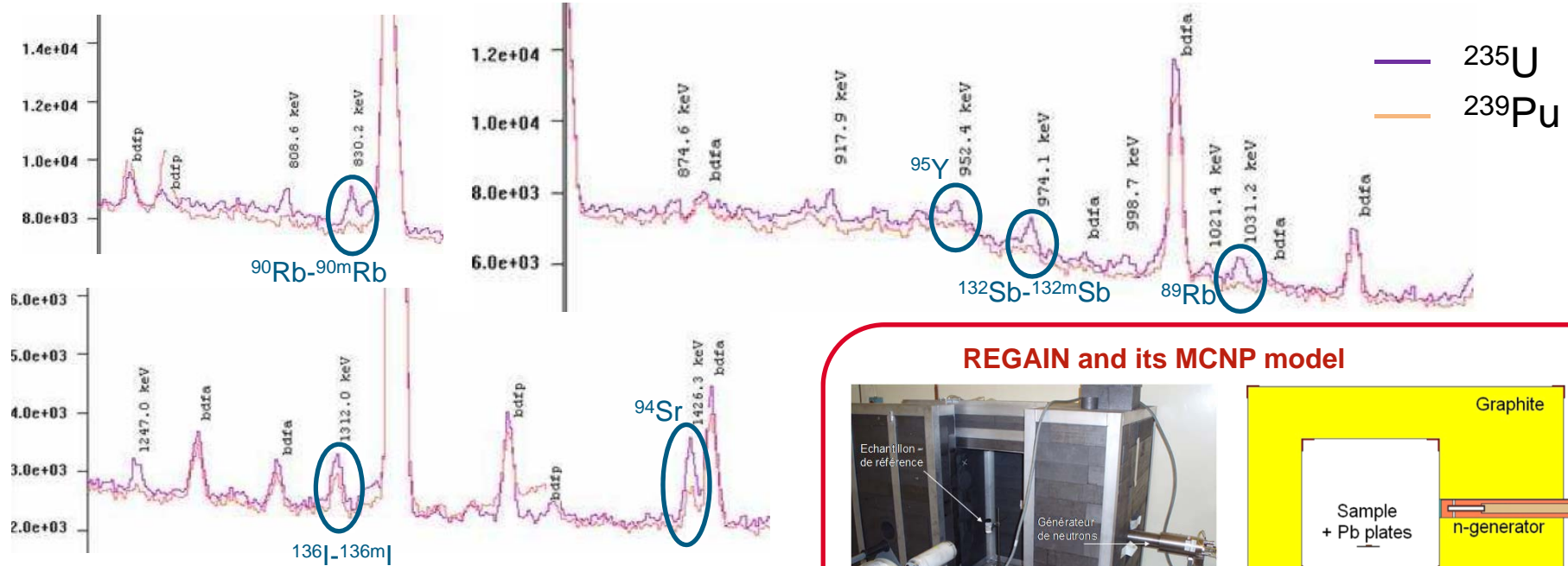


MCNPX model

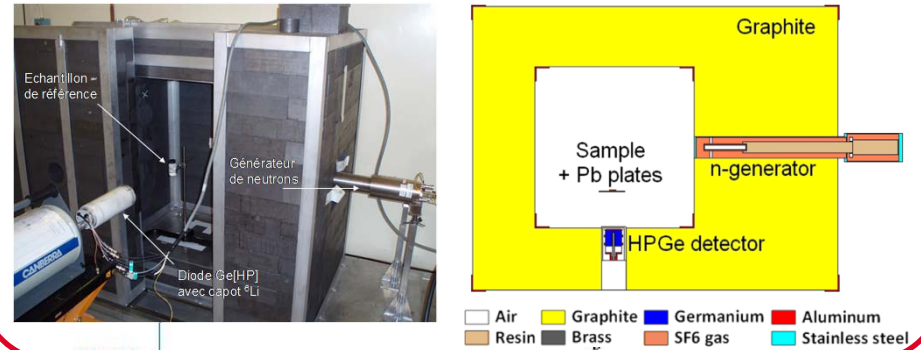


NEUTRON-INDUCED FISSION DELAYED γ -RAY YIELDS

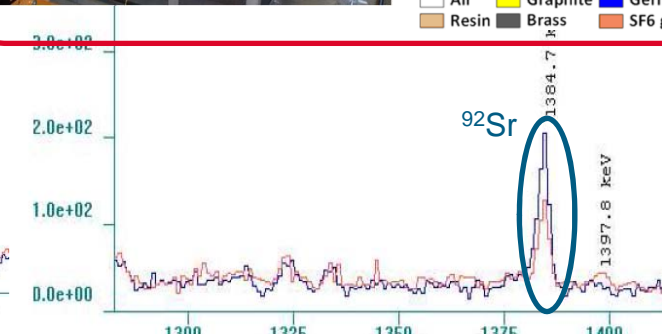
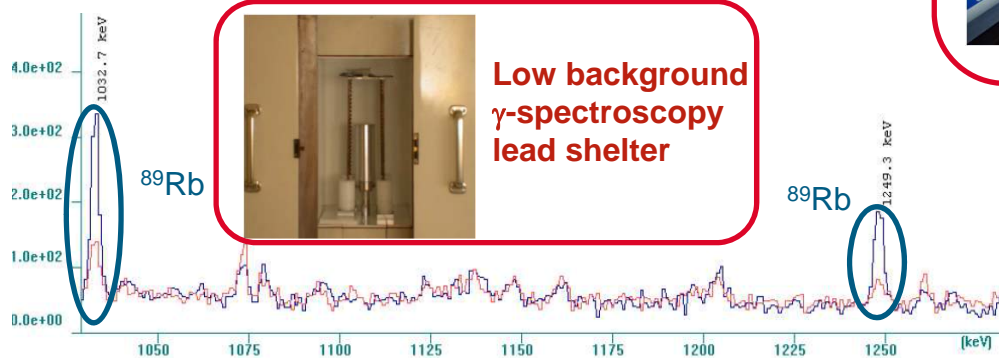
Gamma spectra of ^{235}U and ^{239}Pu samples measured in REGAIN between neutron pulses



