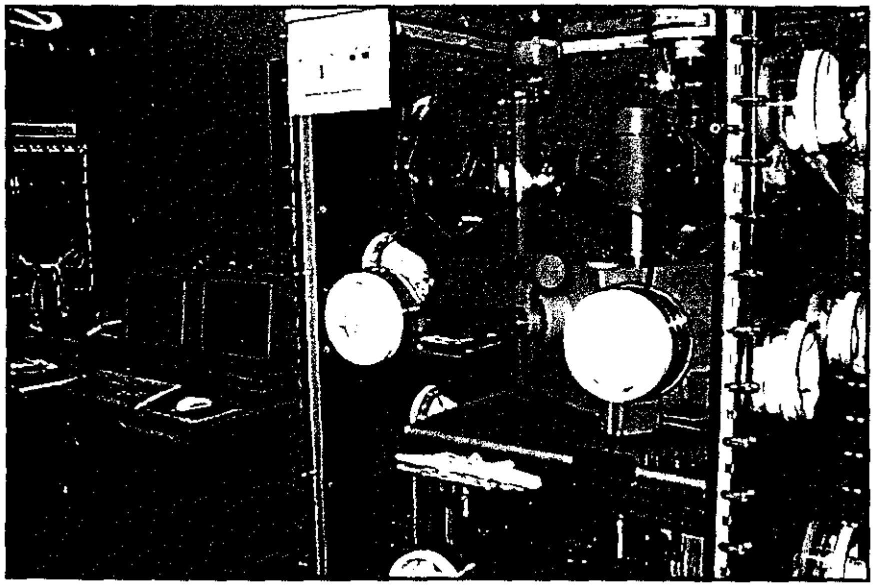
EUROPEAN WORKING GROUP on "HOT LABORATORIES AND REMOTE HANDLING"

# Electron Microscopy for PIE: New Developments

**T. Wiss, I. Ray, H. Thiele, W. Huber and Hj. Matzke**



**Philips XL 40 Scanning Electron Microscope**

From: "A Scanning Electron Microscope for the examination of Radioactive and Contaminated Samples"
I. Ray, W. Huber, H. Thiele, T. Wiss and Hj. Matzke
Submitted to *Review of Scientific Instruments*



Institute for Transuranium Elements
Applied Physics Unit
JOINT RESEARCH CENTRE
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## Outline

General description of the SEM XL40

- Micromanipulators
- Gun Shot Residue software

Examples of investigations

-swipe tests in forensic analysis
-material research
-leaching studies

*SEM combined with TEM*

-cladding analysis
-characterization of $PuO_2$ powder

Summary

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# The SEM Philips XL40

## Imaging

- **Secondary Electrons for imaging.**
- **Backscattered Electrons giving Z contrast (e.g. phase contrast on polished metals, particles detection).**

## Analytical

- **Energy Dispersive X-ray (EDX) for element analysis (qualitative and quantitative).**
- **EDX mapping for element distribution.**

## Technical features

- **Micromanipulators for small particles handling.**
- **Big chamber with automatic stage operation.**
- **Mounting in glove box (to be finalised) for radioactive sample examination.**

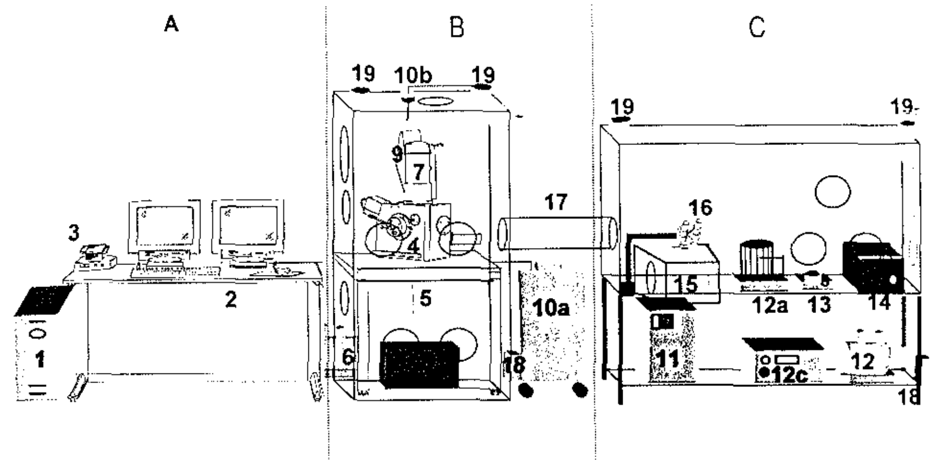
## Technical data

- **Accelerating voltage: 300 V to 30 kV.**
- **resolution: 2 nm.**
- **EDX resolution ~ 100 eV.**





# The SEM XL40 layout




Schematic diagram of the modified SEM showing three major parts of the installation: A *the control console* with: 1 computer unit, 2 photo camera, 3 operating console with monitors, keyboard, mouse and micromanipulator command console; B *the SEM box* with: 4 specimen chamber, 5 turbomolecular pump, 6 rotary vacuum pump, 7 electron column, 8 high tension generator, 9 cooling reservoir on the EDX detector, **10a** liquid nitrogen tank, **10b** feed-through for liquid nitrogen; C *the specimen preparation and instrument servicing box* with: **11** external column water cooling system, **12a** coating container, **12b** coating system console, **12c** coating system vacuum pump, **13** heating plate, **14** ultrasonic bath, **15** additional plexiglas box equipped with **16** stereo microscope for precise manipulations (e.g. Wehnelt filament changing).

The two gloveboxes are connected via a tunnel, **17**, for samples and tools transfer. They are equipped with alarm (fire, flooding, underpressure and operator controlled) boxes, **18** and particle filters, **19**, for connection to the main atmosphere circuit.

