



### **Task 3: Actinide targets properties after irradiation**

The Task 8.3 “Characterisation of irradiated targets in hot cell” has been performed at PSI.

This task include

- dismantling of the irradiated target from the containment in PSI hot cell
- extraction of sub-samples from the UC target for EPMA analysis
- extraction of sub-samples from the UC target for microXAS analysis
- characterisation of the UC material of the different sub-samples by EPMA

Electron Probe Micro analyser (EPMA) has been used in order to characterise specimens extracted from different elevations of the irradiated target. The examination on this task could be completed in September 2014. The results are presented in the released PSI report TM-43-14-07. Following a summary of the PSI report is shown.

In the ENSAR/ActiLab project three uranium carbide specimens irradiated by a proton beam in a graphite tube were prepared in the hot cell for subsequent EPMA analysis of spallation and fission products in the targets.

#### **Specimens**

Three specimens were selected (Tab. 1) and cut out at different positions from the 20 cm long target tube (Fig. 5) near the beam entrance (EPMA/Cer\_E), in the middle (EPMA/Cer\_M) and at the end (EPMA/Cer\_B).

An unirradiated specimen was examined as reference material A.

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**Hot Cell Preparation**

The preparation had to be carried out under nitrogen atmosphere except for the short surface decontamination and sample loading. The irradiated specimens B and M have been prepared in a 17 mm diameter SIMS holder (Fig. 3, 4) and specimen E and the reference specimen A in a one inch steel holder (Fig. 1, 2). The latter one was already pre-mounted on a SEM holder. The preparation was done according to the standard procedure with epoxy resin embedding, grinding and polishing in steps with a final ¼ µm oil-based diamond polishing slurry.

Specimens A, E and M, B are embedded in one inch holders and 17 mm Φ holders, respectively.

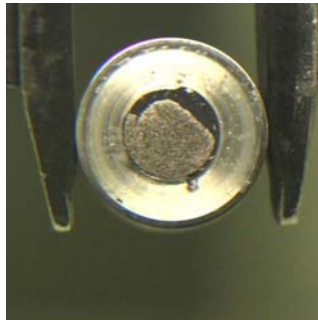


Fig. 1 Reference A

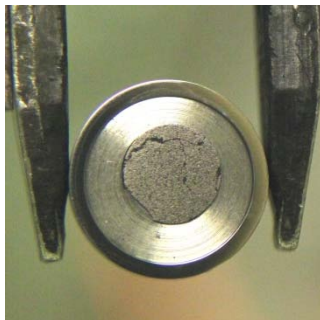


Fig. 2 Specimen E

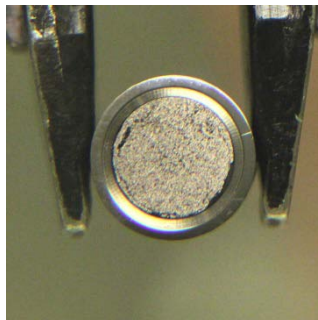


Fig. 3 Specimen M

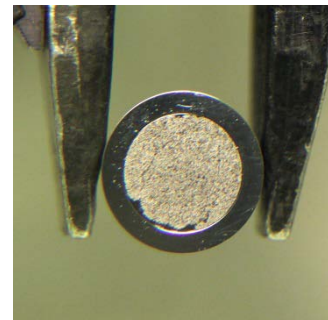


Fig. 4 Specimen B

To assess the background level for EPMA analysis the dose rate is usually measured in a distance of 50 cm. The dose rates for the irradiated specimens were very low (< 1 µGy/h) for this distance. Under these low activity circumstances no contamination level could be determined during cleaning of the samples with ethanol in an ultrasonic bath.

