Detection of uranium contamination in the process of dismantling and decommissioning nuclear facilities

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- Photonic Systems Laboratory (DRT/LETI/DOPT/SISP/LASP)
- Laboratory for System Design and Modelling (DES/DER/SESI/LEMS)

Summary

Context and objectives

- Nuclear decommissioning in France
- Gaseous Diffusion Plant (UDG – Tricastin Nuclear Site)
- Technical challenges and chosen detection methods

Contamination detection modelling

- Beta particles detection
- Gamma rays detection
- Alpha particles detection
- Estimation of acquisition times and conclusions

In-situ measurements at the UDG enrichment facility

- Detectors and inspected zones
- Method for surface activity estimation
- Results and comparison between methods

Conclusions and perspectives

1. Context and objectives

Nuclear decommissioning in France

35 facilities awaiting or undergoing decommissioning in France

Reasons for shutdown :

- Aging components (facilities built in the 1960s with an estimated 40 years of service life)
- Political decisions (nuclear power phase-out)

A lengthy process :

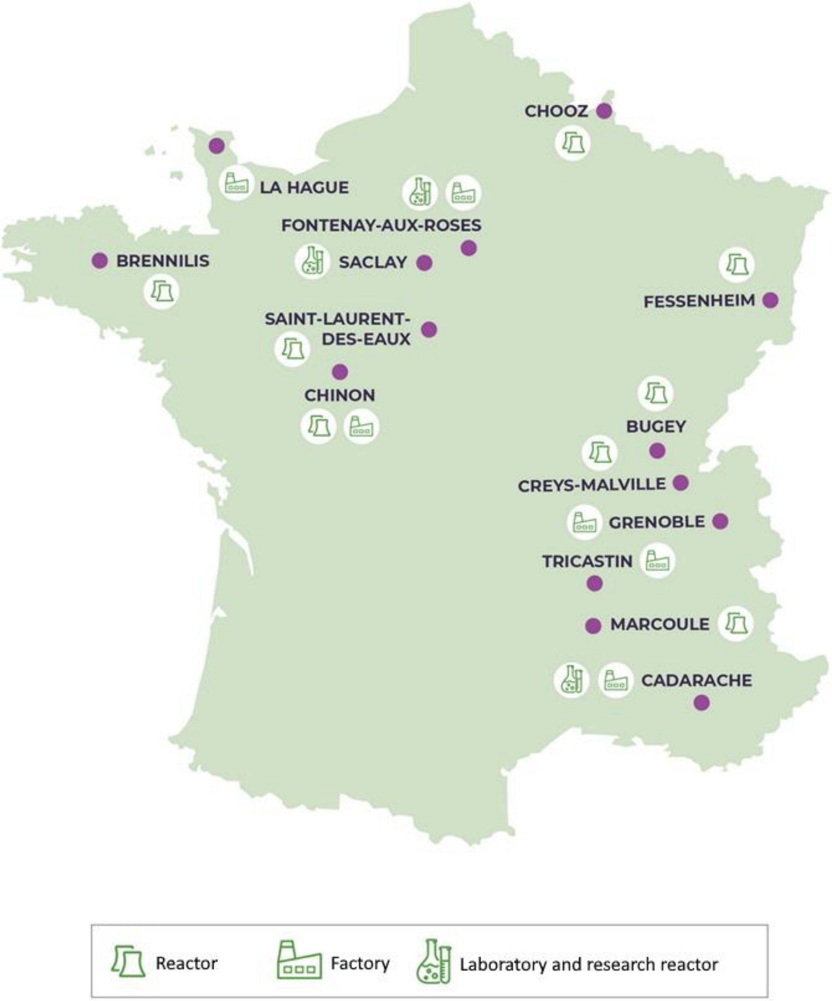
- 15 years to dismantle a Pressurized Water Reactor (PWR)
- 10 years for an enrichment facility

A costly procedure :

- 350-400 million euros to dismantle a PWR

Technical challenges may extend decommissioning time depending on the facility

A critical need to develop new technologies aiming to reduce times and costs of the decommissioning process arises



Map of shutdown nuclear facilities in France
(source : French Nuclear Safety Authority)

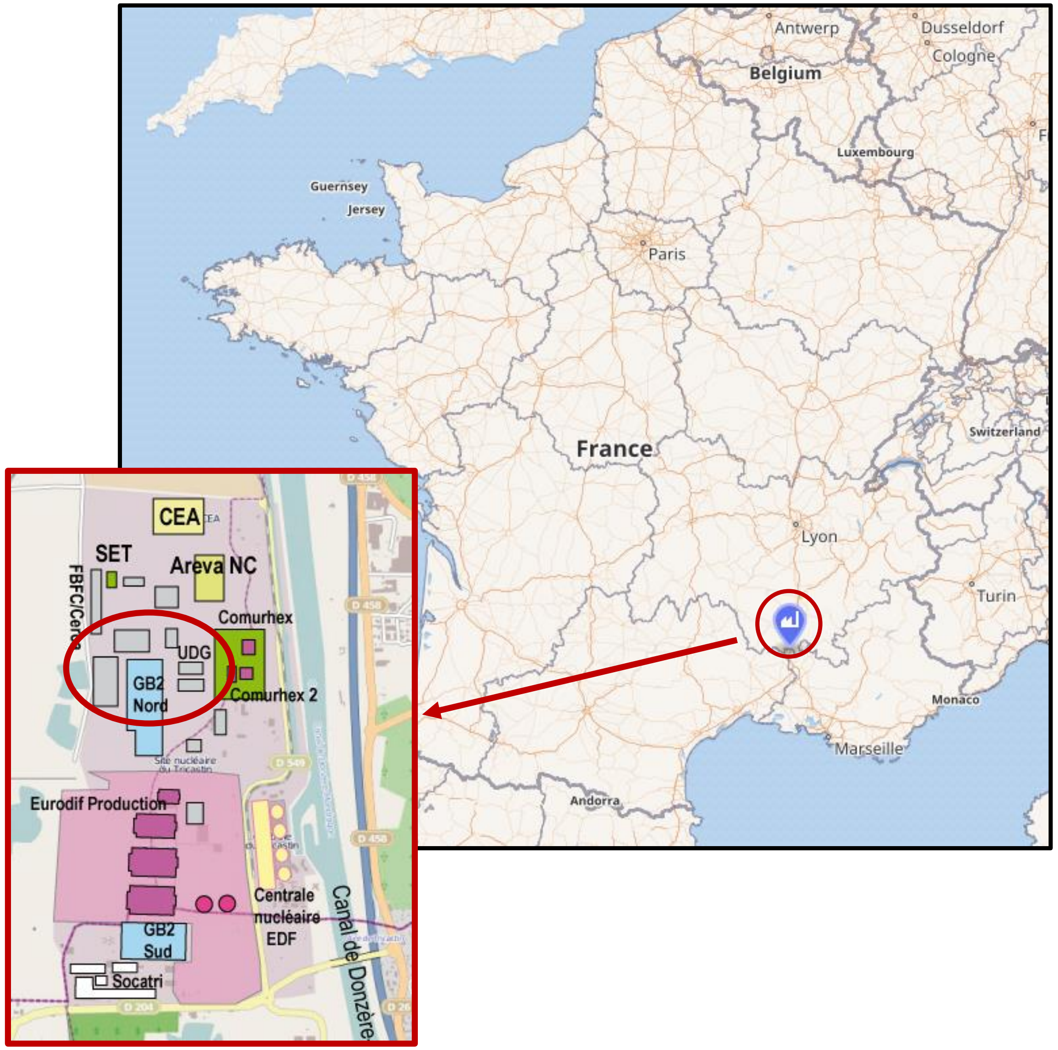
The Gaseous Diffusion Plant (UDG) - Tricastin Nuclear Site

Legacy uranium enrichment facility
→ increase of ^{235}U proportion via gaseous diffusion process

Characteristics of the UDG enrichment facility

- Lifetime = 40 years
- Total surface ≈ 700 000 m^2
- 4 plants
  - Low enrichment plant (LP) > 0,8 % ^{235}U
  - Medium enrichment plant (MP) > 1 % ^{235}U
  - High enrichment plant (HP) > 3 % ^{235}U
  - Very high enrichment plant (VHP1, VHP2, VHP3) > 20 % ^{235}U

Age of the facility and enrichment level play an important role on the type of particles emitted



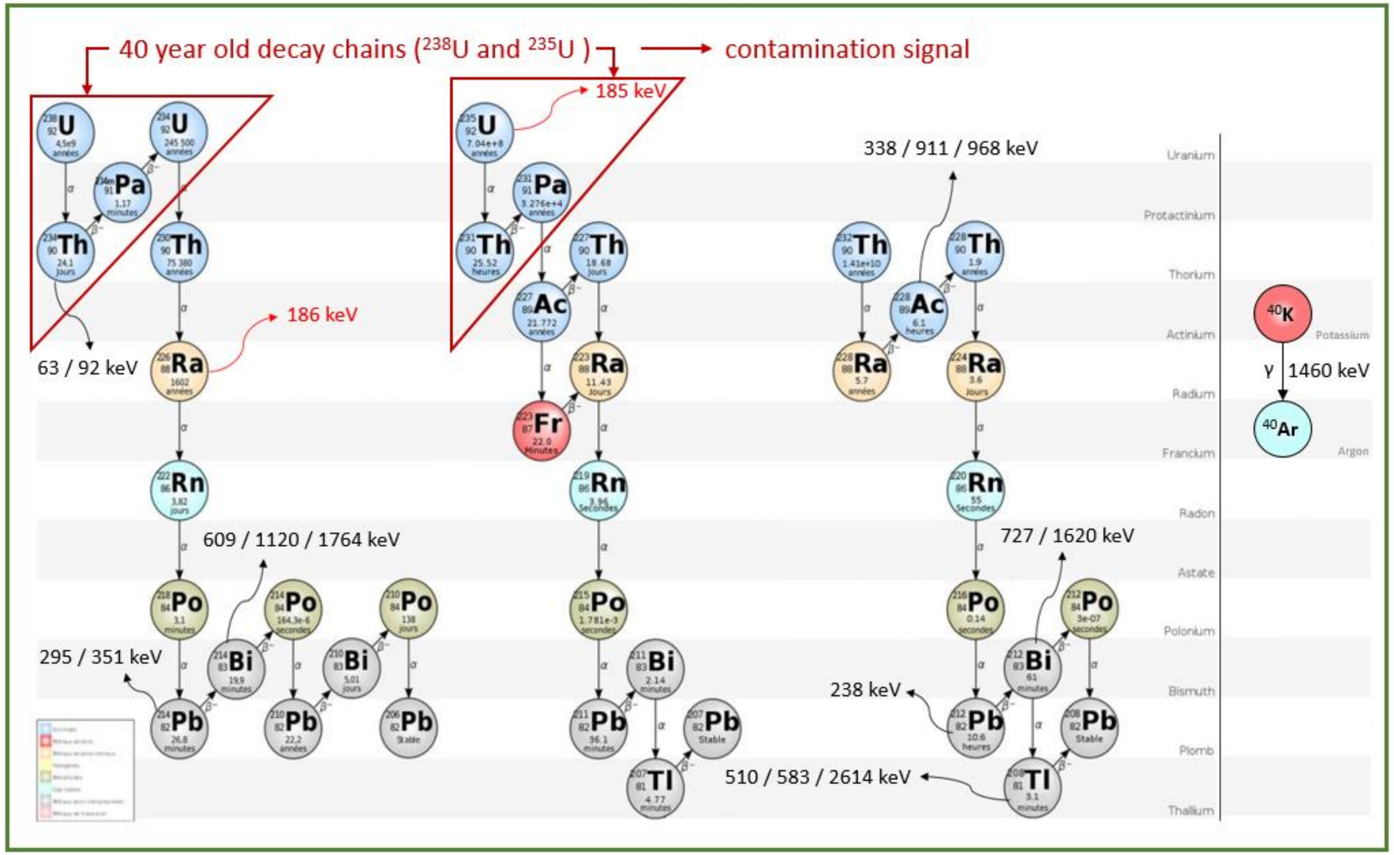
UDG and Tricastin nuclear site location in France

