SEM analyzes of powdered actinide compounds: Implementation in a hot laboratory

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19,000 m² on 3 levels
12 shielded lines
17 laboratories (250 glove boxes)
LCC, laboratory L26 (end of 2017)

L26 ≈ 130 m²

12 agents
SEM-FEG in LES216
for radioactives powders
since 2010
SEM-FEG ZEISS SUPRA 55 WDS

* patent WO2013/068321 A1

glove box specially designed

Existing shield (20 mCi)
Vibrations reduction for imaging

Glove box on Silent blocks

shutter

Vacuum pump away
SEM-FEG ZEISS SUPRA 55 WDS

EDS  INLENS  WDS

Everhart-Thornley

BSE
How to maintain it?
Maintenance

Below

Disconnection without breaking containment

Inflatable joint
Maintenance

SEM Glove box

DEN/DTEC/SECA/LCC
Examples

BSE scintillators

Diaphragms in the column

Magnetic brake ajustement

EDS chip
Sample preparations and imaging examples
Preparations

Glove box

Hot cell

wood alloy
Everhart-Thornley (SE2)

- Sintered pellet of U-AmO₂
- Iron plate lixiviated 1 year with UO₂ pellet

InLens (high resolution)

- Iron plate lixiviated 1 year with UO₂ pellet

BSE (chemical contrast)

- PuO₂/St-Zn mixing
- Dissolution of PuO₂ assisted by US

Plutonium colloid

Images
EDS measurements
Special EDS configuration

Be window used as a shield

+ EDS/sample distance increased

+ 0.5 mm Collimator of (standard at 2 mm)

First element measurable: Na

+ Low counts measured at 15 kV minimum with the largest diaphragm

Bad spatial resolution
Long counting time
Destruction of many samples
Special EDS configuration

- 25 µm Be window
- High distance sample / detector
- 4 years old chip

- 12 µm Be window
- Medium distance sample / detector
- New chip

Calibration is essential!

MAC55 Standards
Home made standard for actinides
EDS analysis examples

Plutonium / Uranium mapping (red/green)

UO₂ / PuO₂ powders mixed

MOX sintered pellet

Spectrum on UO₂ pellet lixiviated in water for 1 year in presence of an iron foil
Images and EDS on Mo/Zr precipitates (raw)

Spent fuel dissolution → Mo/Zr precipitates formed

22h of precipitation (other experiment during 29h et 35h)

<table>
<thead>
<tr>
<th>Standards conditions</th>
<th>Additive before dissolution</th>
<th>Additive through dissolution</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 μm</td>
<td>1 μm</td>
<td>1 μm</td>
</tr>
<tr>
<td>500 nm</td>
<td>500 nm</td>
<td>500 nm</td>
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</tbody>
</table>
The supra 55 is devoted to **powders** characterization

<table>
<thead>
<tr>
<th>Time</th>
<th>Zr</th>
<th>Mo</th>
<th>M</th>
<th>U</th>
<th>Pu</th>
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<tbody>
<tr>
<td><strong>before 22h</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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<tr>
<td>Average</td>
<td>29.1</td>
<td>57.6</td>
<td>6.2</td>
<td>6.2</td>
<td>0.9</td>
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<tr>
<td>SD</td>
<td>1.40</td>
<td>2.76</td>
<td>0.62</td>
<td>2.93</td>
<td>0.16</td>
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<tr>
<td>RSD</td>
<td>5%</td>
<td>5%</td>
<td>10%</td>
<td>72%</td>
<td>18%</td>
</tr>
<tr>
<td><strong>Before 29h</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Average</td>
<td>29.8</td>
<td>59.8</td>
<td>5.4</td>
<td>4.3</td>
<td>0.8</td>
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<tr>
<td>SD</td>
<td>1.79</td>
<td>1.59</td>
<td>0.91</td>
<td>1.12</td>
<td>0.23</td>
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<tr>
<td>RSD</td>
<td>6.0%</td>
<td>2.6%</td>
<td>17.7%</td>
<td>24.9%</td>
<td>30.9%</td>
</tr>
<tr>
<td><strong>Trough 22h</strong></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Average</td>
<td>37.6</td>
<td>55.0</td>
<td>4.5</td>
<td>2.1</td>
<td>0.9</td>
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<tr>
<td>SD</td>
<td>7.14</td>
<td>7.07</td>
<td>0.91</td>
<td>0.49</td>
<td>0.32</td>
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<tr>
<td>RSD</td>
<td>17%</td>
<td>14%</td>
<td>22%</td>
<td>21%</td>
<td>30%</td>
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</table>

EDS results are average on numerous spectrum
Number of spectrum depends of the SD and RSD
Images and EDS on Mo/Zr precipitates (raw)

Time influence on composition is insignificant

<table>
<thead>
<tr>
<th>%at*</th>
<th>Mo</th>
<th>Zr</th>
<th>M</th>
<th>Mo/(Mo+Zr)</th>
<th>Zr/(Mo+Zr)</th>
<th>(Mo+M)/(Mo+Zr+M)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1 - through</td>
<td>56</td>
<td>36</td>
<td>6</td>
<td>61%</td>
<td>39%</td>
<td>63</td>
</tr>
<tr>
<td>2 - before</td>
<td>58</td>
<td>30</td>
<td>5</td>
<td>66%</td>
<td>34%</td>
<td>68</td>
</tr>
<tr>
<td>3 - without</td>
<td>70</td>
<td>27</td>
<td>0</td>
<td>72%</td>
<td>28%</td>
<td>72</td>
</tr>
</tbody>
</table>

* (% incertitude not include)

Microstructure and composition linked

Mo substitution?
- close ionique radius
- Octahedral coordination for both

XRD mesurements to confirm!
WDS measurements
Examples on dissolution residues of spent fuel

EDS

WDS

1 µm
Americium detection in PuO$_2$ powders
Pu oxalate

Thank you for your attention