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Table 1: Sample cutting for microXAS beamline and EPMA examination



Sample cutting for microXAS beam line October 2013 and EPMA

Table 1: Samples fabricated out of the CERN target

Sample name	Number of pellets	Responsible target area	comments
CERN_EPMA/Cer_B	1	Sample from the area of the graphite plug	EPMA examination, sort of ceramography
CERN_XAS_BC	0.5	Sample from the area of the graphite plug	microXAS sample take from the pellet center of the half pellet (FIB)
CERN_XAS_BR	0.5	Sample from the area of the graphite plug	microXAS sample take from the pellet rim area of the half pellet (FIB)
CERN_EPMA/Cer_M	1	Sample from the center of the target looking from graphite plug	EPMA examination, sort of ceramography
CERN_XAS_MC	0.5	Sample from the center of the target looking from graphite plug	microXAS sample take from the pellet center of the half pellet (FIB)
CERN_XAS_MR	0.5	Sample from the center of the target looking from graphite plug </td <td>microXAS sample take from the pellet rim area of the half pellet (FIB)</td>	microXAS sample take from the pellet rim area of the half pellet (FIB)
CERN_EPMA/Cer_E	1	Sample from the end of the target looking from graphite plug	EPMA examination, sort of ceramography
CERN_XAS_EC	0.5	Sample from the end of the target looking from graphite plug	microXAS sample take from the pellet center of the half pellet (FIB)
CERN_XAS_ER	0.5	Sample from the end of the target looking from graphite plug	microXAS sample take from the pellet rim area of the half pellet (FIB)
CERN_Test	1		Control of the oxidation conditions

→  
→  
ob./p.s. →

+ Anzahlprobe UC<sub>2</sub> (Nr. A)

➤ Sample cutting for microXAS beam line and EPMA examination

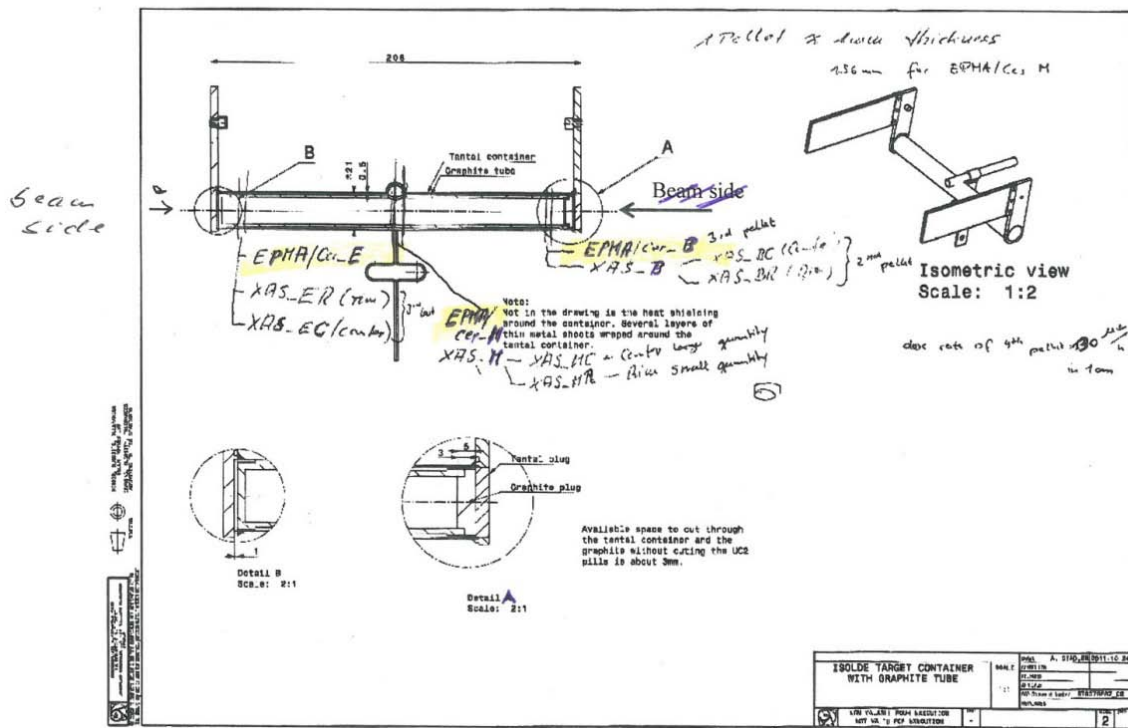


Fig. 5: Cutting plan for microXAS and EPMA examinations of irradiated UC-target



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**Conditions for EPMA Analysis**

The electron probe microanalyser (EPMA) is a shielded JEOL 8500F with field emission gun (FE-EPMA) and four wave length dispersive spectrometers (WDS). Extensive scans on 4 spectrometers were carried out in the centre and at the border of the specimens to track possible trace elements with the diffracting crystals TAP, LIF, PET and LD1 to be able to detect all elements from carbon on.

