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NON DESTRUCTIVE CONTROLS OF RADIOACTIVE WASTE AT CEA

8 th International Summer School on Nuclear Decomissionning and Waste Management

12-16 SEPTEMBER 2016, ISPRA

OUTLINES

- **Nuclear Waste Classification**
- **The Characterization on Nuclear Waste at CEA**
- **The 2nd level controls or "Supercontrols"**
- **The Legacy Waste**
- **R&D on measurement technics**
- **Conclusion**

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NUCLEAR WASTE CLASSIFICATION

2nd Level controls by CEA under ANDRA Spécification

Massic Activity (Bq/g)	lower than 100	100 to 10^5	10^5 to 10^9	Higher than 10^9
Activity level and repository	**(1) VLLW** *Very low level wastes* **(Storage at Centre de l'Aube -CIRES)**	**(2) LIL-SL$t_{1/2}$<31 y (Storage at Centre de l'Aube-CSA)** *low-level and intermediate-level, short life* **(3) LA-LL** *low-level and intermediate-level, long life* Intermediate storage *final storage (project)*	**(4) IL-LL** *CIGEO (project)*	**(5) HA** *High Activity* Producer intermediate storage *-> CIGEO (project)*
Type of solid wastes	Debris, scrap iron, plastics,… mainly from the dismantling	Gloves, coats, glasses, scrap iron, …	Cladding, hulls and end caps from spent fuel, Wastes coming from glove boxes and hot cells, filters, …	Vitrified Fission Products coming from the fuel reprocessing
% of volume of French radioactive waste	20,1%	LIL-SL : 68,8 % LA-LL : 7,2 % IL-LL : 3,6%		0,2 %
% of activity	0,000003%	LIL-SL < 0,03% LA-LL < 0,009% IL-LL : 4,98%		94,98 %

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OBJECTIVES : Check Conformity versus interim storage, transport and final disposal specifications -> SAFETY AND PUBLIC ACCEPTANCE

- Radiological specifications
  - α, βγ activities
  - α after 300 years
  - Fissile matter amounts
- Geometrical specifications
  - Sizes and envelope thickness
  - Outside containers
  - Waste centering
- Physical specification
  - Free space remaining
  - Homogeneity, porosity
  - Local defects
- Chemical specification
  - Amount of limited materials
  - Forbidden materials

Nuclear Measurement
Sampling +
Chemical Analysis

- - - -> = partial answer to characterization needs

SEVERAL LEVELS OF CHARACTERIZATION :

- **During Waste Production (AREVA, EDF, CEA, …)**
  - **During fabrication**
  - **Quality control and final characterization**

- **During interim storage (producers) or before final disposal (ANDRA)**

- **2nd level controls or “Supercontrôles” : for LIL-SL waste : specified by Andra for few tens of WP, “blind” controls performed by CEA : Expert labs for destructive and non destructive measurements.**

THE CHARACTERIZATION OF WASTE PRODUCED CURRENTLY BY CEA : CASE OF IL-LL WASTE

Content : Waste from operation of nuclear installation, Waste coming from the dismantling

For most WP, characterization is done :

- **Either by destructive measurements on samples (case of homogeneous WP),**
- **Either by non destructive measurements on primary WP or non conditioned WP.**

Primary WP : 100 liters (measurable) -> compaction or concreted - > final WP : 870 liters (difficult to measure)

Measurable primary WP are:

- **Either 100l ou 118l drums (final WP : 870 l αPu),**
- **Either 20 to 70l containers (final WP : 500 l MI).**

Once in their final packaging, characteristics of the waste (physical, chemical, radiological ...) will be difficult (or impossible) to obtain with non-destructive methods

THE CHARACTERIZATION TECHNICS:

- **Radiological Characterization (Activity, fissil mass)**

Dose Rate measurement + nuclide spectra, gamma spectrometry, passive neutron measurement (sometime) and active (rarely)

- **Physical characterization (material)**

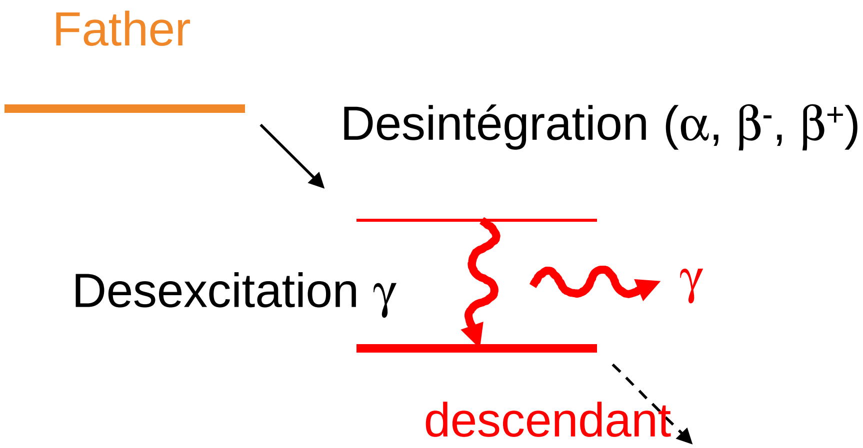
X ray imaging: radiography et tomography

