Multi Analyses on Radioactive Samples

A BEAM LINE DEDICATED TO RADIOACTIVE SAMPLES AT SOLEIL

RADIATION SAFETY ISSUES
Contents

• Presentation of the Mars Beam Line
• Radioactives Samples
• Containment
• Specific Shieldings
• Status and forecast
Characterizing radioactive samples for environmental, health and energy applications:

- **Solution chemistry** (organic and inorganic ligands and natural organic matter)
- **Solid state physics and chemistry** (structural properties of nuclear fuel, matrix for nuclear waste storage, stability of actinide alloys, study of material for current and future nuclear facilities)
- **Interface chemistry** (RN retention in nuclear waste storage context, RN migration in the geosphere)
- **Biology** (retention & migration of RN in nuclear waste storage context, study of complexes of medical interest, biological effects due to radiations, nuclear toxicology studies)
THE MARS BEAM LINE

- Set on a SR Bending Magnet, electron beam $E=2.75$ GeV, $B_z=1.71$ T (critical energy : 8.6 keV)
- Synchrotron Xray Energy range 3.5 keV – 36 keV
- Two experimental stations
  - CX2: High Resolution Diffraction spectroscopy (HRXRD)
  - CX3: Absorption spectroscopy (XAS, TXRD) and
  - Xray Fluorescence (XRF)
THE MARS BEAM LINE

- Versatile sample environment during experiment → 5 experimental families
  - F0: Normal conditions, no external constraint ($P_{\text{atm}}, T$);
  - F1: High temperature and pressure (<2000K, up to 100GPa);
  - F2: Low temperature (10K to 300K);
  - F3: Chemical and Redox reactions on liquid samples (300K to 450K)
  - F4: High temperature (300K to 1800K);

- Total activity able to be hosted on MARS BL: 185 GBq (max per sample: 18.5 GBq)
GENERAL LAYOUT OF THE MARS BEAMLINE
GENERAL LAYOUT OF THE MARS BEAMLINE

CX2: diffraction station

CX3: absorption & fluorescence station
2 EXPERIMENTAL STATIONS
SPECIFIC EQUIPMENTS REQUIRED

• Special fireproof layer on metallic walls and ceilings
• Fireproof airtight feedthroughs for cables and pipes
• Glued metal sheets, special paint and separate gantry supporting servitudes
SPECIFIC EQUIPMENTS REQUIRED

• Absolute filters barriers on both ventilation networks and outlets ventilators connected to external chimney on the top of synchrotron building

• Dynamic containment from 2 independant ventilation networks (breathable air and process)

• Complex fire detection system connected with ventilation system and able to close fire shutters and doors
SPECIFIC EQUIPMENTS REQUIRED

• Changing room and primary decontamination block (hands & arms)
• Specific working suite to be wear in controlled area
• Users and staff control before exiting controlled area by surface contamination monitors ($\alpha, \beta\gamma$) for cloths, hands and feet
SPECIFIC EQUIPMENTS REQUIRED

• Sample-holder control desk (low activity samples)

• Glove box for safety controls and operation on sample holders

(eventual change of 2\textsuperscript{nd} containment barrier for specific experiment)
SPECIFIC EQUIPMENTS REQUIRED

• Emergency calling network system closed to each working station

• Continuous air area monitoring system for alpha and beta particles

ICAM™ probes and RADACS™ Display (CANBERRA)
GENERAL RADIATION SAFETY GOALS
AT SOLEIL

• SOLEIL Staff and Users are Non classified workers regarding ionizing radiation hazards in the Experimental Hall

• Assess that in all conditions the radiation doses outside the tunnels are below the maximum dose authorized for non classified people:
  - Annual Dose < 1 mSv ⇔ average dose rate < 0.5 μSv/h

• Exposed workers limited to the one accessing tunnels during shutdown → below B class annual dose limits (6 mSv/y)
RADIATION SAFETY GOALS SPECIFIC TO MARS BEAM LINE

- The experiments on radioactive samples at MARS should have no effect on SOLEIL operations for both machine and BL at any time.
- No direct contact between SOLEIL staff or users and the samples. All samples preparation has to be done in user’s laboratory.
- No contamination hazard affordable for MARS operation → particular attention on containment!
- MARS team permanent members should’t be classified higher than B category.
RADIATION SAFETY GOALS SPECIFIC TO MARS BEAM LINE

• What ever the radioactivity welcomed on MARS, doserates outside the BL rooms have to be below 0.5 µSv/h all around
• Containment designed and tested by users to ensure efficiency during experiment in all conditions
• Acceptance given by SOLEIL radiation safety and clearance officially given after Regulation Authority green light
• Individual annual dose objective is to reach the same as SOLEIL exposed staff eg. < 2 mSv/year
RADIOACTIVE SAMPLES

- Only Solids and liquids are allowed on MARS beam line (incl. sintered powder)
- Gaseous samples and powder are strictly forbidden on MARS
- About 400 different isotopes with activity limited up to 18.5 GBq per sample / sample holder
- Limitation also coming from corresponding dose rates and contamination levels in case of leakage of the sample containment
RADIOACTIVE SAMPLES

Typical examples:

