SHIELDED ELECTRON PROBE MICROANALYSERS (EPMA) – FROM CAMECA TO JEOL PRELIMINARY COMPARISON OF DESIGN AND PHYSICAL PROPERTIES

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ABSTRACT

PSI operated for more than 20 years a shielded EPMA CAMECA SXR-SX50 for the surface examination of irradiated specimens. This method allows the characterisation of the sample surface morphology by scanning electron microscopy (SEM) and both the high resolution elemental composition and its spatial distribution by wavelength dispersive X-ray spectroscopy (WDS).
This instrument has been replaced this year by a standard JEOL 8500F field-emission EPMA which has now been modified to enable the investigation of high radioactive samples including fuel rods segments. This induces a different arrangement of the electronics for remote control and the addition of a tungsten alloy shielding both on the specimen stage and between the spectrometers and the specimen chamber. The objective of the acquisition of this new instrument is to widen the scope of investigations from polished specimens to fractography samples and to enhance the SEM and X-ray resolution. It is achieved by a field emission gun which provides generally a much smaller probe diameter than a tungsten hairpin cathode and allows working at low accelerating voltage with thus a smaller analysis area. The open design of the spectrometers towards the specimen chamber poses a challenge in the shielding of the β,γ -radiation, bremsstrahlung and reflection.

1. Introduction

Electron Microscopy is widely used in post-irradiation examinations for materials characterisation. The most common methods are Scanning Electron Microscopy (SEM) and Electron Probe Microanalysis (EPMA). Both techniques allow the imaging of solid surfaces at high magnification with a large field of depth and resolution of both topography and microstructure of the specimen. These instruments are additionally equipped with analyses devices which deliver the quantitative elemental composition of a micrometer sized volume. The methods measure the characteristic X-rays generated by the ionisation of atoms with excitation of the K-shell or L-shell through the applied electron beam. The wave length dispersive (WDS) technique of the EPMA allows analysing also high active materials if a shielding of the X-ray counter against γ-radiation is provided.
PSI had in operation for more than twenty years a CAMECA SX50R Microanalyser especially for the analyses of irradiated fuel rods. It was decided in 2006 to replace the ageing machine with an instrument of the last generation. Considering the rising scope of active specimen examinations that our unshielded SEM cannot cover, the future EPMA has to satisfy new and enhanced features. These are to provide better scanning electron imaging including the possibility to observe fractography and a higher lateral X-ray resolution.

2. Decommissioning

The disassembly of the EPMA SX50R started in July 2008 and took about 4 man-weeks. An additional 4 man-weeks workload was dedicated to control and classify the non contaminated parts of the instrument. These were e.g. the circuit boards, supply units, motors, valves, spectrometers with crystals, counters and windows, W-cathode, anode and beam regulator of the column, penning gauge, rotary pumps and tungsten shielding. Only slight contamination (up to 10 Bq/cm^2) was measured in the recipient, sample stage, electron column optics and vacuum system which could or can easily be decontaminated. Decontamination of these parts was made in October and only some parts of the column and vacuum system will need special treatment in a chemical bath.
The highest contamination was of spot size and located at the e-beam exit of the poleshoe with about 300 Bq/cm^2. This part will be isolated so that there are very little active waste assets.

3. EPMA

The selected new EPMA is a standard JEOL 8500F with a Schottky field emission gun (FEG). It provides high resolution SE-images and an improvement of the X-ray resolution (e.g. X-ray elemental maps) thanks to the small electron beam diameter and the high beam density at low accelerating voltage. Besides the examination of polished specimens the instrument can also be equipped with a rotation and tilt holder for fractography. There are four spectrometers: two with LIF/PET crystals and one with TAP/LDE1 and one with TAP/LDE2, the last ones being dedicated to the analysis of light elements. The spectrometers are open to the recipient (no windows). They have only a 140 mm Rowland circle so they are smaller than the ones of the CAMECA machine (180 mm). Therefore the crystals and counters are nearer to the specimen. This should be a benefit for the signal intensity but can be impaired by the closeness of the active sample that implies a higher radioactive background on the counter.
The installation of the new EPMA began in January 2009 in the same lead shielded environment as before. It is in the edge part of a lab with about 6 m^2 spacing which can be accessed through an extra door. It had first to be electrically/electronically customised for remote control and measurement by JEOL. remX is the project leader and is making the tungsten alloy shielding of the instrument and the sample holder capable for manipulator handling.
The main shielding is positioned between spectrometers and specimen chamber with a notch in the upper level for the crystal drive towards the poleshoe, as shown on Fig. 1. Two stacked W-plates encase the end piece of the poleshoe to shield the upper part of the chamber and partly the electron optics.

