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

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 PuO₂ powder

Summary
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. MgAl₂O₄, CeO₂, UO₂, ZrO₂,...
- 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.

The Kammrath & Weiss Micromanipulator Module mounted inside the specimen chamber of the Philips XL 40 Scanning Electron Microscope. The two independent micromanipulators 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.
Micromanipulation in the Philips XL 40 SEM

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

Selection of a \( \text{UO}_2 \) particle (marked at A) for transfer.

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

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

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 stuck on a TIMS filament.

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

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.
BSE and SE micrographs of a UO₂ particle on IAEA swipe test

Mapping and magnified SE micrograph of a UO₂ particle
EDX spectra and SE image of a UO₂ particle

EDX analysis on Yttria-stabilized ZrO₂ with La-content
SEM micrograph and mappings of Yttria-stabilized ZrO₂ 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

SIMFUEL with 3% burnup leached at 200 °C for 102 h in demineralized water in air.
Hitachi STEM H700 ST for radioactive sample examination

Operating voltage: 200 kV
Resolution: 2 Å
EDX detector
System of glove boxes

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 intermetallic particles in a sample of irradiated zircaloy cladding. The low magnification TEM micrograph A shows a high density (2.55 μm⁻¹) of Fe-Cr-rich and FeNi-rich particles within a grain of the zircaloy. The TEM micrograph B reveals Fe-Cr-rich particles showing traces of dissolution (red arrows), a large FeNi-rich particle and grain boundary decorated by small precipitates (blue arrow). The EDX spectrum is from a typical Fe-Cr-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 850 nm.
Characterization of a PuO₂ Powder Sample

Comparison of Plutonium Oxide Platelets

EDX

SEM

TEM

Comparison

Platelet Size (µm)

Log. Grain Size

Reference SM3-1
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 information on:
  
  Microstructure, surface morphology, chemical composition, phases distribution, ...

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