Microbeam Analysis of Irradiated Nuclear Materials

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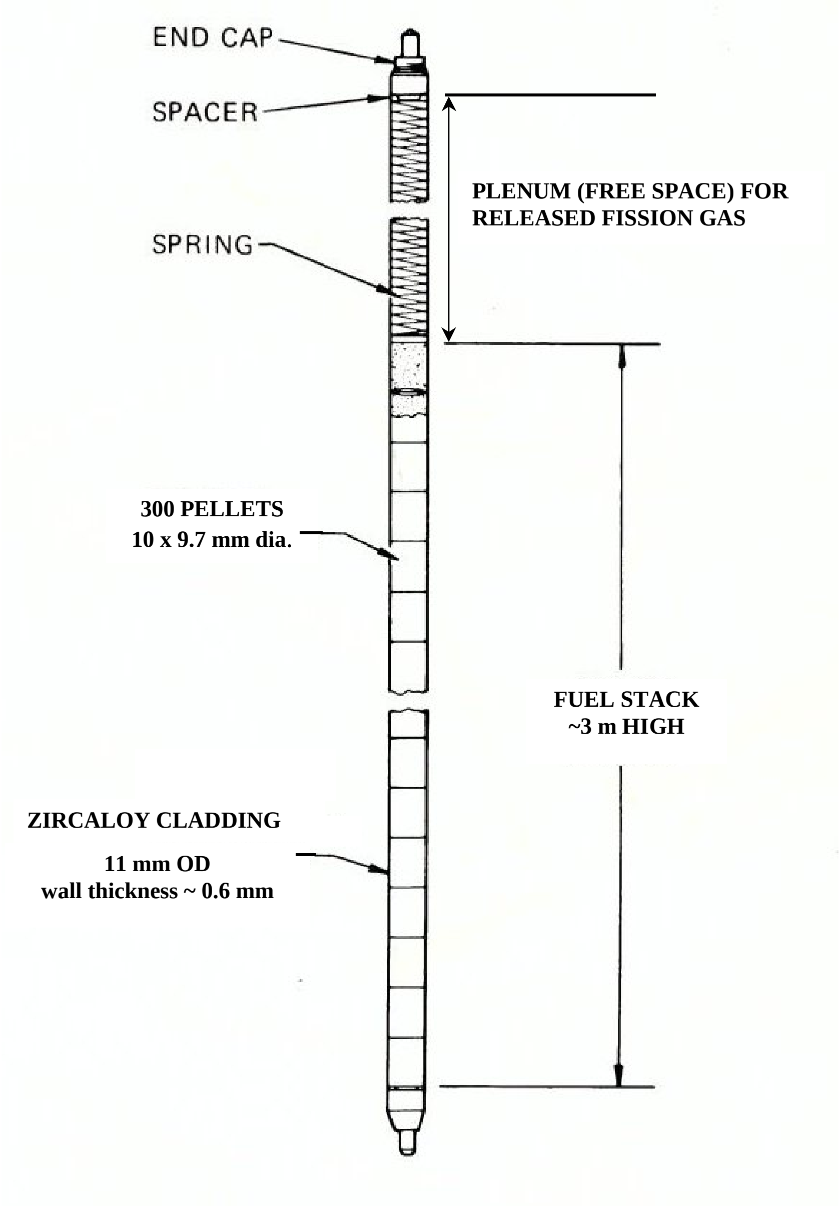
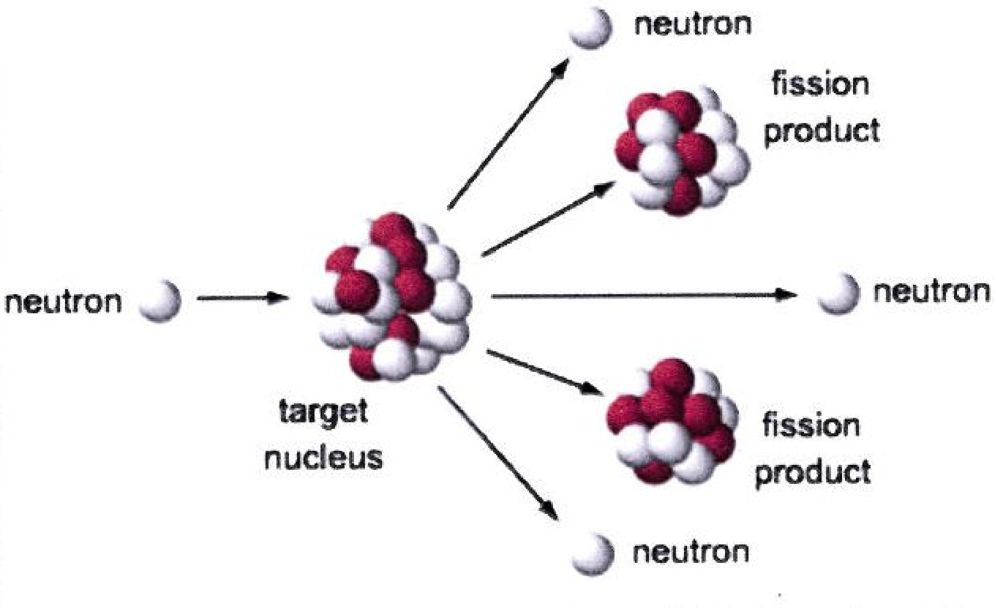


The fuel rod is filled with helium up to a pressure of 25 bar. The gas provides a path for heat conduction across the gap between the fuel pellets and the Zircaloy cladding.