Technical challenges and chosen detection methods

Two main challenges

Discriminate the contamination signal from the background noise

Background noise



^{238}U chain ^{235}U chain ^{232}Th chain ^{40}K chain

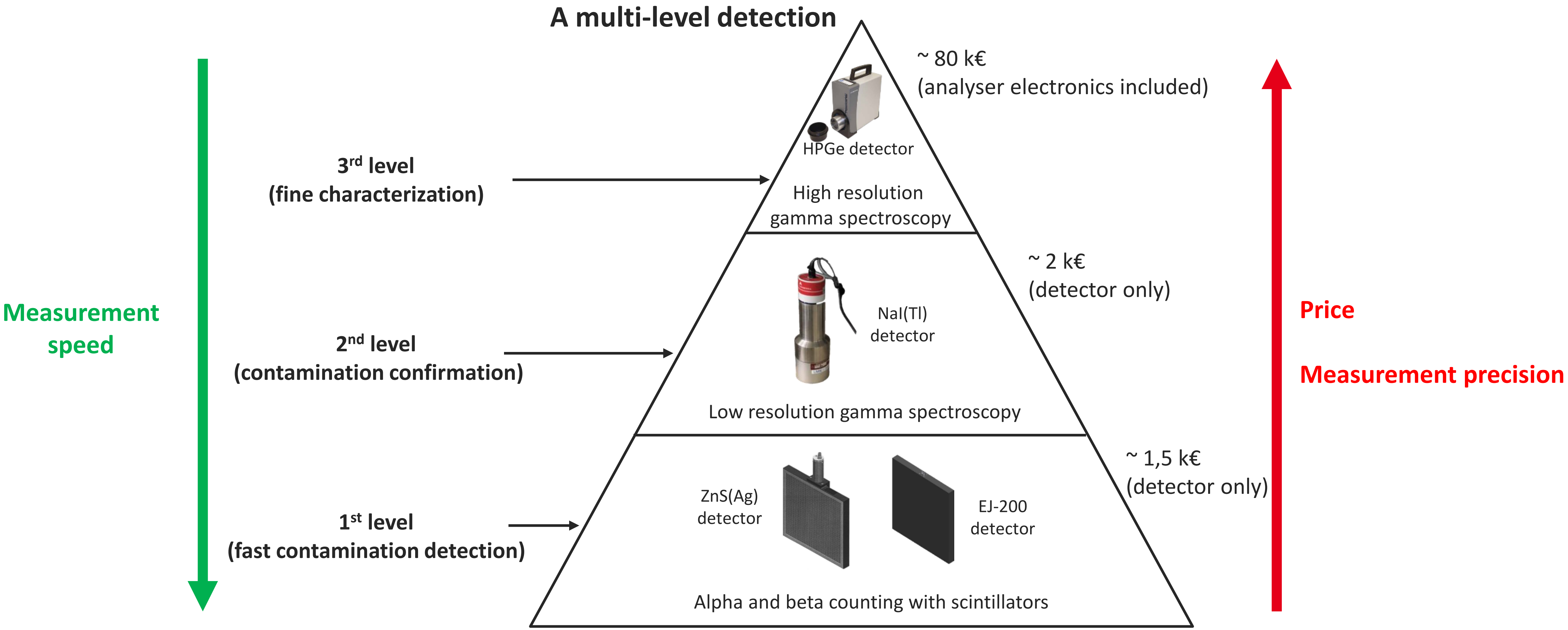
Detect very low activity contaminations with fast counting times

- Total area of 700 000 m^2 ⇨ measuring time as low as possible
- Decommissioning threshold ⇨ 0,4 Bq.cm^{-2} (α activity)

UDG corridor
Source : https://www.francetnp.gouv.fr/Pierrelatte-l-usine-d

Gamma and beta measurements sensitive to enrichment

Technical challenges and chosen detection methods



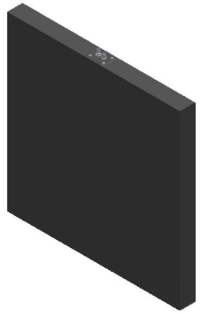
Goals :

- Demonstrate the feasibility of uranium contamination detection with each of these methods
- Propose a prototype for surveying the floors and walls inside the facility

2. Contamination detection modelling

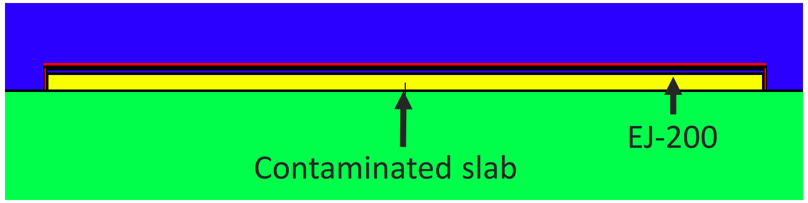
Beta particle detection

Detect the beta particles produced by ^{234m}Pa (^{238}U) within a region of interest between 300 and 2500 keV

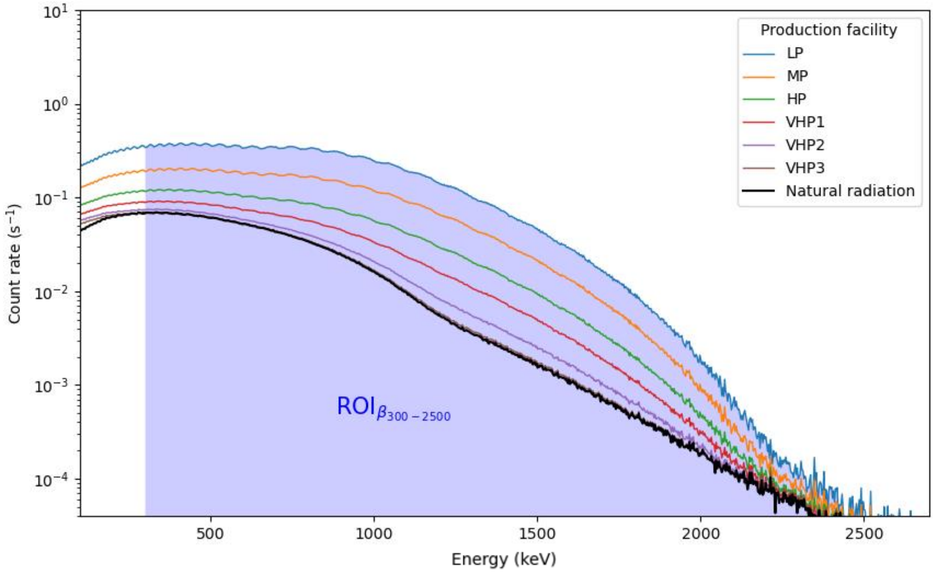


EJ-200 detector

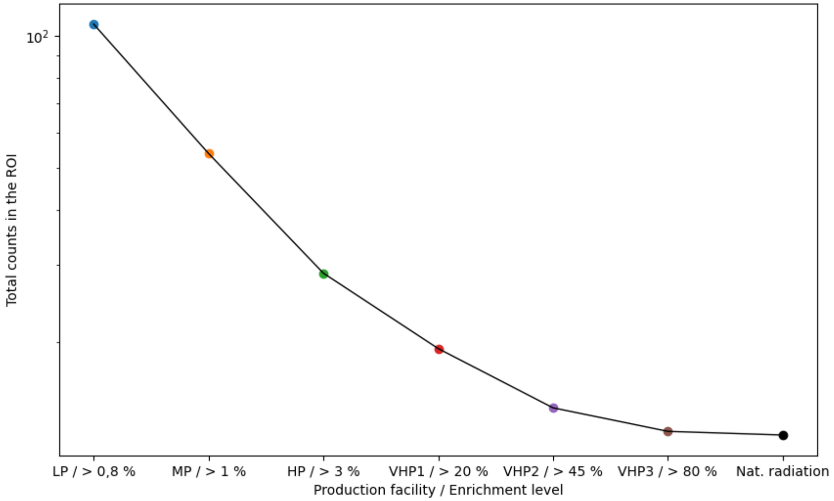
Simulation of a EJ-200 detector over a contaminated concrete slab



MCNP model



- EJ200 detector size : 32 x 32 x 0,4 cm^3
- 1 mm thick aluminum cover surrounds the scintillator
- Contamination characteristics
  - 1000 Bq distributed homogenously inside a 32 x 32 x 0,1 cm^3 concrete slab (1 $Bq.cm^{-2}$)



⊕ Moderate attenuation in concrete. Detection possible up to mm deep

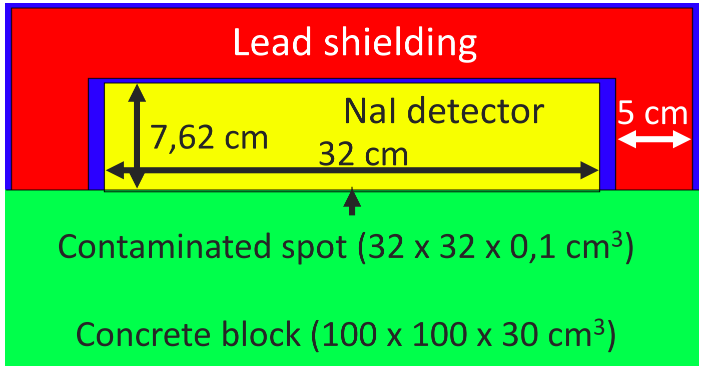
⊖ Highly dependent on ^{235}U enrichment percentage

Gamma ray detection

Detect the gamma rays produced by both ^{238}U and ^{235}U isotopes within a region of interest between 50 and 220 keV

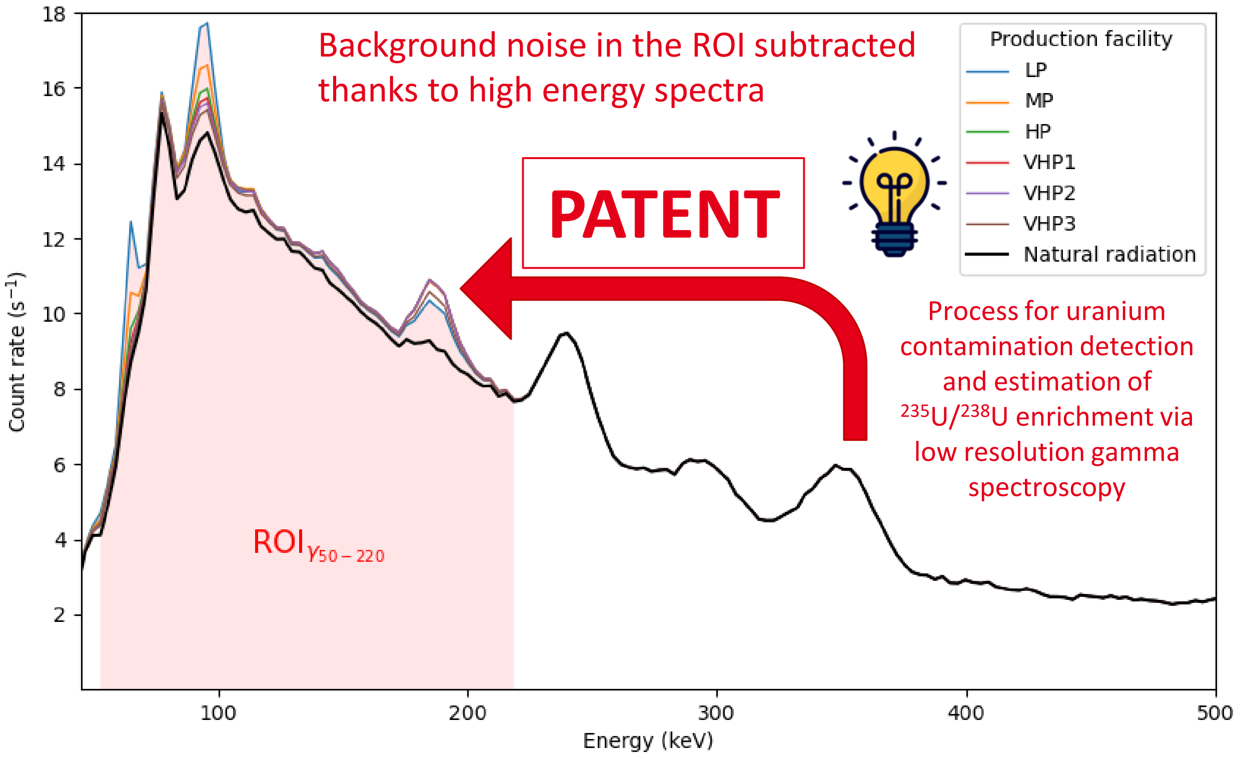
Simulation of a NaI(Tl) detector over a contaminated concrete slab

