Standard Electron Probe Microanalysis of Irradiated Fuel at PSI

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Contain of the presentation

- Presentation of our shielded Electron Micro-Probe (EPMA)
  - Short description of the machine
  - Main characteristics of the machine

- Description of the measurement procedures developed for the characterisation of irradiated fuel with our probe
  - X-Ray spectrum of a fuel specimen
  - Quantification difficulties
  - Typical measurement procedure
The LWV Micro-Probe
CAMECA SXR-SX50 shielded instrument

Specimen holder for the analysis of fuel specimens

Specimen holder
(Specimen and U standard)

EPMA massive stage

Shipping container
(with sealing)
**X-Ray spectrum of high burn-up MOX fuel**

- Interference between Pu and U lines
- U M\(_{\alpha}\)
- Pu M\(_{\alpha}\)
- Nd L\(_{\alpha}\)
- Ba L\(_{\alpha}\)
- Cs L\(_{\beta}\)
- Xe L\(_{\alpha}\)

**Interference between fission products lines**
- U M\(_{\beta}\)
- Pu M\(_{\beta}\)
- Ru L\(_{\alpha}\)
- Pd L\(_{\alpha}\) (Ru, Rh)
- Pd L\(_{\beta}\)

**Radioactive background**

**Emission lines for PET monochromator**

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**X-Ray spectrum of UO\(_{2}\) standard material**

**Influence of U on the measurement of Pu**

- U M\(_{\alpha}\)
- U M\(_{\beta}\)

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(Instrument: CAMECA SXR-SXSO; HV: 20keV, Current: 150 nA)

**Position of Pu M\(_{\beta}\)**

**Emission lines for PET monochromator**
### X-Ray spectrum of irradiated UO₂ and MOX fuel

Influence of U on the measurement of Pu

UO₂ fuel 18 GWd/t = 0.5% Pu

MOX fuel, 59 GWd/t = 5% Pu

![X-Ray spectrum images](image)

(Instrument: Cameca SXRL320; HV: 20 kV; Current: 150 nA)

Emission lines for PET monochromator

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### Spectrometer settings for fuel analysis

<table>
<thead>
<tr>
<th>Element/line</th>
<th>Crystal</th>
<th>Peak 18° sin θ</th>
<th>Background range in 18° sin θ (°)</th>
<th>Standard used</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>U M₁</td>
<td>PET</td>
<td>44604</td>
<td>+1800 to +12800</td>
<td>UO₂</td>
<td>Interference from Pu</td>
</tr>
<tr>
<td>Pu M₁</td>
<td>PET</td>
<td>40069</td>
<td>+1800 to +12800</td>
<td>UO₂ or U₂PuO₄</td>
<td>Count rate of U M₁ is 1.5x that of Pu M₁ at U₂PuO₄</td>
</tr>
<tr>
<td>Ge L₃</td>
<td>LiF</td>
<td>50560</td>
<td>+1700 to +8000</td>
<td>U₂PuO₄</td>
<td>Interf. from U₂Mo, CsI at Ge L₃</td>
</tr>
<tr>
<td>O K₂</td>
<td>PCI</td>
<td>39440</td>
<td>+300 to +10000</td>
<td>U₂Mo</td>
<td>I.e. high activity at U₂Mo</td>
</tr>
<tr>
<td>Na L₃</td>
<td>PET</td>
<td>37990</td>
<td>+2250 to +2660</td>
<td>Na₂Mo</td>
<td>High activity at Na L₃</td>
</tr>
<tr>
<td>Co L₃</td>
<td>PET</td>
<td>33840</td>
<td>+270 to +3700</td>
<td>Co₃O₄</td>
<td>Interference from Co L₃</td>
</tr>
<tr>
<td>K L₂</td>
<td>PET</td>
<td>54470</td>
<td>+2000 to +2500 or +2590 to +3000</td>
<td>Na₂Mo, CsI</td>
<td>High activity at K L₂</td>
</tr>
</tbody>
</table>

