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Magnox Generation

Developments in the use of SEM and SIMS for the study of Irradiated Fuel at Berkeley

- R C Corcoran and W J Stephen

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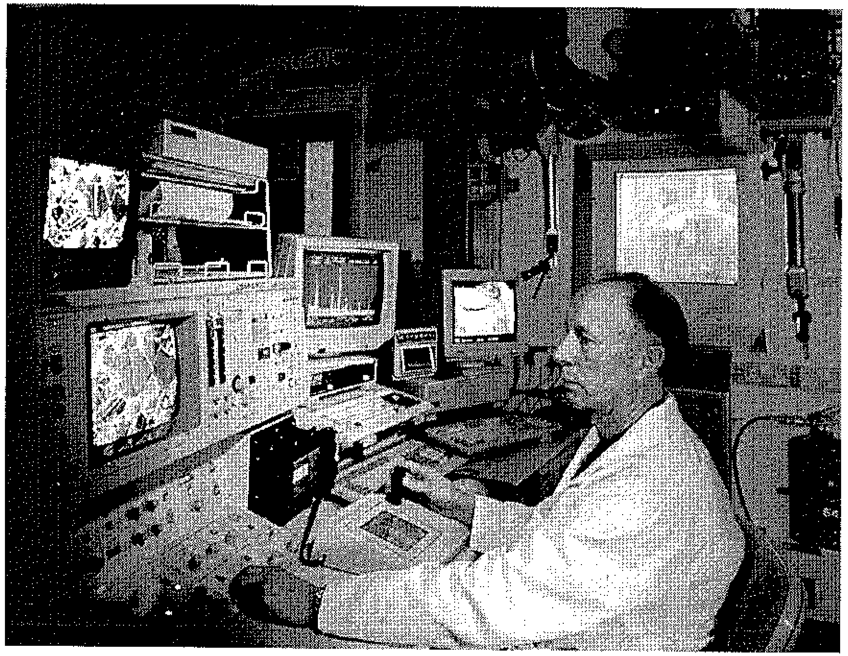
Berkeley Shielded SEM



- JEOL 6100 - FULLY SHIELDED (With analysis chamber in cell).
- EDX/WDX for chemical analysis.
- Adjacent cells for sample preparation.

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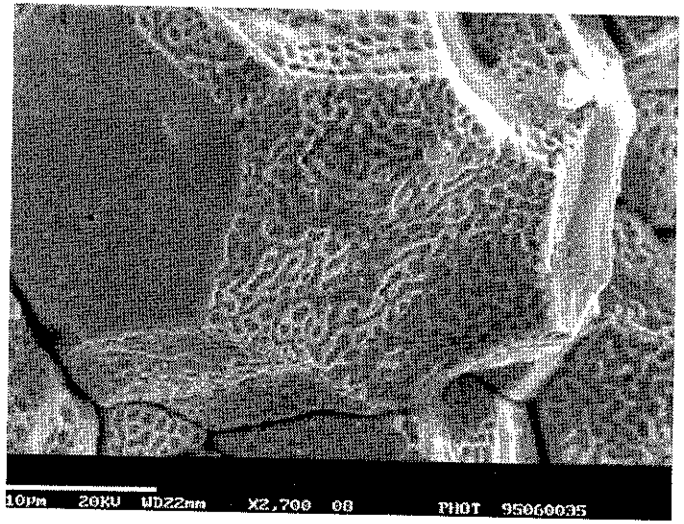
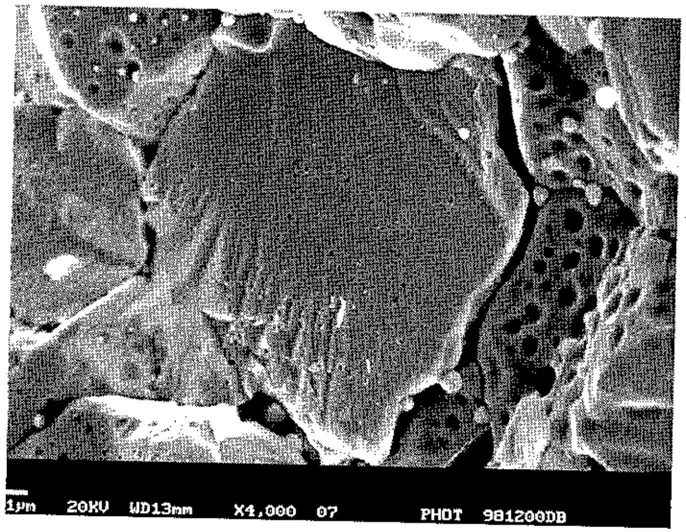
Fuel SEM

- Fractography performed on irradiated fuel.
- Analysis of intergranular and intragranular fission gas bubble populations
- Distribution of fission gas related to position across fuel pellet.



Intergranular



Intragranular

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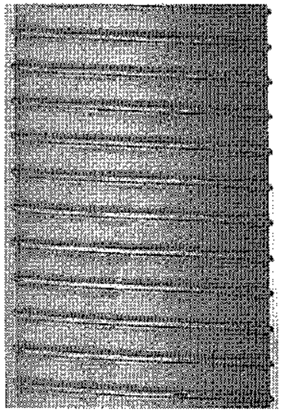
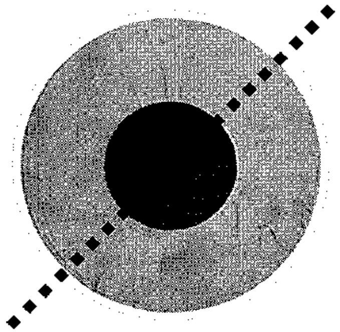
Fuel SEM