Diagram of a PWR Fuel Rod

Nuclear Fission



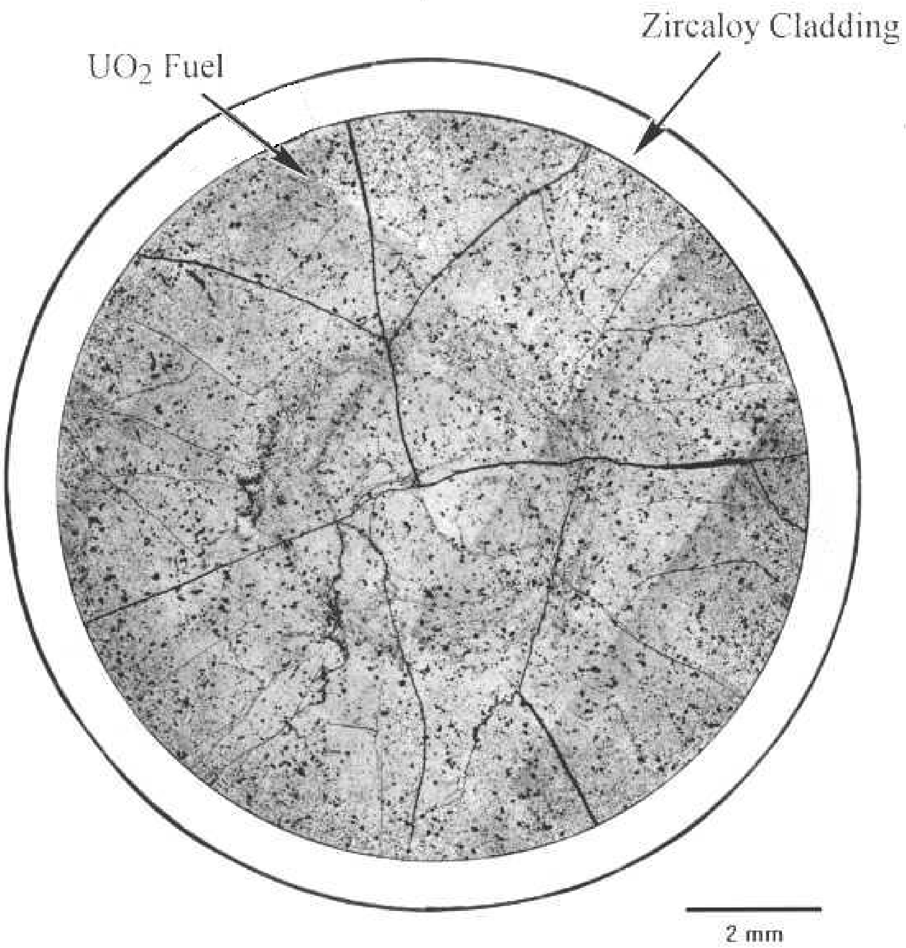
Typical reaction:

$$ {}^{235}_{92}U + {}^{1}_{0}n \rightarrow {}^{236}_{92}U^{*} \rightarrow {}^{147}_{57}La + {}^{87}_{35}Br + 2\,{}^{1}_{0}n + 200\,MeV $$

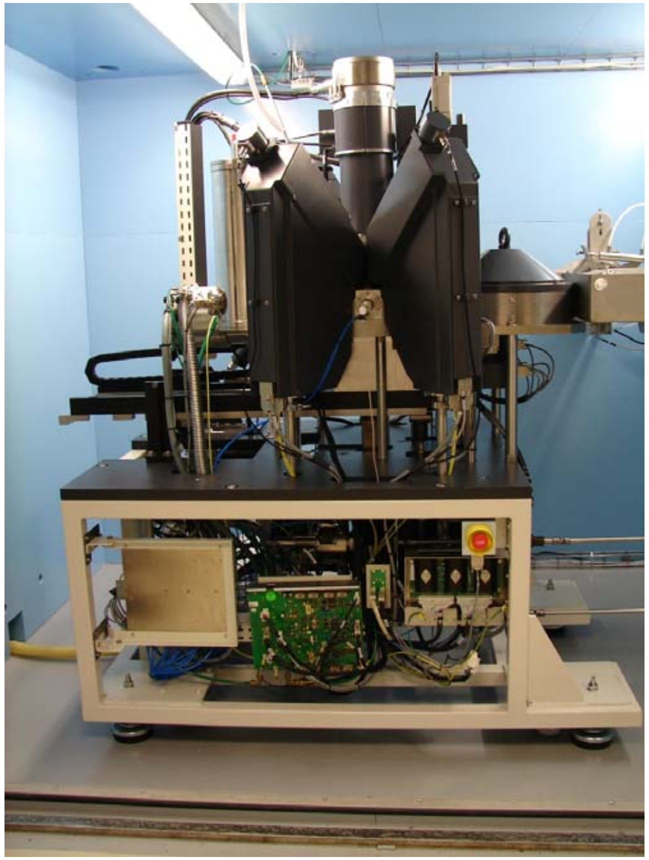
Thermal Neutron Cross-Sections

Nuclide	Cross-Section (barns)	
	Fission	Capture
^{232}Th	$3.9\mathrm{x}10^{-5}$	7.4
^{235}U	577	101
^{238}U	$1.1\mathrm{x}10^{-5}$	2.7
^{239}Pu	741	274
^{240} Pu	0.03	290
^{242}Pu	<0.2	18.5

The nuclides can be divided into fertile and fissile isotopes. Fertile isotopes have an even mass number (e.g., ^{238}U), whereas fissile isotopes have an odd mass number (e.g., ^{235}U) .



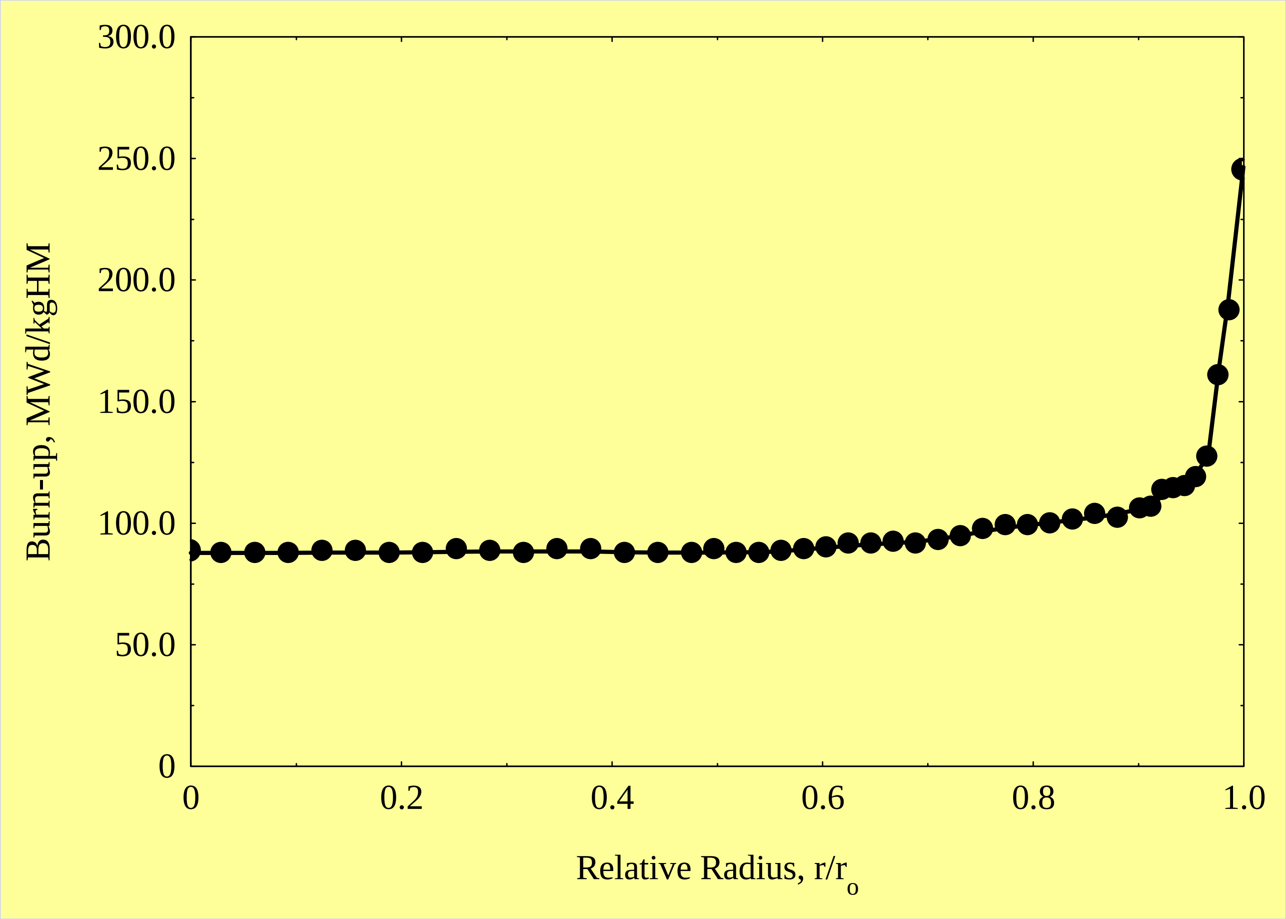
A sample of UO_2 nuclear fuel prepared for EPMA. The sample is from a fuel rod that had seen a reactor power excursion to 42 kW/m.