# Scanning Electron Microscope Investigations on Materials

## Objectives:

Characterisation of the microstructure (porosity and grain size), of the surface morphology, of particle shape, dimension, element analysis (qualitative, quantitative) and distribution,
**In:**
Nuclear Fuel, Inert Matrices for the transmutation of minor actinides
-Virgin materials (as fabricated) e.g. $MgAl_2O_4$, $CeO_2$, $UO_2$, $ZrO_2$,…
-Ion irradiated materials
-Reactor irradiated fuels, inert matrices.
-Leached specimens

Other materials
-Cladding
-Thin layers
-Particles in swipes tests

## Techniques:

SEM equipped with **Secondary Electron Detector** for imaging, **Backscattered Electron Detector** for Z contrast, **Energy Dispersive X-ray Analysis Detector** for elemental analysis and **micromanipulators** for small sized particle handling under the electron beam.



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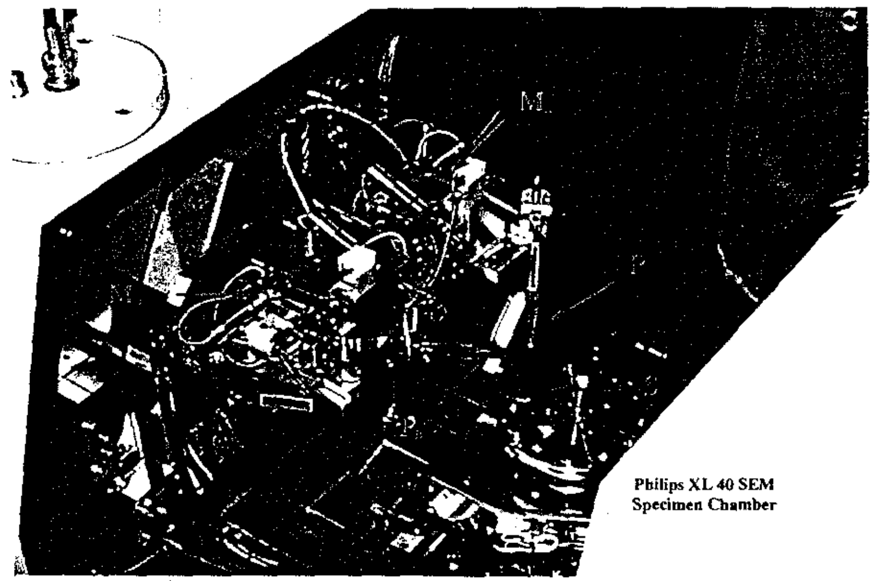
# Micromanipulator Module in the Philips XL 40 SEM



**Philips XL 40 SEM Specimen Chamber**

The Kammrath & Weiss Micromanipulator Module mounted inside the specimen chamber of the Philips XL 40 Scanning Electron Microscope. The two independent microtweezers $P_A$ and $P_B$ are driven in the X, Y, and Z axes by step motors and piezo-electric crystals in the control modules $M_A$ and $M_B$, to perform manipulations on the specimen which is shown indicated at S.

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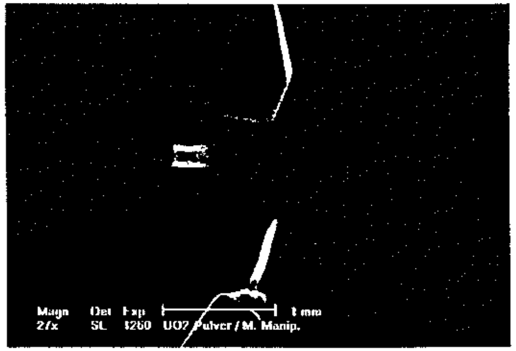
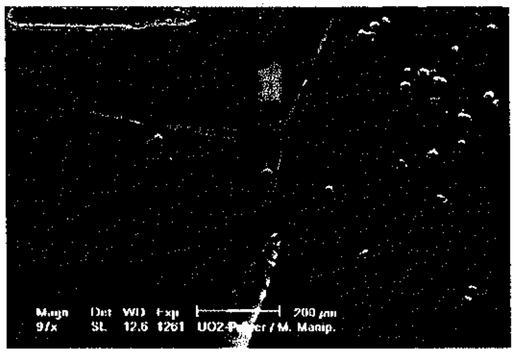
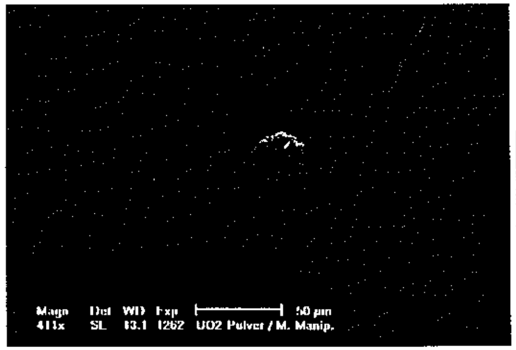
# Micromanipulation in the Philips XL 40 SEM



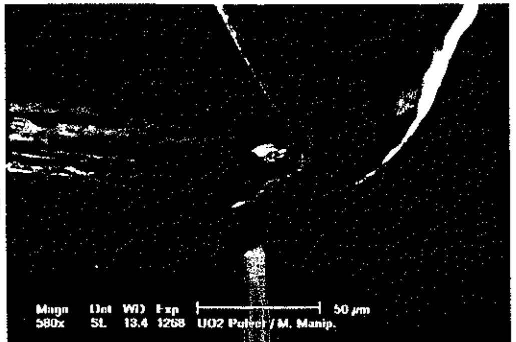

SEM micrograph of the tweezer (up) and the helping hand (bottom) above the specimen.




Selection of a $UO_2$ particle (marked at A) for transfer.