Fractography

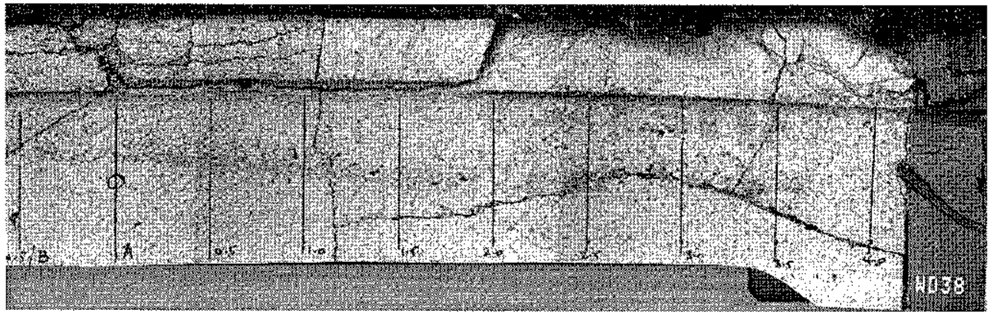


AGR Fuel Pin

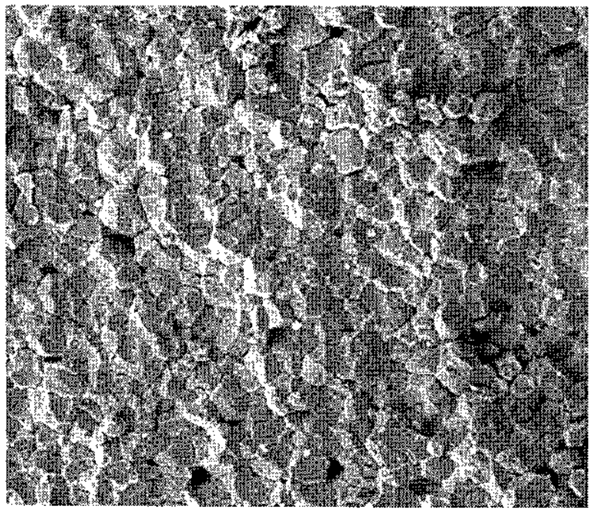


Slice of fuel cut and notched across diameter for fracture.

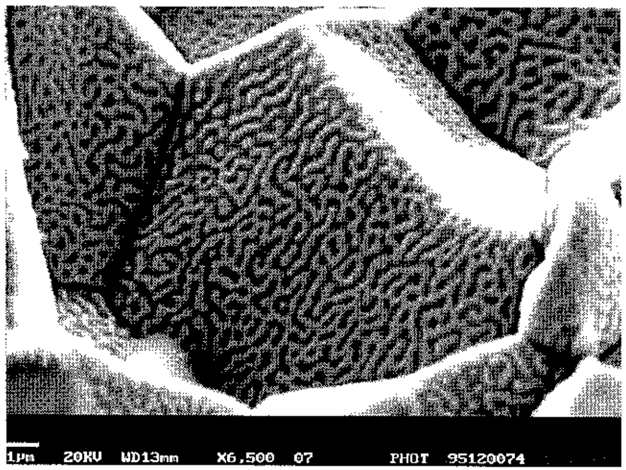
Fractured fuel slice.



Position of inner bore



Low magnification image of fracture surface



High magnification image of intergranular facet

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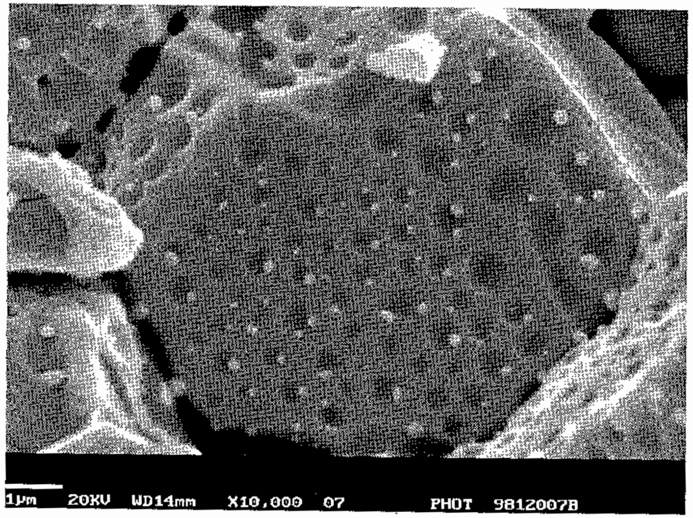
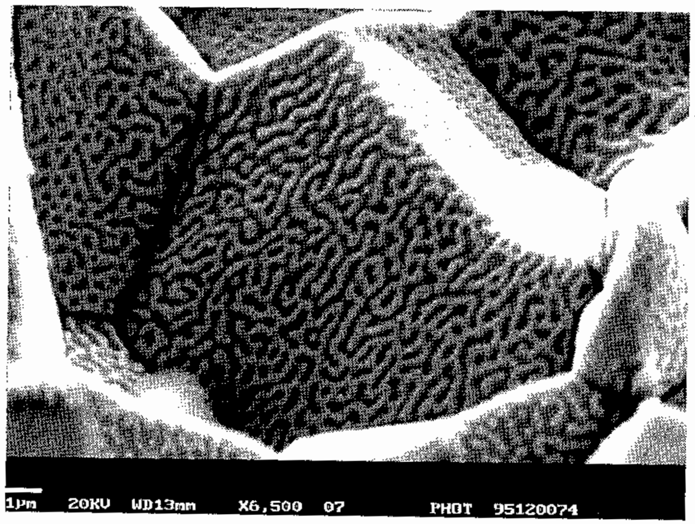
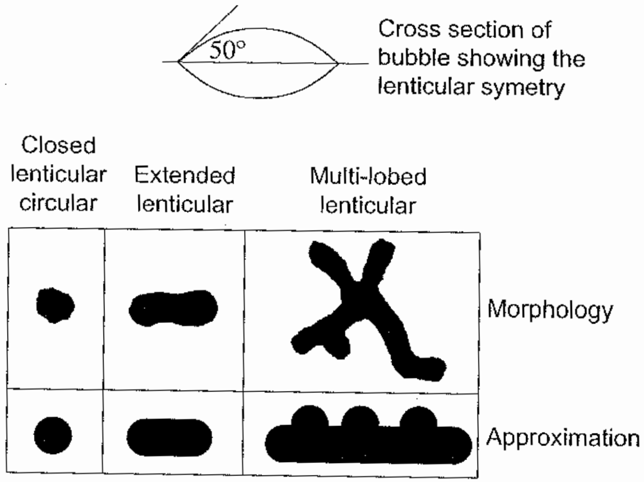
Intergranular Volume Swelling





