Treatment of the Thetis reactor graphite at Belgoprocess

INGSM 2019
Belgoprocess

- Operational link in the waste management of radioactive waste in Belgium
  - Treatment, conditioning & temporal storage

- Mission:
  - Processing of radioactive waste
  - Decommissioning of obsolete nuclear sites and installations
  - Decontamination of materials
  - Developing and valorisation of nuclear know-how
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1. Thetis introduction

- Nuclear sciences institute of the Ghent University
- LEU fuel type UO$_2$ (max. operating power 250 kW)
- Production of radio-isotopes and activation analysis
- In service from 1967 until 2003, decommissioned in 2015
- Neutron reflector graphite was treated by Belgoprocess
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2. Irradiated graphite characteristics

- 8 graphite blocks
  - Average dimensions: 0.5m x 0.5m x 0.7m
  - Gross mass ~ 300 kg each
  - Aluminium cladding ~ 2 to 5 mm

- Characterization (by SCK•CEN)
  - Stored Wigner energy ≈ 100 to 250 J/g
    - No significant amount allowed by regulator
  - Maximum dose rate < 2 mSv/h
    - $^{60}$Co, $^{152}$Eu, $^{154}$Eu, $^{134}$Cs
    - Major contribution in (activated) aluminum
  - Estimation of radiological content
    - Nuclide vector developed through modelling
    - Safety-relevant for disposal/treatment: $^{3}$H, $^{14}$C, $^{36}$Cl
2. Irradiated graphite characteristics

- Limited specific measurements of some important parameters
  - Wigner energy
  - Radioactivity content

- Simple approach for sampling
  - Limited amount of graphite allows evaluation through conservative samples
    - No safety plates
    - Maximum neutron flux 10 cm
2. Irradiated graphite characteristics

- **Wigner energy**
  - According to IAEA TECDOC-1521 (2006):
    
    "It is not acceptable to store or dispose of graphite containing significant releasable stored energy."
  
- **DSC-analysis (Differential Scanning Calorimetry) for 2 samples**
  - Measurement
    - \( \text{N}_2 \) @ 20 °C/min
    - Up to 600 °C
    - Repeat (2\textsuperscript{nd} run)
  - Stored energy eliminated at [120°C - 550°C]
  - Maximum stored energy \( \approx 250 \text{ J/g} \)
2. Irradiated graphite characteristics

- **Radioactivity content**
  - Specific radiochemical analysis $^3$H, $^{14}$C and $^{36}$Cl
    - Conservative sampling at 10 cm depth (at hotspot area)
      - Assumption of simplified activation profile for mean activity content
      - E.g. $^{14}$C through dominant formation mechanism $[^{13}\text{C} (n,\gamma) ^{14}\text{C} @ 200 \text{ keV}$

![Graph showing activity content vs depth in graphite matrix](image-url)
2. Irradiated graphite characteristics

- Radioactivity content (cont’d)
  - Evaluation shows underestimation
  - Impact needs to be evaluated
    - Take this into account in treatment scenario
    - Representativeness (conservative?)

<table>
<thead>
<tr>
<th></th>
<th>Bq/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>H-3</td>
<td>Declaration: 7.15E+04</td>
</tr>
<tr>
<td></td>
<td>C-14: 2.31E+03</td>
</tr>
<tr>
<td>Cl-36</td>
<td>7.84E+01</td>
</tr>
</tbody>
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3. Evaluation of possible treatment pathways

- Most relevant pathways (literature review)

<table>
<thead>
<tr>
<th>Scenario</th>
<th>Installation available?</th>
<th>Wigner energy</th>
<th>Volume reduction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Conditioning as such</td>
<td>Yes</td>
<td>Still present</td>
<td>No</td>
</tr>
<tr>
<td>Conditioning after annealing</td>
<td>No</td>
<td>Eliminated</td>
<td>No</td>
</tr>
<tr>
<td>Incineration</td>
<td>Yes</td>
<td>Eliminated</td>
<td>Yes</td>
</tr>
</tbody>
</table>

- Incineration was considered best option
  - No investments/licensing required
  - Stored Wigner energy is eliminated
  - Limited volume reduction
  - Radiological content in agreement with license of LLW incinerator (CILVA)
  - Size reduction of graphite blocks necessary
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- LLW incinerator (CILVA)
4. Treatment

- Incineration in LLW incinerator (CILVA)
  - Graphite packages of max 5 kg
  - Treated together with standard burnable waste
  - Mixing needed in order to avoid
    - Fluctuations in temperature of primary combustion chamber
    - Difficulties in ash removal (blockage of augers)
4. Treatment

- Need for pre-treatment to fit in existing process
  - to eliminate aluminum cladding
  - to feed the incinerator
  - to avoid operational issues
  - optimize combustion process

- Cutting and size reduction
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5. Conclusion and operational feedback

- Cutting and size reduction ...
  - is absolutely necessary to avoid process issues
  - is hard work
    → it takes about 2-3 days to do 1 block
- No incidents occurred during treatment
- Major advantages of incineration
  - Stable and homogenous waste form, ready for surface disposal
  - Wigner energy was eliminated
  - Volume reduction
5. Conclusion and operational feedback

- Monitoring during incineration campaign
  - $^3$H is captured (99.5%) in scrubber
  - $^{14}$C is released (> 99%) in the stack
  - All safety, operational and radiological limits were respected

- Incineration of small amounts of graphite with stored Wigner energy and limited amounts of $^{14}$C will result in final waste packages that comply with the waste acceptance criteria of surface disposal
Thank you for your attention