REGAIN and its MCNP model

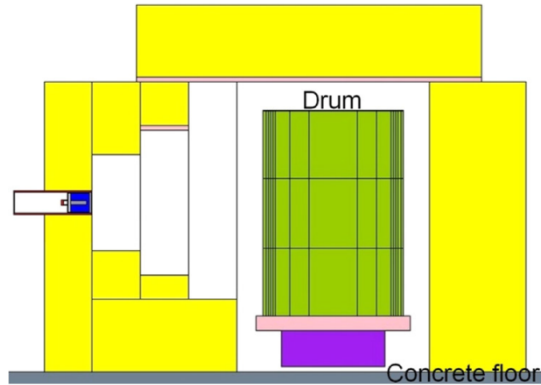


Gamma spectra after irradiation

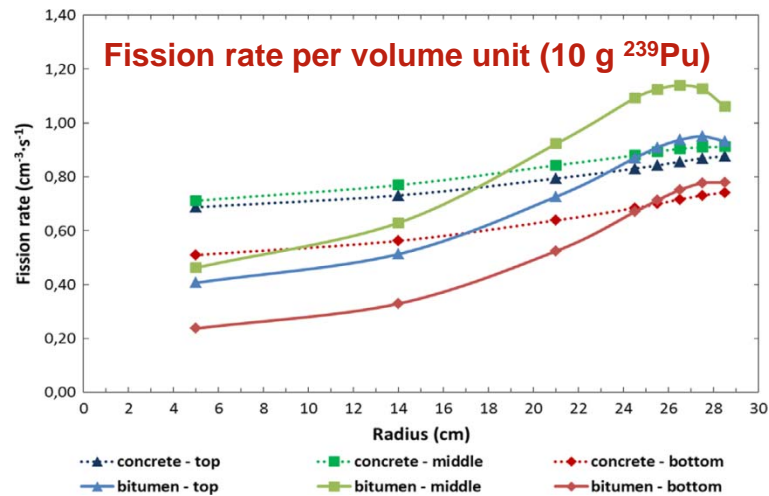


FISSION DELAYED γ -RAY DETECTION LIMITS

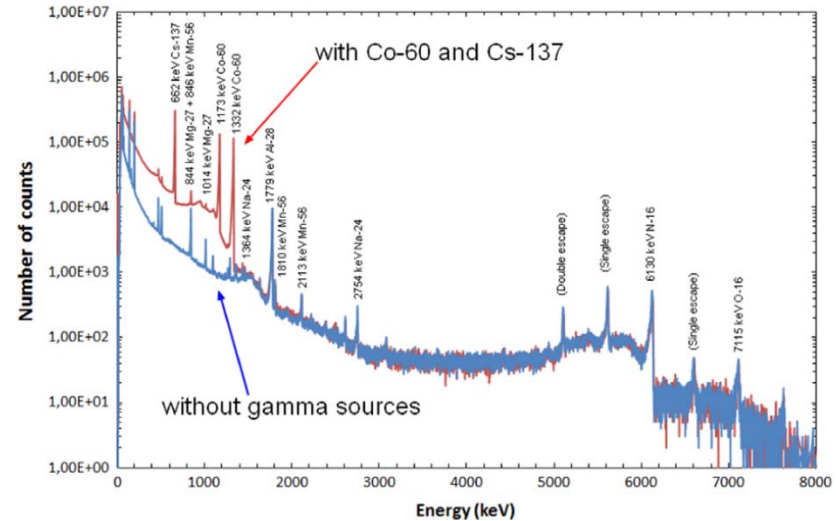
MCNP calculations (MEDINA, 225 L drums)



Air
 Graphite
 Concrete
 Germanium
 Iron
 Carbon-Fiber Reinforced Polymers
 Aluminum
 Matrix (concrete, bituminized, ...)



Active background (MEDINA, 225 L concrete mockup drum, measurement between pulses)

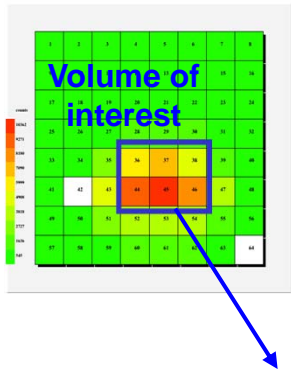


Detection limits in a 225 L concrete drum, with a $2 \cdot 10^8$ n/s emission, 7200 s irradiation, 900 s post irradiation counting

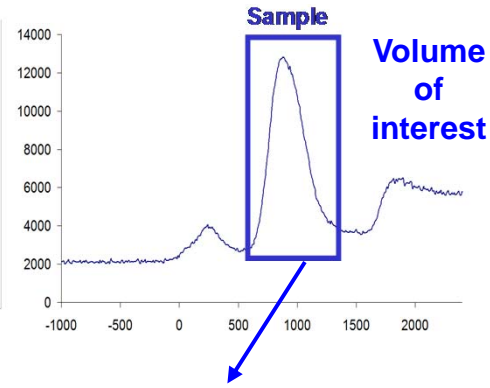
E (keV)	Precursor	$T_{1/2}$ (s)	DL ^{239}Pu (g)	DL ^{235}U (g)
830	^{90}Rb - $^{90\text{m}}\text{Rb}$	156-258	154.2	71.3
952	^{95}Y	618	192.2	197.2
973	^{132}Sb - $^{132\text{m}}\text{Sb}$	168-252	66.3	86.7
1032	^{89}Rb	924	287.3	202.3
1312	$^{136\text{I}}$ - $^{136\text{m}}\text{I}$	84-47	67.9	84.2
1384	^{92}Sr	9756	24.1	19.0
1427	^{94}Sr	75	21.5	18.7
1614	^{104}Tc - ^{134}I	1092-3156	54.8	108.5

THE ASSOCIATED PARTICLE TECHNIQUE (APT)