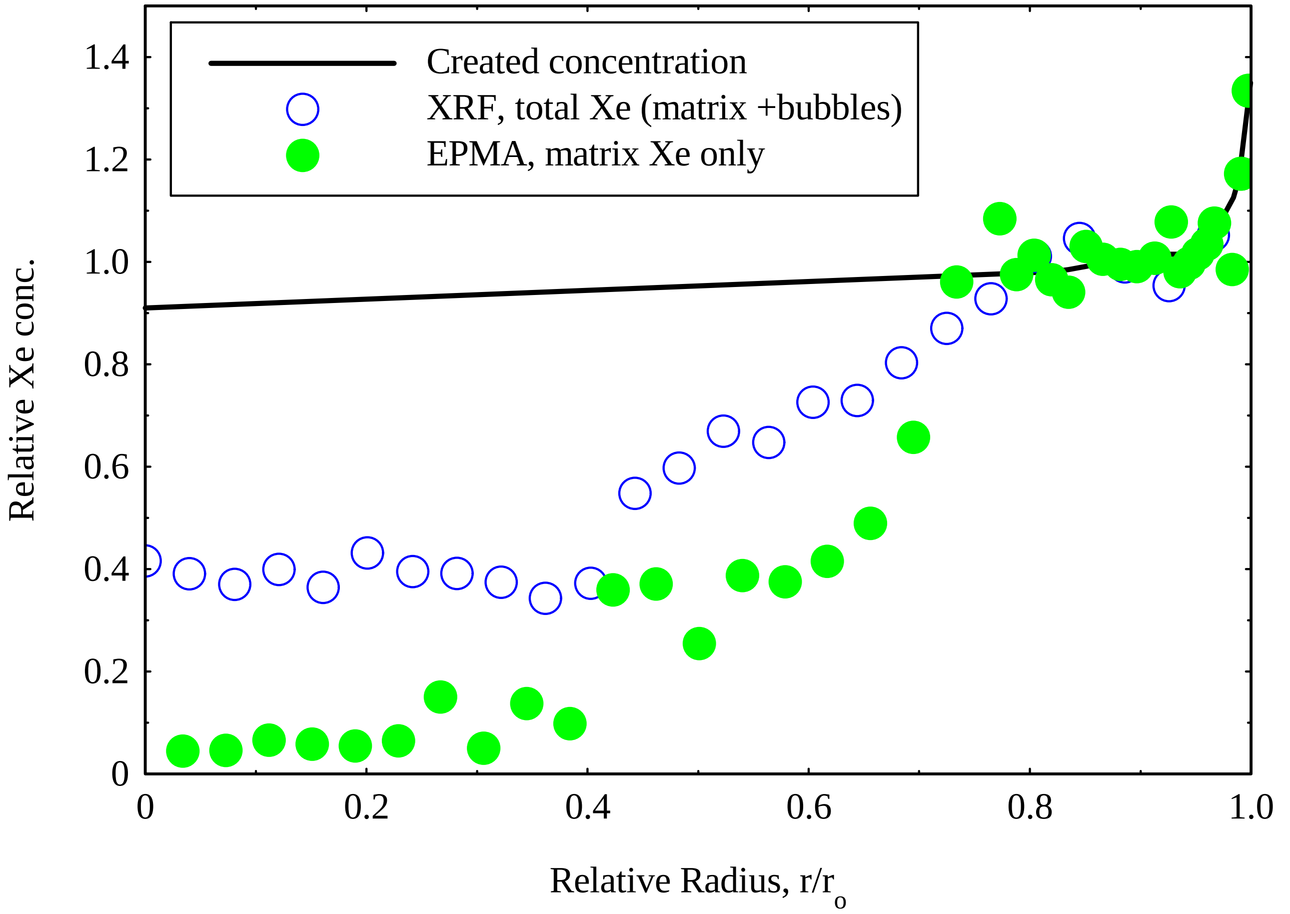
Views of the Cameca SX100R electron microprobe analyser for the analysis of highly irradiated materials at the Institute for Transuranium Elements.

Applications Presented

- Measurement of the local burn-up in irradiated nuclear fuel.
- Fission gas release.
- Investigation of the recrystallisation of high burn-up UO_2 fuel.
- Hydrogen pick-up by Zircaloy cladding
- Effect of water chemistry on the corrosion rate of Zircaloy cladding.
- Characterisation of corium.



