Shielded Electron Probe Microanalysers (EPMA) – from CAMECA to JEOL

Preliminary comparison of design and physical properties

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Outline

• Synopsis of microanalysis with example of application.
• Previous microanalysis instrument for irradiated materials at PSI with decommissioning results.
• Features, installation and shielding of the new EPMA.
• Comparison of X-ray intensity signals and radioactive background with previous EPMA.
• Summary and work progress.
Principles of Electron Probe Microanalysis

HV electron beam
backscattered electrons
secondary electrons
X-rays

cathodoluminescence

Sample

2-20 μm³ excitation volume

X-ray continuum

→ SEM image

→ Element concentration and mapping

WDS-spectrometer for characteristic X-rays
X-ray counter
Faraday cup
0.1 μm Sample stage

Electron gun
40 keV GUN

Optical microscope
Condenser lenses
High sensitivity diffracting crystal (1:1 to 4)

Seibert, JNMT, 32(2004)

Courtesy of Cameca

Seibert, JNMT, 32(2004)
EPMA Mapping of irradiated Inert Matrix Fuel (YSZ-IMF)

Scanning Electron Micrograph (SEM) of porous pellet border (100μm x 100μm; bright porous phase Pu rich)

Pu distribution at the border and point analysis
[range: 1 - 50wt%; av.:10wt%, 2.5at%]

R. Restani et al., JNM 385(2009) 435
CAMECA SXR-50 at PSI (1988-2008)

Courtesy of CAMECA
Sample Loading on CAMECA SXR-50
Active waste of decommissioned EPMA for decontamination

- Sample stage (denal block)
- Sample chamber, objective lens
- Vacuum system
- Electron column parts
- 20 cm

HOTLAB Division - Nuclear Energy and Safety Research Department
Goal and purpose of future use of new EPMA

- Replacement of 21 year old shielded EPMA CAMECA SXR with the same performance and enhanced SE-, BSE- and X-ray imaging capabilities.
- Optimal political-financial situation and keeping to the budget.
- Examination and quantitative elemental microanalysis of radioactive materials as polished metallo-ceramographic specimen.
- and new of fractography specimen (qualitative) as there is only an additional unshielded Scanning Electron Microscope (SEM) available in the lab.
- Manipulator handling and remote control of the instrument as before.
EPMA JEOL JXA-8500F (PSI-Configuration)

- **Field Emission Electron Gun (FEG)**
  HT: 1-30 keV; Beam 0.01 nA – 500 nA (variation ≥ 0.5%/h)

- **Small probe diameter** (typically 20-100 nm between 1 nA and 100 nA) gives high intensity X-ray signals from a microarea and high resolution SE images.

- **4 Spectrometers**: Rowland circle radius of 140 mm! No large crystals!
  SP1/SP3 (left): TAP/LDE2 and LIF/PET (Ar/CH₄ flow and Xe closed counter).
  SP2/SP4 (right): LIF/PET (Xe counter) and TAP/LDE1 (Ar/CH₄ counter).
  Crystal-sample distance for Cs Lα (PET): 9 cm
  PP-Counter-sample distance for Cs Lα (PET): 19 cm
  No spectrometer windows (chamber vacuum)!

- **8 x 12 cm size W-alloy block of sample stage** with sample holder for two 1 inch and two 17 mm samples. X,Y,Z-mov. in 1 μm steps (35x35x7mm).
  Additional **rotation and tilt sample holder** for 1 inch samples available (mounting and shielding of active samples on these stages difficult).

- **SUN-Workstation**, transformation of data on peripheral PC!
Ground plan of laboratory and installation
JEOL JXA-8500F in PSI: Remote control
JEOL JXA-8500F in PSI (front view through window)
JEOL JXA-8500F (FEG EPMA) in PSI (back view)
Sample transport through box port and EPMA airlock
Shielding of JEOL 8500F

Cross-section

Distances specimen to crystal (mm) with counter positions

Courtesy of remX GmbH
JEOL JXA-8500F: Poleshoe with 1st shielding

- Gear screw for crystal
- Electron filter
- BSE
- W-alloy
- SE Faraday cage

Courtesy of JEOL & remX
JEOL JXA-8500F: Poleshoe with 2nd shielding

Courtesy of remX
JXA-8500F: Shielded sample holder remX

Courtesy of JEOL & remX
Shielded sample holder remX in EPMA chamber

- Objective lens
- Crystal drive of spectrometer 4
- X-ray exit port
- Sample holder
- Sample holder shielding
- Faraday cage of SE detector → SEM image
- Sample stage

Courtesy of JEOL & remX
JXA-8500F: Shielded spectrometer remX

Crystal start position (75 mm from sample)  Crystal end position (250 mm from sample)

Courtesy of JEOL & remX
CAMECA SXR-50: Shielded spectrometer
Comparison of spectrometer performances *(inactive specimens)*