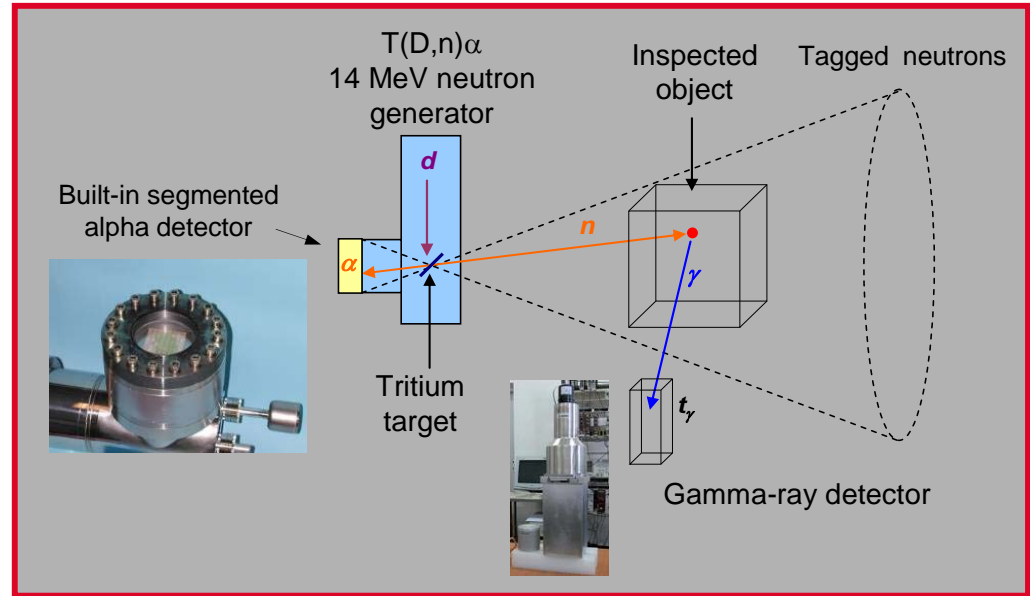
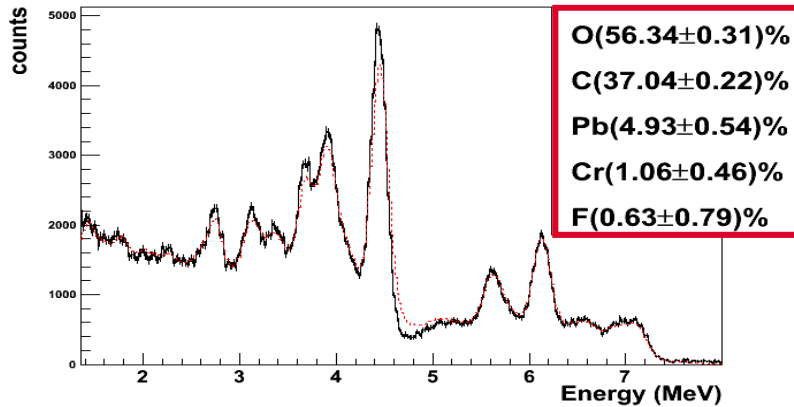
Alpha detector
(position sensitive)



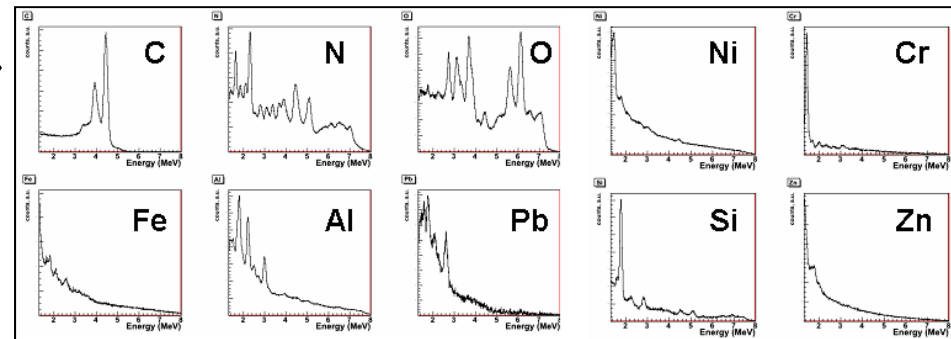
Neutron TOF
(alpha-gamma coincidence)

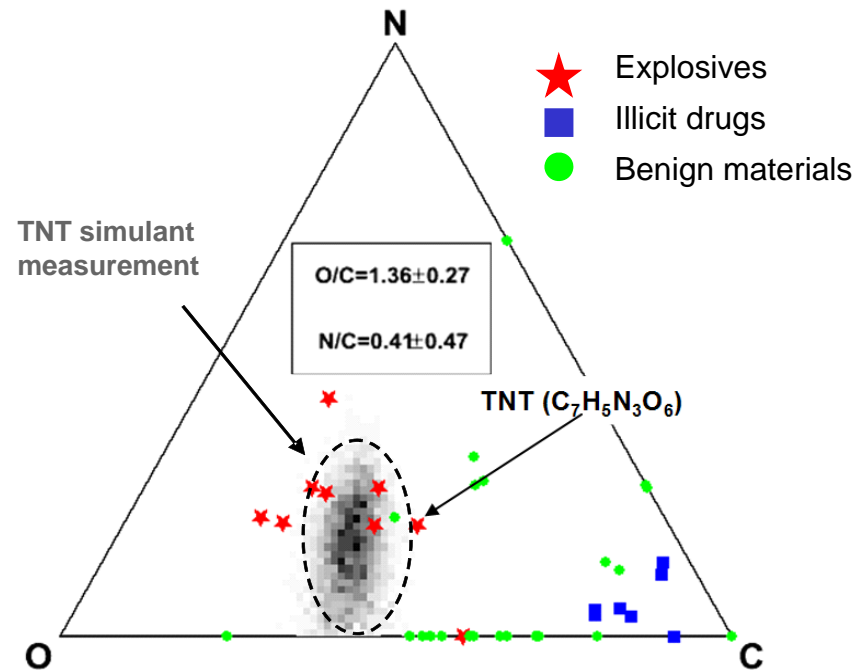
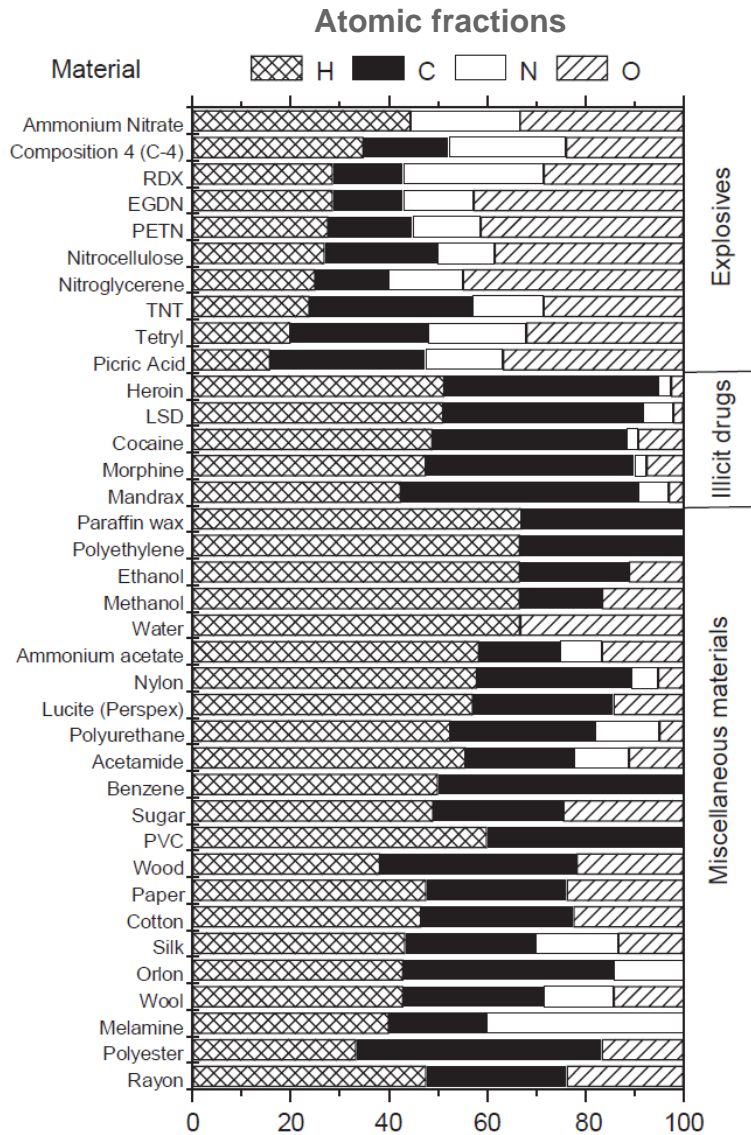