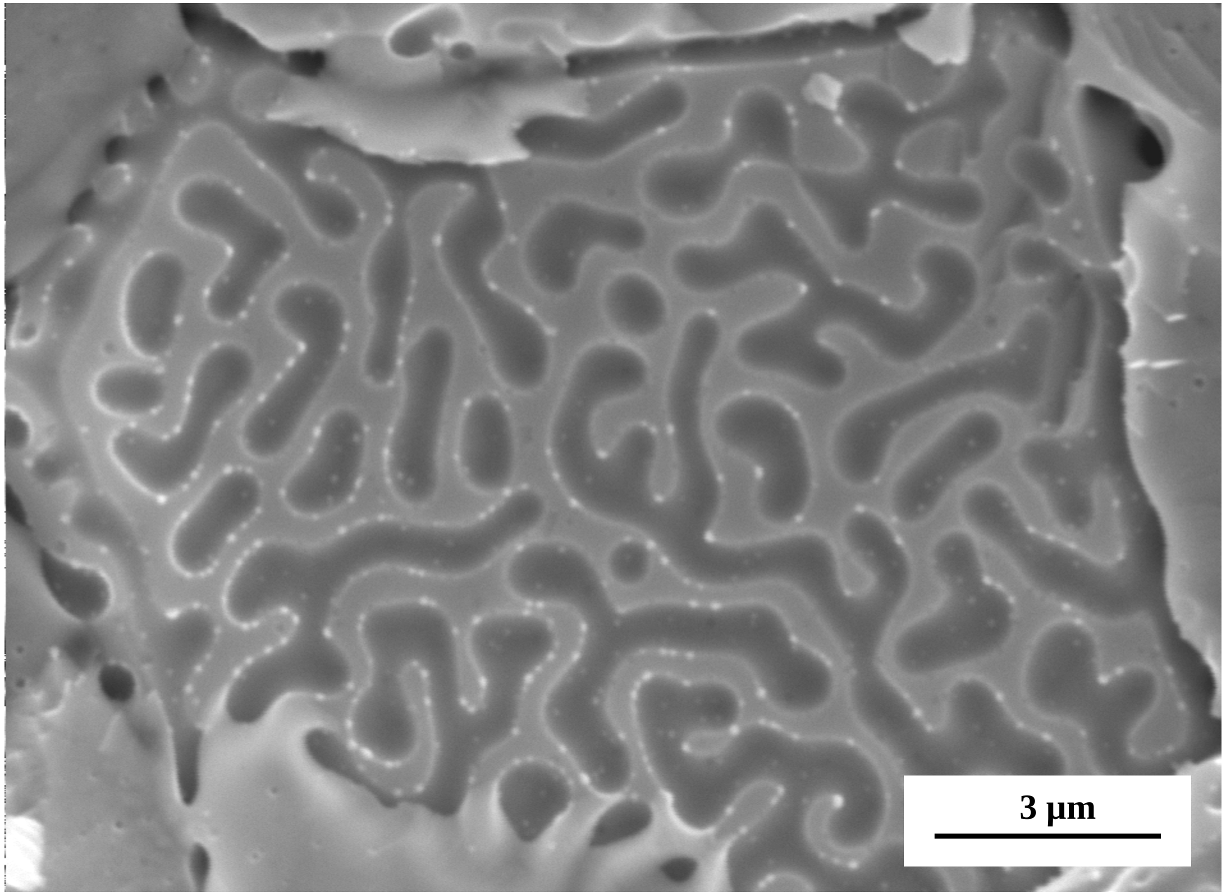
The radial burn-up distribution in a UO_2 nuclear fuel pellet with an average burn-up of about 100 MWd/kgHM. The local burn-up increases sharply at the fuel surface due to the fission of plutonium created from uranium by neutron capture.



The radial distribution of xenon retained in a UO_2 fuel subjected to reactor power excursion to 40 Wm^{-1} at a burn-up of 50 MWd/kgHM as revealed by EPMA and XRF. The difference between the two profiles corresponds to the amount of gas in bubbles and bubbles.

Consequences of Gas Bubble Formation in Nuclear Fuel

- **The formation of gas bubbles causes the fuel to expand, or swell. At high burn-up, swelling can result in increased mechanical interaction between the fuel and its containment which can eventually lead to rod failure.**
- **The presence of gas bubbles decreases the thermal conductivity of the fuel (similar effect to porosity) which leads to an increase in the fuel operating temperature.**



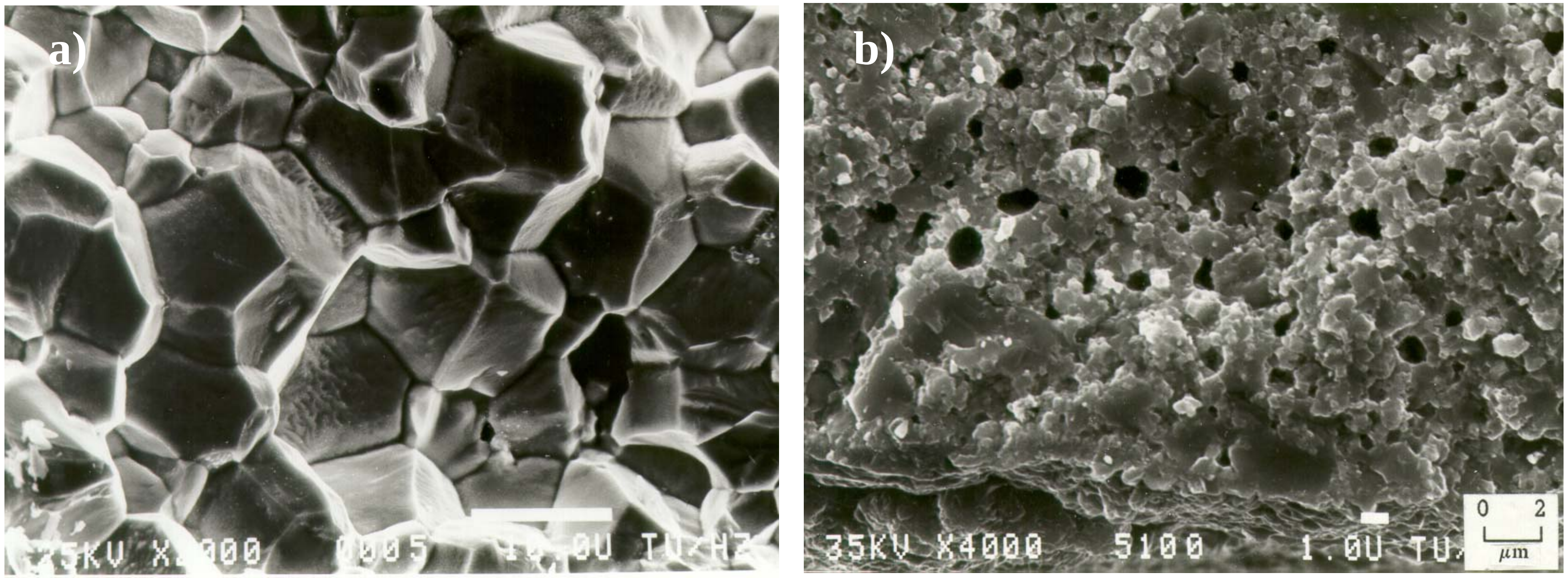
SEM micrograph of vermicular gas pores on a grain face in a UO_2 nuclear fuel formed by the interconnection of individual spherical bubbles.