+ coupling with non destructive measurements

- **Chemical Characterization (forbidden or limited materials)**

Sampling + chemical analysis

GAMMA SPECTROMETRY



Identification and quantification of radionuclides through his descendants

Standard gamma spectrometry device
100 liters WP

- **Global Measurement**
- **Segmented Measurement**
- **Better resolution with Germanium Detectors**

THE ADVANTAGES AND DRAWBACKS OF γ SPECTROMETRY

Advantages :

- Easy to implement
- Activity of numerous β/γ emitteurs
- Adapted for low density WP (d<1,5)

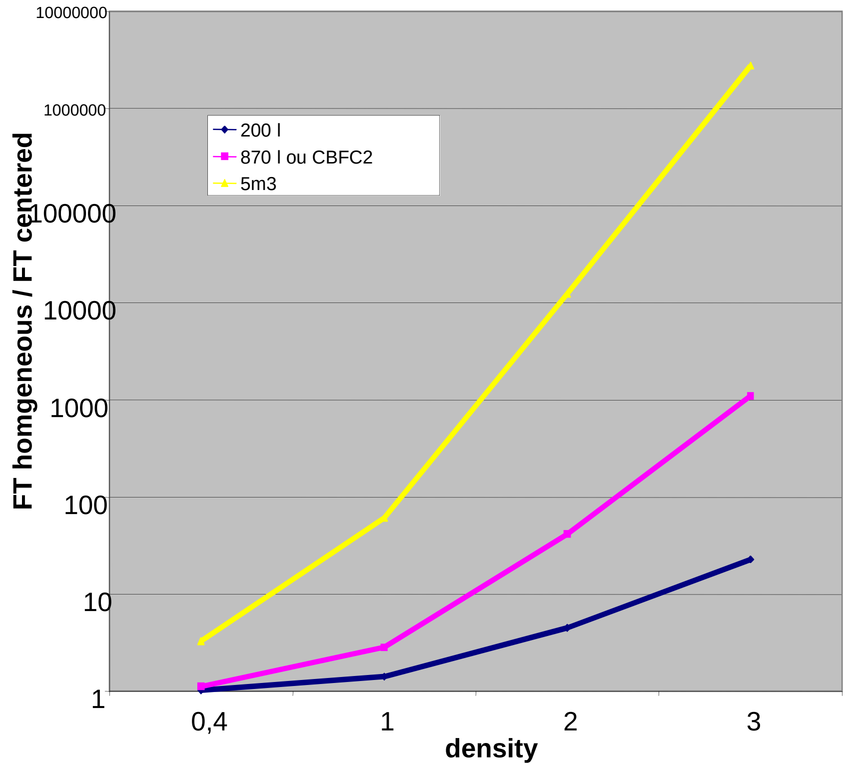
Drawbacks :

- Unsuited for high volumes and/or high density WP
- Unsuited for low enegy gamma raies (case of actinides U and Pu)
- Needs a transfert function to take into account :
  - Density distribution
  - Activity distribution

$$A_{WP} = A_{\text{measured outside WP}} / FT(E_{\gamma}, WP)$$

Uncertainties

Effect of activity distribution (500 keV)



PASSIVE NEUTRON MEASUREMENT

Global measurement of neutron emission : (spontaneous fission + (α, n) reaction) : suited to Pu measurement

En ~ 2MeV => Slowing down – Thermalisation - detection

Indirect measurement of total Pu
: $^{238}Pu + {}^{240}Pu + {}^{242}Pu + ({}^{244}Cm, {}^{241}Am$ …)!
•En (^{240}Pu)= 1020 n.$^{s-1}$.g^{-1}
•En (^{238}Pu)= 2590 n.s^{-1}.g^{-1}
•En (^{244}Cm)= 1,08 10^{7} n.s^{-1}.g^{-1} -> a small quantity of Cm can hide Pu isotopes !!!

Needs Isotopic Composition (CI)
-> coupling with gamma spectrometry or nuclide spectra

Passive neutron measurement device PEGASE

- **Global counting or coincidence ((α, n) rate)**

ACTIVE NEUTRON MEASUREMENT

Global measurement of neutron emission after activation : induced fission by <u>thermal neutrons</u>
-> suited to the measurement of fissil isotopes of U and Pu

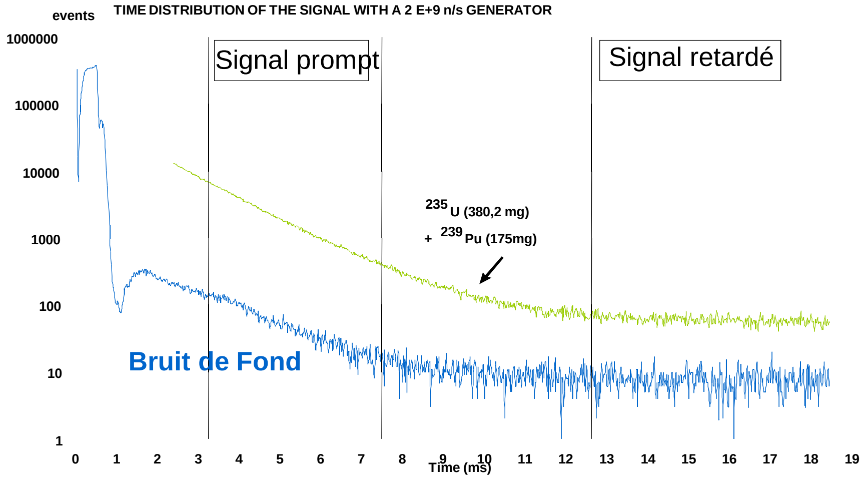
Neutron d'émission = 14 MeV (2.10^9 s^{-1}) -> Thermalisation -> Fissions -> fast neutron production -> Thermalisation -> Detection of prompt ou delayed neutrons

Indirect measurement of Pu :

- **Only fissiles isotopes**
  ^{235}U + ^{239}Pu + ^{241}Pu (no more problems with Cm!)