Gamma spectrum (selected volume)

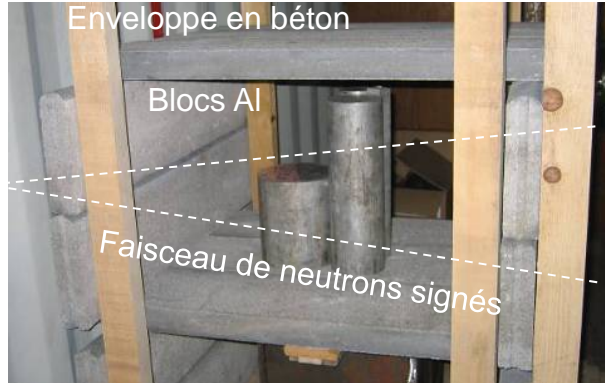


Unfolding (with a calibration database)

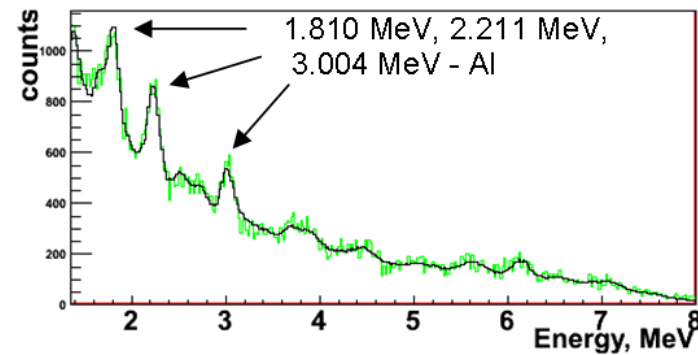




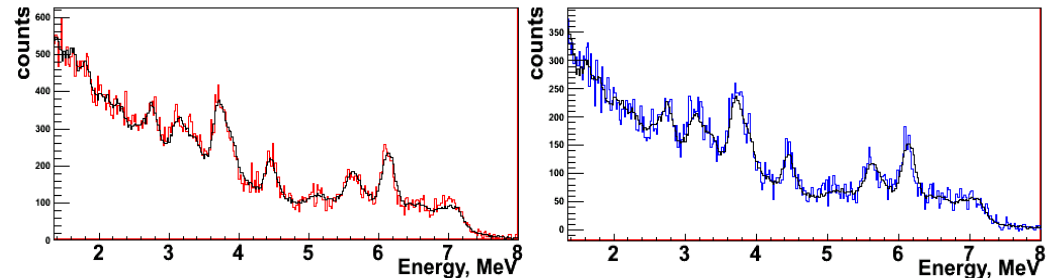
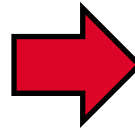
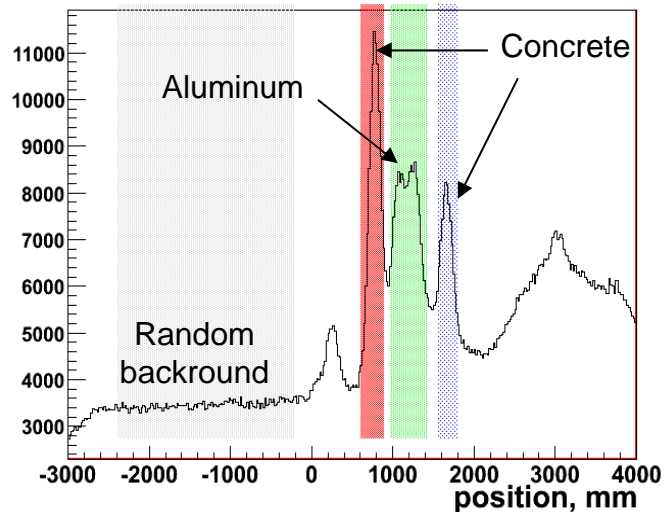
□ Feasibility study on non-radioactive materials (experiments)