Qualitative linescans across the specimens were made with an almost focused beam in order to find out a gradient in uranium, carbon and oxygen. At a few points also qualitative mappings were acquired to illustrate the distribution of uranium carbide and graphite.

Analysis conditions were chosen as follows:

High voltage electron beam	20 kV
Beam current	200-210 nA ± 0.5 nA
Beam diameter	≈ 0.2 μm
Lateral resolution for X-rays	≈ 1 μm
Beam	1 μm beam φ for each point of linescan, 2 μm beam φ for spectrum scan and focused beam for mapping.
Elements	U, Cl with PET diffracting crystals of spectrometer 3 (SP3) and 2 (SP2). O and C with SP4/LD1 and SP1/LD2.
Measuring time for peak or point	2.5 s for linescans. 3.5 s for spectrum acquisition.
X-ray mappings	18 x 18 μm <sup>2</sup> up to 50 x 50 μm <sup>2</sup> with beam scanning (256 x 256 pixels). Acquisition time: typically 110 min. per element (100 ms/pix.).
Step size in spectra	0.11 mm which corresponds to sin Θ= 0.00039 (see below)

**Results**

The x-axis scale of the given spectra is normally given in Bragg's angle sin Θ. To convert to wave length λ (Å) or energy E (eV) the following equations must be applied:

$$n * \lambda = 2d * \sin \Theta; \quad \lambda \text{ and } d \text{ in } (\text{\AA}), \quad \text{integer } n / \text{ Bragg's diffraction law,}$$
$$E = 12398 / \lambda$$

Lattice plane 2d values (Å): TAP: 25.745,            LIF: 4.0267,    PET: 8.75

L (mm) = 280 \* sin Θ; for JEOL 8500F with distance L between specimen and crystal



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**Results of the reference specimen A**

The SEM images are on Fig. 6 – 7. The porosity of the sphere is very high. Twinning is observed on Fig. 6. On the spectra the uranium peaks and the X-ray line positions of the possible spallation products of the irradiated specimens are marked. It helps to detect the presence of these elements.

Interferences with different degree for the following main characteristic X-ray lines with uranium lines are found: CsL $\alpha$ , NdL $\alpha$  (not on LIF), TeL $\alpha$ , ThM $\beta$  (not M $\alpha$ ), BiM $\beta$  (not L $\alpha$ ), WM $\alpha$  (not L $\alpha$ ), PuM $\alpha\beta$ , AmM $\alpha$ , NpM $\alpha\beta$ , PoM $\alpha$  (not M $\beta$ ), PaM $\alpha$ .

For PuM $\beta$  the interference of uranium is about 0.9 wt%. Other than uranium interferences are not shown and must be checked as well (e.g. ClK $\alpha$  on PoM $\beta$  or TcL $\alpha$  on BiM $\alpha$ ).

Based on the spectra of one site at the fuel periphery there is an indication that the oxygen content at the specimen surface could be lower than in the centre whereas for carbon it is the other way round. The evaluation of point analyses along a line (linescan) could maybe prove it (see as example specimen E).