- **Needs isotopic composition**

Symetric cell – Chicade facility Cadarache



Achievable performances with passive neutron measurement

(Source at the center of a 118 liter drum - 30 minutes)

Matrice	Empty drum	Cellulose d=0,14	PVC d=0,18	PVC d=0,25	Metal d=0,26
ε (%)	22,9	19,1	19,0	17,2	18,9
CE ^{240}Pu (c/s/g)	39,6	27,5	27,2	22,3	27,0
Detection limit (g ^{240}Pu)	$1{,}7.10^{-3}$	$2{,}5.10^{-3}$	$2{,}5.10^{-3}$	$3{,}1.10^{-3}$	$2{,}6.10^{-3}$

Detection limit about
1 mg of Pu
in the best conditions

Achievable performances with active neutron measurement

(Source at the center of a 118 liter drum - 15 minutes)

Matrice	Cellulose d=0,14	PVC d=0,25	Metal d=0,26
CE ^{239}Pu (c/s/mg)	12	0,3	4,2
Detection limit(mg ^{239}Pu)	0,09	3,4	0,3

Detection limit = few 100 µg of Pu
in the best conditions

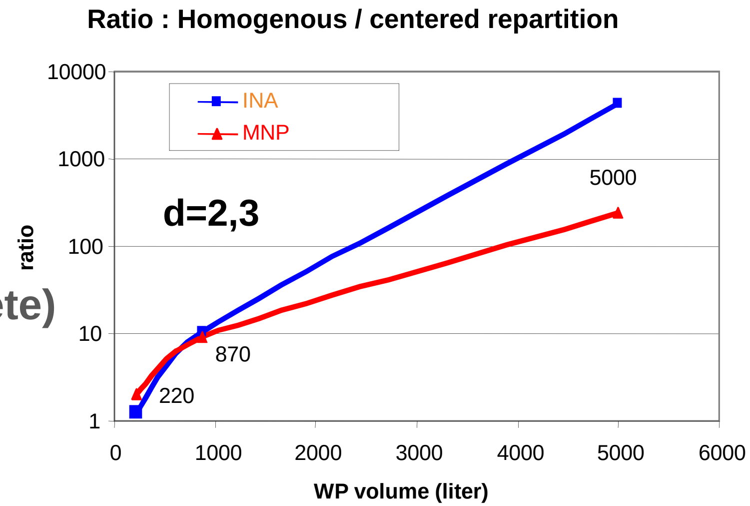
THE ADVANTAGES AND DRAWBACKS OF PASSIVE AND ACTIVE NEUTRON MEASUREMENT

Avantages :

- Direct measurement of U and Pu
- Suited to high density WP (metallic)
- Suited to irradiating WP
- Up tu 870 liters drums
- Possible to localize

Drawbacks :

- Needs isotopic composition
- Perturbated by (α,n) reactions and Cm (passive measurement)
- Impact of activity distribution
- Sensitive to Hydrogène (light materials and concrete)
- Sensitive to neutron absorbers (B, Cl)
- Expensive (case of active measurement)



X IMAGING

- **The X imaging allows an examination of the internal structure of the waste package to check :**

Geometric criterias: Thickness, centering, shielding, filling level
- Spatial resolution 2 mm (drums) 1 cm (bulk containers)

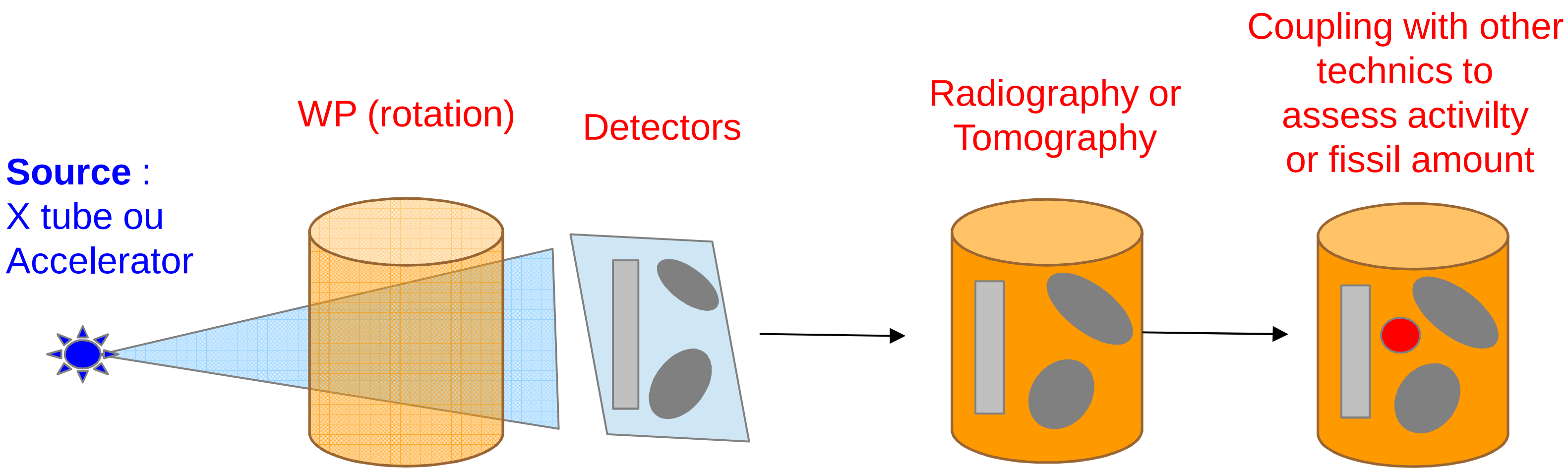
The homogeneity, the presence of defects:
- Detection levels : void (cm3), cracks (2mm * cms)
- Density discrimination : few % (drums) to 10% (bulk containers)