The microtweezer is lowered to contact the particle, and closed to grip it.




Lift off! The particle is lifted clear of the surface.



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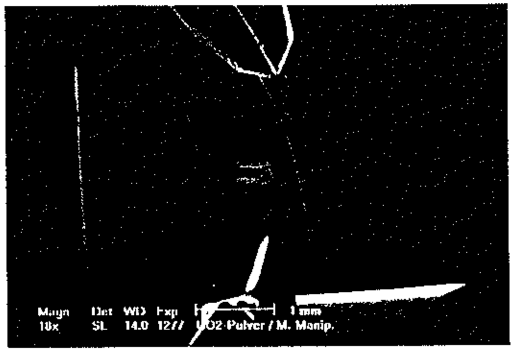
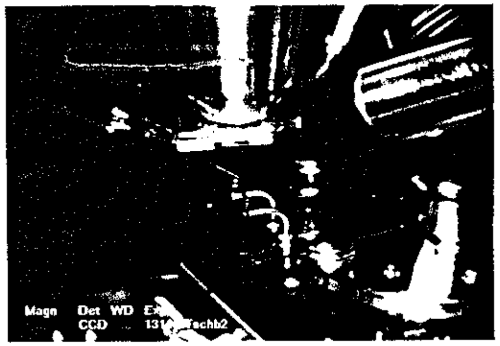
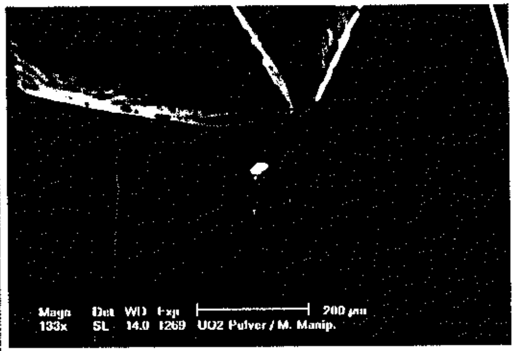
Micromanipulation in the Philips XL 40 SEM



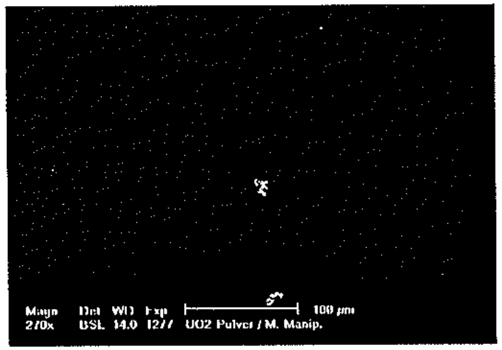

The particle is deposited on the new specimen holder with the help of a very fine needle.



View of the specimen chamber during particle deposition on a MS filament (B).




The particle is sticked on a TIMS filament.




A BSE micrograph confirms the nature (U) of the particle.

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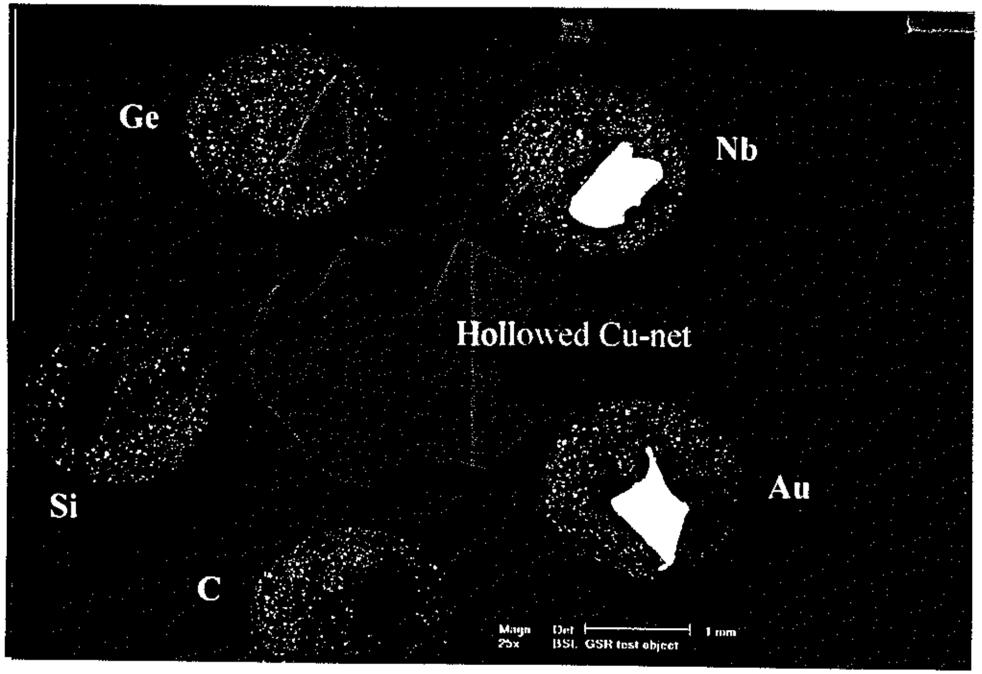
# Gun Shot Residue analysis

EDAX DxGSR allows analysis of particles completely automatically. It controls SEM column, its motor stage, the EDAX DX4 X-ray spectrometer and the BSD detector.