NaI(Tl) detector



MCNP model

- NaI(Tl) detector size : 32 x 32 x 7,62 cm^3
- Natural radiation emitted by a 100 x 100 x 30 cm^3 concrete block (density : 2,35 $g.cm^{-3}$)
- Contamination characteristics
  - 1000 Bq distributed homogenously inside a 32 x 32 x 0,1 cm^3 concrete slab (1 $Bq.cm^{-2}$)



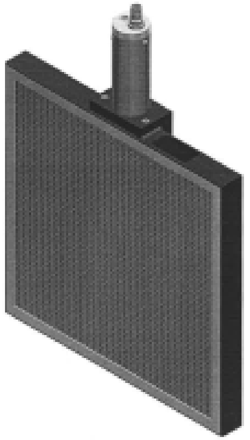
⊕ Able to detect deeper contaminations, patented method allows to determine the contamination enrichment

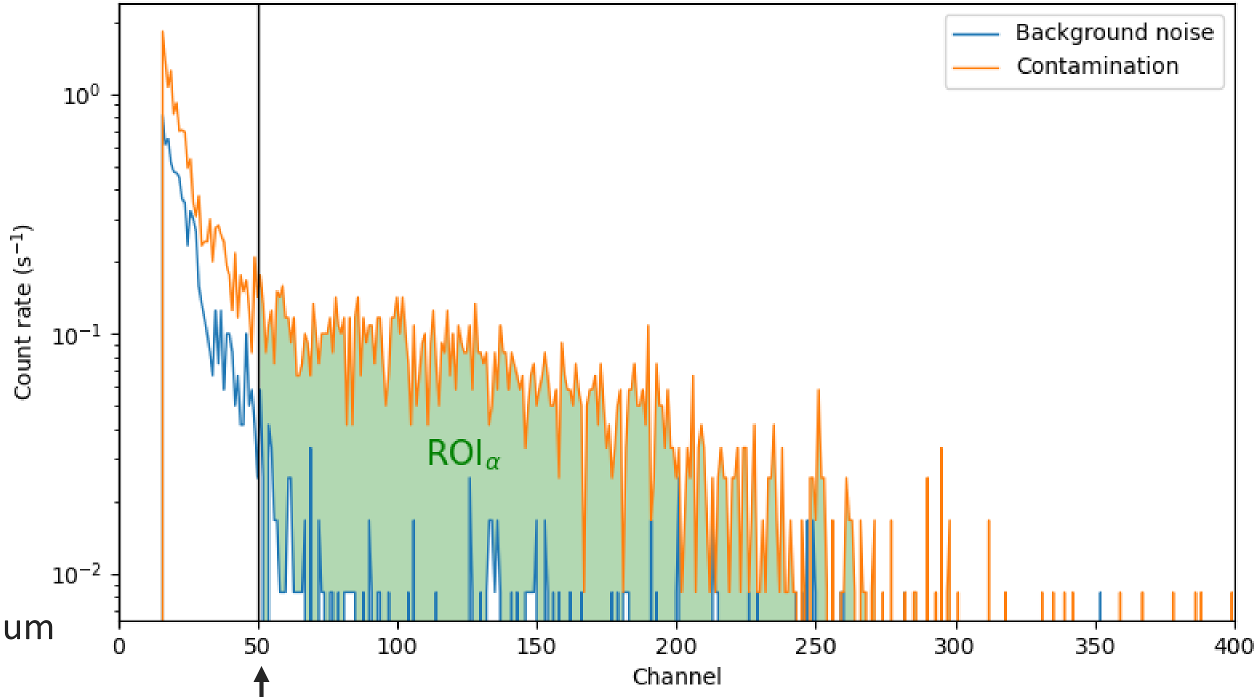
⊖ Slower detection because of the high sensitivity to background noise, moderate dependence to enrichment

Alpha particle detection

Detect the alpha particles emitted by the three uranium isotopes ^{238}U, ^{235}U and ^{234}U

ZnS(Ag) detector





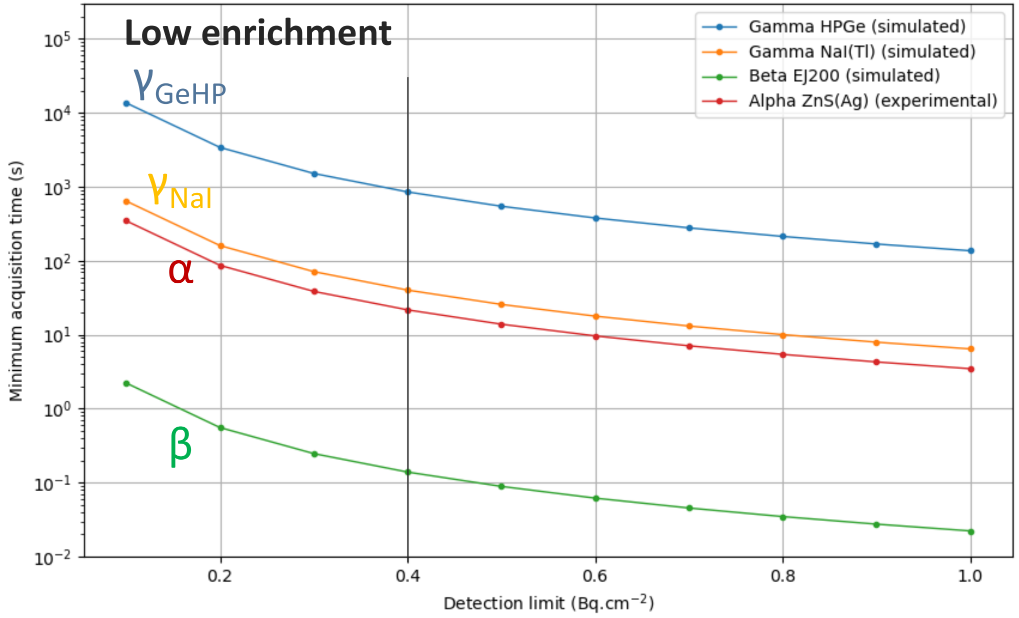
- ZnS(Ag) detector size : 25 x 25 x 2,2 cm^3 (23 x 23 cm^2 window)
- Measurements over two concrete blocks :
  One block is clean, the other is contaminated with uranium

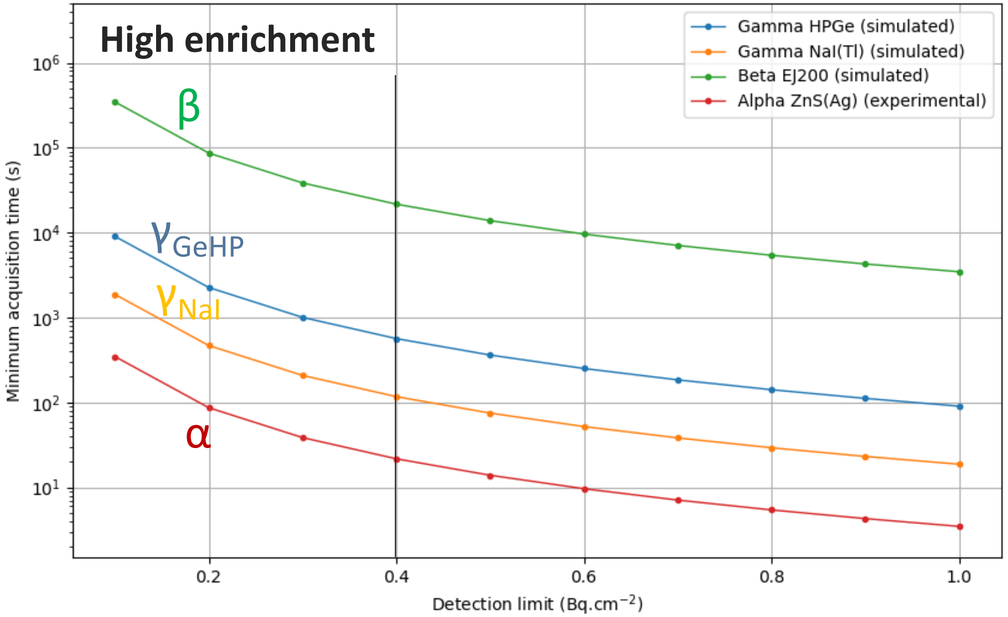
A threshold has been set to reduce the background contribution to the signal

⊕ Independent of ^{235}U enrichment, counting rate directly proportional to the contamination activity

⊖ Can only detect surface contamination. High dependence on the state of the inspected area (dust, rough surface)

Estimation of acquisition times and conclusions





Estimations for a 1 mm deep contamination

	Minimum acquisition time to reach the 0,4 Bq.cm^{-2} DL			
^{235}U enrichment	Gamma HPGe	Gamma NaI(Tl)	Beta EJ200	Alpha ZnS(Ag)
Low enrichment	854 s	40 s	14 x 10^{-2} s	22 s
High enrichment	565 s	117 s	22 x 10^{3} s	22 s

Detection time estimation

Low enrichment ⇨ beta < alpha < gamma

High enrichment ⇨ alpha < gamma < beta

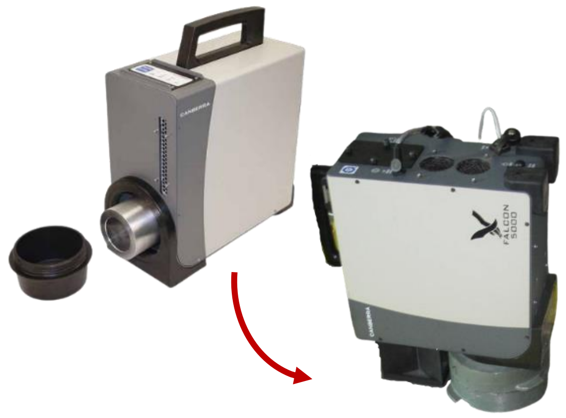
3. In-situ measurements at the UDG enrichment facility

Detectors and inspected zones

γ spectrometry

High resolution

Falcon 5000



$S_{det} = 28\ cm^2$

- Semi-conductor detector (HPGe)
- Cylindrical crystal : 3 cm radius and 3 cm thick
- Electrical cooling
- Lead disks surround the detector entrance window