Gamma spectra for different TOF windows

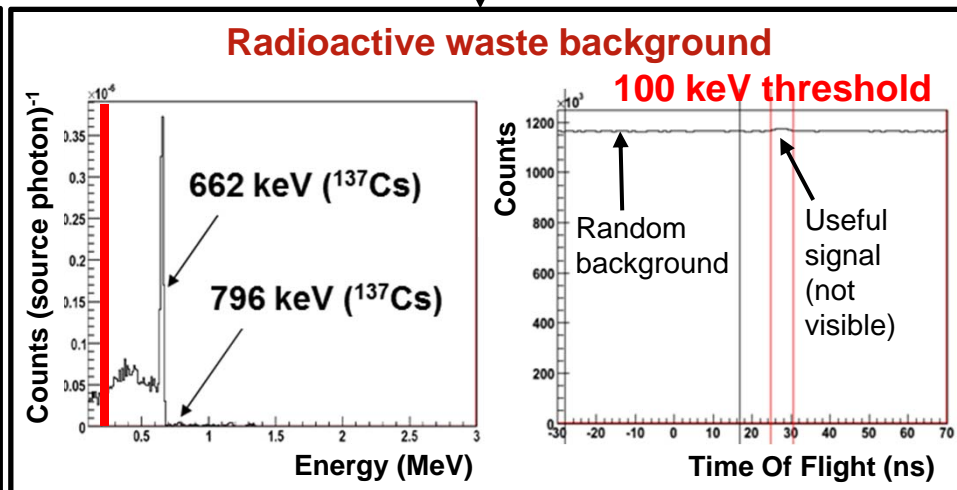
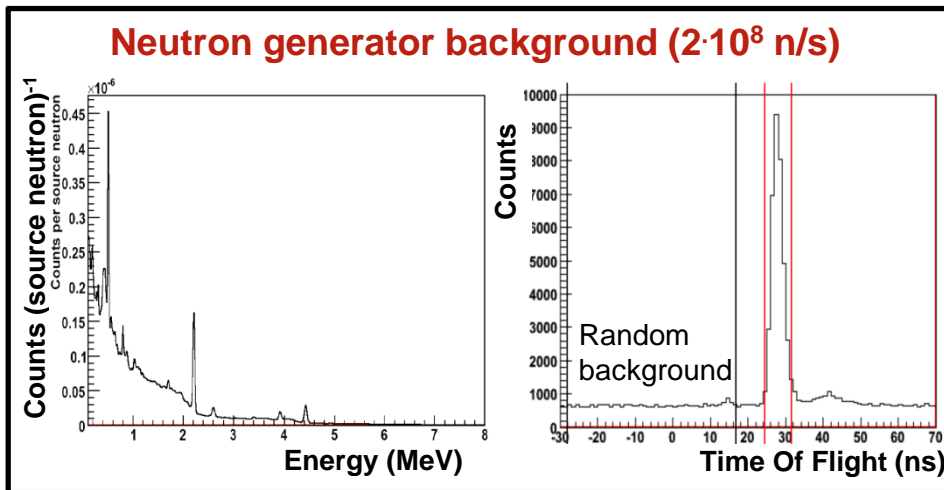
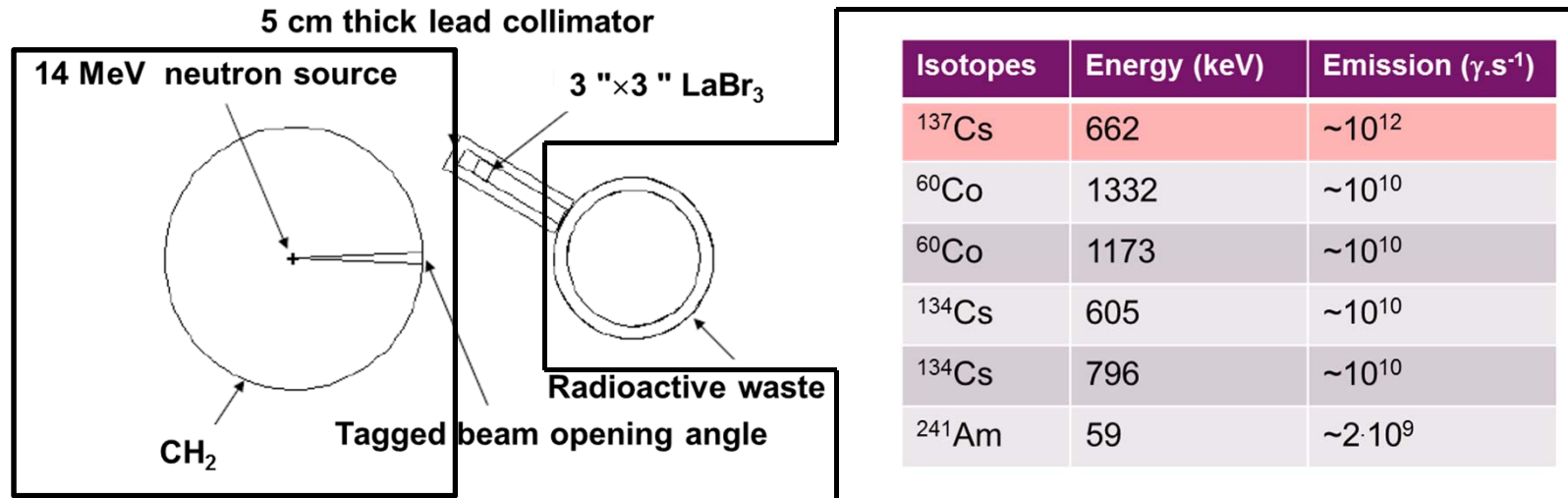


Time-of-flight (TOF) spectrum



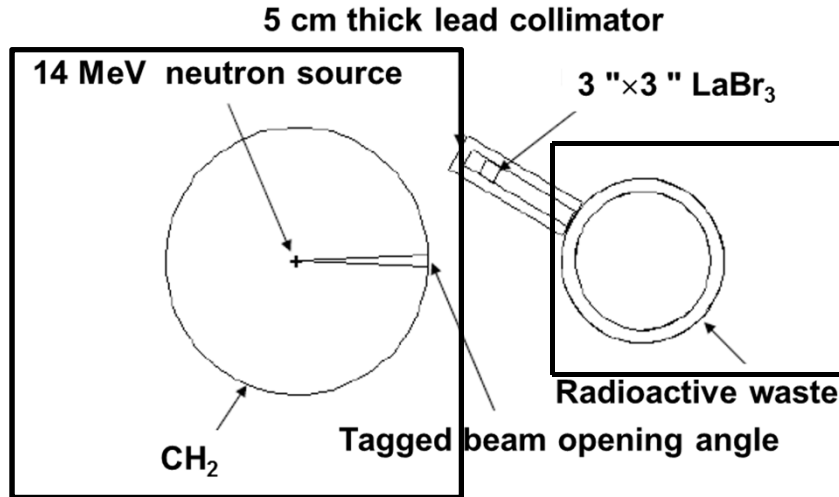
PERFORMANCES FOR RADIOACTIVE WASTE PACKAGES

MCNP simulations for 225 L bituminized waste drums (~ 1 TBq ¹³⁷Cs)

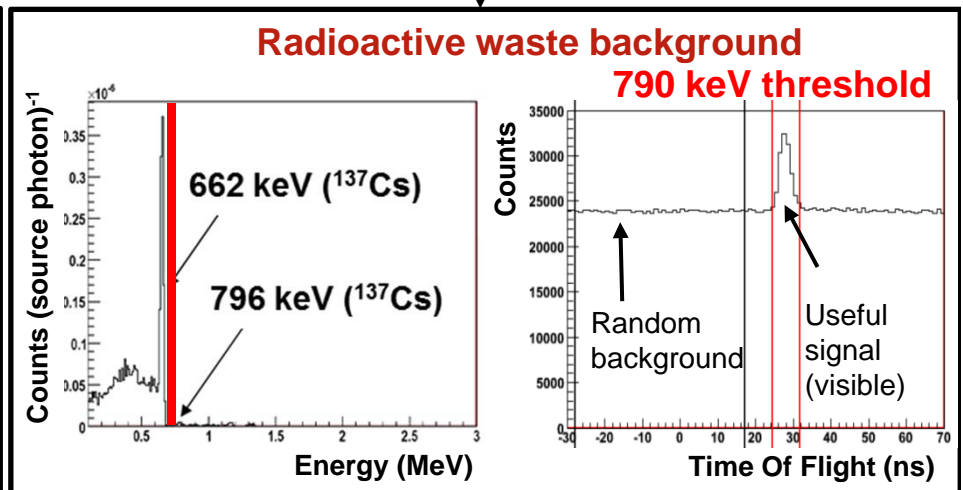
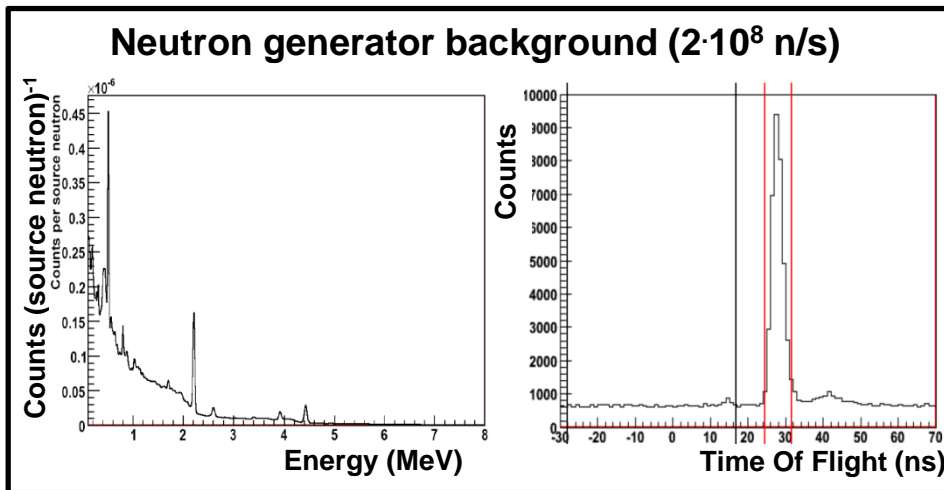