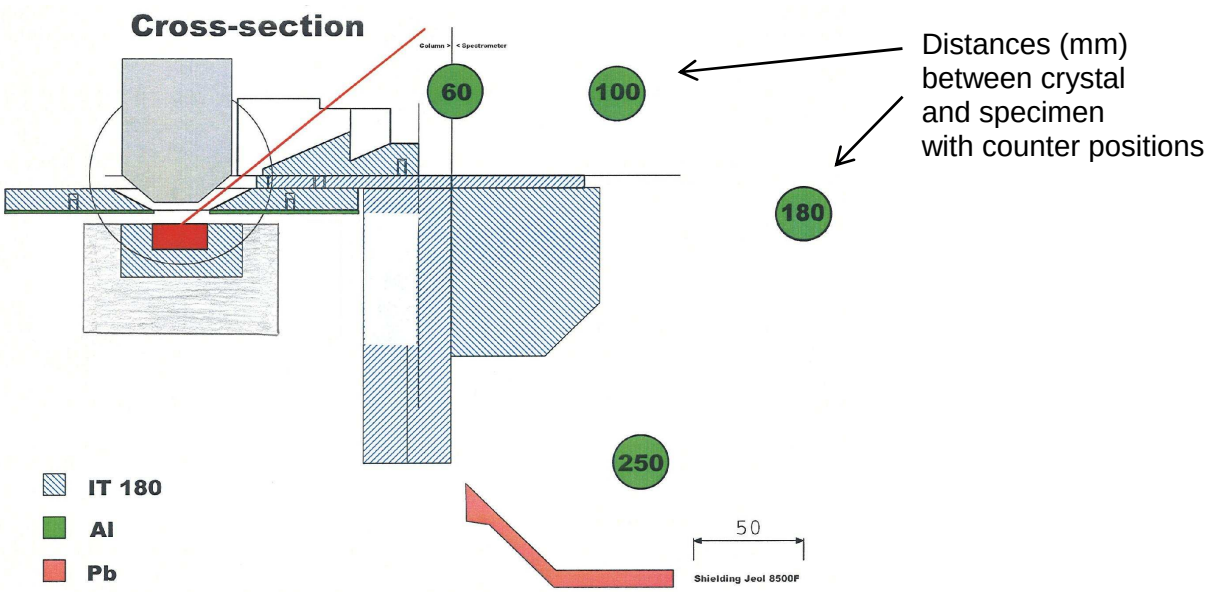

Fig. 1: Schematic of the W-shielding in JEOL 8500F made by remX GmbH (1st concept).

The sample holder is also made of tungsten alloy in the upper part and in the lower part of aluminium in order to not exceed 3.5 kg for manipulator handling. It can accept two 1-inch and two 17 mm samples. The sample container is pushed through an airlock system on the sample stage into an annular W-shielding (Fig. 2). The size of the whole block is 8 cm x 12 cm x 4 cm (w x l x h).