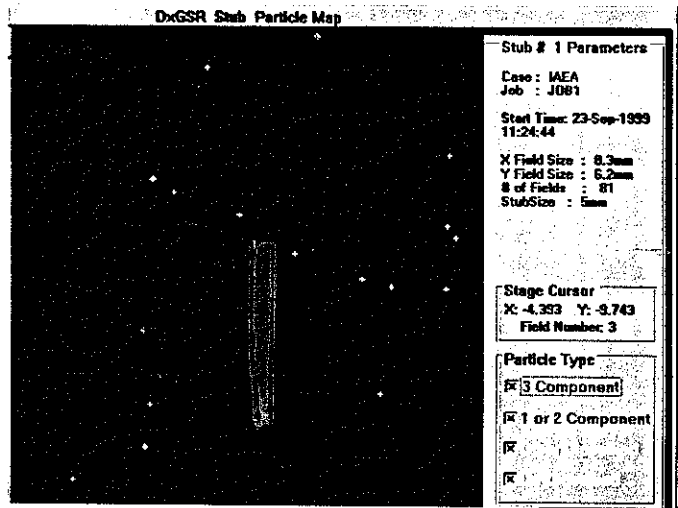

**BSE micrograph of calibration stub for GSR**

Calibration of the grayscale of the video signal is required for Gun Shot Residue analysis. Setting of that parameter allows for specific element/element-group detection.

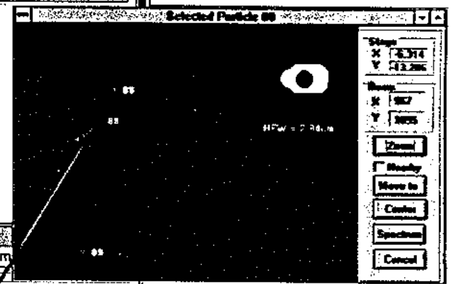


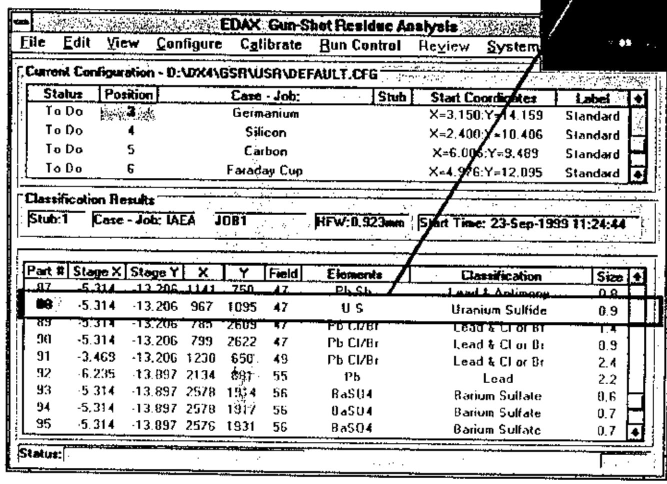


# GSR: Reviewing Stub Particle Map and Particle



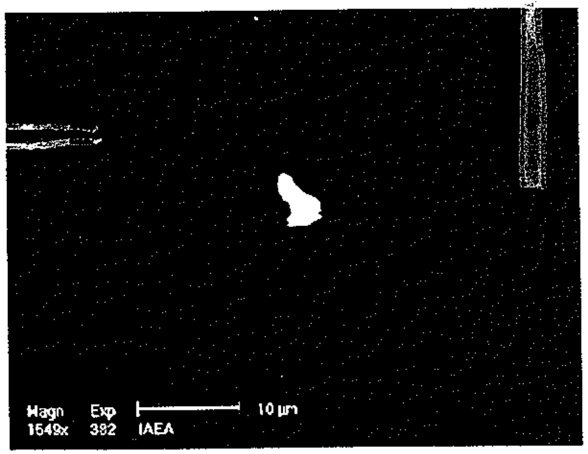

**Map of the search area with location of the detected particles.**

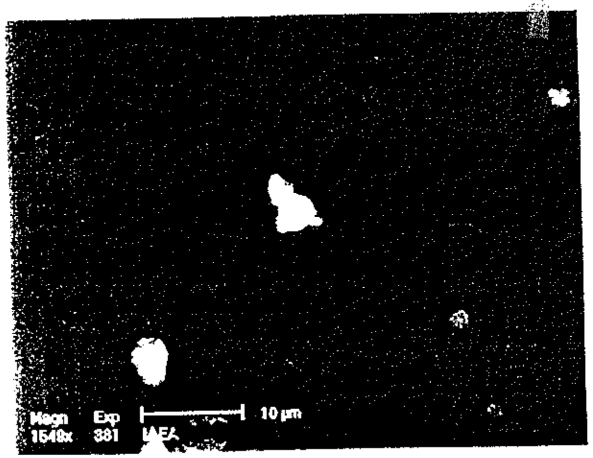



**List of the detected particles indicating their position, composition and size.**

# BSE and SE micrographs of a $UO_2$ particle on IAEA swipe test



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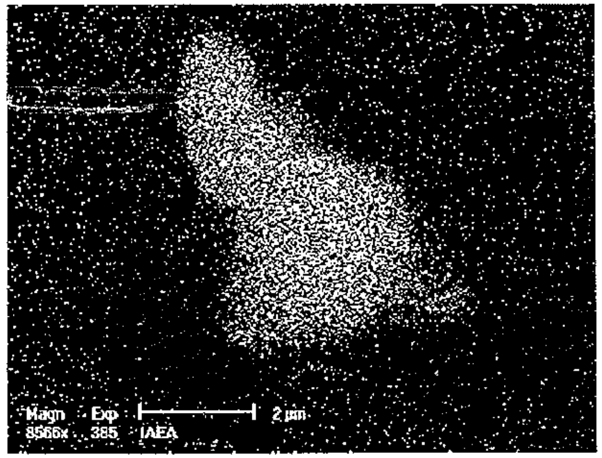
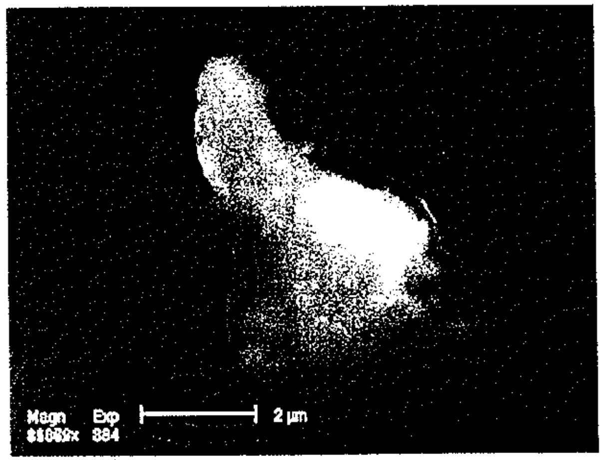
# Mapping and magnificated SE micrograph of a $UO_2$ particle