cea PERFORMANCES FOR RADIOACTIVE WASTE PACKAGES

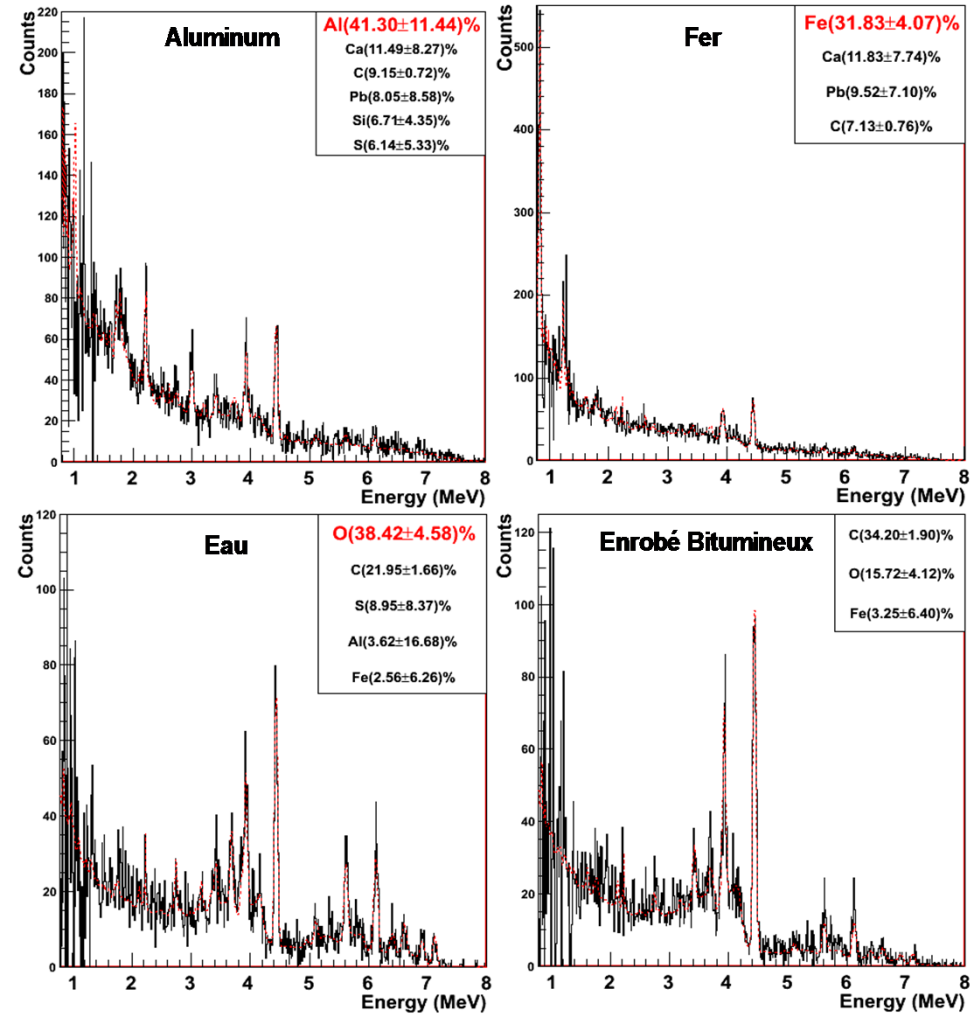
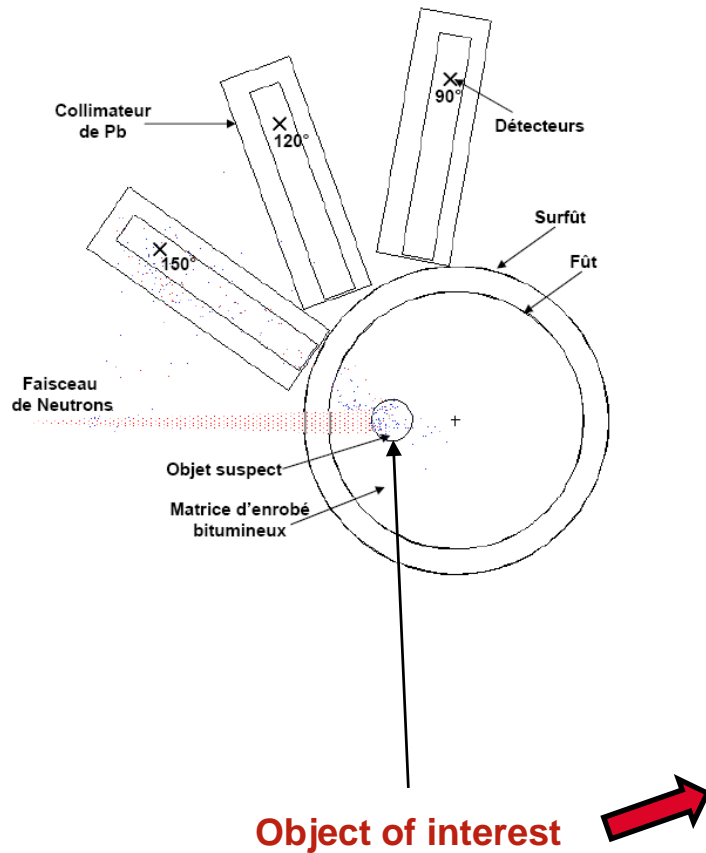
MCNP simulations for 225 L bituminized waste drums (~ 1 TBq ¹³⁷Cs)