The absence of forbidden wastes (limited to the recognition of form, density) : wood, batteries, liquids, ...

- Information on the whole volume of the WP
- Allows the reduction of uncertainties of radiological measurements
- If destructive analysis: Guide for coring or cutting

PRINCIPLE OF X IMAGING

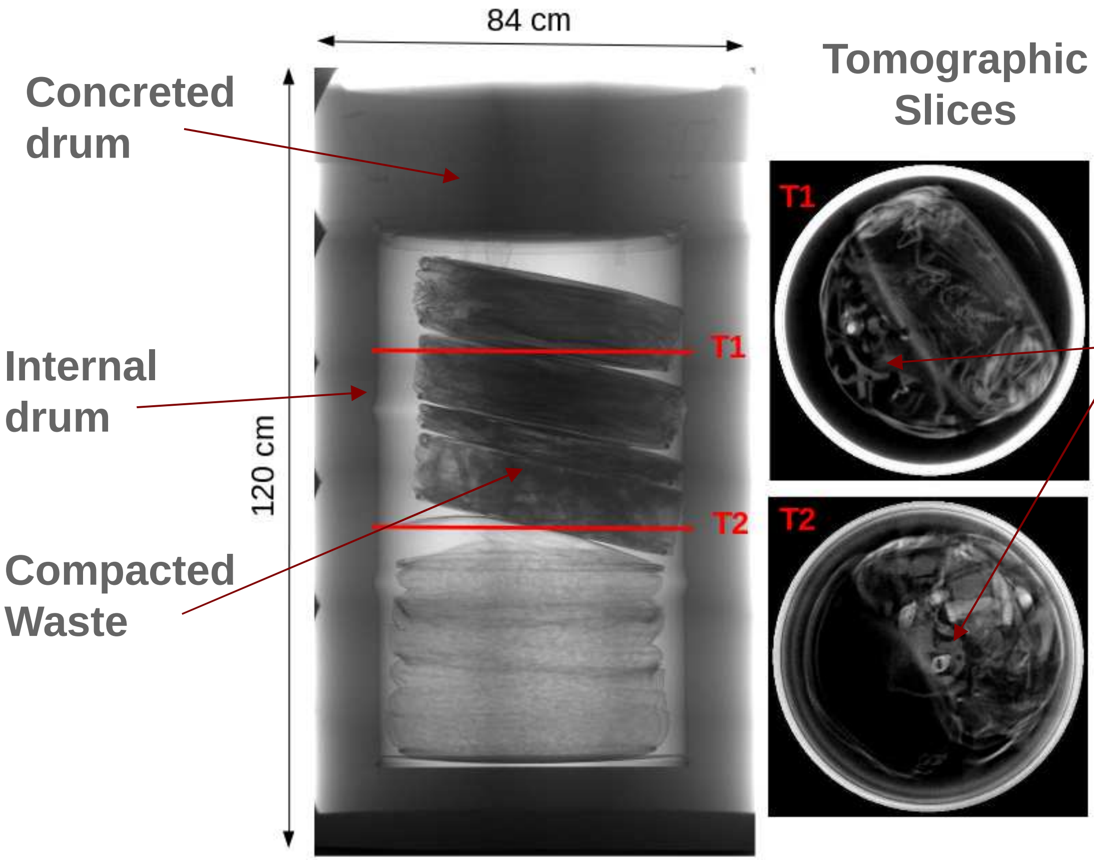
- **Measurement of the exponential attenuation of X Ray inside the Waste Package : attenuation factor μ linked to density**