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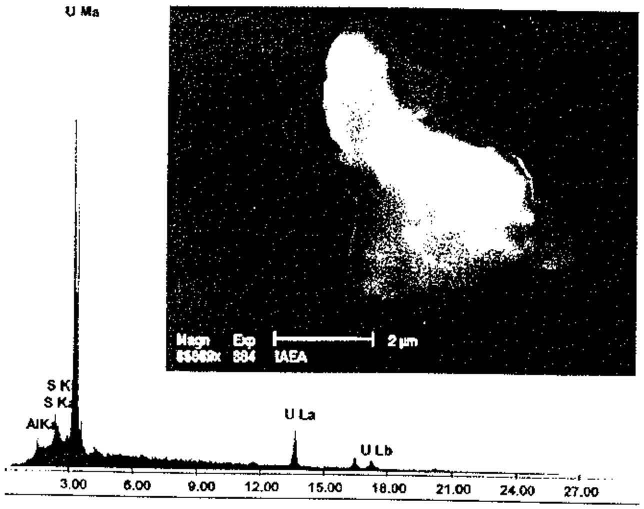
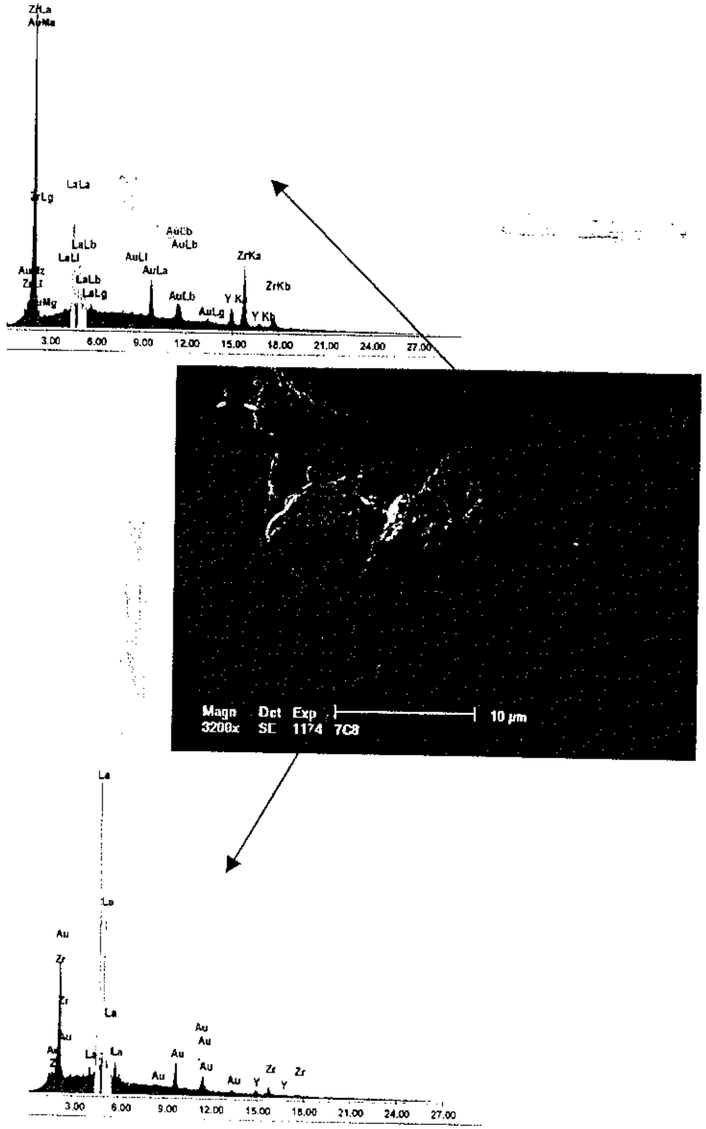
# EDX spectra and SE image of a $UO_2$ particle