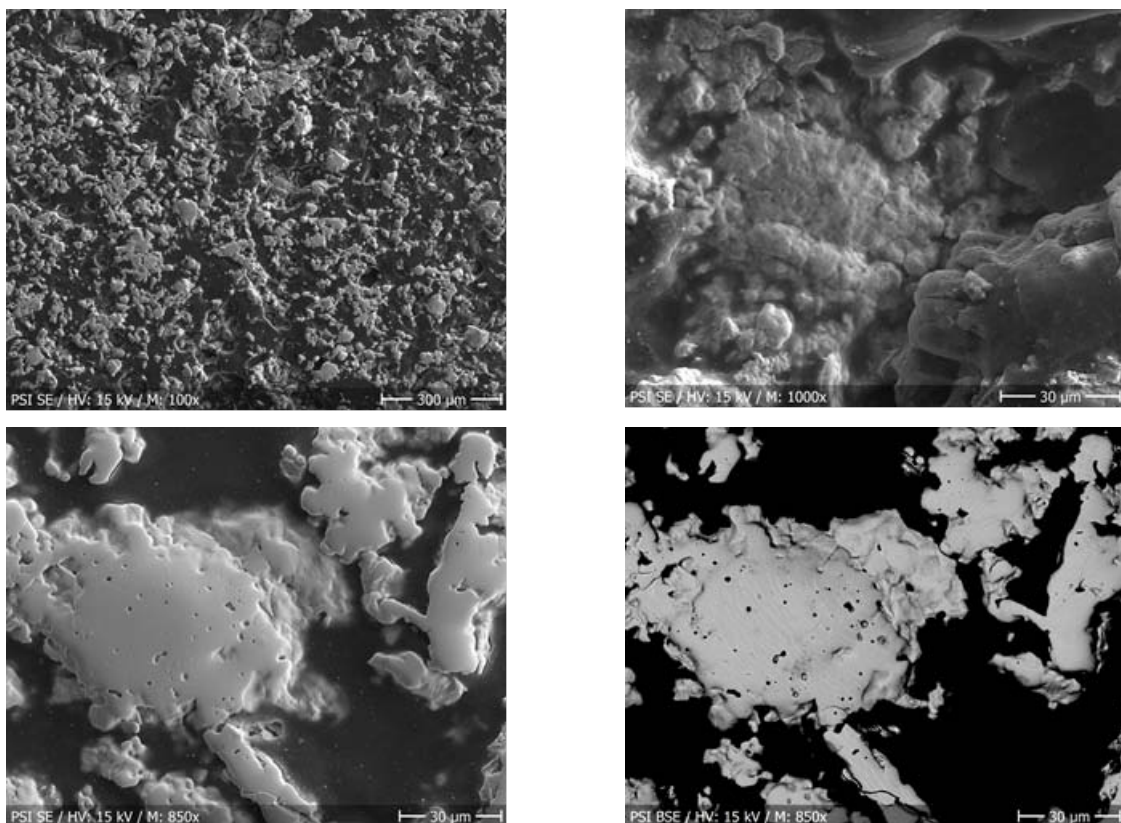


Fig. 6: Reference A: SEM, BSE, pellet center

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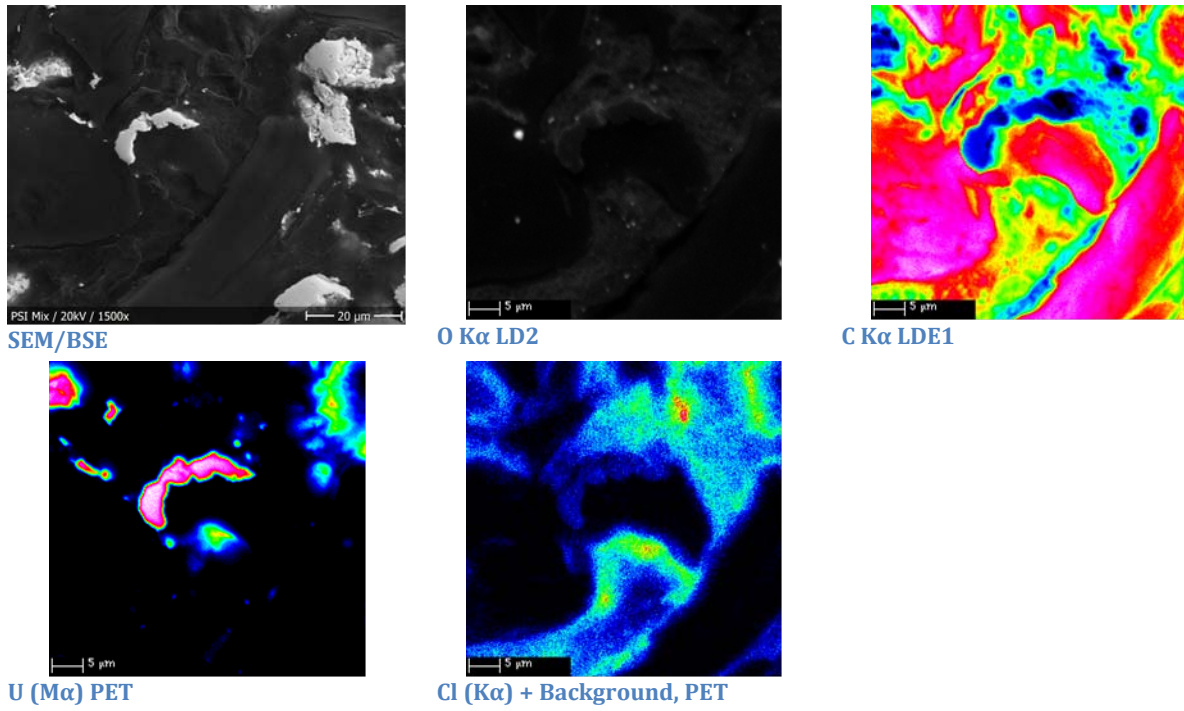