Low resolution

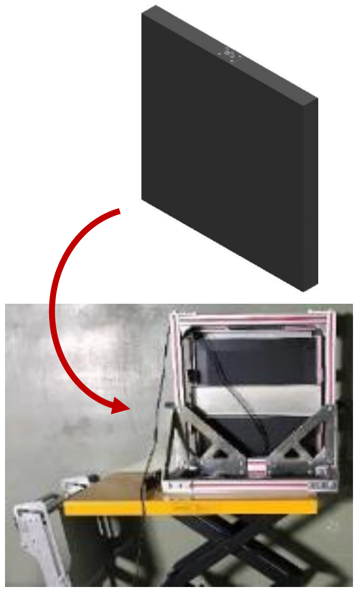
NaI 3"x3"

$S_{det} \approx 530\ cm^2$

- Scintillation detector NaI(Tl)
- Cylindrical crystal : 7,62 cm diameter and 7,62 cm thick
- $D_{detector-floor} = 10,2\ cm$
- Lead disks around the detector + forklift for transportation

β counting

Scintillation detector

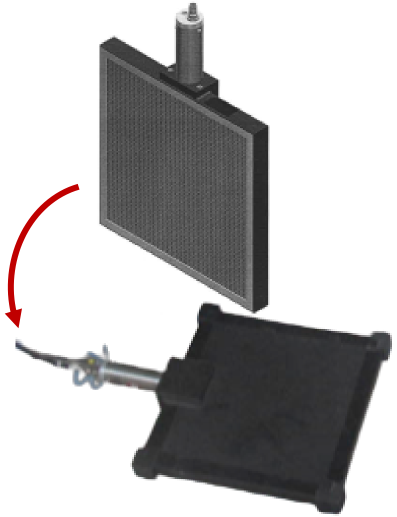


$S_{det} = 2430\ cm^2$ or $625\ cm^2$

- Plastic scintillator EJ-200
- Aluminized PET entrance window
- $D_{detector-floor} = 7,5\ mm$

α counting

Scintillation detector ZnS(Ag)



$S_{det} = 529\ cm^2$

- ZnS(Ag) layer spread over a plastic sheet (EJ-440)
- $D_{detector-floor} = 2\ mm$
- Aluminized PET entrance window protected by a steel mesh

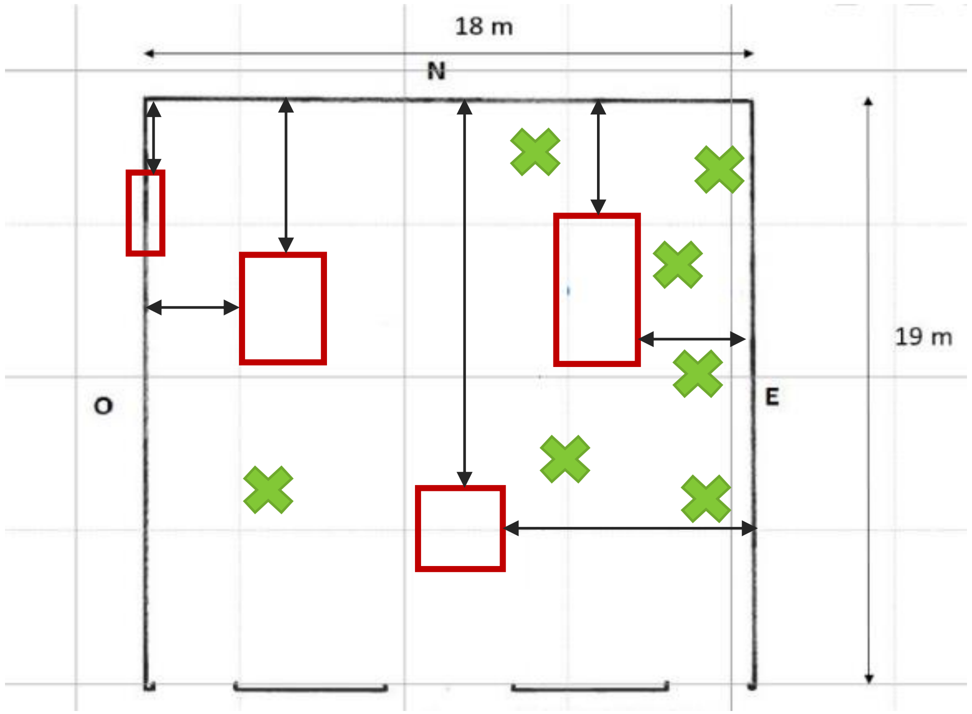
Detection surface compatible between the detection methods

Detectors and inspected areas

Low enrichment plant

Counting time per point : α = 2 to 3 min | β = 2 min | γ = 15 min

→ Measurement areas composed of multiple 25 x 25 cm^2 measurement points



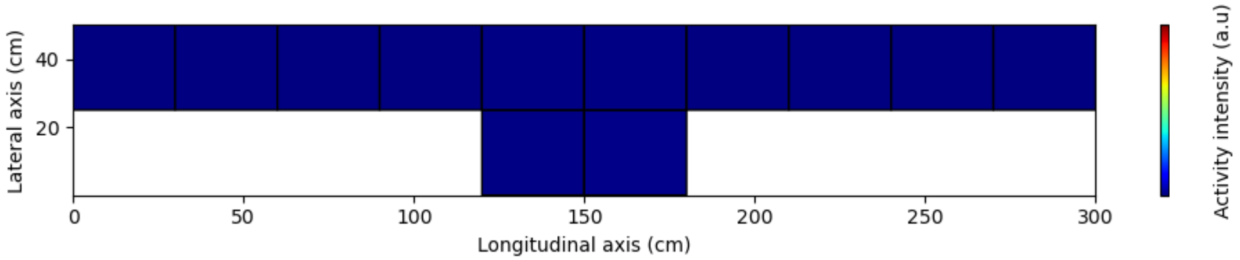
Total area analyzed ≈ 13,3 m^2

- low enrichment plant ⇨ 9 m^2
- high enrichment plant ⇨ 4,3 m^2

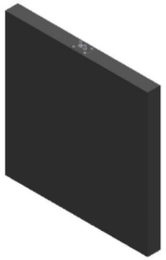
Results for the low enrichment plant

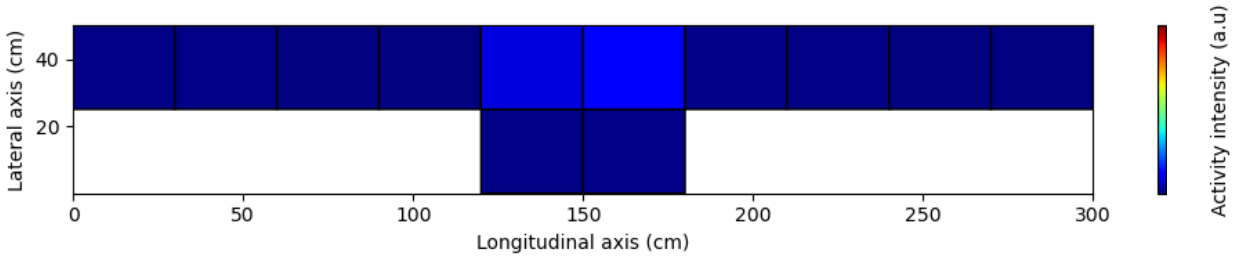
Hypothesis of contamination migration depth : **10 µm**

α



β

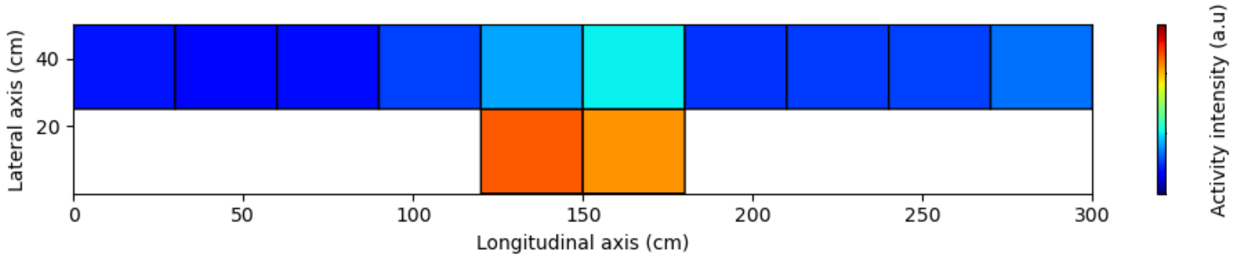




Not contaminated

Contaminated

γ

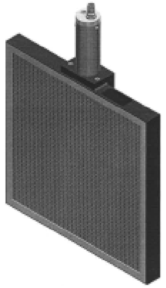


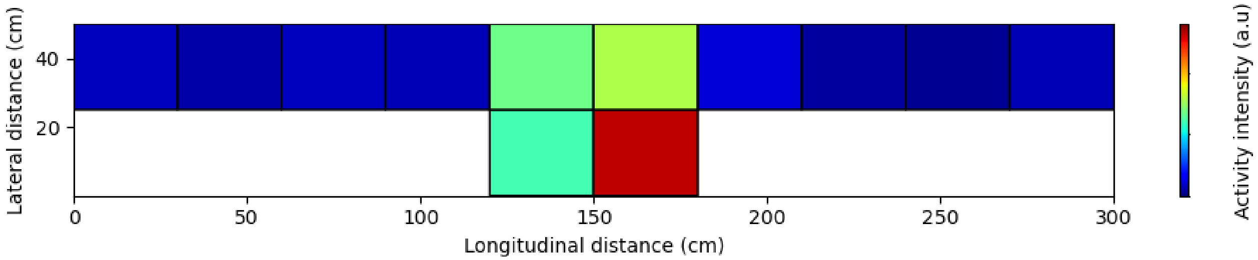
Activity measured by α and β techniques < 0.4 Bq/cm² with the hypothesis of a surface contamination (< 10 µm)

Results for the low enrichment plant

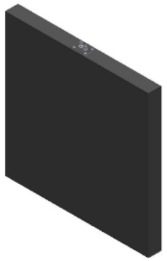
Hypothesis of contamination migration depth : **1 mm**

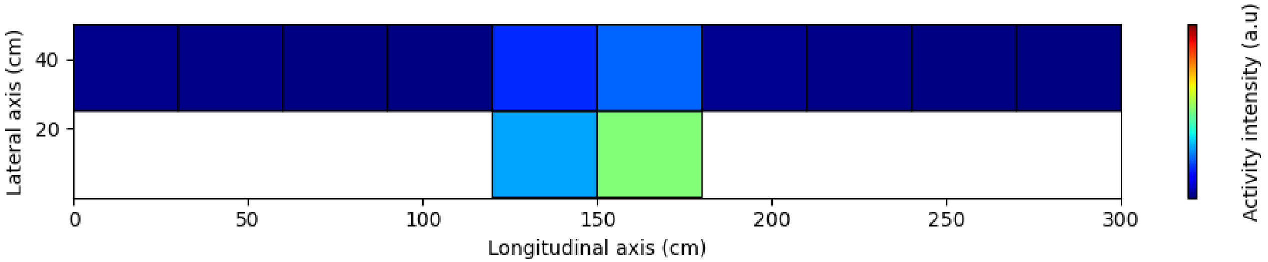
α



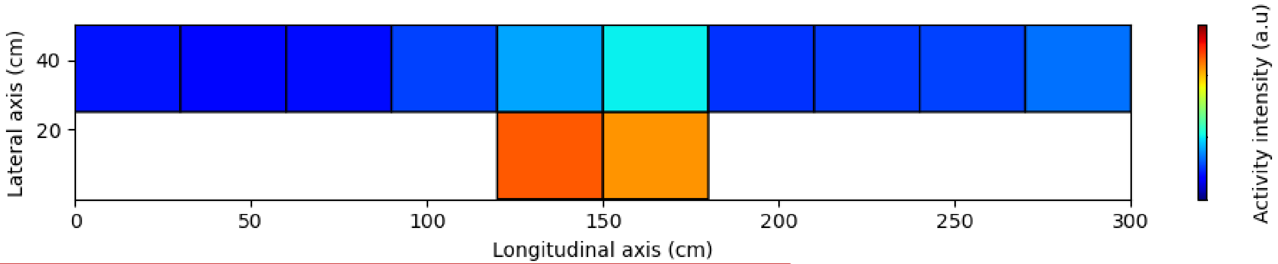


β





γ



Not contaminated

Contaminated

Contamination depth is an important factor for alpha and beta detection from 10 µm to 1 mm