The High Burn-up Structure

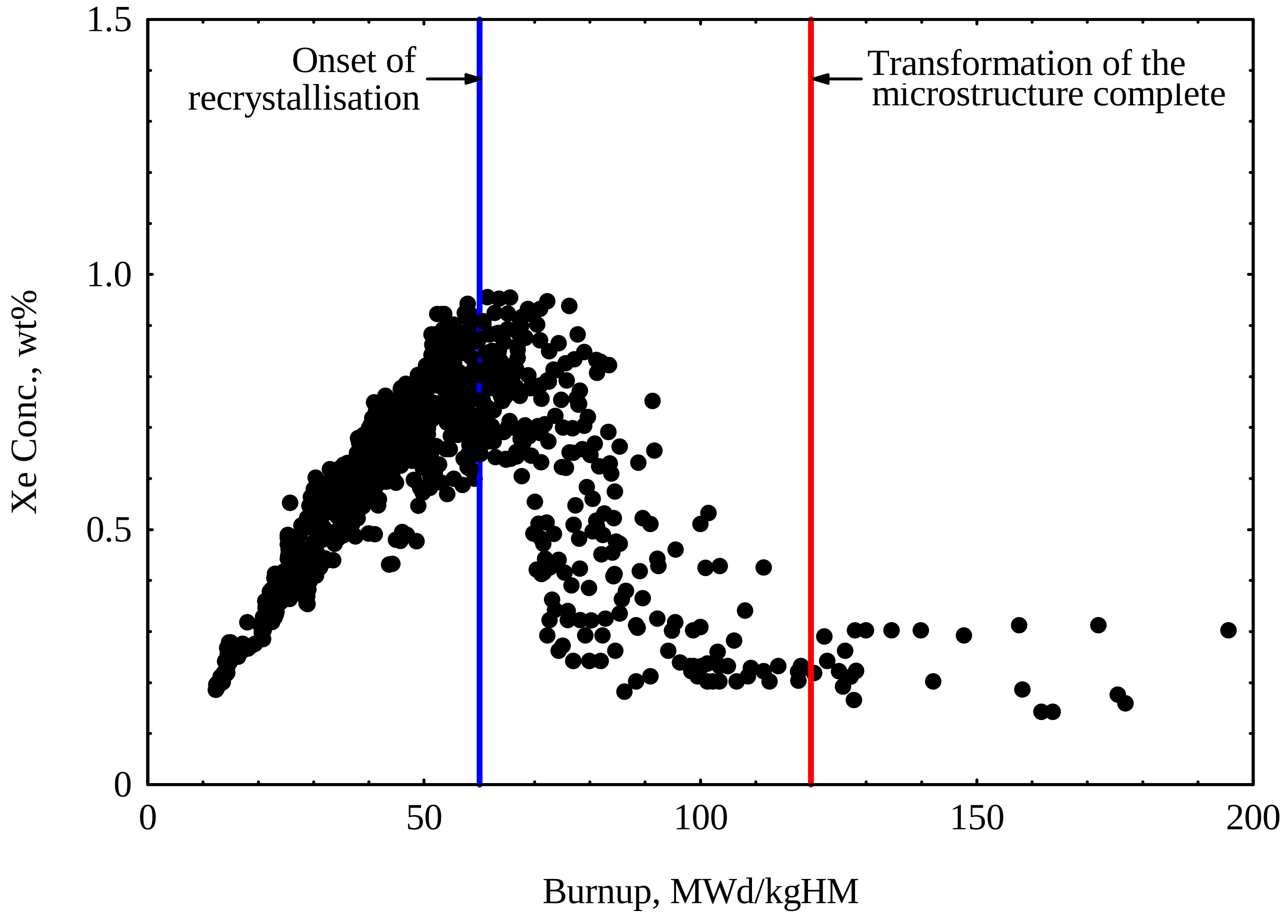
Characteristics:

- **Pronounced decrease in grain size from 10 µm to typically 0.15 µm.**
- **High concentration of small gas pores of typical diameter 1 to 2 µm.**
- **Considerable loss of fission gas from the fuel matrix. The concentration of xenon falls from about 1 wt% to 0.2 to 0.3 wt%.**

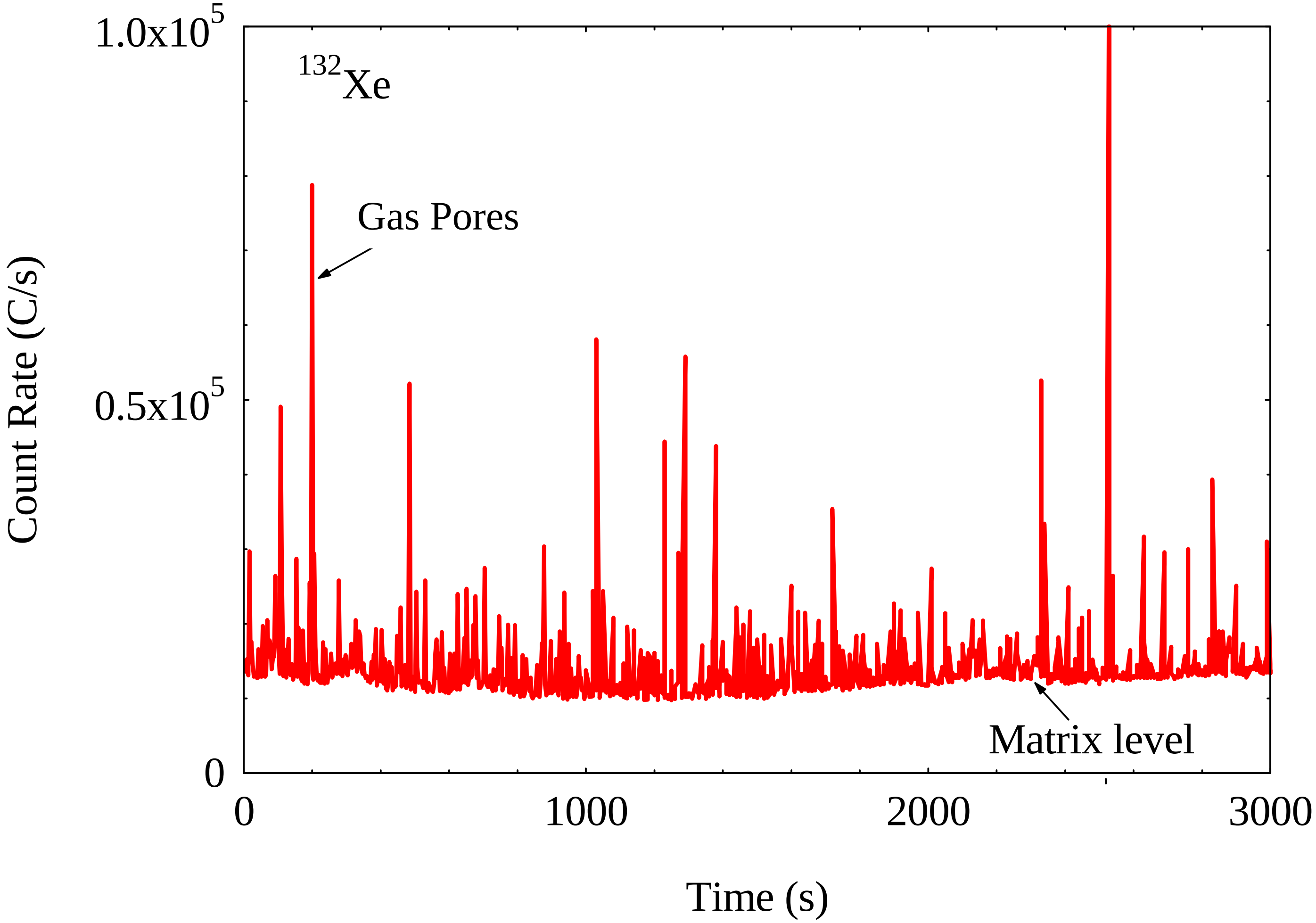
Almost all that is known about the high burn-up structure: its characteristics, the mechanism of its formation, and the processes contributing to its evolution have been obtained from studies employing microbeam analysis techniques.