Net-intensities with PET diffracting crystals (20 keV beam)

<table>
<thead>
<tr>
<th>ELEMENT / Line in ref. material</th>
<th>CAMECA SXR-50  [cts/s/nA]</th>
<th>PET (Ar/CH₄) [cts/s/nA]</th>
<th>PET (Xe) [cts/s/nA]</th>
</tr>
</thead>
<tbody>
<tr>
<td>V Kα in V</td>
<td>1435</td>
<td>1705</td>
<td></td>
</tr>
<tr>
<td>Ti Kα in Ti</td>
<td>1355</td>
<td>1450</td>
<td></td>
</tr>
<tr>
<td>Ca Kα in Wollastonite</td>
<td>345</td>
<td>350(Ar); 500(Xe)</td>
<td></td>
</tr>
<tr>
<td>Ce Lα in CeAl₂</td>
<td>410</td>
<td>515</td>
<td></td>
</tr>
<tr>
<td>Ba Lα in BaF₂</td>
<td>420</td>
<td>475</td>
<td></td>
</tr>
<tr>
<td>Sb Lα in Sb</td>
<td>415</td>
<td>395(Ar); 570(Xe)</td>
<td></td>
</tr>
<tr>
<td>Au Mα in Au</td>
<td>71</td>
<td>63(Ar); 80(Xe)</td>
<td></td>
</tr>
</tbody>
</table>

*(1) partly shielded by remX*
Comparison of spectrometer performances *(inactive specimens)*

Net-intensities with LIF diffracting crystals (20 keV beam)

<table>
<thead>
<tr>
<th>ELEMENT / Line in ref. material</th>
<th>CAMECA SXR-50 LIF (Ar/CH₄) [cts/s/nA]</th>
<th>JEOL 8500F (¹) LIF (Xe) [cts/s/nA]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Zn Kα in Zn</td>
<td>380</td>
<td>530</td>
</tr>
<tr>
<td>Fe Kα in Fe</td>
<td>455</td>
<td>470</td>
</tr>
<tr>
<td>Cr Kα in Cr</td>
<td>360</td>
<td>310</td>
</tr>
<tr>
<td>V Kα in V</td>
<td>255</td>
<td>230</td>
</tr>
<tr>
<td>Ti Kα in Ti</td>
<td>180</td>
<td>145</td>
</tr>
</tbody>
</table>

(¹) partly shielded by remX
Comparison of spectrometer performances (inactive specimens)

Net-intensities with TAP / LSM diffracting crystals (20 keV beam)

<table>
<thead>
<tr>
<th>ELEMENT / Line in ref. material</th>
<th>EPMA</th>
<th>CAMECA SXR-50 [cts/s/nA]</th>
<th>JEOL 8500F (1) [cts/s/nA]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si $K\alpha$ in Jadeite</td>
<td></td>
<td>300</td>
<td>575</td>
</tr>
<tr>
<td>Al $K\alpha$ in Al$_2$O$_3$</td>
<td></td>
<td>610</td>
<td>1260</td>
</tr>
<tr>
<td>Na $K\alpha$ in Jadeite</td>
<td></td>
<td>38</td>
<td>69</td>
</tr>
<tr>
<td>O $K\alpha$ in Al$_2$O$_3$</td>
<td></td>
<td>195</td>
<td>187</td>
</tr>
</tbody>
</table>

(1) partly shielded by remX
\(\gamma\)-radiation impact on proportional counters

*(encapsulated irradiated fuel sample, 8 mSv/h/0.5m)*

### Comparison of Ar/CH\(_4\) with Xe counter

- **Ar/CH\(_4\)-counter/SP1**
- **Xe-counter/SP2**

*recorded with JEOL 8500F*
\( \gamma \)-radiation impact on proportional counters
*(encapsulated irradiated fuel sample, 8 mSv/h/0.5m)*

Dependance on spectrometer, crystal/counter position and sample position

recorded with JEOL 8500F
### Preliminary comparison of background measurements between CAMECA SXR-50 and JEOL 8500F (interpolation from different fuel samples, basis: 5 mSv/h/0.5m)