→ Factor 80 for alpha detection
→ Factor 2 for beta detection
→ No impact for gamma detection

Background noise corrections are also an important aspect

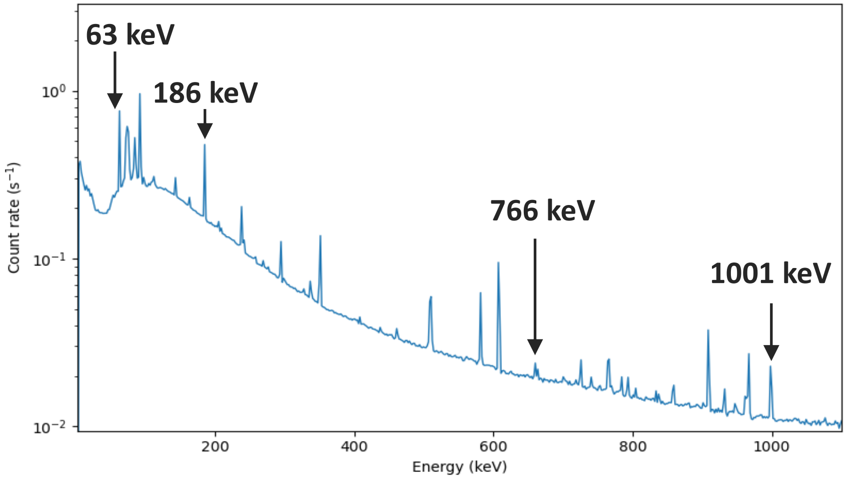
→ Activity estimations rely on correct background subtractions
→ Background fluctuates from one point to another

Uranium enrichment estimation

Measurements done over the same contaminated area

HPGe

64,3 hours measurement



Emitter isotope	Peak energies (keV)
^{238}U	63
	112
	766
	1001
^{235}U	144
	163
	186
	205

Estimated surface activities :

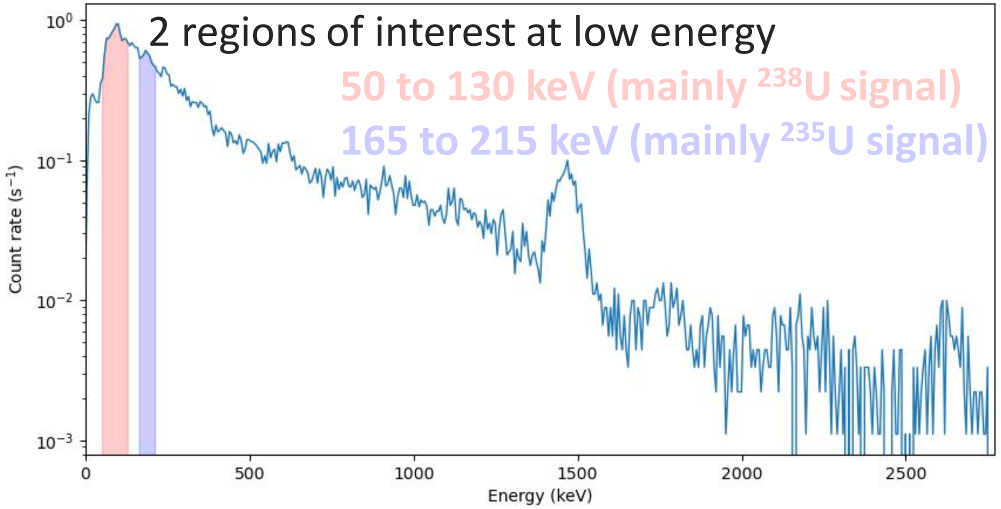
- ^{238}U : 8,7 ± 1,1 Bq.cm^{-2}
- ^{235}U : 0,45 ± 0,05 Bq.cm^{-2}

After conversion to masses

$$\%_{Enrichment} = \frac{M_{235}}{M_{238} + M_{235}} = 0{,}79 \pm 0{,}07\ \%$$

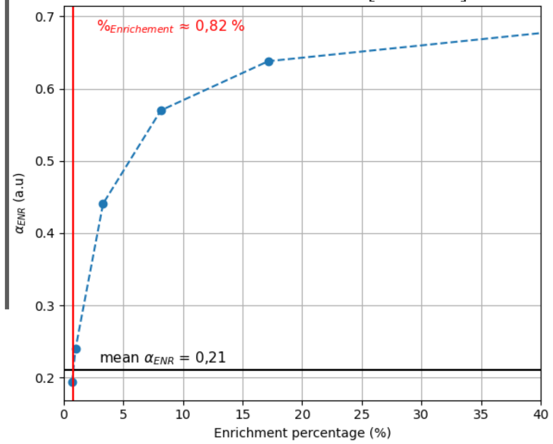
NaI(Tl)

15 minutes measurement



Calculation of a enrichment indicator using experimental data :

$$\alpha_{ENR} = \frac{C^{NET}_{[165-215]}}{C^{NET}_{[50-130]}} = \frac{C^{RAW}_{[165-215]} - C^{NOISE}_{[165-215]}}{C^{RAW}_{[50-130]} - C^{NOISE}_{[50-130]}} = \mathbf{0,21}$$



Calibration curve between α_{ENR} and enrichment level determined via simulation

Enrichment estimation : 0,82 % ranging from 0,62 % to 1,3 % when considering statistical uncertainties

4. Conclusions and perspectives

Conclusions and perspectives

Conclusions drawn from the measurements at the UDG enrichment plant

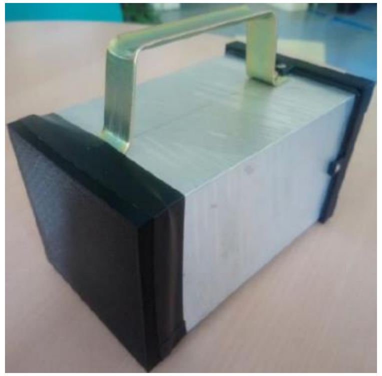
- The migration depth must be a well-known parameter for correct activity estimations
- The background noise must be finely characterized for a good correction of raw counts
- Measurement times differ depending on the method

Particle detected	Impact of migration depth	Impact of ^{235}U enrichment	Impact of background noise fluctuation	In-situ measurement time (for 1000 cm^2)
Alpha	High	No impact	High	~ 1 min.
Beta	Moderate	High	High	~ 1 min.
Gamma	No impact	Moderate Estimate via patented method	High	~ 15 min.

Perspectives

- Large NaI (~ 30 x 30 cm^2) for increased detection efficiency
- New measurements for background noise characterization (2023)
- New contamination detection tests (2024)
- Ongoing feasibility studies for uranium detection by active XRF (K and L lines)

NaI(Tl) 4"x4"



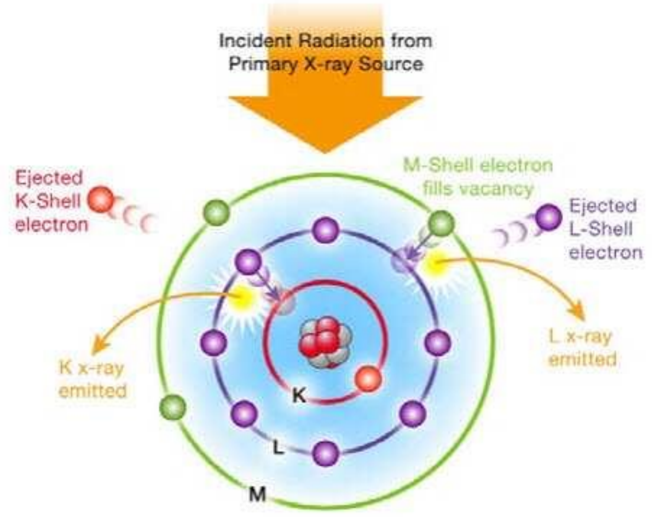
Gamma camera for x-ray imaging

Thank you for your attention

5. Annexes

X-Ray fluorescence

Working principle :



Source : https://www.thermofisher.com/blog/ask-a-scientist/what-is-xrf-x-ray-fluorescence-and-how-does-it-work/

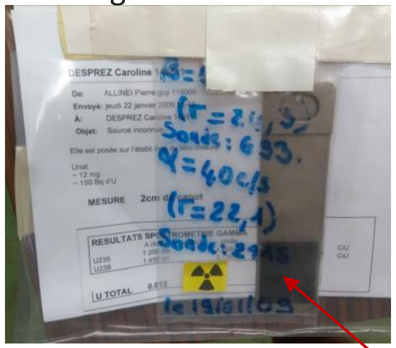
Uranium L-ray energies :

L-ray	Energy (keV)
L_l	11,6
$L_{\alpha 1}$	13,6
$L_{\beta 2}$	16,4
$L_{\beta 1}$	17,2
$L_{\gamma 1}$	20,2

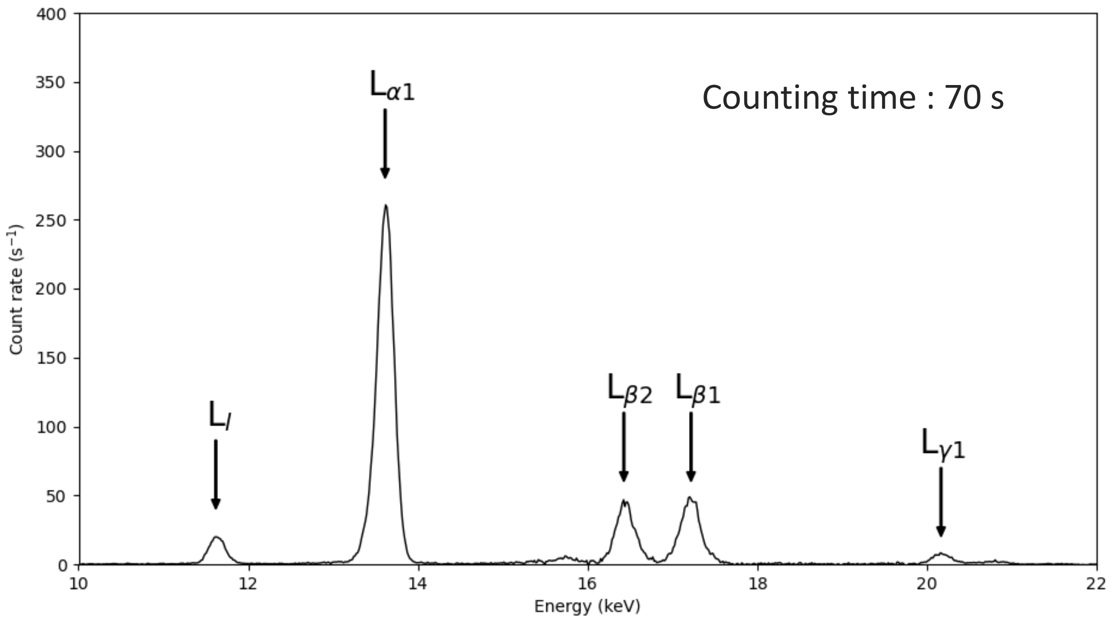
Application to uranium detection :