HIGH ENERGY X IMAGING ON LARGE VOLUME WP

- Radiographies & Tomographies with 2D scintillant screen

Radiographies



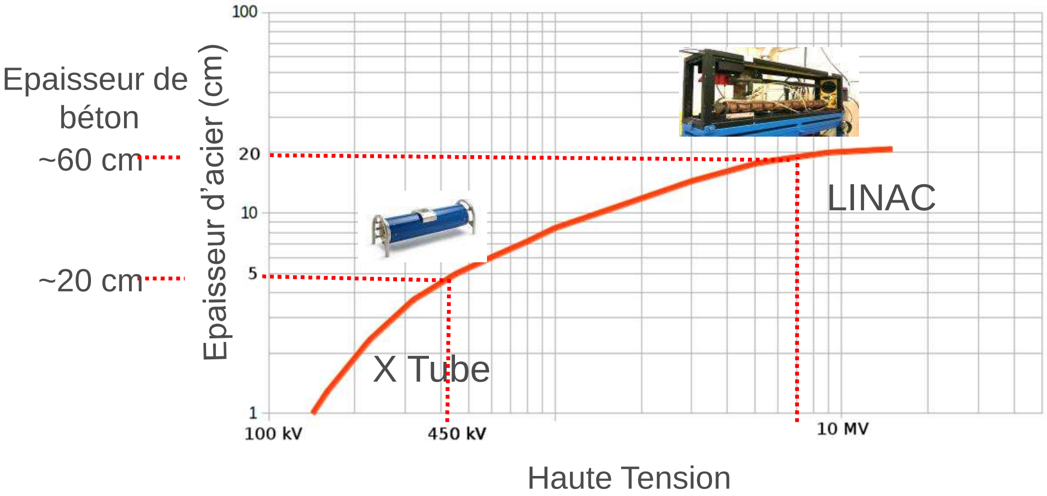
Mechanical parts identified

No density measurement (incomplete projections due to field-of-view

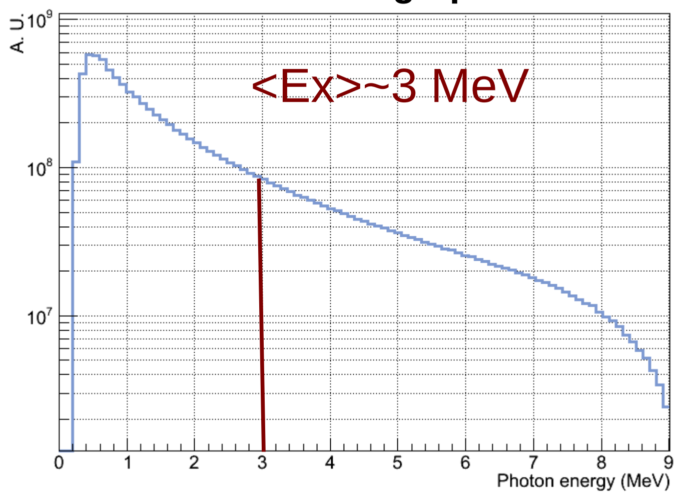
IMAGING SOURCE: X TUBE LINEAR ACCELERATOR (LINAC)

- **For medium (60 cm diam.) and large (> 1m diam.) waste drums, MegaVoltage source mandatory**

Max. Thickness with standard Imaging setup



Varian MiniLinatron 9 MeV
Eq. Dose rate: **20 Gy/min**
Pulse Freq : 300 Hz
Bremstrahlung Spectrum:



- **Radiological Safety : imaging setup placed in underground irradiation cell (ex : Cinphonie)**

THE ADVANTAGES AND DRAWBACKS OF X IMAGING

Advantages :

- Non destructive measurement to obtain a global view of the inside of a WP
- Under certain conditions : access to density, Z (R&D)
- Suited to high density and/or high volumes if high energy source available
- Suited to irradiating WP

Drawbacks :

- Mainly qualitative measurement
- High cost with high energy : source (LINAC) and underground cell
- Radiological constraints : underground cell, surveillance system

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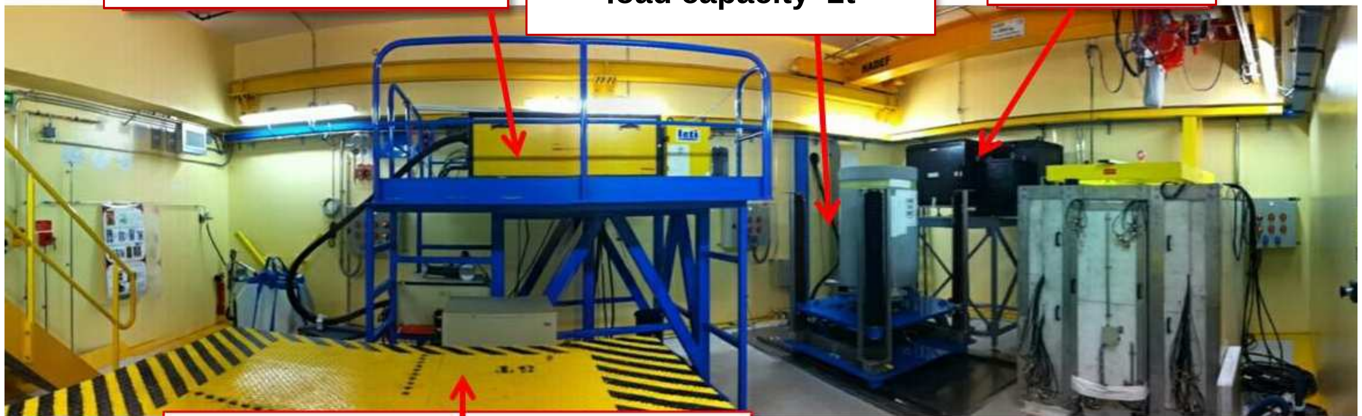
SOME SPECIFIC CONTROLS : THE "SUPERCONTROLS"

