Improving thermal power measurements of large radioactive waste packages before repository in burial sites

B. Hay

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- **Nuclear energy in France**
  - 58 nuclear reactors providing 77% of the electricity
  - 3 main “high level and long lived waste” generators (EDF, AREVA and CEA) representing 92% of the total radioactive waste produced
  - Over 1,100 small generators (hospitals, universities, industry…)
  - ≈ 2 kg of radioactive waste produced annually per inhabitant
- Nuclear waste management in France
  - Andra: National Radioactive Waste Management Agency
    - Collect radioactive elements and assess nuclear waste
    - Manage and monitor the storage for all the nuclear waste
  - Classification of French radioactive waste

### Classification of French radioactive waste

<table>
<thead>
<tr>
<th>ACTIVITY</th>
<th>Very short-lived Half-life &lt; 100 days</th>
<th>Short-lived Half-life ≤ 31 years</th>
<th>Long-lived Half-life &gt; 31 years</th>
</tr>
</thead>
<tbody>
<tr>
<td>Very low level</td>
<td>Stored to allow radioactive decay on the production site then disposed of adopting conventional solutions</td>
<td>Surface disposal facility (Very-low-level radioactive waste disposal facility in the Aube district)</td>
<td>Shallow disposal facility (studied in accordance with the Act of 28 June 2006)</td>
</tr>
<tr>
<td>Low level</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Intermediate level</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>High level</td>
<td></td>
<td></td>
<td>Reversible deep geological disposal facility (studied in accordance with the Act of 28 June 2006)</td>
</tr>
</tbody>
</table>
Nuclear waste management in France

- Highly radioactive long-lived (HLW) and intermediate-level long-lived (ILW-LL) waste
  - French Parliament chose in 2006 deep disposal for ensuring the long-term safety of radwaste
  - Cigéo geological disposal facility will serve as a repository for HLW and ILW-LL waste in 2025
  - Wastes will be buried some 500 meters below ground in an impermeable argillaceous rock
**HLW and ILW-LL quality control**

- **Inspection on radwaste package before storage in Cigéo (radioactivity, thermal power...)**
  - Safety of radwaste repository underground facility depends on waste thermal power
  - *Avoid temperature above 100 °C in geological medium to maintain argillite properties*

- **Acceptance criteria for repository**
  - Intermediate-level long-lived waste (ILW-LL) : Thermal power from 1 W to 50 W per package
  - Highly radioactive long-lived (HLW) : Thermal power up to 500 W per package

<table>
<thead>
<tr>
<th>HLW</th>
<th>ILW-LL</th>
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<tbody>
<tr>
<td>175 l</td>
<td>2 m³</td>
</tr>
<tr>
<td>h = 1.015 m</td>
<td>4500 kg to 6500 kg</td>
</tr>
<tr>
<td>φ = 0.498 m</td>
<td>h = 1.300 m</td>
</tr>
<tr>
<td>300 kg to 450 kg</td>
<td>φ = 1.400 m</td>
</tr>
<tr>
<td>Stainless steel</td>
<td>Concrete</td>
</tr>
</tbody>
</table>

⇒ Thermal power should be known with an uncertainty better than 5 %
General background: Project “MetroDecom”

- Thermal power measurement of radioactive waste packages - State of the art
  - Evaluation by applying non-destructive radioassay methods
    - Calculation of the thermal power of HLW and ILW-LL packages from their radioactive spectra
      - Methods sensitive to attenuation and heterogeneity problems due to matrix effects
  - Thermal characterization by direct measurement methods (calorimetry)
    - Some measurements are already performed using commercial calorimeters (Setaram...)
      - Optimized for low thermal power (less than 1 W)
      - Not designed for large samples
      - Uncertainties of measurements are not known

Necessity to improve the metrology of thermal power measurements by calorimetry for this type of applications
« **MetroDecom** » **Project**

European joint research project aiming to solve metrological issues related to the decommissioning of nuclear sites

Develop methods and facilities traceable to SI for:

- Characterisation and selection of radioactive wastes coming from decommissioning process of nuclear facilities
- Monitoring of radioactive waste repositories

**Partners** (14)

<table>
<thead>
<tr>
<th>Partner</th>
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<tbody>
<tr>
<td>CMI</td>
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<td>LNE</td>
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<td>ANDRA</td>
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<td>EDF</td>
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<td>STUK</td>
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<td>ENEA</td>
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<td>IFIN-HH</td>
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<td>NPL</td>
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<td>PTB</td>
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<tr>
<td>SCK·CEN</td>
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<tr>
<td>CEA</td>
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<tr>
<td>MIKES</td>
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<td>ENVINET a.s.</td>
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<td>JRC</td>
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Porto, 31 August - 04 September 2014
**MetroDecom** Project

- **Structure of the project**
  - WP1: Characterisation of materials present on decommissioning sites
  - WP2: Measurement facility for waste segregation
  - WP3: Implementation of free release measurement facility on decommissioning site
  - WP4: Radioactive waste repositories monitoring
  - WP5: Measurement facility for waste segregation
  - WP6: Creating impact
  - WP7: Management and coordination

**Scientific and technical objectives of the task 4.5**

- Design a calorimeter for the measurement of thermal power of nuclear waste packages
  - *Method for on-site measurements traceable to SI*
  - *Calorimetric method for real size packages*
  - *Thermal power range: 1 W to 500 W*
  - *Uncertainty of measurements: < 5%*
First calorimetric measurements (1780) \[1\]