Source + detector :
Niton GOLDD XL2

Sample :
12 mg of natural uranium



Uranium distributed over a 11,1 cm^2 surface



Emission rates of alpha, beta and gamma radiation

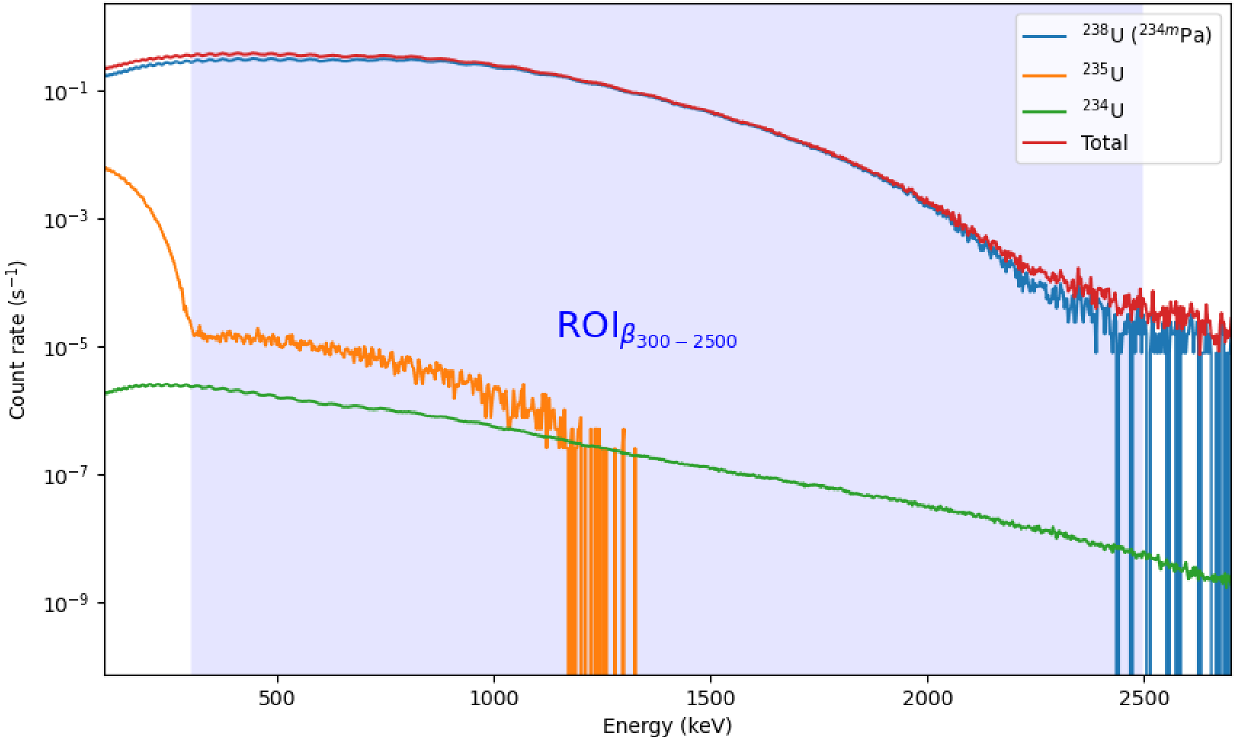
Gamma, alpha and beta emission rates for a uranium contamination with a total activity of 1000 Bq

Facility	Emission rate γ (/s)				Emission rate α (/s)				Emission rate β (/s)		
	^{238}U	^{235}U	^{234}U	Total	^{238}U	^{235}U	^{234}U	Total	^{238}U	^{235}U	Total
LP	109,2	58,9	59,6	227,7	400,0	26,0	574,0	1000,0	800,0	26,0	826,0
HP	19,1	90,7	92,4	202,1	70,0	40,0	890,0	1000,0	140,0	40,0	180,0

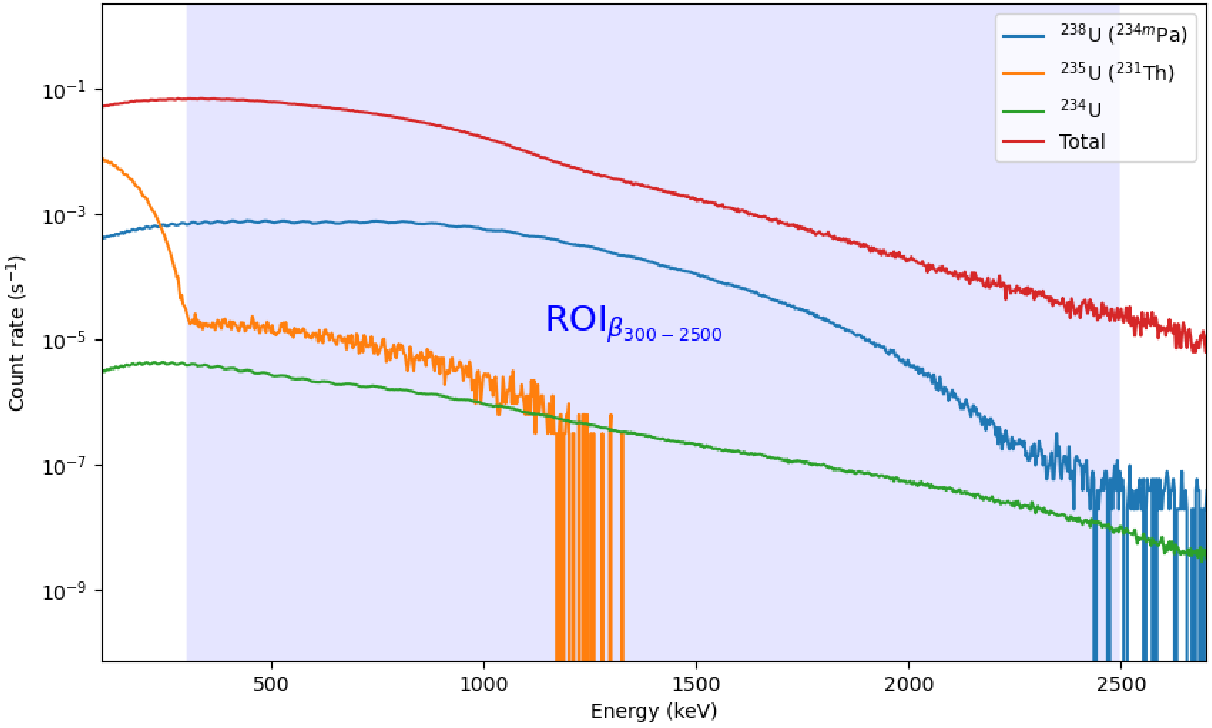
Beta particles detection

Decomposition of ^{238}U, ^{235}U and ^{234}U contributions to the signal (without natural radiation) in **1 % and 80 % enriched contamination**

1 % enrichment

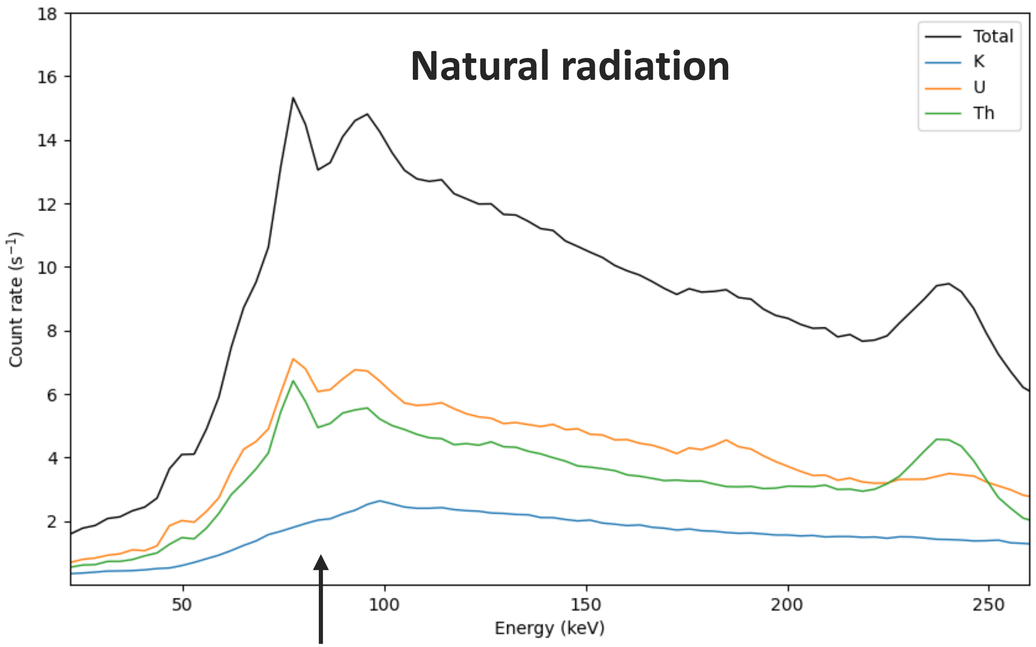


80 % enrichment



Decreased ^{238}U beta signal at 80 % enrichment

Gamma rays detection



Most natural gamma signal in the ROI comes from uranium and thorium

Natural abundances :

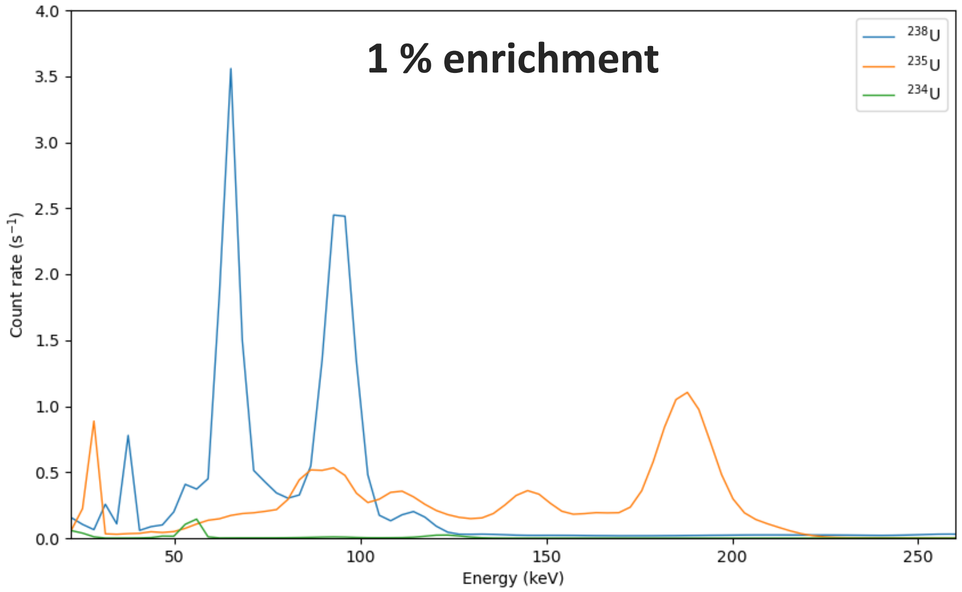
Uranium → 99,27 % ^{238}U | 0,72 % ^{235}U | 0,005 % ^{234}U