- Depending on Irradiation History bubbles form:

  - Circular Lenticular Bubbles
  - Extended Lenticular Bubbles
  - Multi-lobed Lenticular Bubbles



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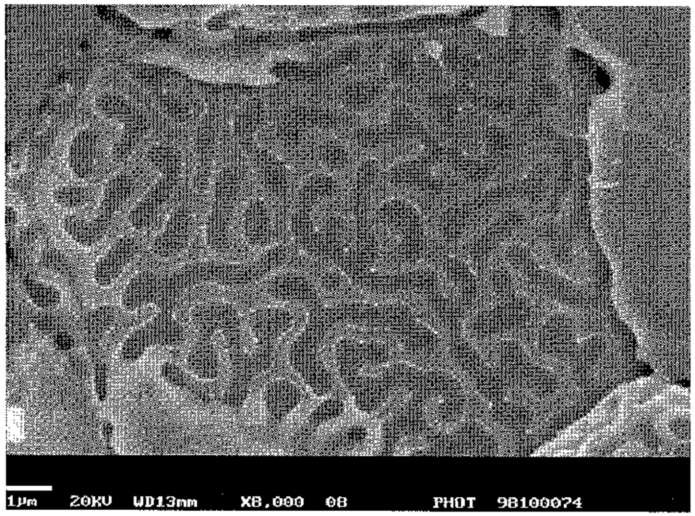
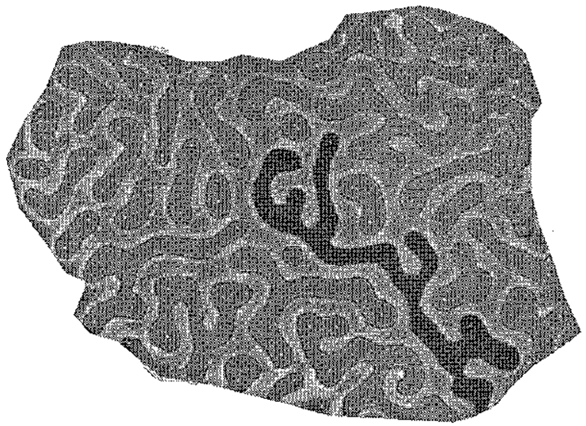
Intergranular Volume Swelling

In-House Software Developed for Image Analysis



SEM Image of grain face



Grain area extracted
- Bubbles Outlined

Measure:
- Perimeter (P)
- Areas (A)
- Length (L)
- Number of nodes (n)

Get Volume (V_{nl})

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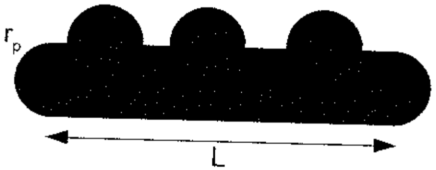
Intergranular Volume Swelling

In-House Software Developed for Image Analysis

Extracted Feature
Measure A and P

Skeletonised Feature
Determine L and n

Model of Extended Pore



$$P = 2L + (n\pi + 4 - 2n)r_p$$

$$A = \frac{n}{2}\pi r_p^2 + 2Lr_p$$

$$V_{nl} = n\frac{2\pi}{3}\left(\frac{r_p}{\sin\theta}\right)^3 f_f(\theta) + L\left(\frac{r_p}{\sin\theta}\right)^2 f_p(\theta)$$

- Volume of porosity obtained
- L, r_p and n allow bubble morphology to be examined

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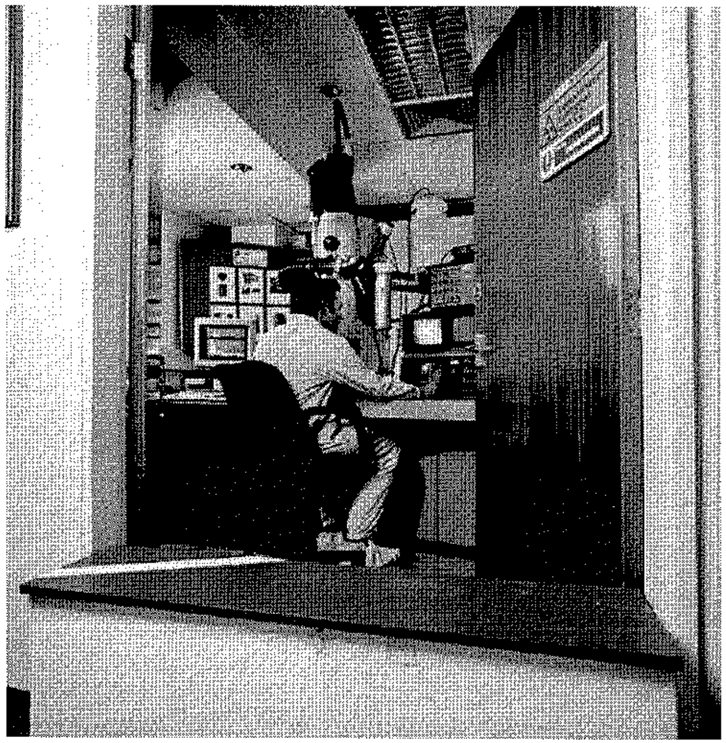
Berkeley Fuel TEM