Isotopes	Energy (keV)	Emission (γ.s ⁻¹)
¹³⁷ Cs	662	~10 ¹²
⁶⁰ Co	1332	~10 ¹⁰
⁶⁰ Co	1173	~10 ¹⁰
¹³⁴ Cs	605	~10 ¹⁰
¹³⁴ Cs	796	~10 ¹⁰
²⁴¹ Am	59	~2·10 ⁹

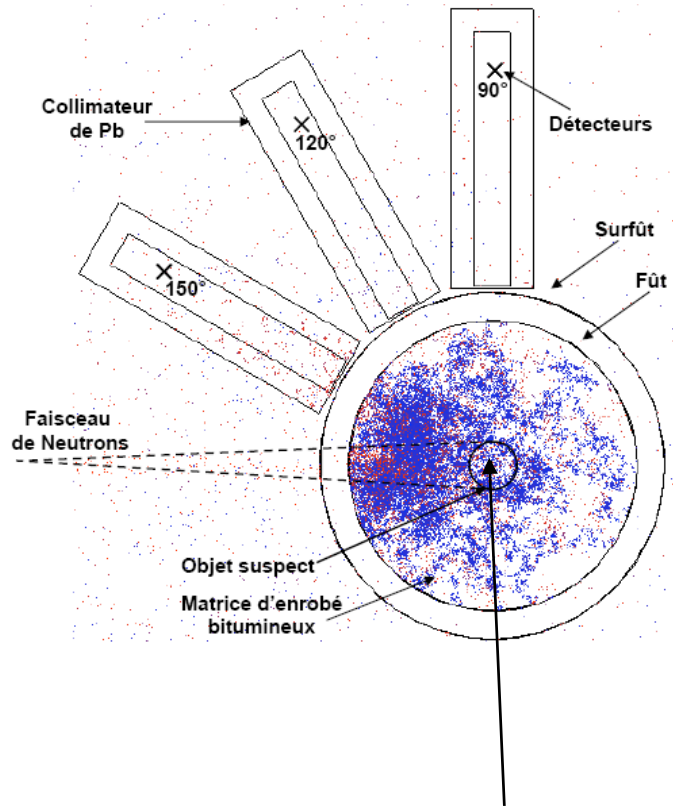


SIMULATION OF OBJECTS IN THE 225 L BITUMINIZED DRUM



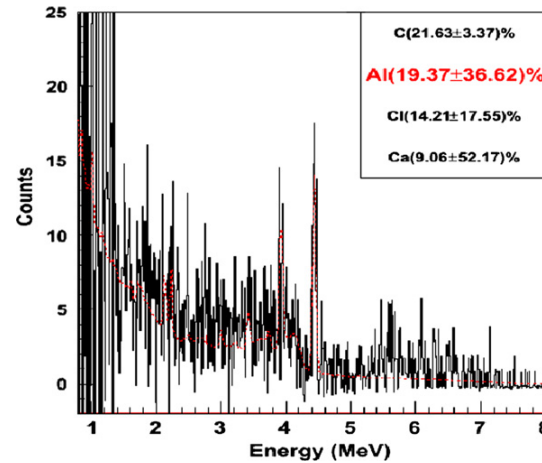
W. El Kanawati, B. Perot, C. Carasco, Monte Carlo Simulation of High-Level Radioactive Waste Characterization with the Associated Particle Technique, Nuclear Instruments and Methods in Physics Research A 705 (2013) 61–73.

OBJECT IN THE CENTER OF THE BITUMINIZED DRUM (MCNP)

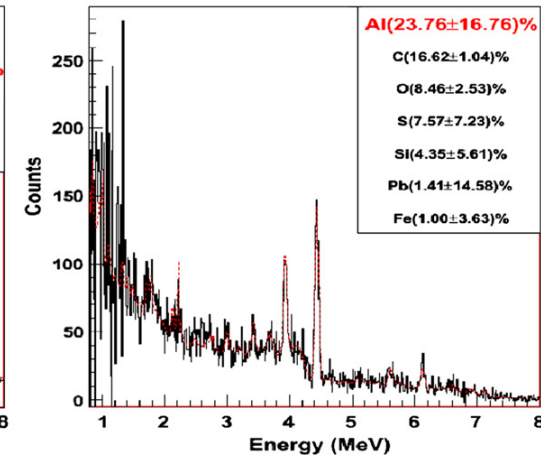


Object of interest →

1 detector, 60 min →



10 times more counts
(10 detectors or 600 min)



W. El Kanawati, B. Perot, C. Carasco, Monte Carlo Simulation of High-Level Radioactive Waste Characterization with the Associated Particle Technique, Nuclear Instruments and Methods in Physics Research A 705 (2013) 61–73.

SCINTILLATORS TO REPLACE ^3He DETECTORS

^3He proportional counters



GOOD EFFICIENCY



EXPENSIVE (since 09/11/2001)



SLOW RESPONSE ($\sim \mu\text{s}$)
(thermalization needed)



UNSENSITIVE TO γ RAYS AND CROSS TALK

Plastic Scintillators



GOOD EFFICIENCY



CHEAP (\sim factor 5 wrt. ^3He)



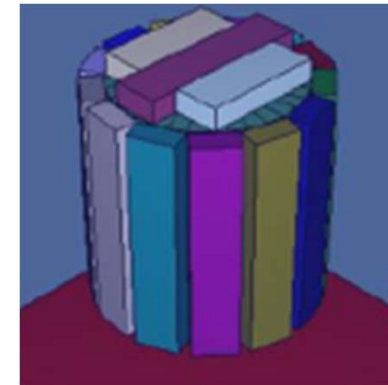
FAST RESPONSE ($\sim \text{ns}$)
(recoil proton)



VERY SENSITIVE TO γ RAYS AND CROSS-TALK

Passive neutron coincidence counting for radioactive waste drums - MCNP Polimi simulation

- 118 L waste drums
- 10 cm thick plastic detectors (with lead shields)
- ~ 0.5 g of $^{240}\text{Pu}_{\text{eq}}$
- 1500 s acquisition time