• Environmental samples (solid in solution and liquid) from geological and biology studies, from rivers, mines, volcanos, etc) \( \rightarrow \) few Bq to \( \sim \) 1-2 kBq U and/or Th

• Doped glass for Nuclear Waste management studies, long live and high activity \( \sim \) 100 kBq to 2 GBq

• Structure material in nuclear power plants (activated metallic alloys samples) \( \rightarrow \) few kBq up to \( \sim \) 1GBq

• New and spent fuel samples (sections of \( \text{UO}_2 \) fuel pellets, 23mg) \( \rightarrow \) up to 3 GBq
ACTIVITY THRESHOLDS PER ISOTOPES

- Doserate calculations performed with MERCURAD™ and MCNPX codes for unshielded and shielded sample holders to define maximum acceptable activity to stay below 0.5 µSv/h outside MARS enclosure

- Positions investigated:
  - Storage in LEN;
  - CX2 station;
  - CX3 station
# Activity Thresholds per Isotopes

**ACTIVITE MAXIMUM AUTORISEE SUR LA LIGNE MARIE PAR RADIOISOTOPES**

<table>
<thead>
<tr>
<th>Radioisotope</th>
<th>Activity Threshold</th>
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<tbody>
<tr>
<td>$^{60}$Co</td>
<td>25 MBq</td>
</tr>
<tr>
<td>$^{137}$Cs</td>
<td>3.8 GBq</td>
</tr>
<tr>
<td>$^{239}$Pu</td>
<td>18.5 GBq</td>
</tr>
</tbody>
</table>

![Graphical representation of activity thresholds](image-url)
SAMPLE CONTAINMENT PRINCIPLE

- If contamination risk is present because of the sample itself and/or because of the experience, then 3 independent containment barriers are required between the sample and the environment.
  - 2 1st around the sample
  - 3rd beam line airtight rooms
SAMPLE CONTAINMENT EXAMPLE 1

- PEEK dome sample holder (Brucker) for diffraction experiments
- Can be loaded with up to four samples
- For solid samples, mainly of low activity level and for normal conditions experiments (F0 family)
SAMPLE CONTAINMENT EXAMPLE 2

F3: liquid samples and redox reactions

10cm³ cell containing the sample solution
Sealed electrodes
Top cover

Complete 10cm³ cell
External support with 3 kapton windows for absorption and fluorescence measurements

1st barrier
2nd barrier
Both able to resist high acid concentration

Complete setup mounted on CX3 station
SAMPLE CONTAINMENT EXAMPLE 3

- Multi samples holder loaded with up to 12 different samples, liquid or solid, or both
  → Typical example: natural or depleted uranium solutions ($10^0$ to $10^2$ Bq/sample) with concentration gradient
SAMPLE CONTAINMENT EXAMPLE 4

- Nuclear fuel pellet sample (PWR UO₂), new or spent (up to 90 GW.d/t after at least 1(5) years cooling)
- Typically cylindrical, 2mm diameter and 0.7mm thick
- About 23mg – 3 GBq
- The Highest dose rate source:
  - Up to 2 mSv/h (gamma H*(10) – 30cm)
  - Up to 100 mSv/h (beta Hp(0.07) – 30cm)
SAMPLE CONTAINMENT EXAMPLE 4

• No direct contact to the sample holder for both transfer and radiological controls
• Necessity to have a shielding around the sample holder during the whole experiment
BASIC SCHEME OF MEASUREMENT

Sample or station shielding

Synchrotron beam

Radioactive sample

Analyser cristal

détecteur

External shielding
EXPERIMENTAL STATION SHIELDING

- CX2
Small movable lead blocks push away by the analyser and recovering initial position by a spring to close the shielding opening.
EXPERIMENTAL STATION SHIELDING

Diffractomètre

Compléments latéraux arrière

Only able to be open if sample holder shielding is locked on

CX2 shielding sliding door

analyser

Sample holder shielding
EXPERIMENTAL STATION SHIELDING

- Sample holder
- Analyser opening
- CX2 shielding door
- SH shielding door
- SH shielding
- Plateau 2 theta
- 3cm Pb diffractometer shielding
- Diffusemè}

RadSynch’13 – NSLS / BNL
8-10th May 2013
Jean-Baptiste PRUVOST
ADDITIONAL SHielding REQUIREMENTS

• Assuming Safe Storage of a maximum of 6 spent fuel samples at same time with their transport shielding (3cm lead)
• MERCURAD™ calculations
• Safe shielded with 5cm of lead panels and a 6.5cm on the SW
OPERATION STATUS

• MARS is already running operation with radioactive samples below exemption limit thresholds (mainly F0 but also few F1, F3 and F4) since 2010 with specific autorisation from Regulation Authority limited to few isotopes (~20)

• 27 experiments achieved up to now (12/12) with radioactive samples with satisfying scientific results (more than 12 pub.)
Next:

- In September 2013, first experiment above exemption threshold is scheduled if ASN autorisation is given.
- Nominal operation with radioactive samples is foreseen before the end of 2015 with 28 experiments per year representing about 100 samples hosted and analysed on MARS beamline.
THANK YOU FOR YOUR ATTENTION