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Spectrometer settings for fuel analysis

<table>
<thead>
<tr>
<th>Element/line</th>
<th>Crystal</th>
<th>Peak Position</th>
<th>Backgrounds (range in 10^-4 of Q) and/or Slope Factor</th>
<th>Standard used*</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zr Lα</td>
<td>PET</td>
<td>69436</td>
<td>- (650 to 1650) + (280 to 1000)</td>
<td>Zr, ZrO₂</td>
<td>Interference from Ca on BC</td>
</tr>
<tr>
<td>Be Lα</td>
<td>PET</td>
<td>31725</td>
<td>asymmetric BG absorbance</td>
<td>Be₃O₃</td>
<td>Small interference on peak, interference on BeG (U, Ne Lα) interference on BeCl smaller</td>
</tr>
<tr>
<td>Ru Lα</td>
<td>PET</td>
<td>55400</td>
<td>asymmetric BG absorbance</td>
<td>Ru₂(C₂O₄)</td>
<td>Interference from Ru Lα on BG</td>
</tr>
<tr>
<td>Te Lα</td>
<td>PET</td>
<td>28480</td>
<td>- (550 to 650) + (250 to 650)</td>
<td>Te₂O₃</td>
<td>Small interference from TeLα₂</td>
</tr>
<tr>
<td>Mo Lα</td>
<td>PET</td>
<td>61790</td>
<td>- (700 to 850) + (700 to 850)</td>
<td>Mo</td>
<td>Small interference from Mo Lα</td>
</tr>
<tr>
<td>Pt Lα</td>
<td>PET</td>
<td>27490</td>
<td>- (1000 to 1100) + (1000 to 1100)</td>
<td>Pt</td>
<td>Interference from Ru/Rh and U on Pt Lα and Ru BC and Pt BC (from U)</td>
</tr>
</tbody>
</table>

* Background (BG) correction handled for peaks Zr Lα, Be Lα through specimen analysis, depending on sample construction and microstructure background.

** Spectrometer settings are recommended for the sample to be analyzed.
Fuel analysis with EPMA - Autoradiography

Cross section of a high burn-up sphere-pac MOX fuel (NOK-M308)

α-autoradiography

β/γ-autoradiography

Fission products

Fuel analysis with EPMA - U and Pu distributions in UO₂

UO₂ pellet fuel with 18GWd/t burn-up

Radio chemical analysis
Fuel analysis with EPMA - Nd distribution in UO₂

UO₂ pellet fuel with 18GWD/t burn up

![Nd distribution graph](Image)

- \( \text{Nd}_{\text{RC}} = 0.18\% \)
- \( \text{Nd}_{\text{EPMA}} = 0.14\% \)

Fuel analysis with EPMA - Cs distribution in UO₂

UO₂ pellet fuel with 18GWD/t burn up

![Cs distribution graph](Image)
Fuel analysis with EPMA - Pu distribution in MOX

High burn-up MOX fuel (NOK - M308)

![Pu distribution graph](image)

Fuel analysis with EPMA - Nd and Zr distributions in MOX

High burn-up MOX fuel (NOK - M308)

![Nd and Zr distribution graph](image)
Fuel analysis with EPMA – Cs and Nd distributions in MOX

High burn-up MOX fuel (NOK – M308)

- Secondary phase
- Pores
- Cs, Nd

Pore / intermetallic precipitates

micrometers from fuel center

Fuel analysis with EPMA – Cs, Ba and Xe distributions in MOX

High burn-up MOX fuel (NOK – M308)

- Pores
- Cs, Ba, Xe

Pore / intermetallic precipitates

micrometers from fuel center
Conclusions

- EPMA is a very powerful technique for the characterisation and the analysis of high burn-up fuel specimen
- The measurement and analysis procedures must be carefully set up and checked in order to insure a good quantification of the measurements