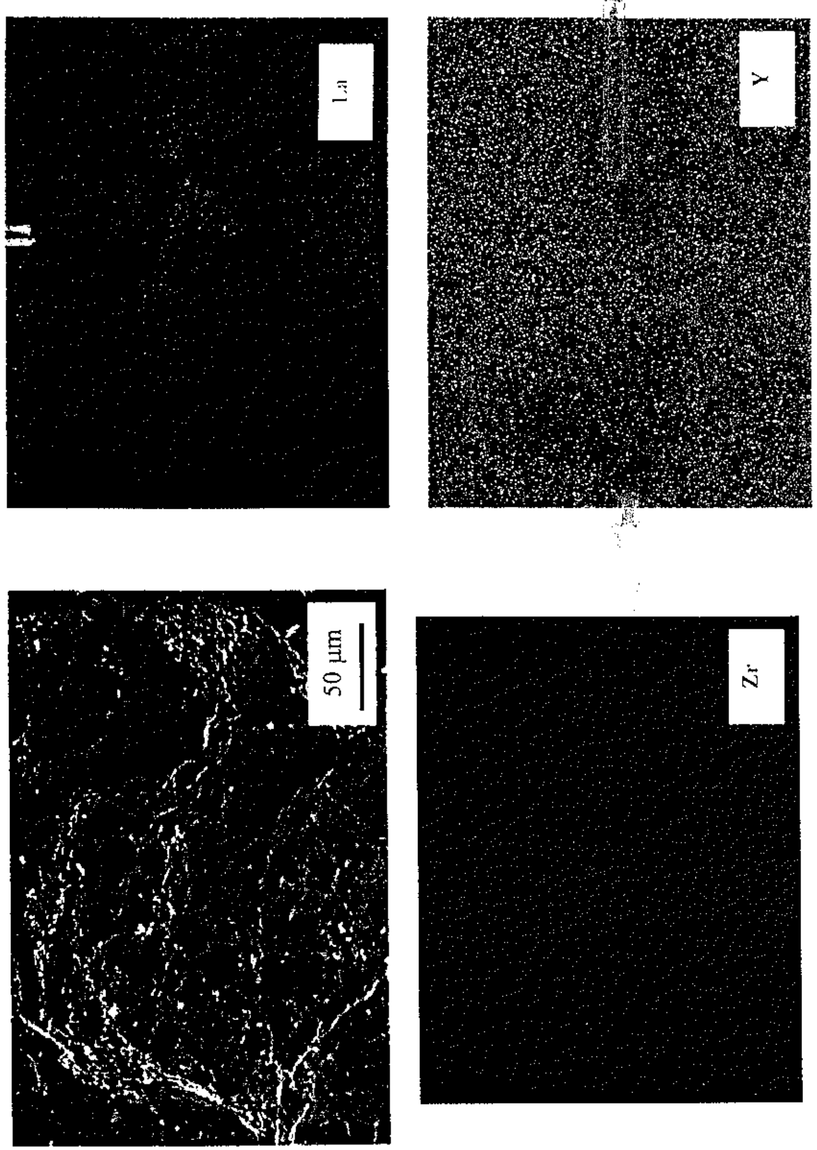





# EDX analysis on Yttria-stabilized $ZrO_2$ with La-content



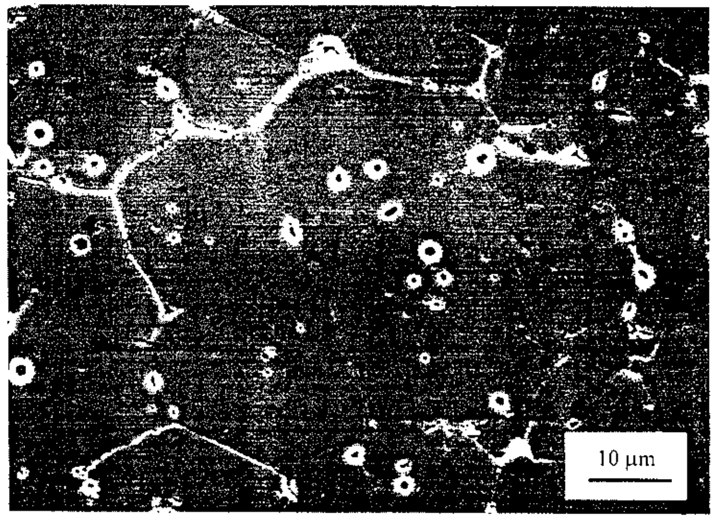
## SEM micrograph and mappings of Yttria-stabilized $ZrO_2$ with La-content