<table>
<thead>
<tr>
<th>Element-Line (crystal)</th>
<th>( \lambda ) [Å]</th>
<th>Sample-Crystal (JEOL) [mm]</th>
<th>Position</th>
<th>EPMA</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nd L( \alpha ) (PET)</td>
<td>2.37</td>
<td>75.9</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Cs L( \alpha ) (PET)</td>
<td>2.89</td>
<td>92.6</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pu M( \beta ) (PET)</td>
<td>3.51</td>
<td>112.5</td>
<td></td>
<td></td>
</tr>
<tr>
<td>U M( \alpha ) (PET)</td>
<td>3.91</td>
<td>125.2</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Zr L( \alpha ) (PET)</td>
<td>6.07</td>
<td>194.4</td>
<td></td>
<td></td>
</tr>
<tr>
<td>O K( \alpha ) (PC1/LDE1)</td>
<td>23.6</td>
<td>110.2</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th></th>
<th>CAMECA(^{(1)}) SXR-50 (Ar) [cts/s]</th>
<th>JEOL 8500F (shielded)(^{(1,2)})</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ar/( \text{CH}_4 ) [cts/s]</td>
<td>2700</td>
<td>2000</td>
</tr>
<tr>
<td>Xe [cts/s]</td>
<td>2900</td>
<td>1500</td>
</tr>
</tbody>
</table>

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\(^{(1)}\) Radioactive background depends on sample position (variation 20-40%) and spectrometer!

\(^{(2)}\) partly shielded, \( \gamma \)- and \( \beta \),\( \gamma \)-sources employed (share due to \( \beta \)-radiation: 5-35%).

JEOL always requires a window setting in the pulse height analyser (differential mode)!
Summary and Conclusions

• High versatility of the EPMA with respect to examination capabilities and types of specimens.
• Superior SEM and X-ray imaging. High(er) lateral resolution (FE-gun!). Performance for high $\beta,\gamma$-active samples must be verified.
• The light microscope has only a focussing function.
• Small efficiency differences between JXA-8500F (140 mm R) and SXR-50 (180 mm R) with advantages for Xe counter:
  \[ \text{LIF}(\text{JXA-Xe}) \approx \text{LIF}(\text{SX-Ar}); \]
  \[ \text{PET}(\text{JXA-Xe}) > \text{PET}(\text{JXA-Ar}), \text{PET}(\text{SX-Ar}); \]
  \[ \text{TAP}(\text{JXA}) > \text{TAP}(\text{SX}); \]
  \[ \text{LDE1 (JXA)} \approx \text{PC1 (SX)} \]
• Spectral resolution has to be verified. It is expected to be that SXR-50 is marginally better than JEOL 8500F (bigger Rowland circle and measurements on similar EPMAs of F. Bussy (Univ. Lausanne), SAMx workshop Nice 2007).
Summary (continued)

• Performance with respect to microanalysis of high active materials is promising (good shielding of $\beta, \gamma$-radiation).
• Sample loading convenient for embedded flat samples.

Open questions – with need for adaptations in work procedure
• Contamination risk (compact, open design).
• More components are exposed to radiation. Their lifetime cannot be ascertained (e.g. electronics, cables, crystals, counter windows – source for leak)? Effect of higher $\gamma$-radiation in surrounding area?
• Sophisticated vacuum system. Delicate to radiation?
• User friendliness of the control system (parameter adjustments, data acquisition, file transfer and handling)?
• Maintenance: remX together with JEOL, but JEOL makes it at the moment only on non contaminated components.
Work progress

- Installation of the basic instrument in the lab by JEOL starting in January 2009 with electronic adjustments.
- First active material examined in June (irradiated cladding).
- Technical approval of the EPMA with shielded sample holder in July 2009.
- Main shielding of the instrument in August 2009 with subsequent active measurements. Changes in spectrometer configurations.
- Mounting of additional shieldings, upgrade light microscope (Sept. - Oct.).
- Additional lab infrastructure and emergency tests (Sept. – Nov.).
- New shielded sample holder for 4 specimens (October). Mounting and shielding of additional rotation holder is in state of design or in feasibility study respectively.
- First irradiated fuel rod sample to be examined starting end of October 2009.
Acknowledgments

- To CAMECA, JEOL and remX for the permission for the issue of data and photos.
- To the service engineers of JEOL and remX who have worked hard to maximise the EPMA performance under new and difficult circumstances and the good and very helpful relation with the many CAMECA crew members I had over the lifetime of the SXR-50.
- Our Hotlab maintenance crew in supporting the build-up.
Addendum 1

Target atom

- Electron shells
- Nucleus
- Incident electrons
- Ejected K shell electron
- Characteristic Discrete energy
- Close interaction Moderate energy
- Impact with nucleus Maximum energy
- Distant interaction Low energy
Addendum 2: X-Ray spectrum of high burn-up UO$_2$ fuel

Emission lines for PET monochromator

recorded with SXR-50
Addendum 3: X-Ray spectrum of high burn-up MOX fuel

Emission lines for PET monochromator

- U $M_\beta$
- U $M_\alpha$
- Ba $L_\alpha$
- Cs $L_\alpha$
- Xe $L_\alpha$
- Pu $M_\beta$
- Nd $L_\alpha$
- Pd $L_\alpha$ (Ru/Rh)
- Pd $L_\beta$
- Ru $L_\alpha$
- Ru $L_\beta$

recorded with SXR-50