NON DESTRUCTIVE CONTROLS OF RADIOACTIVE WASTE AT CEA

8 th International Summer School on Nuclear Decommissioning and Waste Management

12-16 SEPTEMBER 2016, ISPRA
OUTLINES

• Nuclear Waste Classification
• The Characterization on Nuclear Waste at CEA
• The 2\textsuperscript{nd} level controls or “Supercontrols”
• The Legacy Waste
• R&D on measurement technics
• Conclusion
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<table>
<thead>
<tr>
<th>Massic Activity (Bq/g)</th>
<th>lower than 100</th>
<th>100 to 10^5</th>
<th>10^5 to 10^9</th>
<th>Higher than 10^9</th>
</tr>
</thead>
<tbody>
<tr>
<td>Activity level and repository</td>
<td>(1) VLLW Very low level wastes (Storage at Centre de l’Aube -CIRES)</td>
<td>(2) LIL-SLt_{1/2}&lt;31 y (Storage at Centre de l’Aube-CSA) low-level and intermediate-level, short life</td>
<td>(3) LA-LL low-level and intermediate-level, long life Intermediate storage final storage (project)</td>
<td>(5) HA High Activity Producer intermediate storage -&gt; CIGEO (project)</td>
</tr>
<tr>
<td>Type of solid wastes</td>
<td>Debris, scrap iron, plastics,… mainly from the dismantling</td>
<td>Gloves, coats, glasses, scrap iron, …</td>
<td>Cladding, hulls and end caps from spent fuel, Wastes coming from glove boxes and hot cells, filters,…</td>
<td>Vitrified Fission Products coming from the fuel reprocessing</td>
</tr>
<tr>
<td>% of volume of French radioactive waste</td>
<td>20,1%</td>
<td>LIL-SL : 68,8 % LA-LL : 7,2 % IL-LL : 3,6%</td>
<td>0,2 %</td>
<td></td>
</tr>
<tr>
<td>% of activity</td>
<td>0,000003%</td>
<td>LIL-SL &lt; 0,03% LA-LL &lt; 0,009% IL-LL : 4,98%</td>
<td>94,98 %</td>
<td></td>
</tr>
</tbody>
</table>

2nd Level controls by CEA under ANDRA Spécification
• Nuclear Waste Classification
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OBJECTIVES: Check Conformity versus interim storage, transport and final disposal specifications -> SAFETY AND PUBLIC ACCEPTANCE

- Radiological specifications
  - $\alpha$, $\beta\gamma$ activities
  - $\alpha$ 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
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, fissile 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

Desintégration ($\alpha$, $\beta^-$, $\beta^+$)

Desexcitation $\gamma$

Identification and quantification of radionuclides through his descendants

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

Standard gamma spectrometry device
100 liters WP
THE ADVANTAGES AND DRAWBACKS OF $\gamma$ SPECTROMETRY

Advantages:
- Easy to implement
- Activity of numerous $\beta/\gamma$ emitteurs
- Adapted for low density WP ($d<1,5$)

Drawbacks:
- Unsuitable for high volumes and/or high density WP
- Unsuitable for low energy gamma rays (case of actinides U and Pu)
- Needs a transfer function to take into account:
  - Density distribution
  - Activity distribution

$$A_{WP} = A_{measured \ outside \ WP}/ FT(E_{\gamma}, WP)$$
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
: $^{238}\text{Pu} + ^{240}\text{Pu} + ^{242}\text{Pu} + (^{244}\text{Cm}, ^{241}\text{Am} \ldots)!$
• $\text{En} (^{240}\text{Pu})= 1020 \text{ n.s}^{-1}.\text{g}^{-1}$
• $\text{En} (^{238}\text{Pu})= 2590 \text{ n.s}^{-1}.\text{g}^{-1}$
• $\text{En} (^{244}\text{Cm})= 1,08 \times 10^7 \text{ n.s}^{-1}.\text{g}^{-1}$ -> a small quantity of Cm can hide Pu isotopes !!!

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

• Global counting or coincidence ((α, n) rate)
ACTIVE NEUTRON MEASUREMENT

Global measurement of neutron emission after activation: induced fission by thermal neutrons
-> suited to the measurement of fissile 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
  235U + 239Pu + 241Pu (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)

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

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)

<table>
<thead>
<tr>
<th>Matrice</th>
<th>Cellulose d=0,14</th>
<th>PVC d=0,25</th>
<th>Metal d=0,26</th>
</tr>
</thead>
<tbody>
<tr>
<td>CE $^{239}$Pu (c/s/mg)</td>
<td>12</td>
<td>0,3</td>
<td>4,2</td>
</tr>
<tr>
<td>Detection limit (mg $^{239}$Pu)</td>
<td>0,09</td>
<td>3,4</td>
<td>0,3</td>
</tr>
</tbody>
</table>

Detection limit = few 100 µg of Pu in the best conditions
The non destructive analysis

The advantages and drawbacks of passive and active neutron measurement

Advantages:
- Direct measurement of U and Pu
- Suited to high density WP (metallic)
- Suited to irradiating WP
- Up to 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 $\mu$ linked to density

**Source:**
- X tube ou Accelerator

**WP (rotation)**

**Detectors**

**Radiography or Tomography**

**Coupling with other technics to assess activity or fissil amount**
HIGH ENERGY X IMAGING ON LARGE VOLUME WP

- Radiographies & Tomographies with 2D scintillant screen

**Radiographies**

Concreted drum

Internal drum

Compacted Waste

84 cm

Tomographic Slices

Mechanical parts identified

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

120 cm

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

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

*Bremstrahlung Spectrum:*

\[ \langle E_X \rangle \approx 3 \text{ MeV} \]

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 2ND LEVEL CONTROLS OR “SUPERCONTROLS”

THE GREAT TOOLS USED FOR SUPERCONTROLS:
CINPHONIE IRRADIATION CELL – CHICADE FACILITY - CADARACHE

Cell view

- LINAC 9 MeV
- Mechanical bench load capacity 2t
- 2D Screen
- Elevator - load capacity 5t

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

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THE GREAT TOOLS USED FOR SUPERCONTROLS:

DRY CORING CELL ALCESTE- CHICADE FACILITY - CADARACHE

--> Homogeneous WP– 2m³ – 10 tons - 11.1TBq

On coring samples:
Check of confinement 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 heterogeneous WP - 16 tons – Max Activity 250 GBq (β, γ)
+ 35GBq (α)

-> Underwater coring and cutting
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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 m³
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 DESTRUCTIVE 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.

\[ \frac{\mu(E_1)}{\mu(E_2)} = \frac{(\frac{\mu}{\rho})(Z,E_1)}{(\frac{\mu}{\rho})(Z,E_2)} = R(E_1, E_2, Z) \]

\( E_{BE} = 4.21 \text{ MeV}, \ E_{HE} = 6.56 \text{ MeV} \)

\( \mu_1/\mu_2 \) measured

Estimated 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 preliminary concentration of Tritium

(*) Cavity Ring Down Spectroscopy
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”

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Diagram:
- Inelastique scattering in the shield ($n,n'\gamma$)
- Crosstalk following elasitique scattering ($n,n'$)
- Crosstalk following Compton effect ($\gamma,\gamma'$)
- Reaction ($n,2n$) in the shield
- Crosstalk following inelastique scattering ($n,n'\gamma$)
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 conjunction 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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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 !!