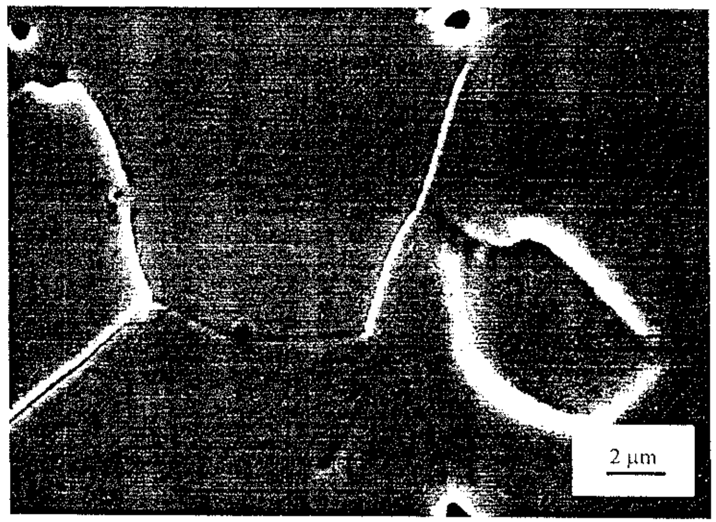




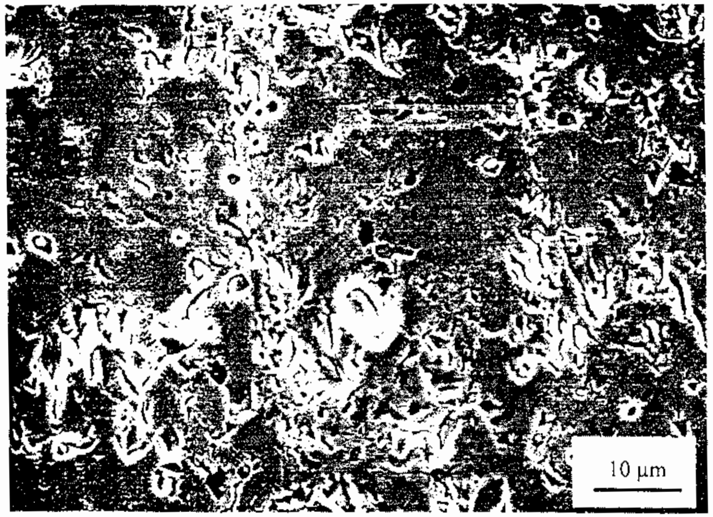



## Surface of SIMFUEL 3% at. unleached



## SIMFUEL 3 % at. leached 1 h in granitic water at 200 °C in presence of granite under argon atmosphere







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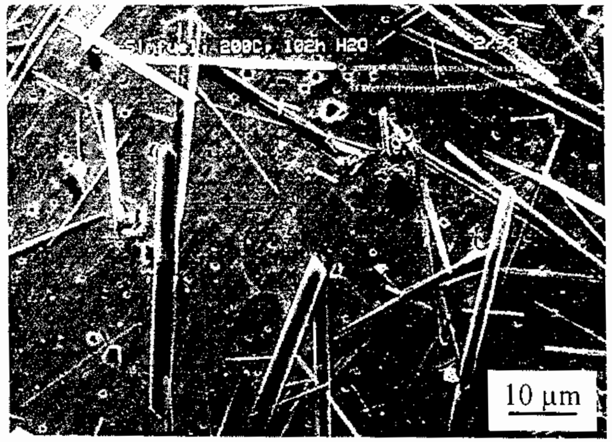


SIMFUEL with 3% burnup leached at 200 °C for 102 h in demineralized water in air.

VVR/HJM-jnm1

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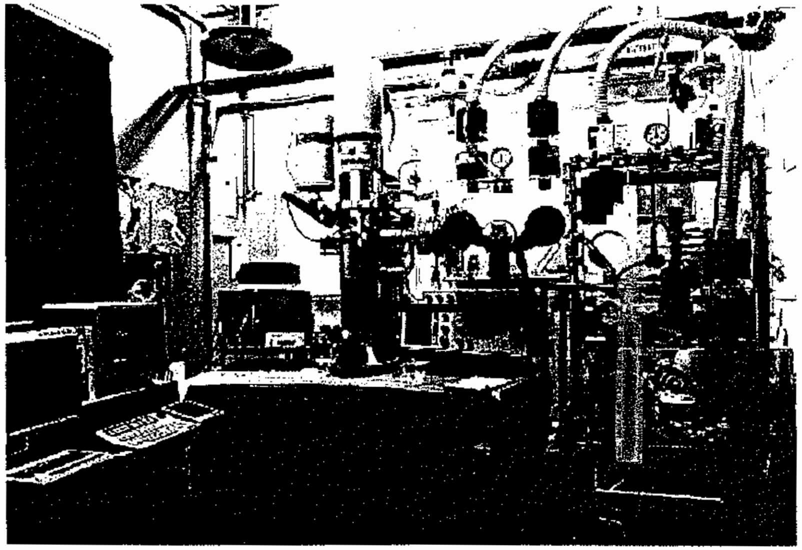
# Hitachi STEM H700 ST for radioactive sample examination



Operating voltage: 200 kV

Resolution: 2 Å

EDX detector

System of glove boxes

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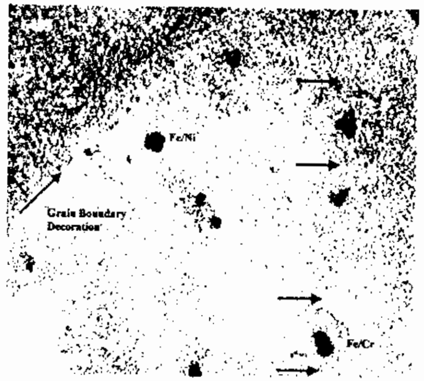
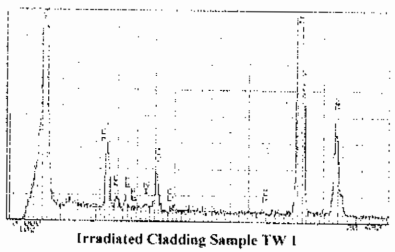
# Analysis of Intermetallic Precipitates in Irradiated Zircaloy Cladding

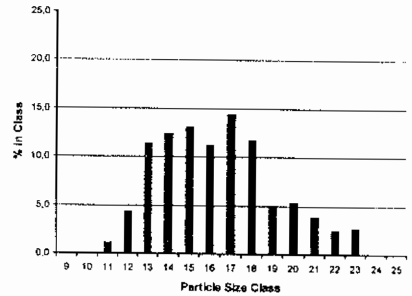

Transmission Electron Micrograph A




Transmission Electron Micrograph B




EDX Spectrum from a Fe/Cr-rich Particle




Intermetallic Particle Size Distribution

**Analysis of the composition, density and size distribution of the intermetallic particles in a sample of irradiated zircaloy cladding. The low magnification TEM micrograph A shows a high density (2.55 $\mu m^{-2}$) of FeCr-rich and FeNi-rich particles within a grain of the zircaloy. The TEM micrograph B reveals FeCr-rich particles showing traces of dissolution (red arrows), a large FeNi-rich particle and grain boundary decoration by small precipitates (blue arrow). The EDX spectrum is from a typical FeCr-rich particle. The intermetallic particle size distribution is based on measurements of 400 particle equivalent diameters. The particle size axis is divided into logarithmic classes from 15 nm to 486 nm.**