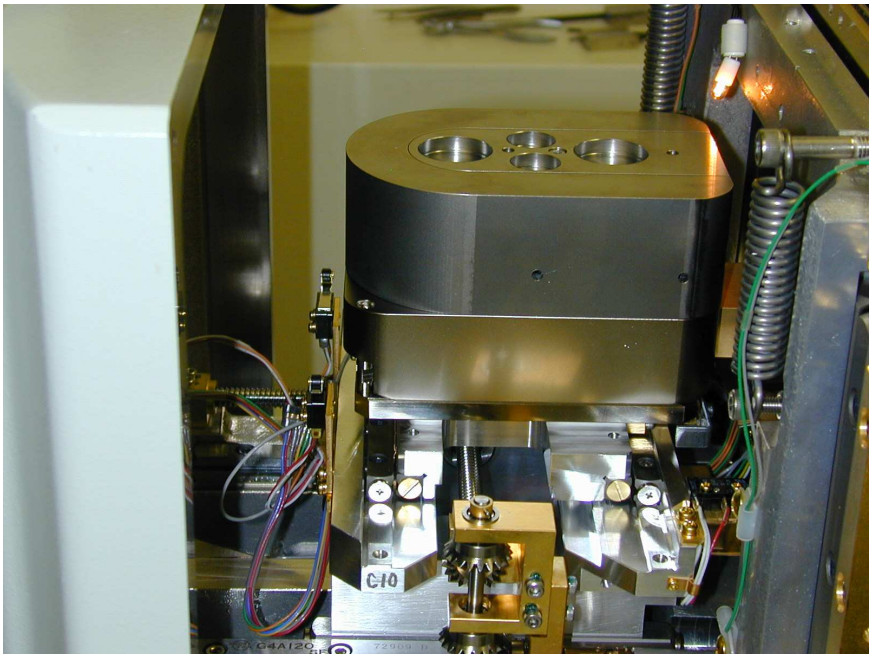

Fig. 2: Sample stage with shielded sample holder in JEOL 8500F (courtesy of JEOL and remX, 1st concept).

4. Performance

The nominal specifications of the EPMA with respect to SE- and BSE imaging as well as X-ray signal intensities and peak/background ratios were tested and obtained with inactive standard materials in July 2009. The shielding concept was assessed by PSI but no guarantee on the part of the manufacturers could be given with respect to the quality of examination for high active materials.

4.1 SE- and BSE-imaging

The quality of the electron microscopy imaging of a high γ-active specimen (encapsulated fuel rod piece) was very good (no excessive noise). The image quality of open specimens (β,γ-active) has now to be checked.

4.2 Intensities

The important features for microanalysis are the spectral resolution, the signal intensities and the peak/background ratio (P/BG). The first one has not yet been checked in detail and is anticipated to be marginally worse than on the old machine because of the small size of the spectrometers. Some P/BG ratios have been measured and found to be very high for standard materials. The comparison of a few signal intensities are reported in table 1.

EPMA		CAMECA SXR-50	JEOL 8500F (partly shielded)	
ELEMENT / Line in ref. material	λ (Å)	**PET** (Ar/CH_4) [cts/s/nA]	**PET** (Ar/CH_4) [cts/s/nA]	**PET** (Xe) [cts/s/nA]
Ti **Kα** in Ti	2.75	1355	-	1450
Ca **Kα** in Wollastonite	3.36	345	350	500
Ba **Lα** in BaF_2	2.78	420	-	475
Au **Mα** in Au	5.84	71	63	80
		LIF (Ar/CH_4)	-	**LIF** (Xe)
Zn **Kα** in Zn	1.44	380	-	530
Ti **Kα** in Ti	2.75	180	-	145
		PC1 (Ar/CH_4)	**LDE1**(Ar/CH_4)	-
O **Kα** in Al_2O_3	23.6	195	187	

Table 1: Net signal intensities of characteristic X-rays from elements measured by bombarding different targets with a 20 keV e-beam. Note: Different diffracting crystals (monochromators) and X-ray counters have been used.

No big differences in the spectrometer efficiencies have been found. The differences are often in the range of 10 to 20% which is similar to the variation observed between spectrometers of the same manufacturer. However the sealed Xe counters which JEOL applies for higher energy X-ray lines, deliver a higher intensity signal than the gas flow counters Ar/CH_4 except for LIF towards lower energies. The JEOL spectrometers with TAPs

are better in all conditions but the crystal is very sensitive to irradiation and its efficiency deteriorates with time.