Fig. 7: Reference A: SEM/BSE image and qualitative mappings (section of SEM). Pos. right.

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**Results of specimen E**

The spectra show no evidence of significant amount of spallation products. Under the applied acquisition conditions the detection limit for spectra with PET crystal is around 0.1 wt% and for LIF crystal even lower without considering the background and interference level. It is therefore not possible to trace elements in the scale of tenth or hundreds of ppm.

CERN made a linear fit of the radial linescans data. There seems to be a decrease in the carbon content from the border to the centre as indicated already for the reference specimen.

The SEM images and mappings are on Fig. 8. No change in the high porosity is observed at first sight. Strong twinning in the grains is generally present.

Graphite phases can be met not only at the border but also in the centre (Fig. 8). Surface oxidation has occurred and seems to be high in the pores.

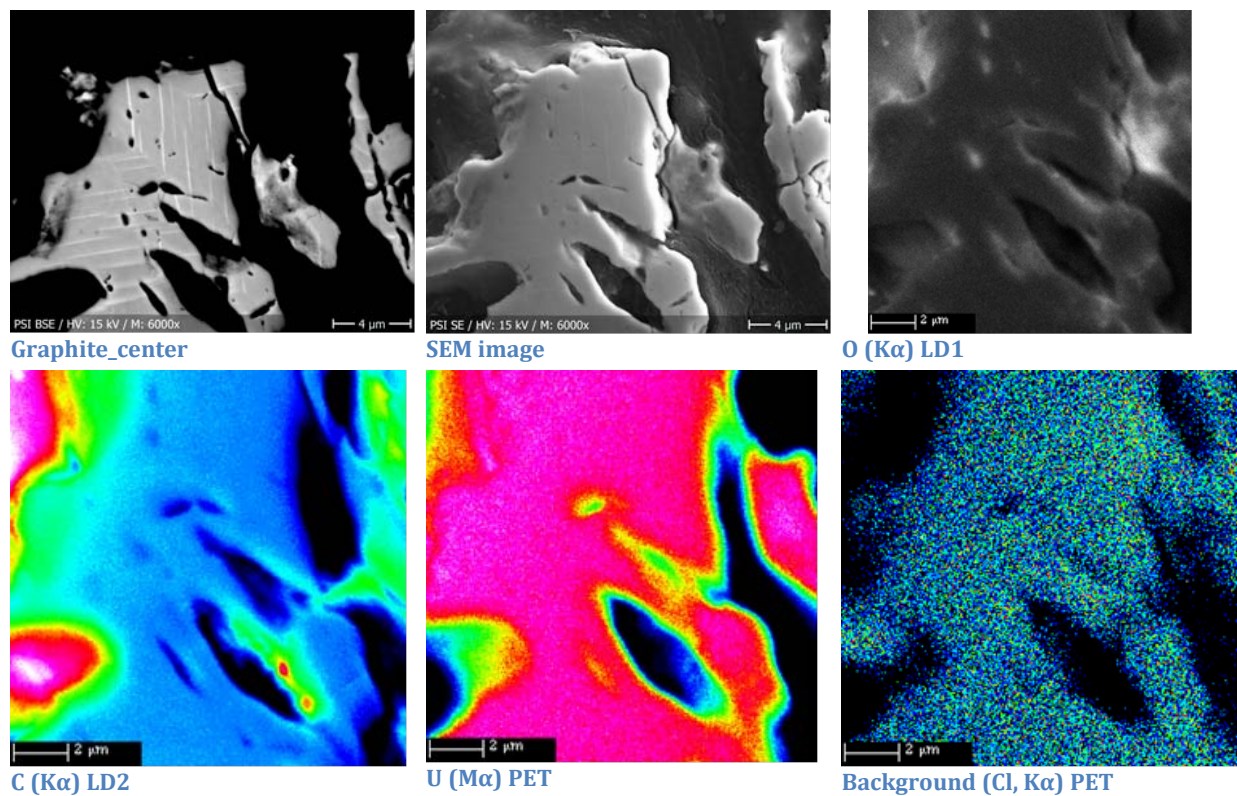


Fig. 8: Specimen E: SEM, BSE image and qualitative mappings (section of SEM). Pos. center



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**Results of specimen M**

No differences as to spectra and SE-Images are seen at a glance in comparison to specimen E. Twinning is again present but not so well seen because of reduced contrast in the images. The elemental linescans have to be evaluated. Cl was not present on the mappings (Fig. 9) as on the reference specimen.

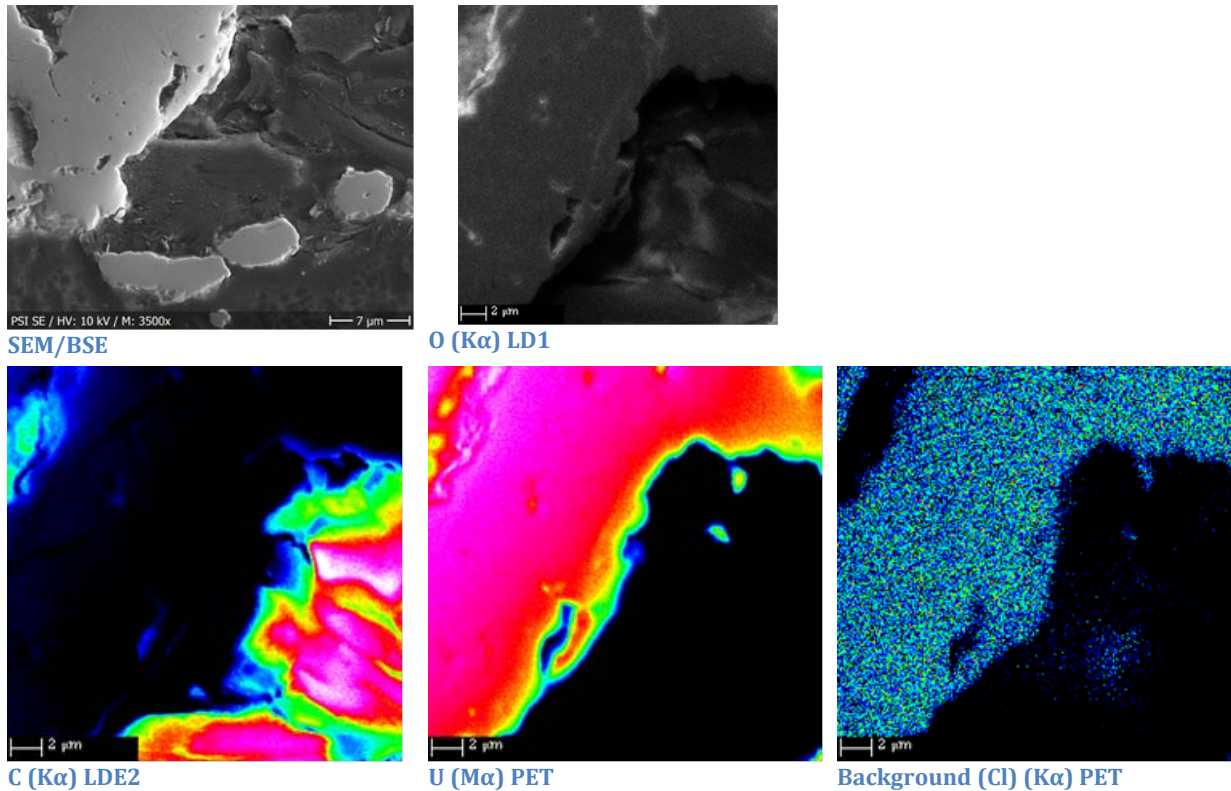


Fig. 9: Specimen M: SEM image and qualitative mappings (section of SEM). Pos. top border.