- Goal : to check the conformity of LIL-SL WP versus storage specification (CSA)
- Realized under Andra specification on samples of LIL-SL WP
- Small quantity : about 30 WP per year but …
- Very detailed controls : non destructives and destructives
- Non destructive controls :
  - X imaging
  - Activity measurement by coupling High Energy X imaging, neutron measurement and gamma spectrometry
  - Outgassing measurement
- Destructive controls :
  - Coring and sampling
  - Proficiency testings on samples : diffusion, permeability, porosity, mechanical resistance
  - Chemical analysis
  - Inventories

THE GREAT TOOLS USED FOR SUPERCONTROLS :

CINPHONIE IRRADIATION CELL – CHICADE FACILITY - CADARACHE

- Cell view



- Detection System

Scintillant screen

Mirror

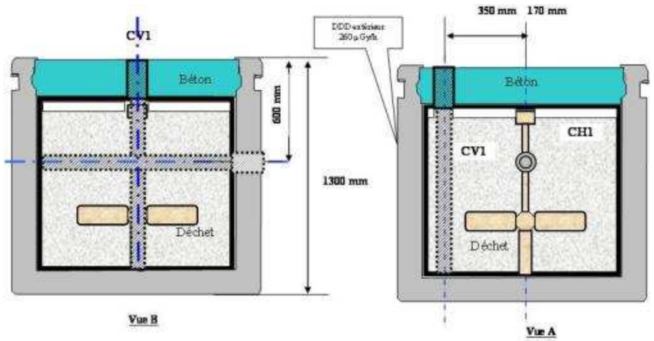
Low noise camera

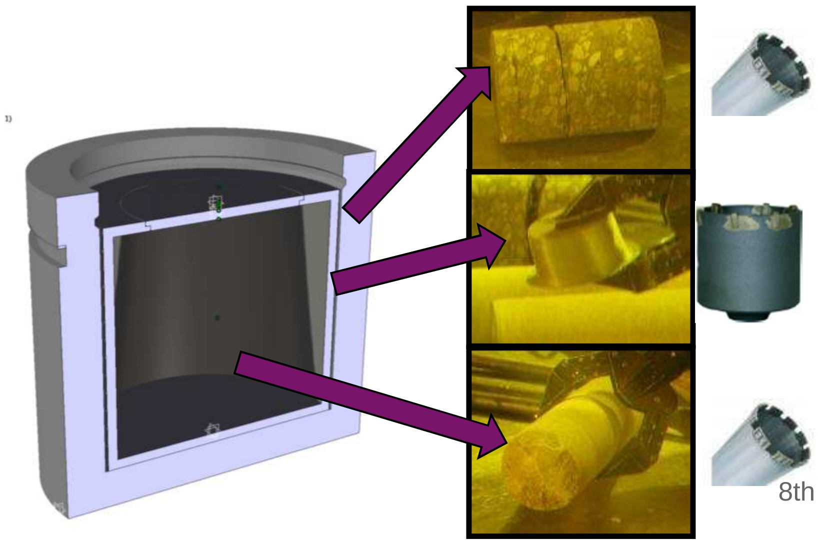
L = 9,8 m
L = 6,5 m
H = 4 m
Upper slab :
thickness = 1,5 m
of reinforced concrete

THE GREAT TOOLS USED FOR SUPERCONTROLS :

DRY CORING CELL ALCESTE- CHICADE FACILITY - CADARACHE

-> Homogeneous WP– 2m^3 – 10 tons - 11,1TBq





On coring samples :
Check of confinment properties of matrices and waste : diffusion measurement, mechanical resistance, porosity, permeability

THE 2ND LEVEL CONTROLS OR "SUPERCONTROLS"

THE GREAT TOOLS USED FOR SUPERCONTROLS :

UNDER WATER CUTTING CELL CADECOL – CHICADE FACILITY – CADARACHE

-> homogeneous or <u>heterogeneous</u> WP - 16 tons – Max Activity 250 GBq (β, γ)

+ 35GBq (α)

-> Underwater coring and <u>cutting</u>

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THE CONDITIONNED LEGACY WP

They are currently stored at Cadarache in several facilities

Mainly large size WP : from 0,5 to 2 m^3

For most of them, it contains bulk or primary waste package blocked in a mortar.

- Case of WP produced after 1990 (870L, 500L MI et coques 500L) : characterization was performed according to the principles of current production.

- For older WP : the characterization may be insufficient and historical knowledge of their production and their contents is insufficient.