- Phillips CM12 - Dedicated Fuel TEM
- EELS for thickness measurements
- EDX for chemical analysis
- Analysis of small intra-granular fission gas bubbles (<50nm diameter)
- General microstructural examination of fuel

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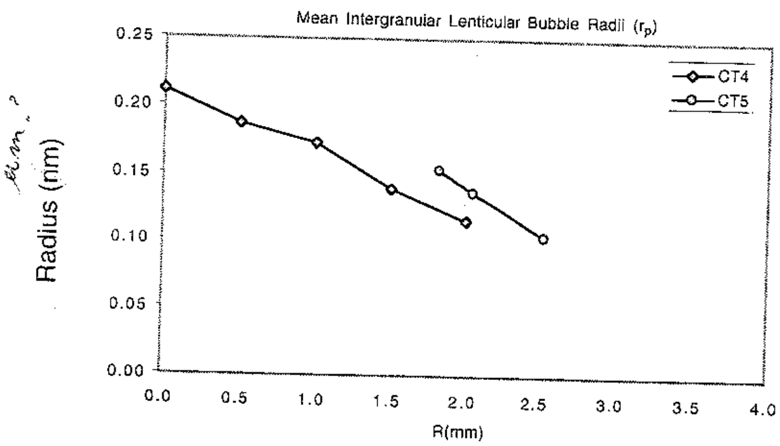

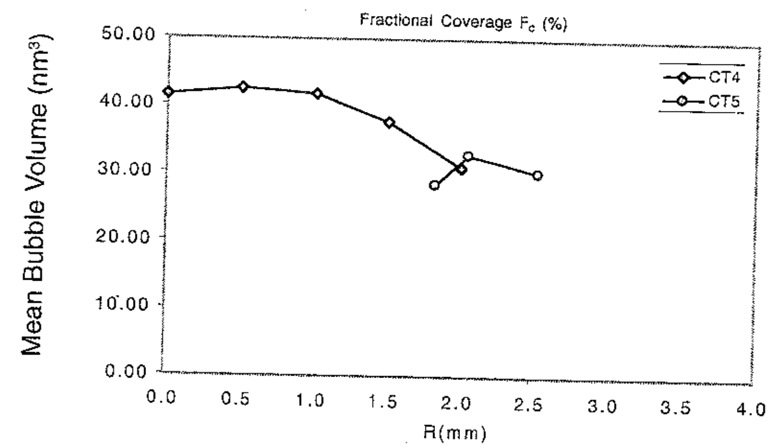

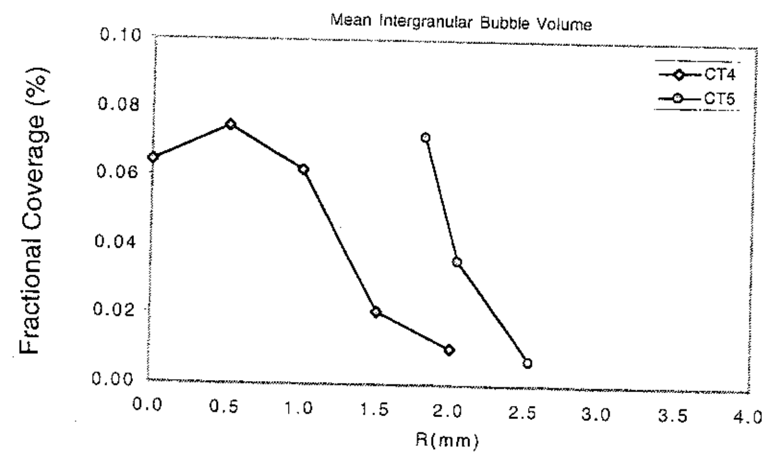

R = Distance from Pellet Centre

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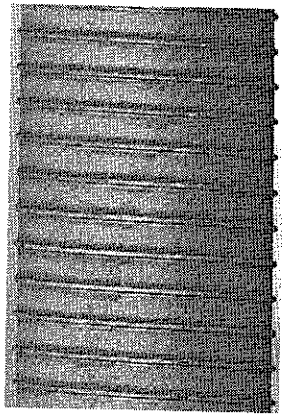
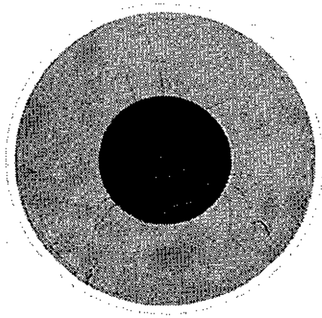
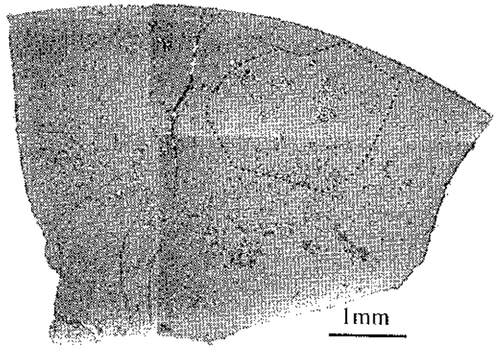
Fuel TEM