0.2 g/cm³ organic matrix

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

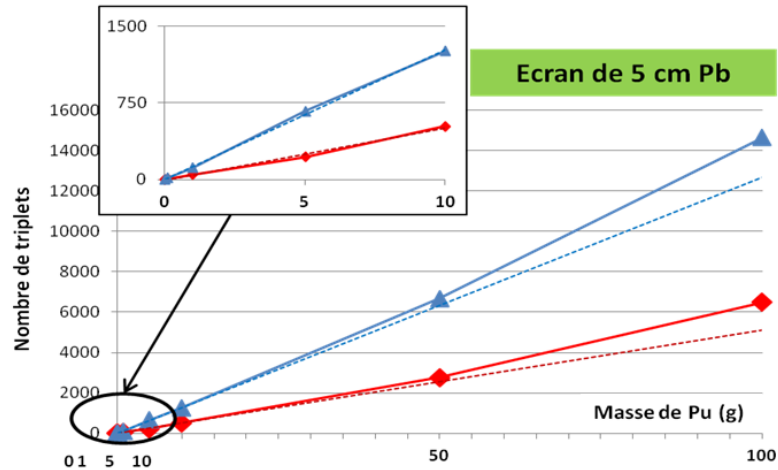
→ Triples (multiplicity 2 coincidences)

0.5 g/cm³ metallic matrix

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

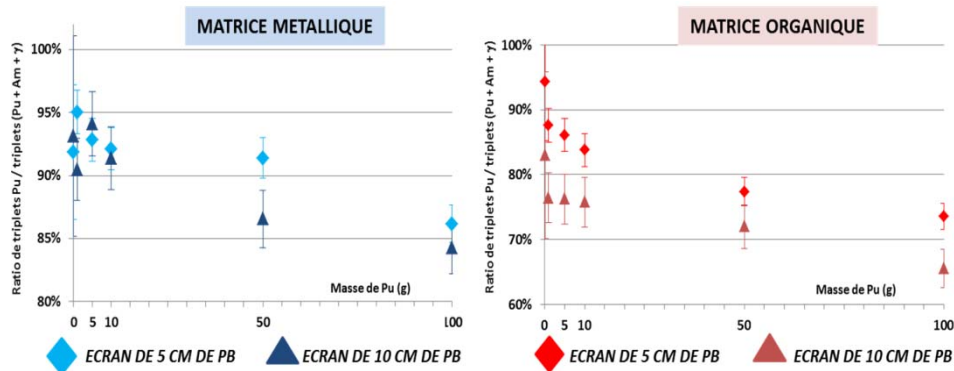
SIMULATED PERFORMANCES (TRIPLES)

❑ Few accidental coincidences up to 10 g of Pu

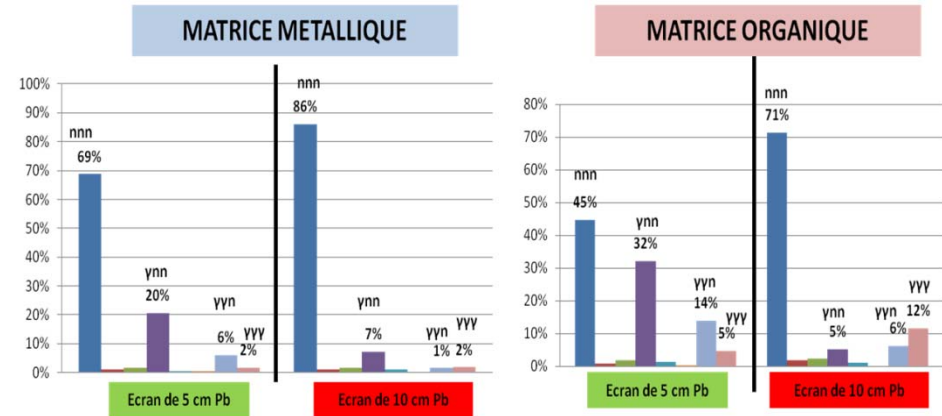


— MATRICE METALLIQUE } AVEC mélange temporel des impulsions
— MATRICE ORGANIQUE } AVEC mélange temporel des impulsions
- - - ALLURE LINEAIRE SANS } SANS mélange temporel des impulsions
- - - COÏNCIDENCES ACCIDENTELLES } SANS mélange temporel des impulsions

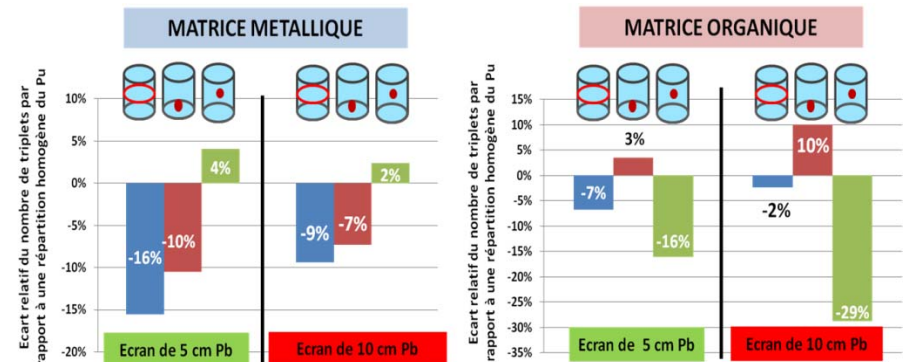
❑ Good signal to noise ratio (Pu/Total)



❑ Neutron vs. gamma events in the triples



❑ Plutonium localization effect



POSTER Benoît SIMONY – EFFMIN 4
benoit.simony@cea.fr

■ Radioactive and nuclear material characterization

- Gamma-ray spectroscopy
- Passive neutron counting
- Neutron and photon interrogation

■ Physical and elemental characterization

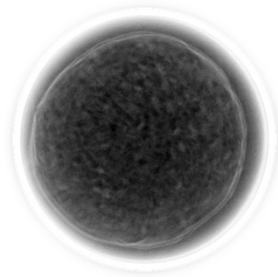
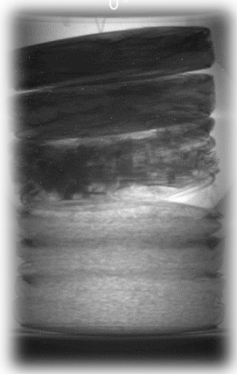
- Imaging techniques: gamma, X-ray
- Neutron activation analysis

■ NDA for D&D and radioactive waste characterization

- Complementary to sampling & analytic methods
- Sensitivity, precision, accuracy Vs. complexity, acquisition time, cost



⇒ The “dream team” = historic operators + NDA experts + D&D operators



EFFMIN4 | MARSEILLE | 18-21 JUILLET 2016

THANK YOU !

bertrand.perot@cea.fr

CEA International Expert



Commissariat à l'énergie atomique et aux énergies alternatives
Centre de Cadarache | F-13108 Saint-Paul-lez-Durance FRANCE
T. +33 (0)1 4 42 25 66 43 | Laboratoire de Mesures Nucléaires

Etablissement public à caractère industriel et commercial | RCS Paris B 775 685 019

Direction de l'Energie Nucléaire
Département de Technologie Nucléaire
Service Mesures et Modélisation des
Transferts et des Accidents Graves