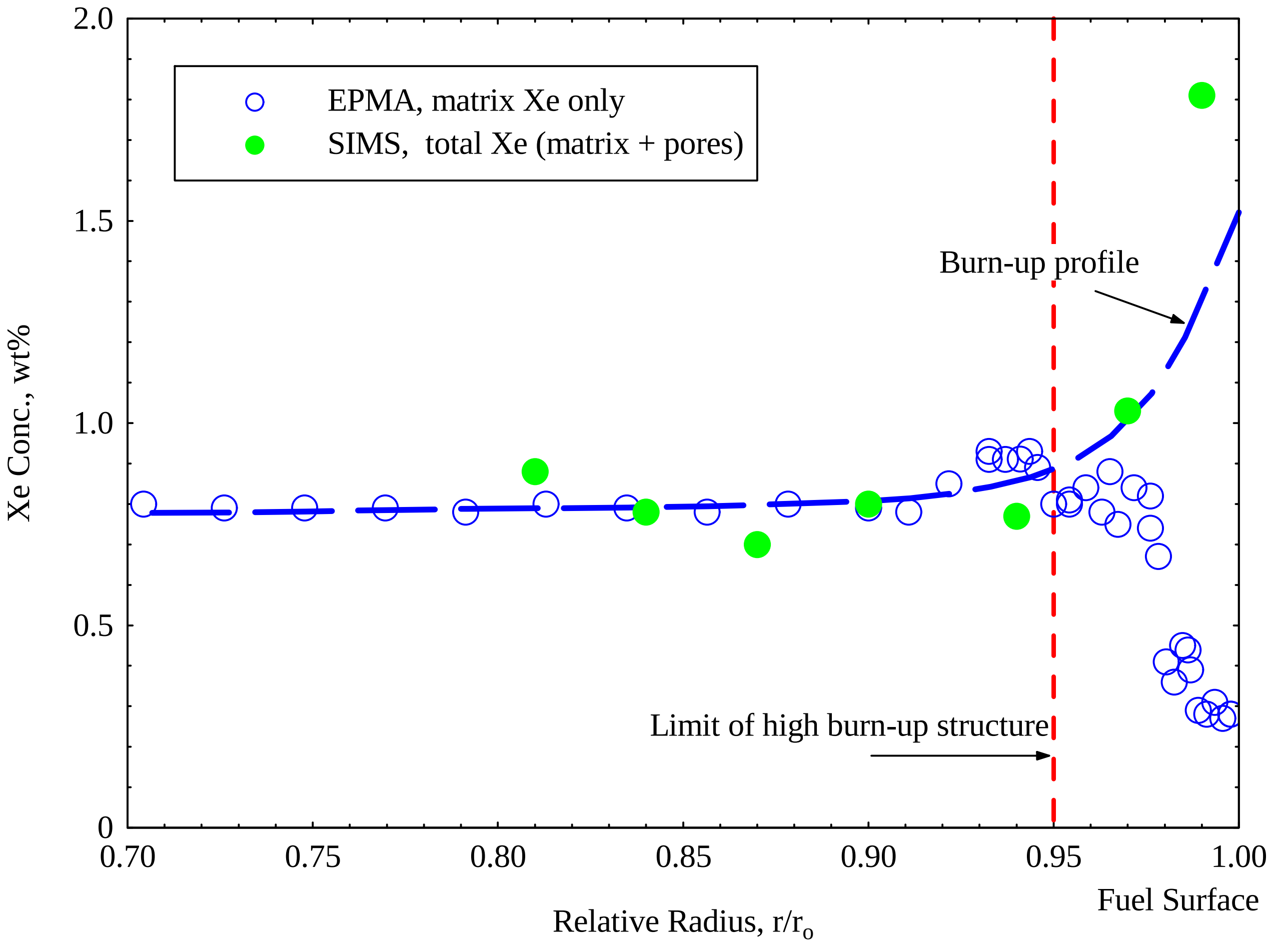
Scanning electron micrographs showing the microstructure at the surface of UO_2 fuel at burn-ups below and above a pellet burn-up of 40 MWd/kgU at which recrystallisation of the UO_2 grains begins. (a) Fuel microstructure at 30 MWd/kgU; (b) fuel microstructure at 45 MWd/kgU.