**Results of specimen B**

Again no differences as to spectra and SE-Images are seen at a glance in comparison to specimen E. The elemental linescans have to be evaluated. Cl is present on all of the five sites examined (Fig. 10).

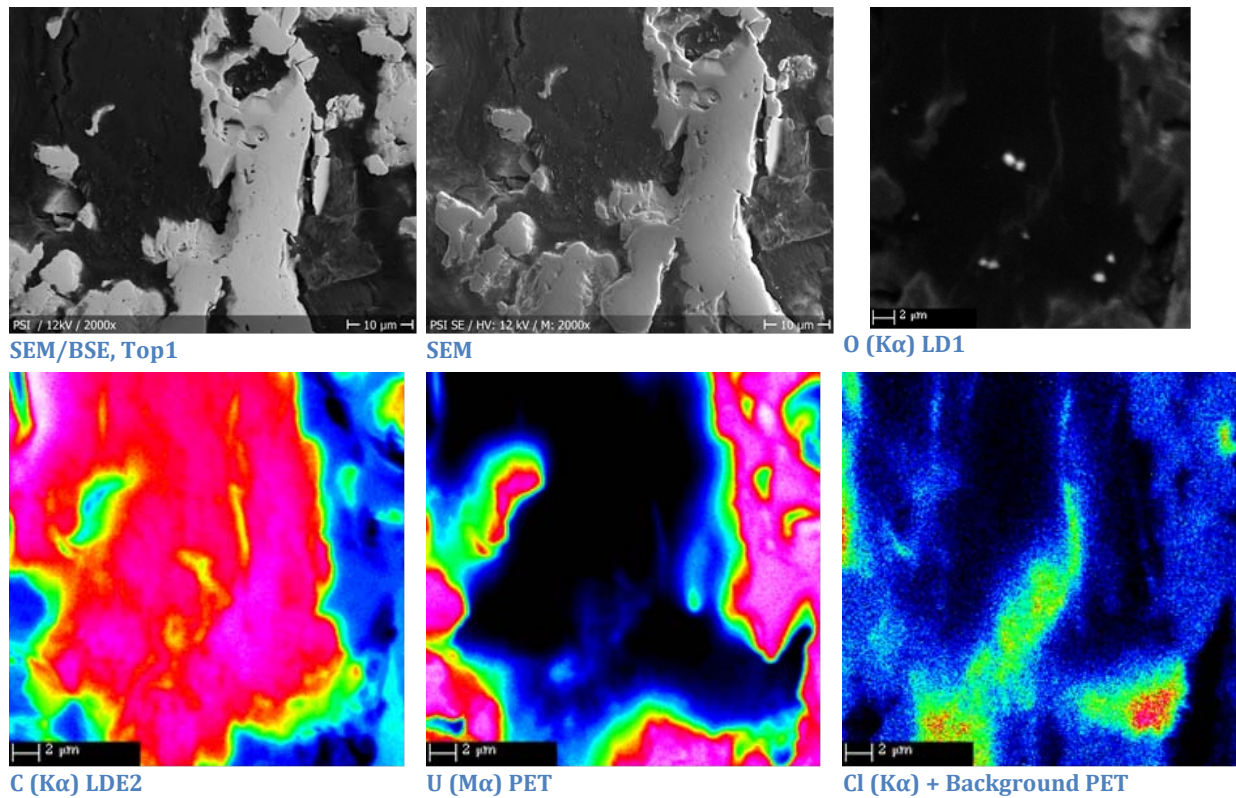


Fig. 10: Specimen B: SEM, Mix (SEM-BSE) image and qualitative mappings (section of SEM). Pos. top border.

**Summary**

No significant changes through irradiation of the examined specimens could be observed and detected. Differences are morphologically in the sub-micron and nano-scale and possible spallation products in the ppm concentration range. This means that crystallographic and stoichiometric information would have to be acquired by e.g. XRD and the content of spallation products by ICP-MS and SIMS.

There seems to be a radial decline in carbon content. Some oxidation of the surface especially of pores during preparation could not be avoided.