NEW DEVELOPMENTS OF AUTORADIOGRAPHY TECHNIQUE TO IMPROVE ALPHA AND BETA MEASUREMENTS FOR DECOMMISSIONING FACILITIES

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WORKSHOP ON “CURRENT AND EMERGING METHODS FOR OPTIMISING SAFETY AND EFFICIENCY IN NUCLEAR DECOMMISSIONING”

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Overview

- Context and LASE laboratory: Analytical Support to Facilities Laboratory, FRANCE

- Digital Autoradiography Technique

- Radionuclide mapping using geostatistics

- Digital Autoradiography in support of sampling processes

- Digital Autoradiography: improvement of the selectivity and new researches

- Conclusions
Overview

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Conclusions
**Who?** The National Radioactive Waste Management Agency **ANDRA** is in charge of the long-term management of radioactive wastes in France

Classification of radioactive wastes as a function of their management

<table>
<thead>
<tr>
<th>Activity - Half-life</th>
<th>Very short-half-life &lt; 100 days</th>
<th>Short half-life ≤ 31 years</th>
<th>Long half-life &gt; 31 years</th>
</tr>
</thead>
<tbody>
<tr>
<td>Very low level waste</td>
<td>Surface disposal facility (CSTFA Aube facility)</td>
<td></td>
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</tr>
<tr>
<td>Low level waste</td>
<td>Management by radioactive decay</td>
<td>Surface disposal facility (CSFMA Aube facility)</td>
<td>Near-surface disposal facility being studied</td>
</tr>
<tr>
<td>Intermediate level waste</td>
<td></td>
<td></td>
<td>Deep disposal facility at 500 meters being studied</td>
</tr>
<tr>
<td>High level waste</td>
<td></td>
<td></td>
<td>Deep disposal facility at 500 meters being studied</td>
</tr>
</tbody>
</table>
CONTEXTE: ORIGINE DES DECHETS NUCLEAIRES FAIBLES ET INTERMEDIERS

- Industries nucléaires (EDF, AREVA)
- Centres de recherche nucléaires (CEA)
- Centres médicaux

Différents types de matériaux
Why? ANDRA requests a characterization of nuclear wastes and specifies acceptance criteria for packages that waste producers have to respect.

Why? Characterization is one of the essential step in decommissioning projects.
NetLab associated to NetLab = A CEA network of analytics and experimental tools (laboratories located on CEA sites) to assist nuclear operators during characterization programs associated to dismantling process.
Different characterization techniques for low and intermediate level wastes.
- Destructive analysis (sample = 1 g)
- Radiochemistry
- Alpha, Gamma, LSC
- Elemental analysis

- In situ technique: Autoradiography
CHARACTERIZATION OF LOW AND INTERMEDIATE LEVEL NUCLEAR WASTES

Mission of Analytical support to facilities Laboratory at CEA-Saclay

ANALYSIS OF ELEMENTS (RADIOACTIVE OR NOT) PRESENT AT TRACE LEVEL IN VARIOUS MATRICES

Chemical and radiological characterization of radioactive materials

Toxic elements, organic ligands, TOC, anions, cations

Radionuclides determined after radiochemistry (A~0.1Bq.g⁻¹)

- $^3$H, $^{14}$C, $^{36}$Cl, $^{55}$Fe, $^{59}$Ni, $^{63}$Ni, $^{69}$Sr, $^{93}$Mo, $^{93}$Zr, $^{93m}$Nb, $^{94}$Nb, $^{108m}$Ag, $^{121m}$Sn, $^{129}$I, $^{153}$Sm, $^{241}$Pu,
- $^{238}$ et $^{240}$Pu, $^{239}$Pu, $^{241}$Am, $^{243}$Am, $^{232}$U, $^{234}$U, $^{235}$U, $^{238}$U

Wipes, technological wastes (tissues, gloves), concretes, ion exchange resins embedded in organic polymers, metals, muds, sludges, oils...
Main Radionuclides that must be investigated in priority