Radiological characterization of these WP is delicate either with passive than active measurements because:

- Interrogators radiation have difficulties to penetrate,

- The measurable emissions (γ or neutron) are strongly attenuated by the WP itself

-> R&D program on non destructive characterization but…

Some data will remain unaccessible

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THE R&D FOR THE NON DESTRUCITVE CHARACTERIZATION OF WP :

In collaboration with Andra :

- **Active Photon Interrogation : fissile mass quantification by photofission delayed gamma rays**
- **High Energy Imaging - bi Energie : quantification of density + mass number Z**
- **the Cavity Ring Down Spectroscopy (CRDS)**

And also :

- **Passive neutron measurement with plastic scintillators instead of ^{3}He counters**

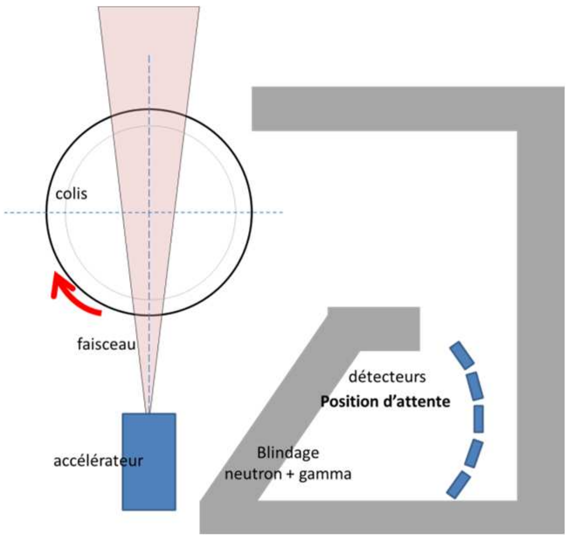
- **The investments for big setups : SATURNE LINAC + 5 tons mechanical bench**

ACTIVE PHOTON INTERROGATION

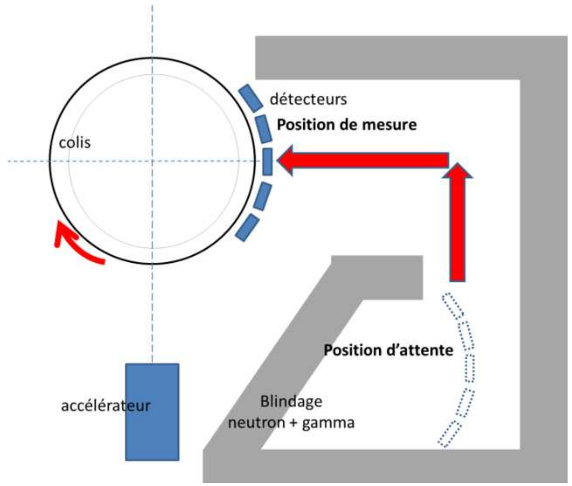
Objective : study the fissile mass quantification in large, long-lived medium activity radioactive waste packages

Principle : Active measurement : Photon interrogation. Photons are highly penetrating and allows to interrogate the centre of the WP and to produce fissions on U and Pu isotopes. Detection is done with photofission delayed gamma rays

Needs a High Energy linear accelerator : X > 15 Mev



Irradiation phase (detectors are protected)



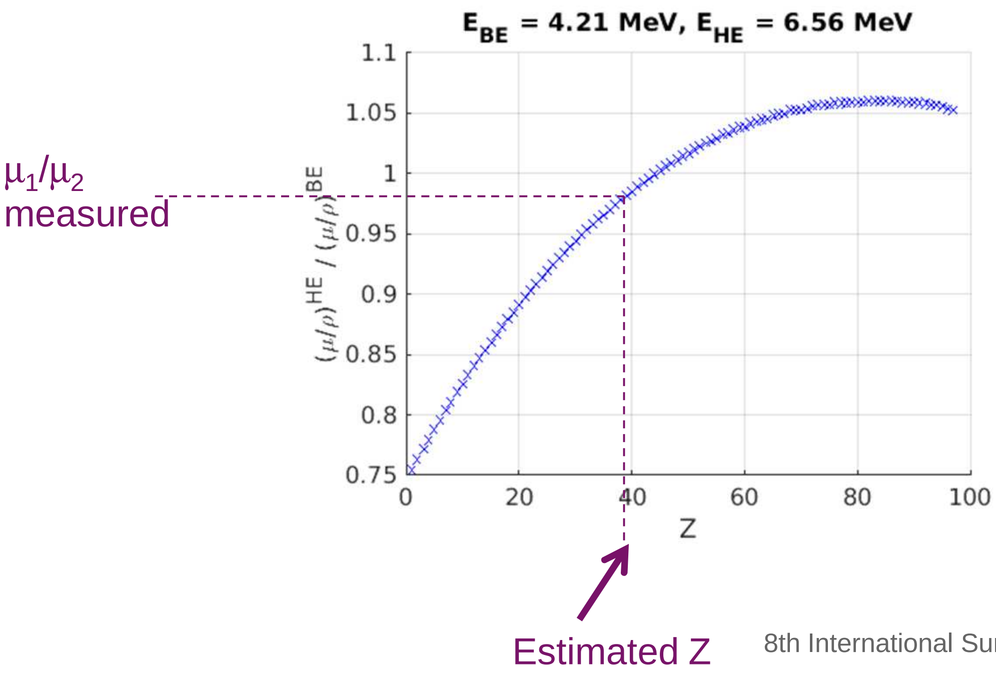
Measurement phase

HIGH ENERGY – BI ENERGY IMAGING

Objective : study the quantification of density and mass number Z in large, long-lived medium activity radioactive waste packages.