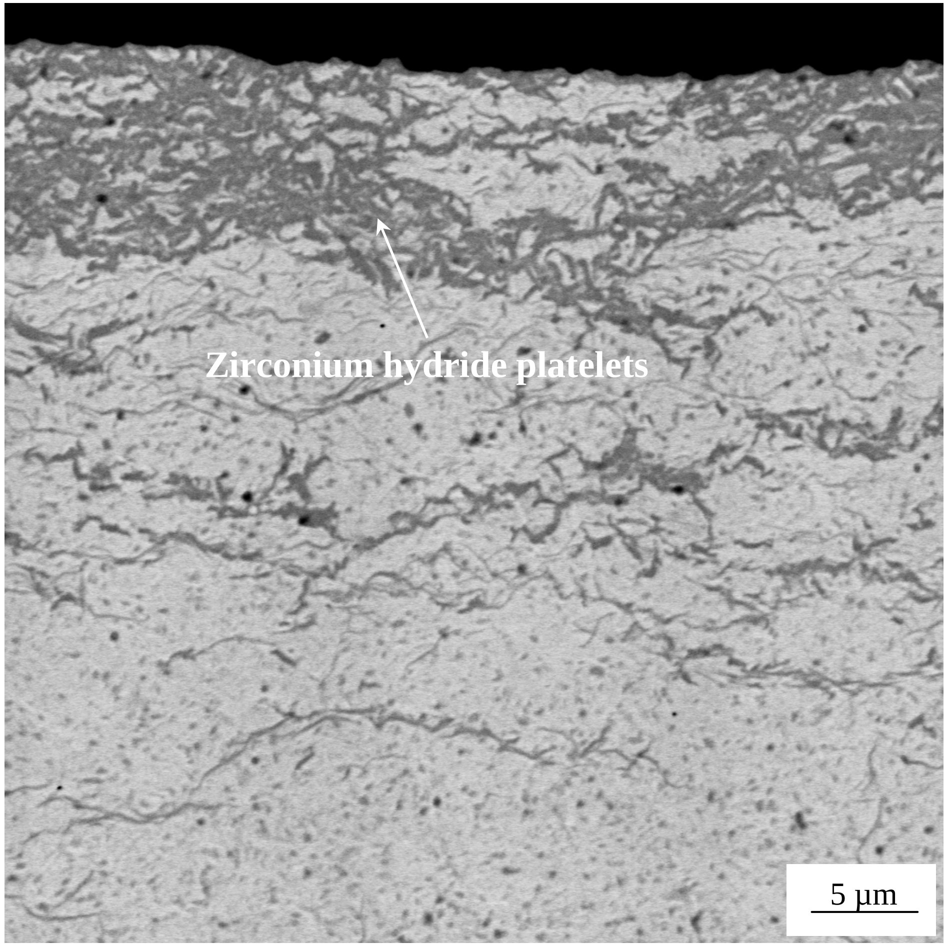
EPMA results for the local concentration of xenon in the outer region of UO_2 fuel related to the local burn-up. The peak concentration at 60 to 75 MWd/kgHM marks the burn-up threshold for the formation of the high burn-up structure.



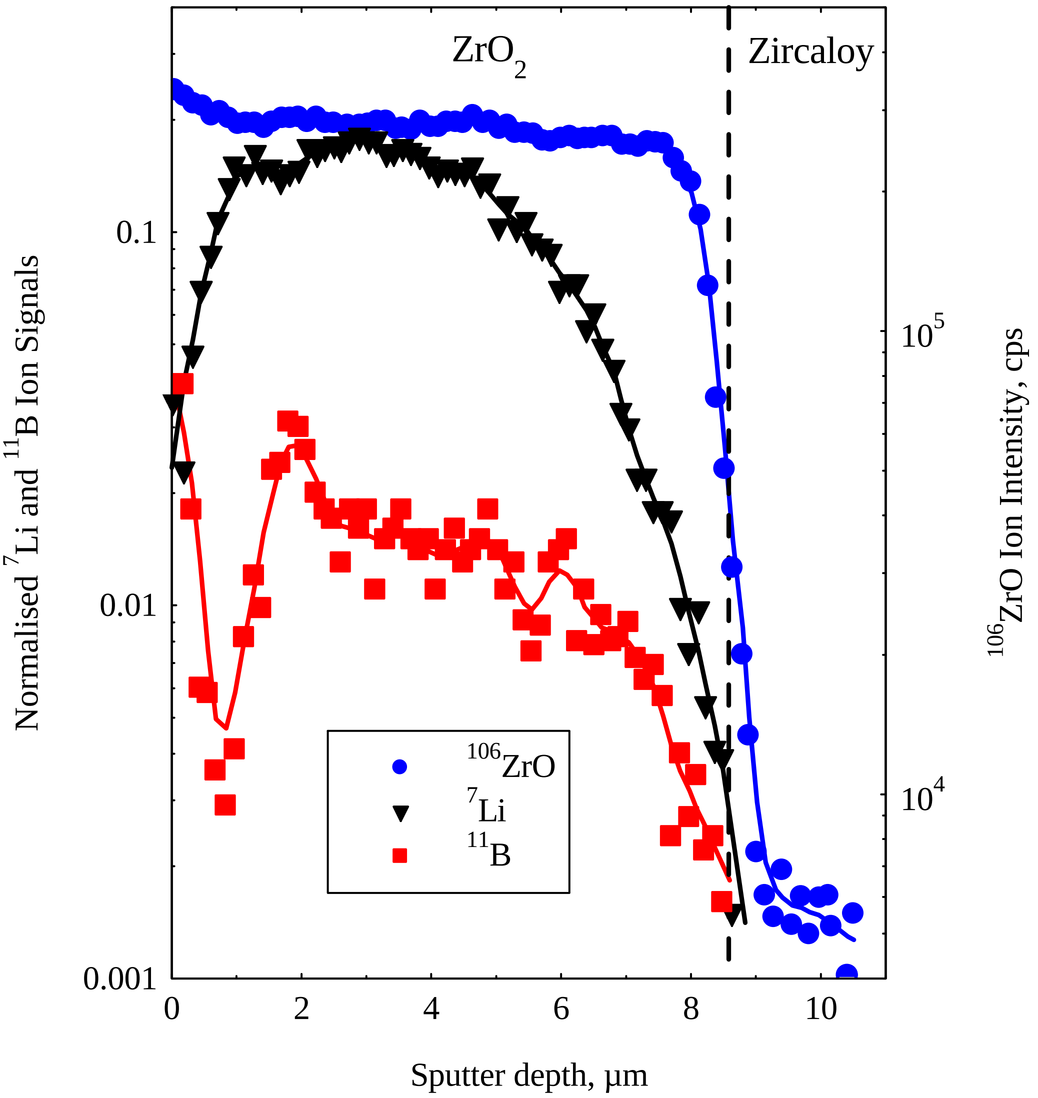
SIMS depth profile for ^{132}Xe in the high burn-up structure produced with a $(^{16}O)^+_2$ primary ion beam at a high current density of 154 mA/cm^2. The intensity spikes are caused by the expulsion of gas from the pores in the microstructure. Time is equivalent to erosion rate.