- Invention of an isothermal calorimeter
  - Measurement of the quantity of melted ice
    - Measurements of specific heat, enthalpy of fusion and of reaction
    - Study of the heat produced by the respiration of a live guinea pig
  - Energy determined by comparison with the quantity needed to lower the temperature of one pound of water (489.5 g) from 80 to 0 degree (Réaumur temperature scale)
  - Accuracy of heat capacity measurements estimated to 1/60\(^{th}\) (1.7 %)

\[1\] A.L. de Lavoisier et P.S. de Laplace « Mémoire sur la chaleur » (1780)
- Reference calorimeter for gas calorific value measurements
  - Measurement of GCV by isoberibolic calorimetry
  - Determination of $C_{cal}$ for each combustion ⇒ Calibration by electrical substitution

$$GCV = \frac{C_{cal} \cdot \Delta T_{ad} + K}{m_{gas}}$$

- Thermistor
- Burned gases
- Burner
- Water
- Heating wire
- Isothermal jacket at 25 °C
- $O_2 + Ar$
- Gas


- LNE mean value = 55518 J.g⁻¹
- Stand. dev. = 0.03 %
- ISO 6976 value = 55516 J.g⁻¹
- U (k=2) = 0.09 %
- Calorimeter for enthalpy of fusion measurement up to 1000 °C
  - Measurement directly traceable to SI with $U(k=2) < 0.5\%$
  - Calibration and $\Delta H_{\text{fus}}$ meas. in the same run ⇒ Conservation of the exp. conditions

- Application to $\Delta H_f$ measurements of In, Sn and Ag

<table>
<thead>
<tr>
<th>$\Delta H_f$ (J·g$^{-1}$)</th>
<th>NIST</th>
<th>PTB</th>
<th>LNE</th>
</tr>
</thead>
<tbody>
<tr>
<td>Indium</td>
<td>$28.51 \pm 0.19$ (0.7 %)</td>
<td>$28.64 \pm 0.11$ (0.4 %)</td>
<td>$28.64 \pm 0.11$ (0.4 %)</td>
</tr>
<tr>
<td>Tin</td>
<td>$60.22 \pm 0.19$ (0.3 %)</td>
<td>$60.24 \pm 0.16$ (0.3 %)</td>
<td>$60.22 \pm 0.22$ (0.4 %)</td>
</tr>
</tbody>
</table>

$\text{Sens (} \mu \text{V} \cdot \text{W}^{-1}) = \frac{A_c (\mu \text{V} \cdot \text{s})}{Q (\text{J})}$

$Q_f (\text{J}) = \frac{A_f (\mu \text{V} \cdot \text{s})}{\text{Sens (} \mu \text{V} \cdot \text{W}^{-1})}$

$e^{\text{m}}$

$\text{Heating rate : } 15 \text{ mK.min}^{-1}$


Calorimeters for the thermal power measurement of real size radwaste packages

- Calorimeters must be “easy to use”, versatile, robust and reproducible
- Heat-flux calorimetry
  - Sensors have to operate in hard radiological environment (gamma dose rate up to 100 Gy/h)
  - Prototype under development
- "Heat balance calorimetry"
  - Measurement of the temperature variation of a fluid circulating around the package
  - Calculation of the thermal power from the increase of air temperature

\[
Q = \dot{m} \cdot C_P \text{air} \cdot (T_3 - T_1) + K \cdot (T_3 - T_{\text{amb}})
\]  

\[
T_3 = (T_2 + T_1) / 2
\]  

- Assumption of a constant power dissipation of the package during the measurements
Development of a “heat balance calorimeter”

- **Determination of the temperatures $\bar{T}_1$ and $\bar{T}_2$**
  - Meas. of the electrical resistance of a Nickel wire ($\phi = 0.05$ mm, $l = 85$ cm) mounted on a grid
    ⇒ Direct determination of the average temperatures $\bar{T}_1$ and $\bar{T}_2$
  - Calibration of these “probes” from $20 \degree$C to $80 \degree$C by comparison with a calibrated SPRT
    ⇒ $T = f(R)$ (Sensitivity $\approx 0.17 \, \Omega/\degree$C)
- Development of a “heat balance calorimeter”
  - Versatile prototype ➞ for different sizes of package
  - Calibration of the calorimeter by electrical substitution (between 0 and 200 W)
Development of a “heat balance calorimeter”

- Versatile prototype ➞ for different sizes of package
- Calibration of the calorimeter by electrical substitution (between 0 and 700 W)

\[ Q = U_{heat} \cdot I_{heat} = \left( U_{heat} \cdot U_s \right) / R_s \]
First results obtained with the concrete “reference package” (ILW-LL)

- Comparison between thermal power measured by the calorimeter and power released by joule effect by the “reference package”
  - For $Q_{heat} \approx 10 \, \text{W}$ and $\dot{m} = 10 \, \text{kg} \cdot \text{h}^{-1}$

<table>
<thead>
<tr>
<th>$U_{\text{heat}}$ (V)</th>
<th>$U_s$ (V)</th>
<th>$R_s$ (Ω)</th>
<th>$Q$ (W)</th>
</tr>
</thead>
<tbody>
<tr>
<td>13.3074</td>
<td>0.0800</td>
<td>0.09996</td>
<td>10.6544</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>$T_1$ (°C)</th>
<th>$T_2$ (°C)</th>
<th>$Q$ (W)</th>
<th>Rel. Dev. (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20.331</td>
<td>24.222</td>
<td>10.8516</td>
<td>-1.85 %</td>
</tr>
</tbody>
</table>
Summary

- Development of large volume calorimeters for the measurement of thermal power of real size radwaste packages
- Preliminary tests performed with a “reference package” ⇒ First results promising

Next steps

- Qualification and calibration of the “air flow” calorimeter
- Development of a specific “heat-flux” calorimeter (study of the aging of sensors)
- Assessment of the uncertainty associated to thermal power measurements

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Thank you for your attention

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