Principle : X HE imaging with two energies. Linear attenuation at 2 energies allows to quantify density and Z -> improve the discrimination of materials

$$\frac{\mu(E_1)}{\mu(E_2)} = \frac{\left(\frac{\mu}{\rho}\right)(Z,E_1)}{\left(\frac{\mu}{\rho}\right)(Z,E_2)} = R(E_1, E_2, Z)$$

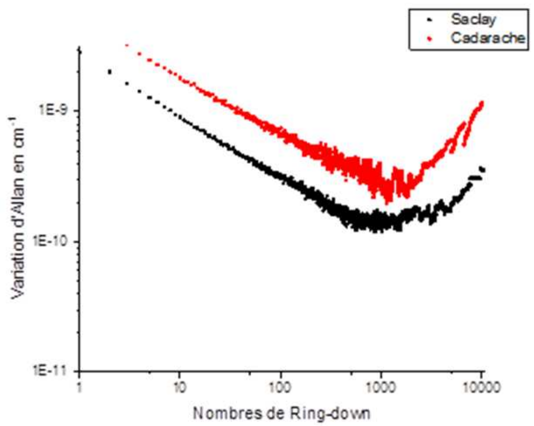


THE CRDS(*) FOR TRITIUM OUTGASSING MEASUREMENT

Objective : Tritium measurement from the degassing of the FMA waste packages by an alternative method compared to liquid scintillation.

Principle : Injection of a continuous laser beam in an optical cavity formed with two highly reflective mirrors. Molecular concentration is calculated from the measured absorption coefficient.

Requires trapping phase and preleminary concentration of Tritium



(*) Cavity Ring Down Spectroscopy

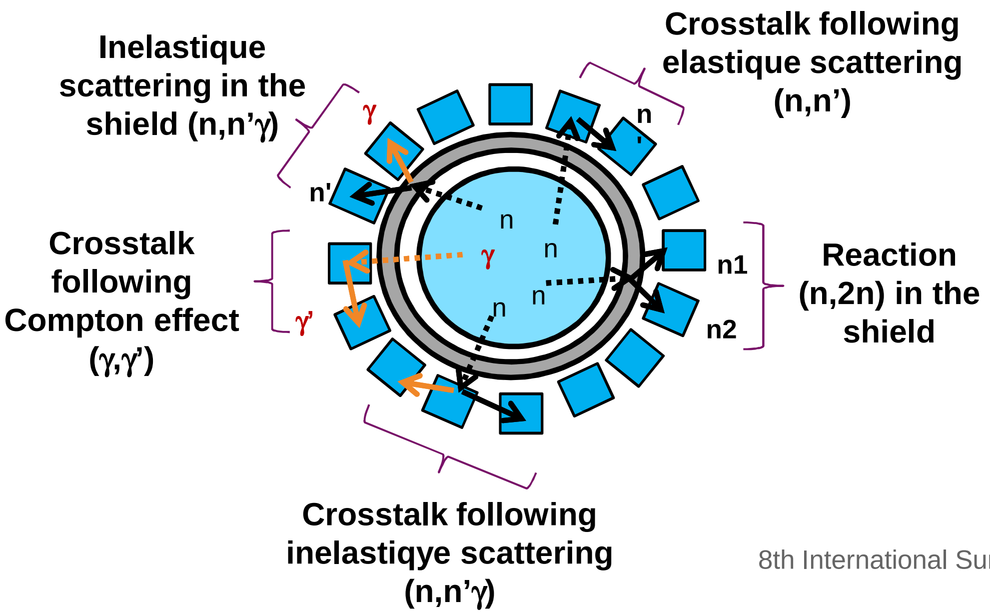
Experimental bench for CRDS

PASSIVE NEUTRON MEASUREMENT WITH PLASTIC SCINTILLATORS

Objective : Consider replacing ^{3}He counters, whose cost increases sharply, with plastic scintillators for passive neutron measurement.

- Advantage of plastic scintillators: reduced cost and sensitivity equivalent to ^{3}He
- Drawbacks : sensitivity to gamma radiation and crosstalk

PhD B.Simony : “Caractérisation du plutonium par analyse de coïncidences avec des scintillateurs organiques »



THE INVESTMENTS IN R&D TOOLS

- **Current features of the platform of imaging Cinphonie :**
  - **single energy LINAC 9 MeV, 20 Gy/min**
  - **Resolution ~ 3 mm**
  - **WP mass < 2 tons**

- **From 2018, in conjonction with R&D on active photon interrogation and bi energy interrogation**
  - **Powerful LINAC until 25 MeV, 250 Gy/min**
  - **Better resolution : 0.5 - 2 mm**
  - **Multi energy beam**
  - **Mechanical bench up to 5 tons**

- **Non-nuclear possible application: control of massive mechanical components**

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CONCLUSION

- **The characterization of nuclear waste is essential for the knowledge, storage, transportation and final disposal of waste, and to check the compliance with waste acceptance criterias.**

- **It is constantly improving:**

- Improvements by the producers by taking into consideration the characterization needs

- Improvement of nondestructive and destructive measurement technics (R&D CEA-Andra)

- **The CEA realizes key investments for R&D : irradiation cell CINPHONIE, high energy tomograph SATURNE and mechanical bench 5 tons**

- **Prospect : increase of requirement by Andra for accepting IL-LL and HA WP at Cigeo: list of 144 RN to be declared, declaration thresholds at 0.1 Bq/g, list of prohibited substances, special, limited, to be declared ……**

THANKS !!

Commissariat à l'énergie atomique et aux énergies alternatives

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