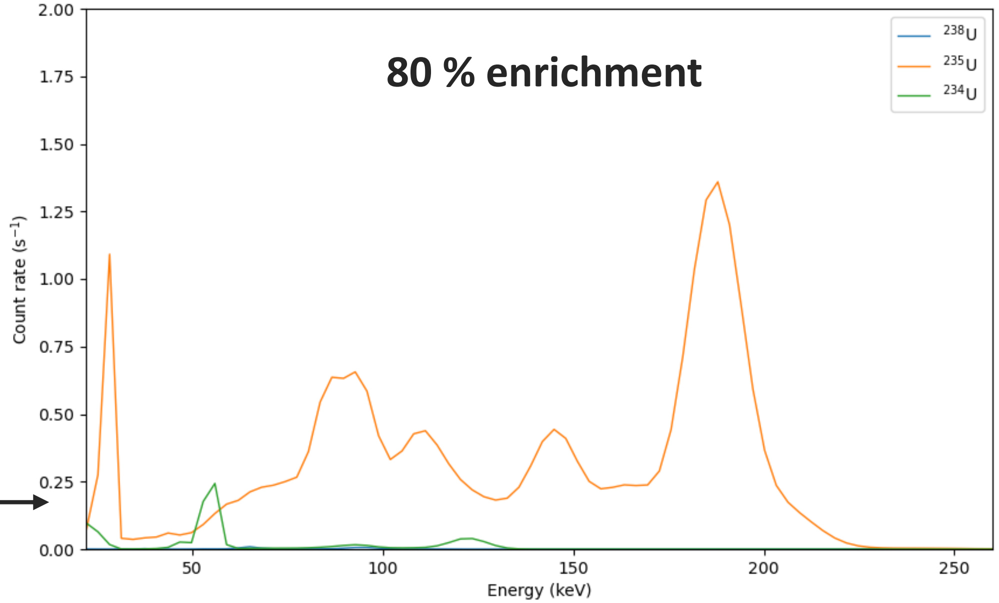
Thorium → virtually 100 % ^{232}Th

Potassium → 93,26 % ^{39}K (stable) | 0,012 % ^{40}K | 6,75 % ^{41}K (stable)

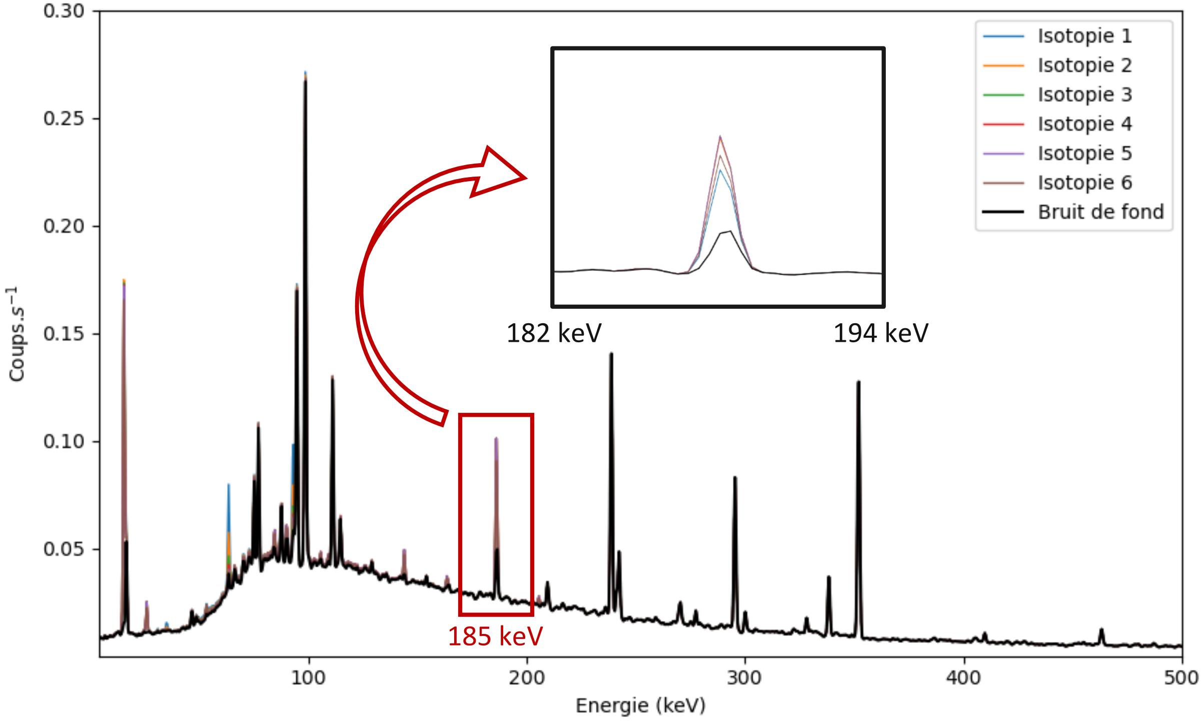
At 80% enrichment :

- The ^{238}U contribution becomes negligible
- Slight increase in the ^{235}U and ^{234}U contributions

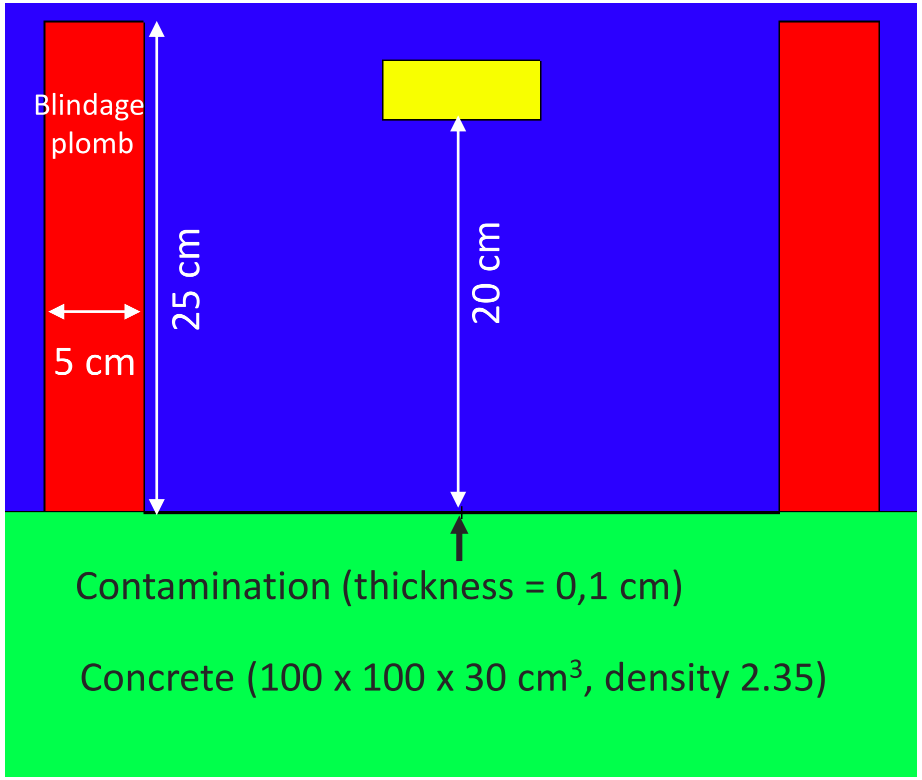




Gamma rays detection (HPGe)



Modèle MCNP BEGe 5030 :



Background noise :

- 2.1 ppm_U, 4.2 ppm_{Th}, 0.78 $\%_K$

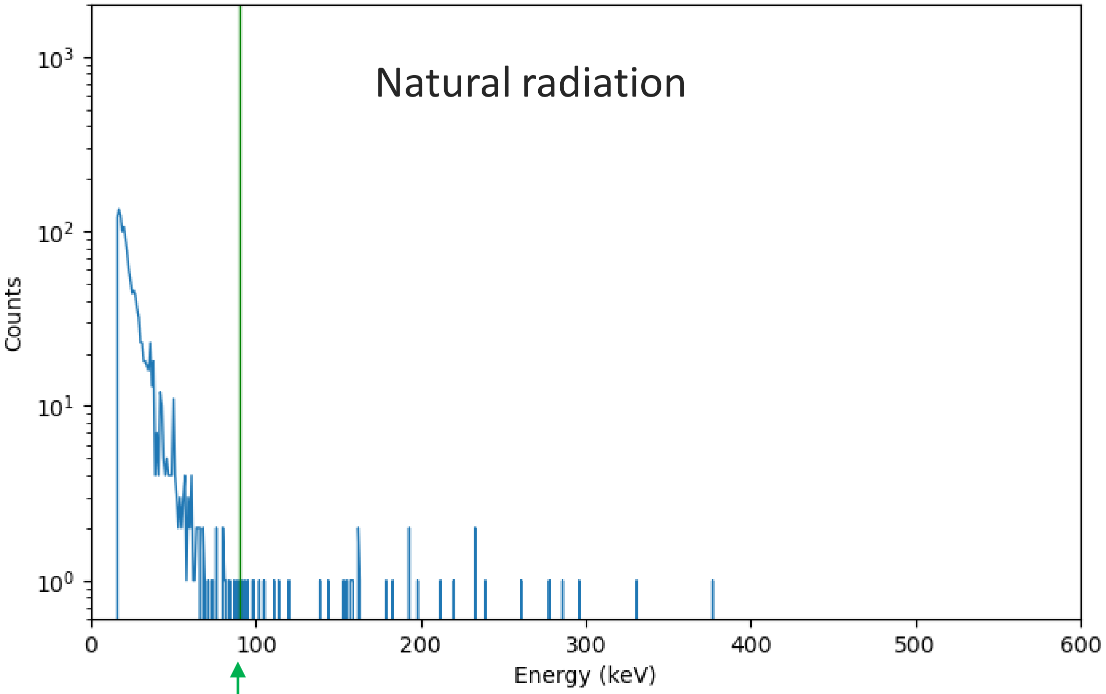
Contamination :

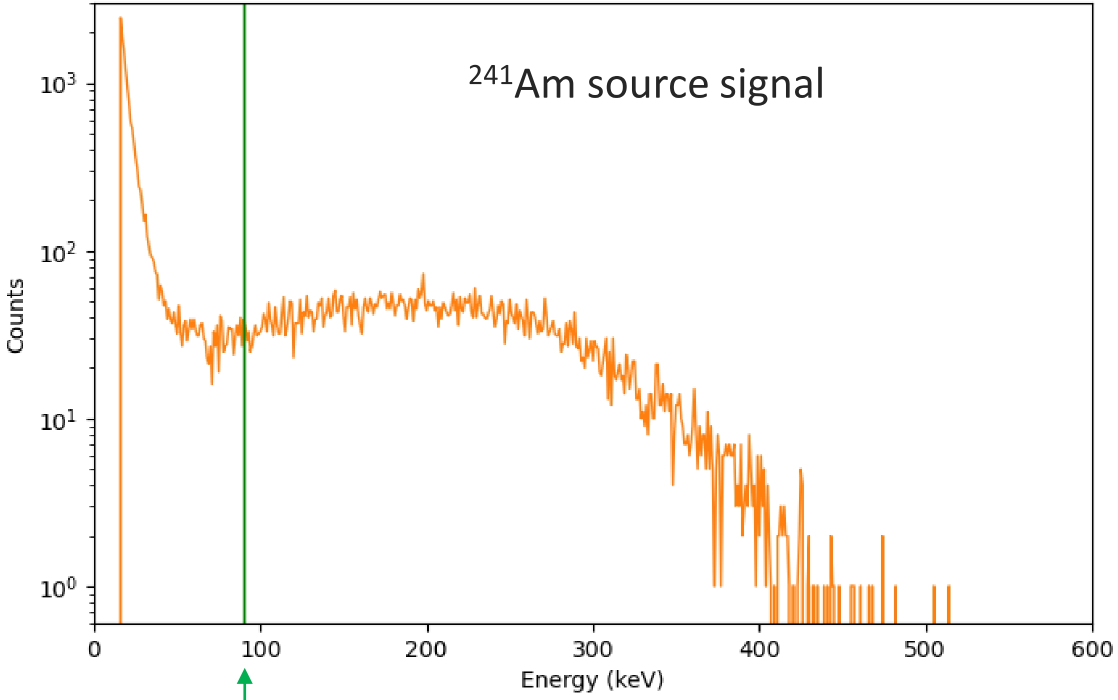
- 1000 Bq d'activité totale
- Distributed in a 32 x 32 x 0.1 cm^3 slab
- Surface = 1024 cm^2 → 0.98 Bq/cm^2