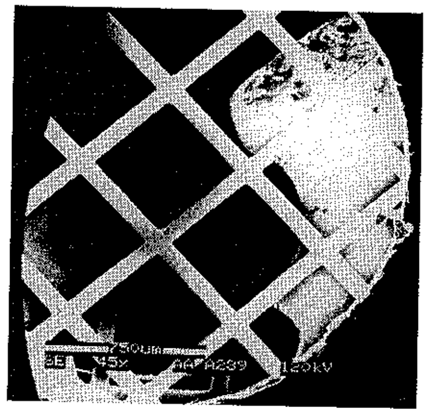
AGR Fuel Pin



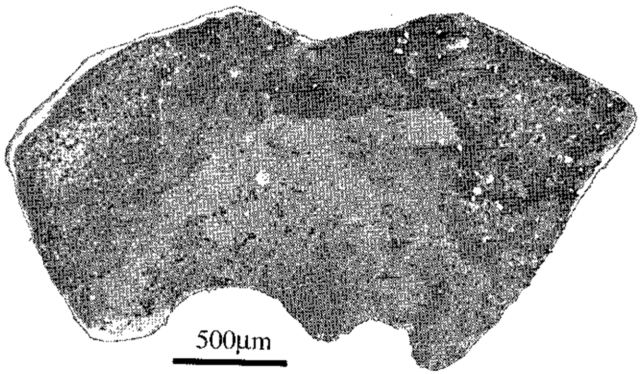
Slice of fuel pin cut



Segment of fuel examined in SSEM



Segment of fuel electropolished and examined in TEM



TEM sample examined in SSEM to locate analysis position

Fuel TEM

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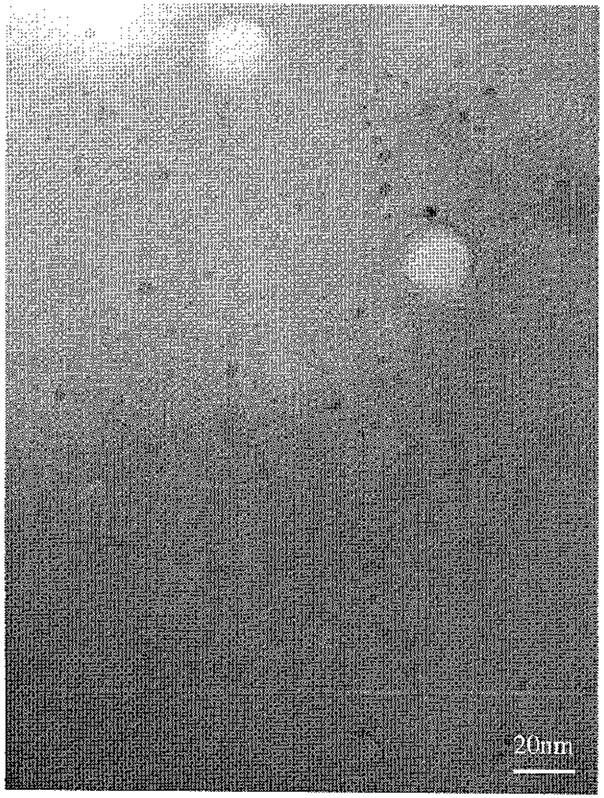
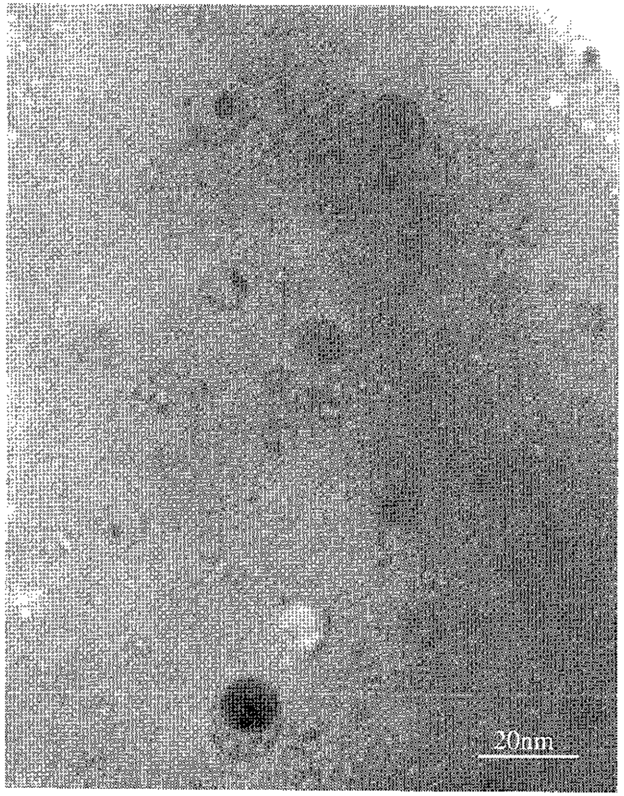


TEM micrograph taken over-focus of 4065 (slow ramp + scram) at 4.1 mm from the clad. The fission gas bubbles image dark, are faceted and have nucleated in lines.



TEM micrograph taken under-focus in 4065 (slow ramp + scram) at 1.45 mm from the clad showing the bimodal fission gas bubble distribution with the larger population all having a solid fission product precipitate inside.