4.3 Radioactive background

The signal to background ratio is especially important for the measurement of active samples. One of the PET/LIF spectrometers was equipped with an Ar/CH_4-flow counter instead of a Xe counter in order to reduce the absorption of the γ-radiation. The difference is significant (Fig. 3) but not so much as it would exceed the higher efficiency of the Xe counter and the probably longer lifetime of the counter and window. The position of the crystal and counter respectively relative to the active specimen is decisive. Related to this is the background dependency on the exact position of the specimen in the holder and therefore on the local shielding environment.

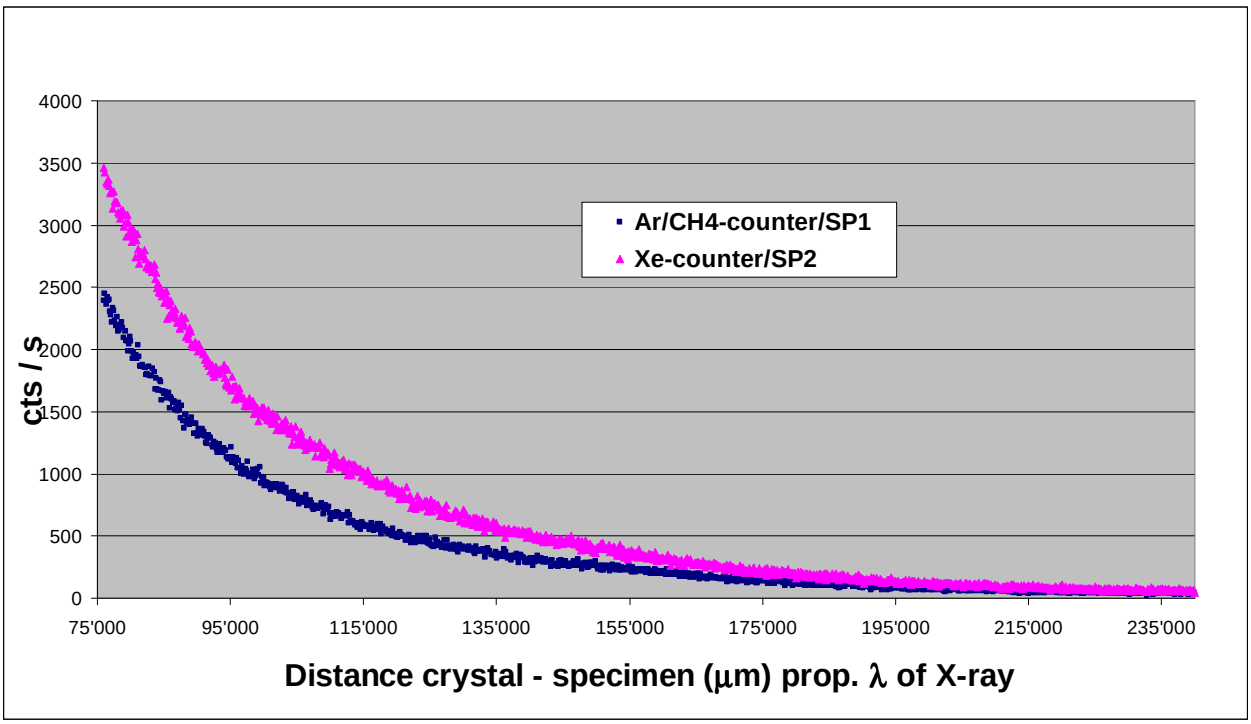

Fig. 3: Radioactive background measurements on two different X-ray counters (Ar/CH_4 gas flow and sealed Xe counters) in front spectrometers of JEOL 8500F. The γ-source is an irradiated encapsulated fuel rod sample with a dose rate of 8mSv/h/0.5m.

The other important impact of the radiation field on the background of the spectrometers in this shielding assembly is the position of the spectrometer itself with respect to the specimen. The front two spectrometers get at the moment about a six times higher background than the two in the rear where the background is very low. For this reason one spectrometer for the measurement of actinides, fission products and metals (PET/LIF) has been exchanged meanwhile and put into the back. An attempt to further shield the front spectrometers is ongoing.