Alpha particles detection (experimental) LMN

Distance to source : 11 cm

Acquisition time : 3 minutes





Cut at channel 90 to remove the natural radiation signal + intrinsic noise

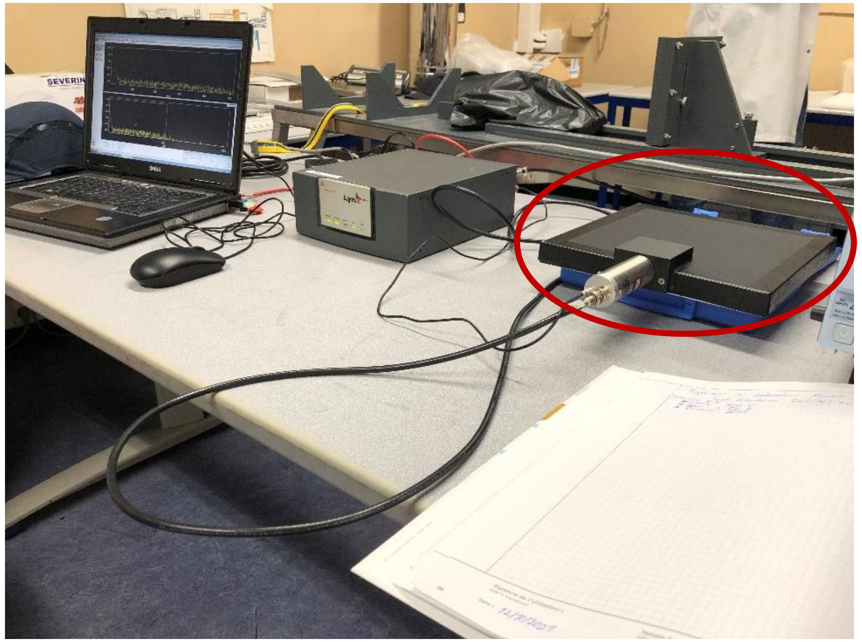
Note : alpha detection does not depend on enrichment
All 3 uranium isotopes decay via alpha emission with a 100 % probability

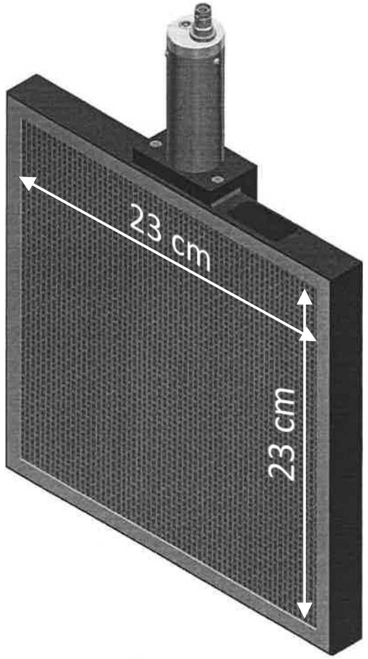
Alpha particles detection (experimental)

Alpha simulation → high uncertainty and resources consuming
Experiments carried out using a ^{241}Am alpha source and a ZnS(Ag) scintillator detector

ZnS(Ag) characteristics :

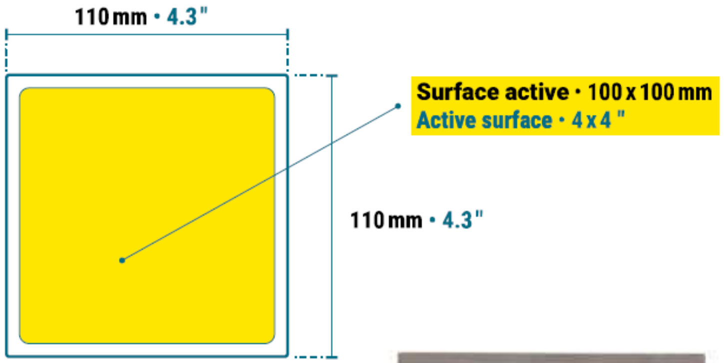
- Detection surface : 23 x 23 cm^2
- 6 µm aluminized PET entry window
- Metal grid for screen protection





^{241}Am source characteristics :

- Alpha emission rate ≈ 376 s^{-1} (4π)
- Active surface = 10 x 10 cm^2
- Emission energy = 5,5 MeV



Estimation of acquisition times and conclusions

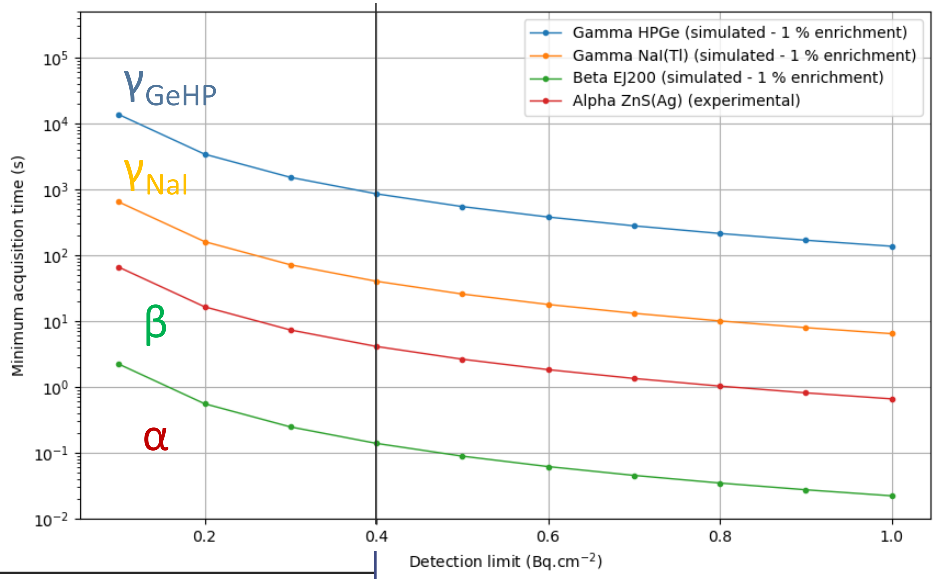
Estimation of required counting time to reach a certain detection limit :

$$DL_{c/s} = 4 \times \sqrt{BN} \times \frac{1}{\sqrt{T_{acq}}}$$

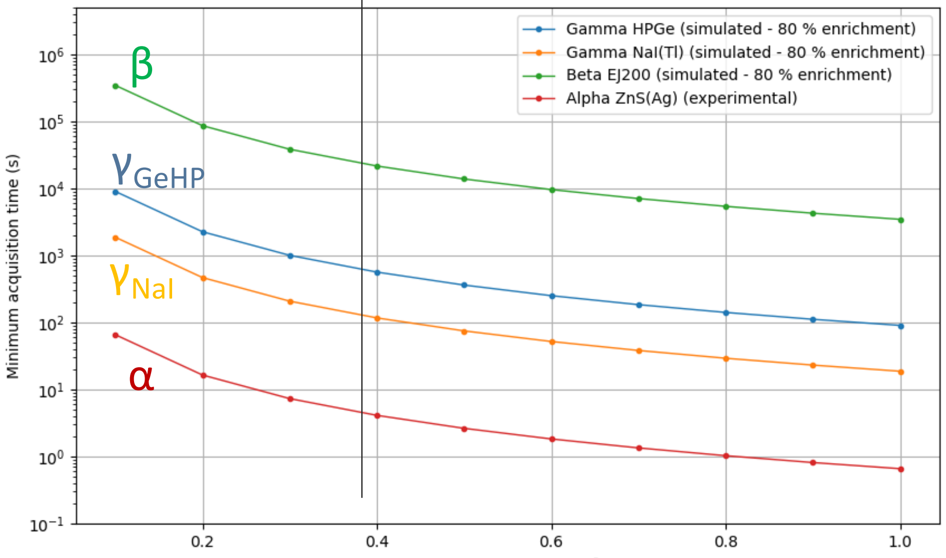
$$DL_{Bq/cm^2} = \frac{DL_{C/S}}{CF}$$

$$T_{acq\ min} = \left(\frac{4 \times \sqrt{BN}}{DL_{Bq.cm^{-2}} \times CF}\right)^2$$

- BN : background noise produced by natural radiation in the ROI (in counts)
- CF : Calibration factor to convert $c.s^{-1}$ to $Bq.cm^{-2}$ (in $c.s^{-1}.Bq^{-1}.cm^{2}$)
- $DL_{Bq.cm^{-2}}$: detection limit to achieve (in $Bq.cm^{-2}$)
- $T_{acq\ min}$: minimum acquisition time required



Minimum acquisition time (s) to reach the 0,4 $Bq.cm^{-2}$ DL				
^{235}U enrichment	Gamma HPGe	Gamma NaI(Tl)	Beta EJ200	Alpha ZnS(Ag)
1 %	854	40,2	13,9 x 10^{-2}	22,9 x 10^{-3}
80 %	565	117	21,6 x 10^{3}	22,9 x 10^{-3}



Detection time estimation allows us to determine the "detection levels" for the detection of uranium contamination (from fastest to slowest)
→ alpha > beta > gamma if the enrichment is low
→ alpha > gamma > beta if the enrichment is high

Gamma detection : counts inside the 50 – 220 keV ROI (NaI) | 185 keV peak area (HPGe)
Beta detection : counts in the 300 – 2500 ROI
Alpha detection : total count after channel 90

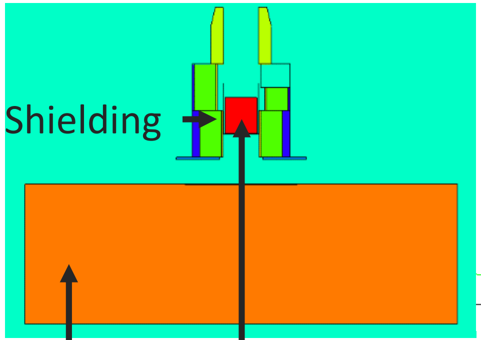
Method for surface activity estimation

Count rate to surface activity estimation

Net counts (C_{net} in $c.s^{-1}$) → Development of a digital representation → Calculation of the calibration factor (CF in $c.s^{-1}.Bq^{-1}.cm^{2}$) → Conversion of net counts → Surface activity ($Bq.cm^{-2}$)

Net counts =
gross counts - background noise counts

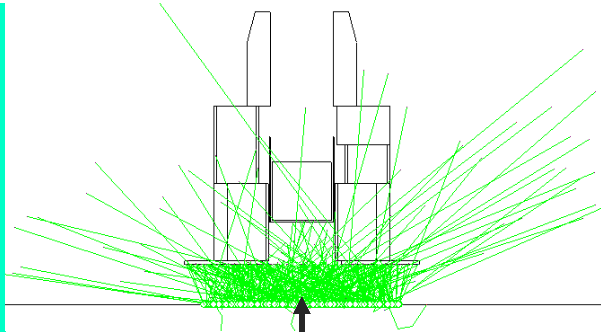
Digital representation of the detector and its shielding



Concrete block

NaI(Tl) detector

Radiation detection simulation.
The source activity is normalized to 1 Bq and its geometry is known



Radiation source

$$A_{surface}\,(Bq.cm^{-2}) = \frac{C_{net}\,(c.s^{-1})}{CF\,(c.s^{-1}.Bq^{-1}.cm^{2})}$$