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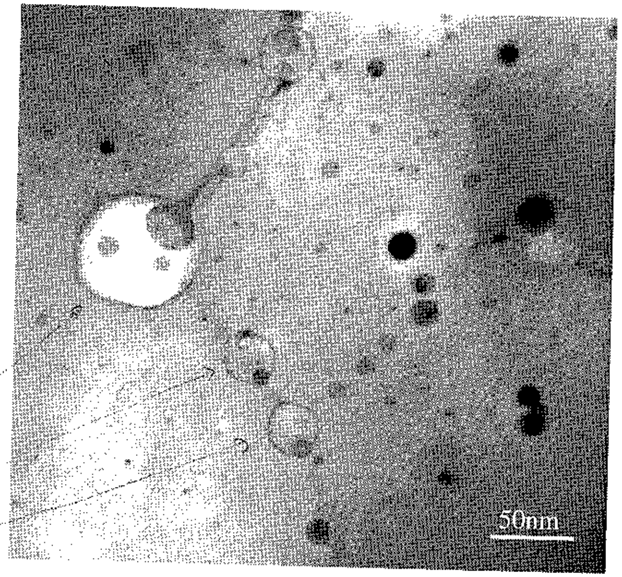
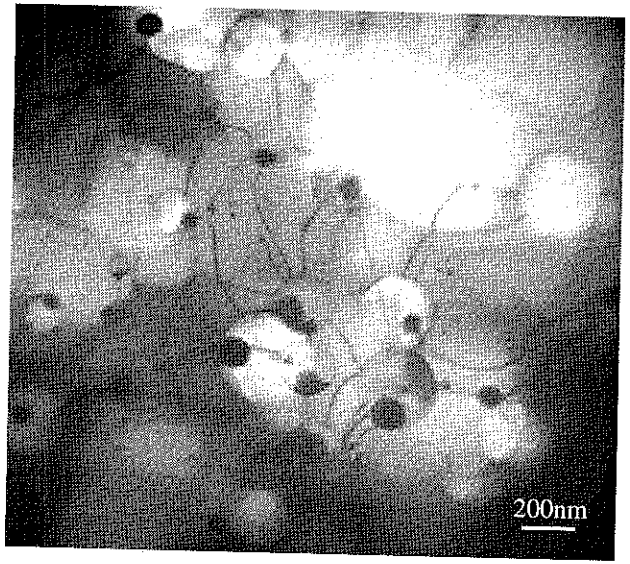
Fuel TEM



TEM micrograph of sample as in Figure 3(b), showing the 3rd and larger population of fission gas bubbles associated with both solid fission product precipitates and dislocations.



TEM micrograph of 4065 (slow ramp + scram) at 3.1 mm from the clad showing the large fission gas bubbles which are those observed by the scanning electron microscope.

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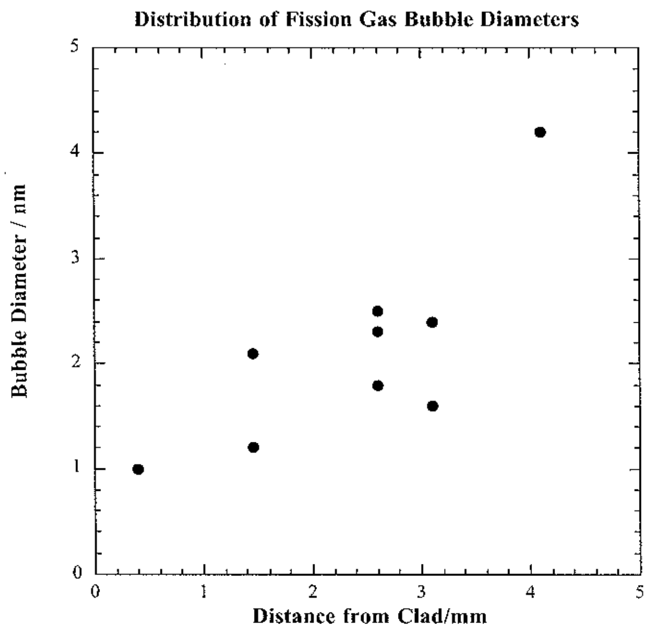
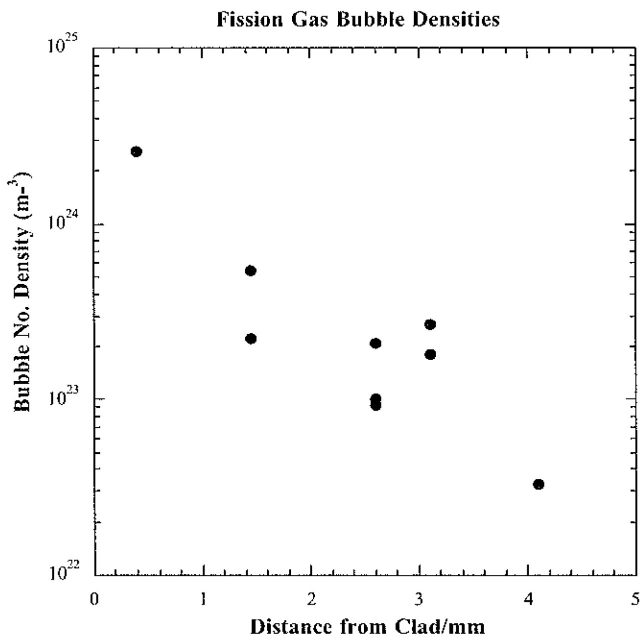
Fuel TEM

(AGR Fuel ramped in Halden)