- Easy to measure:
  - gamma emitters

- Difficult to measure:
  - Alpha emitters
  - Beta emitters
  - H-3
  - C-14
  - Cl-36
  - Sr-90
CHARACTERIZATION IS VERY IMPORTANT FOR D&D PROJECTS

Destructive techniques need less than 1 g of sample for digestion process

Lots of Radwastes must be characterized

Innovation is required for in situ techniques, for techniques allowing a better sampling process
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Conclusions
Radiography

Source → Detector

Autoradiography

Detector

Interests of biologists: H, C, S, ... radionuclides difficult to measure
Screens can be reused hundreds of times

1. Screen initialization
2. Screen Deposit
3. Exposure Time
4. Screen withdrawn
5. Screen scan
6. Calculations
• Semi quantitative values are achievable.

• Repeatability corresponds only to a few percent

Same H-3 analysis
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Decommissioning requires characterization at very low level. Requirement of in situ technique to improve sampling process. 1 g sample needed for destructive analysis.

Decommissioning and dismantling

Mélusine, Grenoble
Sampling process on floor

A grid corresponding to 70 screens = 5 m²

Radioactivity image, here C-14
Localisation of the sample for destructive analysis is clear

Autoradiography with raw images
OTHER EXAMPLE FOR URANIUM MEASUREMENTS

Screen on surface

0.3 m

Contamination shape is also an important parameter for stakeholders.

1.8 m

« Hot » spots clearly appears

Maximum activity found around 6 Bq/cm²

Maximum activity found around 6 Bq/cm²
Current development: geostatistical approach

Current investigation rate:
2 weeks / 100 m²

Conclusion:
Contamination accurate location
Representative and limited sampling enabling
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How to collect 1g or even less?
Preparation of homemade standards for beta analysis: CONCRETES

Concrete spiked at the beginning of the preparation of the cement

Can be spiked with numerous radionuclides H-3, C-14, Gamma emitters…

Homogeneity on concrete surface (here H-3 and C-14)

Raw signal

Numerical treatment to evaluate the homogeneity by digital autoradiography
Sampling process for tritiated wastes

Destructive measurement of H-3 is done by pyrolysis followed by Liquid Scintillation Counting (LSC)

Sample = 1 g
Only 3 wipes among 7 contained Uranium.

After studies with a destructive method: wipe digestion followed by alpha spectrometry and/or ICP-MS, detection limit was determined at 0.2 Bq/wipe for Uranium (more sensitive than alpha spectroscopy).
After surface analysis on D&D sites, determination of 3D contamination

Core made of concrete containing C-14

Activity on surface

Interesting sampling

Activity in depth

Image in real scale
Study of alumina beads containing I-129 located inside a plastic tube

Plastic deposited on a film for 15 minutes

DA as efficient solution for sampling process
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Intermediate precision
example of $^{90}\text{Sr}/^{90}\text{Y}$:
2 point sealed sources from LASE
5 sealed filtrates from LASE
1 point sealed source from INSTN
2 area sources from INSTN

$S_2/S_1 = 33 \pm 6 \%$
$S_3/S_2 = 59 \pm 6 \%$
Screen stacking method (2/2)

Radionuclides “signatures”
NEW RESEARCHES ON CCD TECHNIQUES

- Screen technique
  
  Very easy to use, lots of applications
  The information comes after the exposure time

- CCD development
  
  Technique coming from biological applications
  Main advantage: commercial systems already exist
  Detection is obtained simultaneously
Trace amount of uranium: around 1 Bq/cm²
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Conclusions
Digital Autoradiography was used first because the technique is sensitive to difficult to measure radionuclides.

Geostatistics methods can be very useful to interpret high amount of data and to provide extrapolated data.

Autoradiography appears to be a very interesting technique for sampling processes.
Different developments are currently in R&D for mapping applications, to improve the sensitivity, to try to find possibilities to improve the selectivity and to develop other devices.

The LASE laboratory participates to numerous intercomparison tests where no sampling problem is usually encountered however it is totally different for radiochemical analysis required for solid radwastes matrices.
Thank you for your attention