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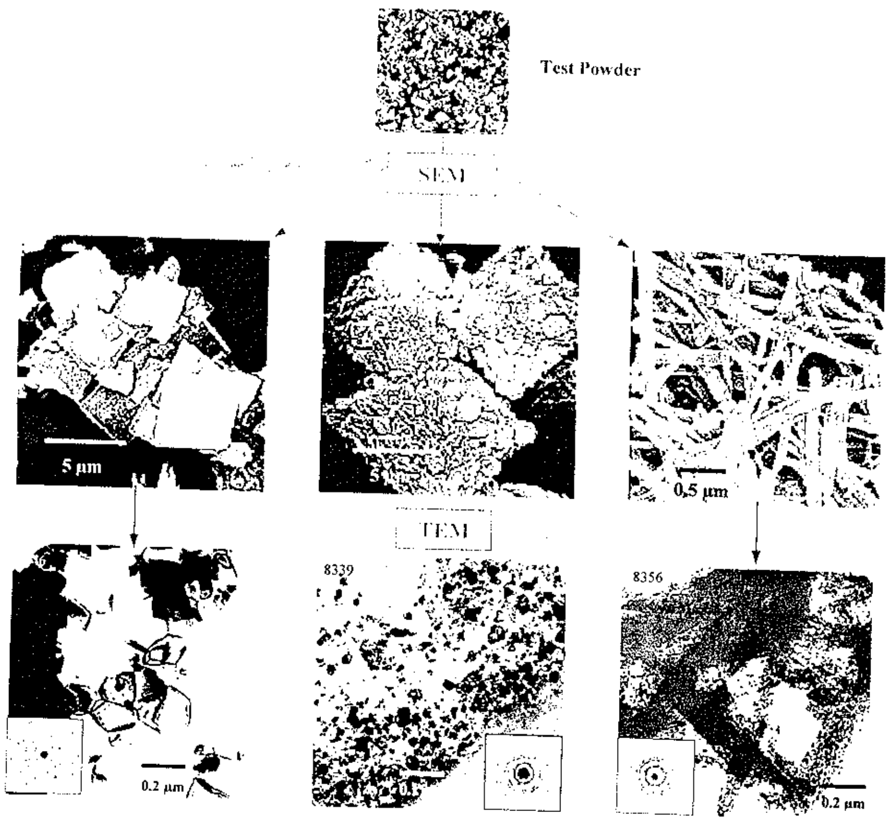
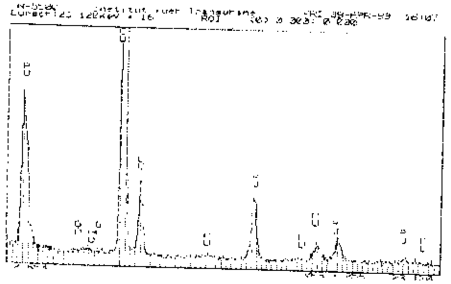
## Characterization of a $PuO_2$ Powder Sample

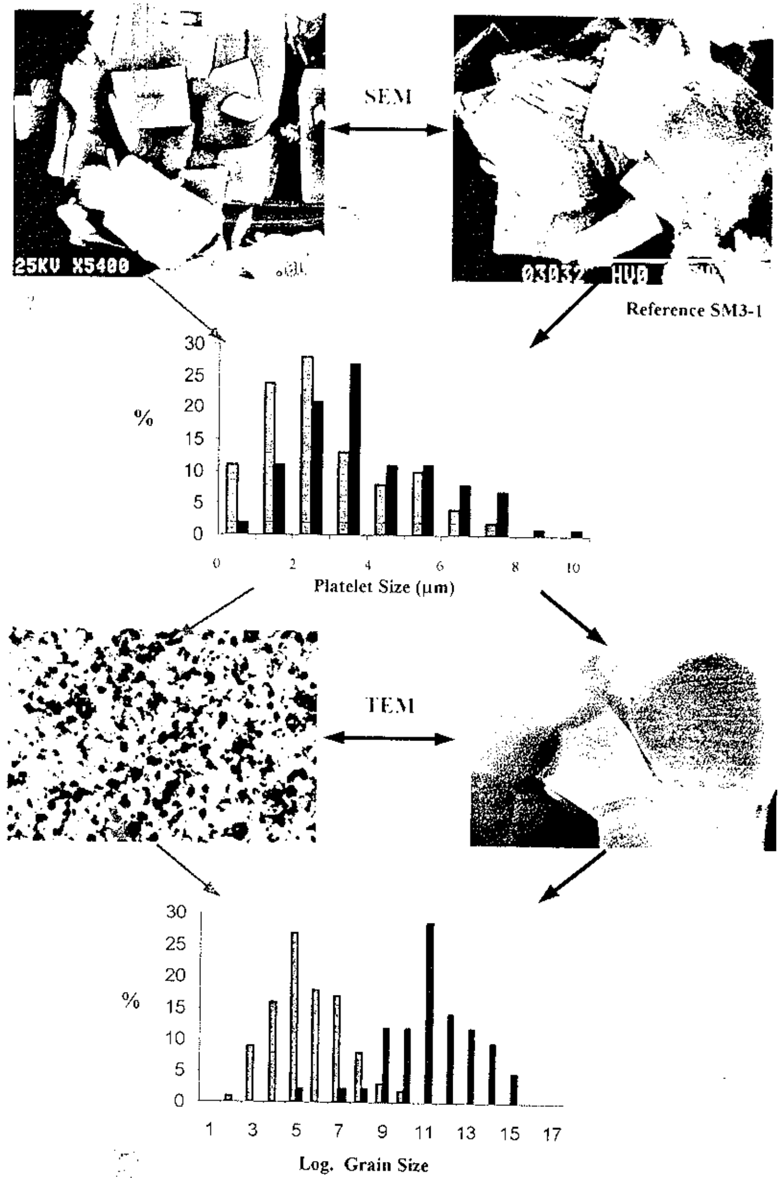



EDX





## Comparison of Plutonium Oxide Platelets



# Summary

- SEM is a versatile instrument
- Many related analysis capabilities

  BSE, EDX, mapping, micromanipulation, automatic particle detection (GRS)
- A large range of materials can be investigated
- Easy and quick techniques to get informations on:

  Microstructure, surface morphology, chemical composition, phases distribution,...

Combination of SEM and TEM provides a powerful tool for sample microstructure characterization.