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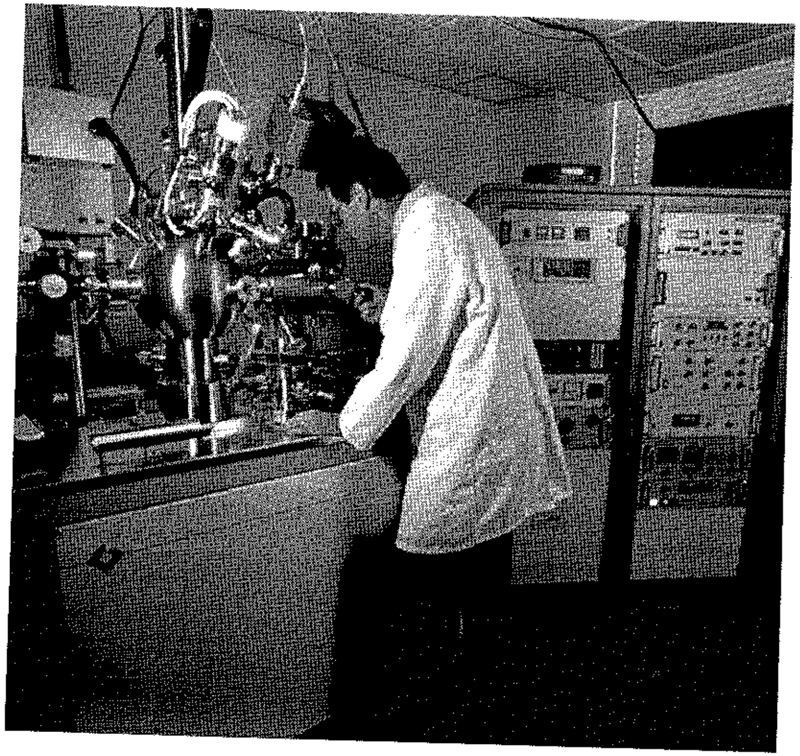
Berkeley SIMS



- Vacuum Generators (VG) MT500 SIMS
- FEI Ga^+ Liquid Metal Ion Gun (LMIG) (Resolution 100nm)
- VG EX05 Ar^+ Ion Gun (Resolution 30μm)

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SIMS

Advantages for Fuel Analysis:

- Isotopic sensitivity e.g. can map/profile ^{239}Pu or ^{240}Pu in MOX
  (Does not have the same peak overlap problems associated with EDX/WDX)
- Increased spatial resolution, 0.1μm relative to large 1μm X-ray generation volumes in EDX/WDX
- Allows depth information to be acquired - Depth profiles used to determine Diffusion Coefficients

Disadvantages:

- Destructive - analysis erodes specimen
- Difficult to quantify
- Requires expensive UHV equipment
- Very surface sensitive

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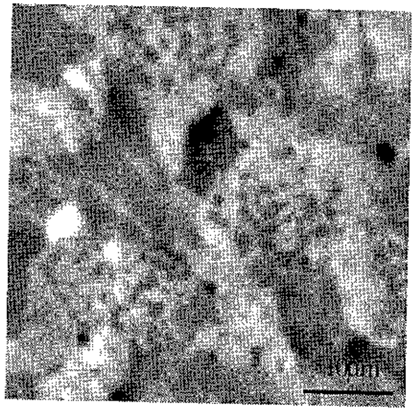
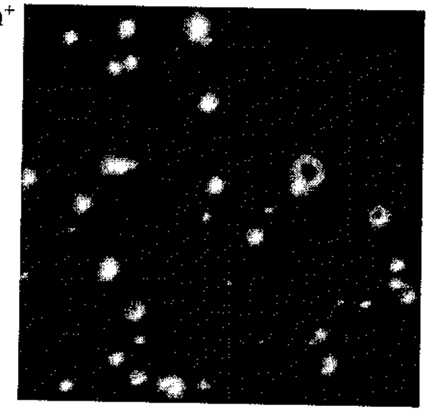
SIMS

SEI and Isotopic maps using Ga^+ LMIG

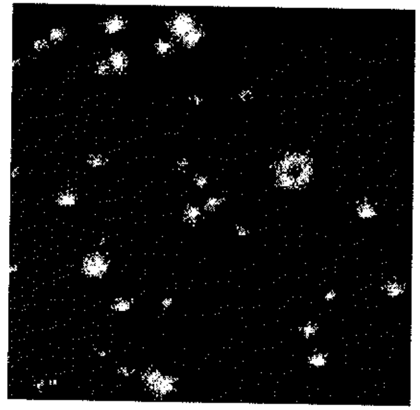


Ion generated secondary electron image (SMA Weld), Maps show Mn/Si inclusions.

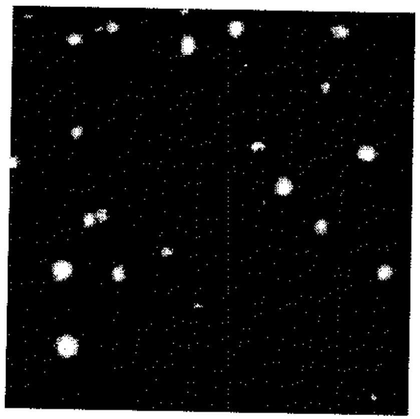
$^{55}Mn^+$

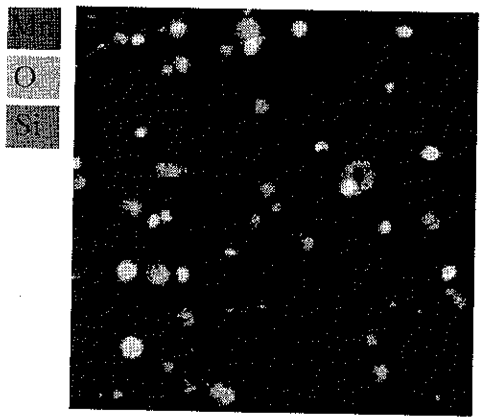


$^{28}Si^+$



$^{16}O^-$





Aim to apply mapping to Pu isotope distributions in irradiated and unirradiated MOX fuel.