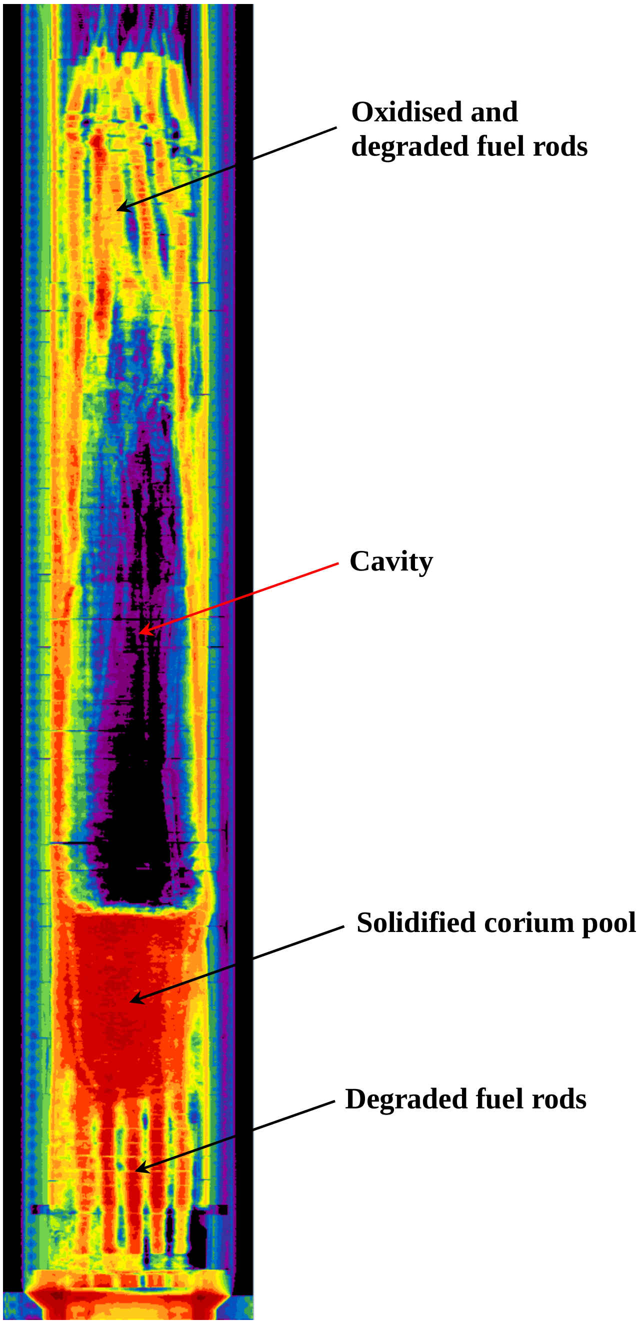
The local concentration of retained xenon in the outer region of UO_2 fuel as measured by SIMS and EPMA. The steep increase in the SIMS profile at the fuel surface indicates that almost all the gas missing from the UO_2 matrix is contained in the pores of the high burn-up structure.



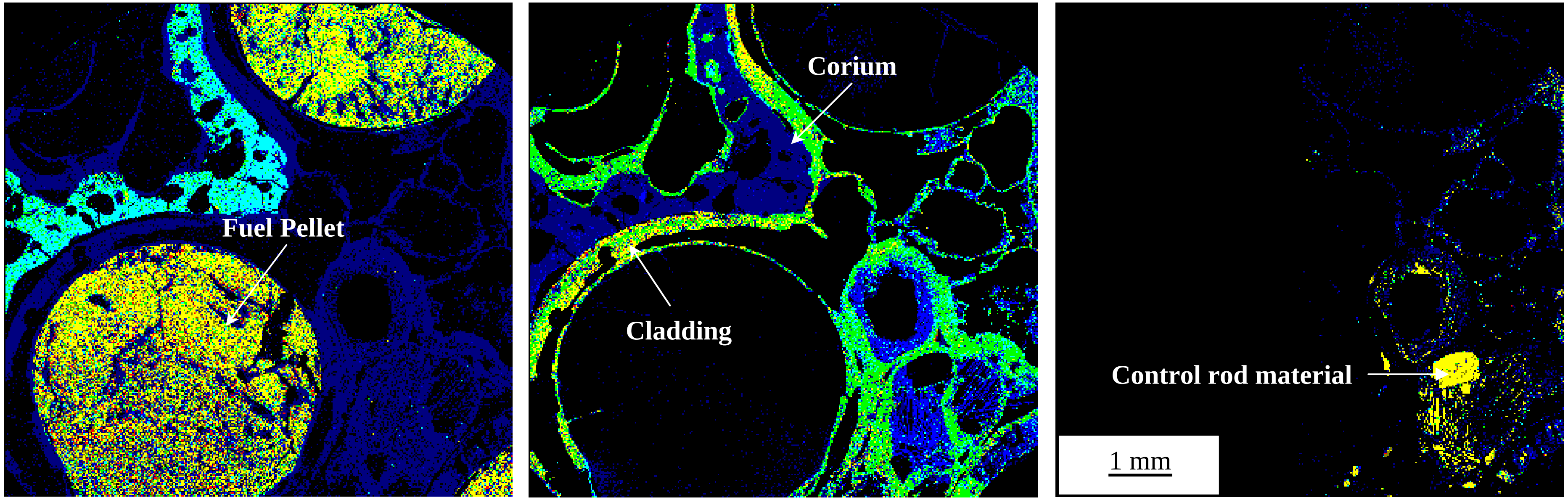
Electron backscatter image of zirconium hydride platelets in Zircaloy cladding hydrided in the laboratory. The accumulation of of hydrides at the sample surface indicates the presence of a steep hydrogen concentration gradient in this region.



SIMS depth profiling results for the distribution of ^{7}Li and ^{11}B in the external oxide layer of the Zircaloy cladding of a PWR fuel rod (after Gebhardt, Fresenius J. Anal. Chem. Vol. 365, 1999).



False colour X-ray radiograph showing the state of the melted, Phebus FPT2 fuel rod assembly at the end of the test (courtesy of the Institut de Radioprotection et de Sûreté Nucléaire (IRSN), Cadarache, France). The material density increases in the order black-blue, green-yellow, red.



Uranium **Zirconium** **Silver**

Fuel rods, Zircaloy cladding (the fuel containment), corium and control rod material in the zone below the corium pool in the melted Phebus FPT2 fuel rod test assembly as revealed by large area, false colour, EPMA X-ray maps for U, Zr and Ag.