An approximate comparison of the radioactive background between the two EPMAs with two spectrometer configuration types is shown in table 2. This assessment comes from different open and closed samples and is interpolated to a typical γ-dose rate. For the JEOL EPMA, quick measurements were made with both a γ- and a β,γ-radiation source. From these the share impact of the β-radiation and subsequent bremsstrahlung has been estimated to be

5 – 35% of the total background depending on the counter position relative to the shielding. As already said, the spectrometer and the sample position play a big role, especially for the shielded JEOL EPMA, the data must therefore be taken with care (see above). Nonetheless these measurements indicate a successful shielding of the JEOL EPMA.

Position			EPMA		
Element/Line (crystal)	λ [Å]	Sample-Crystal (JEOL) [mm]	CAMECA[(1)] SXR-50 (Ar) [cts/s]	JEOL 8500F (shielded)[(1,2)]	
				Ar/CH_4 [cts/s]	Xe [cts/s]
Nd Lα (PET)	2.37	75.9	2700	2000	2900
Cs Lα (PET)	2.89	92.6	1100	1000	1500
Pu Mα (PET)	3.51	112.5	800	500	900
U Mα (PET)	3.91	125.2	500	350	650
Zr Lα (PET)	6.07	194.4	180	65	85
O Kα (PC1/LDE1)	23.6	110.2	700	60	-

Table 2: Preliminary comparison of background measurements between CAMECA SXR-50 and JEOL 8500F (interpolation from different fuel samples, basis: 5 mSv/h/0.5m).
[(1)]The radioactive background depends on the sample position (variation 20-40%) and on the spectrometer used!
[(2)] Partly shielded EPMA, measurement on front spectrometers (PET) with higher background.
γ- and β,γ-sources employed in the test (background proportion due to β-radiation: 5-35%).
JEOL always requires a window setting in the pulse height analyser (differential mode)!

The JEOL EPMA always demands a differential window setting in the pulse height analyser (X-ray energy discriminator) for active specimens otherwise the background would be too high. This covers normally about 60% of the voltage range. This means that the pulse height distribution must always be checked for each element and analysis setup (avoiding of pulse height depression and broadening) but at least higher order reflections from other elements are then normally also suppressed.

5. Summary and conclusions

The new shielded EPMA demonstrates a high versatility with respect to examination capabilities and specimens that can be investigated. It has advanced SEM and X-ray imaging with a high(er) lateral resolution due to the FE-gun. The first test of the machine with active specimens shows:

- The imaging performance for low β,γ-active samples is very good for high active samples it must be verified.
- The spectrometer efficiencies of the CAMECA SXR and JEOL 8500F are comparable with advantages for the Xe counters employed in the JEOL EPMA. Also the spectral resolutions are at first sight comparable despite the small JEOL spectrometers. The first test on lateral X-ray resolution on a low active irradiated cladding specimen was good.

- The performance with respect to microanalysis of high active materials is promising (good shielding of β,γ-radiation). Minor improvements in the shielding geometry are still underway.
- The sample loading for embedded flat samples is convenient. Use and loading of a rotation and tilt sample holder must yet be adjusted.
- Further adjustments on the light microscope, automatic aperture and infrastructure (e.g. emergency specimen recovery) are also in progress.

Resulting from the different technical specifications, the new EPMA requires some modification and adaptation to the analysis and work procedures. In this context some open questions will have to be cleared in near future;

- Contamination risk (compact, open design).
- Lifetime of components can not yet be ascertained. Effect of higher γ-radiation in surrounding area during the measurements.
- Irradiation effect on the large and sophisticated vacuum system.
- The new control system (parameter adjustments, data acquisition, file transfer and handling) based on the operating system UNIX.
- Maintenance of a possibly slightly contaminated machine (service by remX / at the moment JEOL ensures the service of only non contaminated components).

6. Acknowledgments

We wish to thank CAMECA, JEOL and remX for the permission for the issue of data and photos and the support from remX to accomplish these examinations. Further to the service engineers of JEOL and remX who have worked hard to maximise the EPMA performance under new and difficult circumstances and our Hotlab maintenance crew in supporting the build-up.