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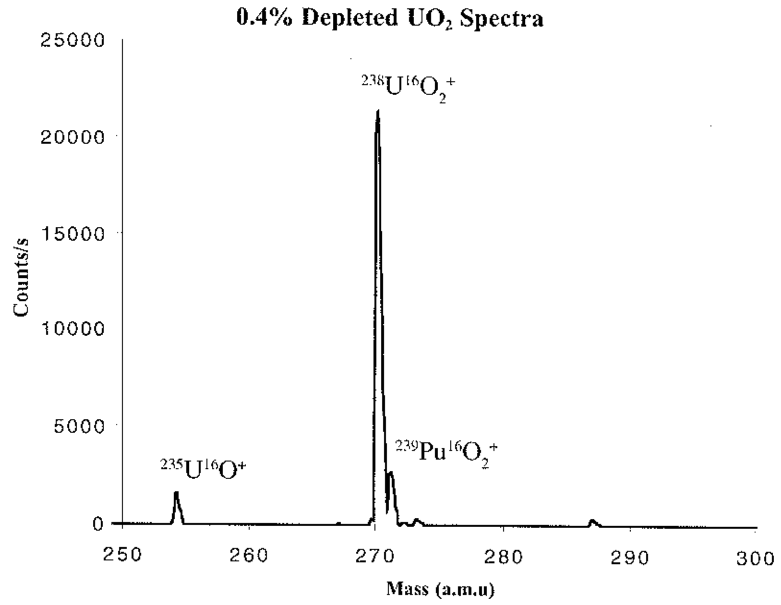
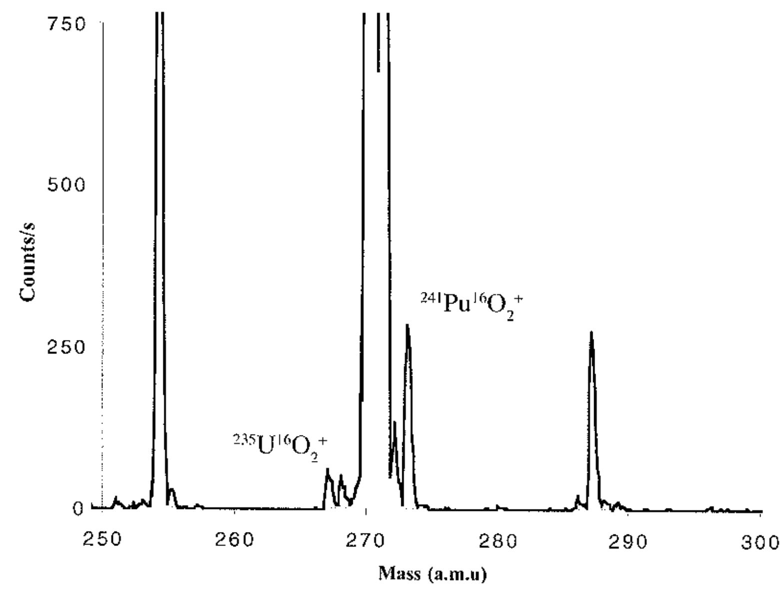
Isotopic Analysis of Fuel - Used to generate isotopic radial profiles





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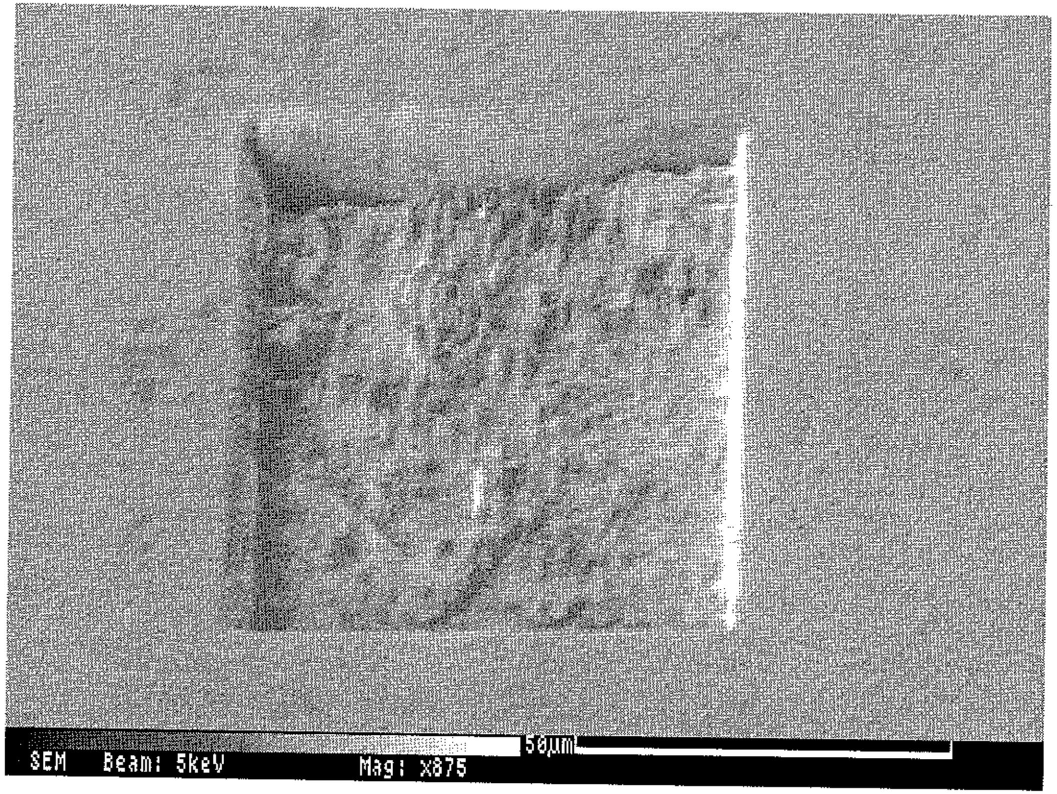
SIMS

Depth Profiles - Iodine Diffusion in Uranium



- Ga^+ ion gun crater
- 60µm x 60µm
- 6.6µm deep
- Crater surface roughness 0.5µm

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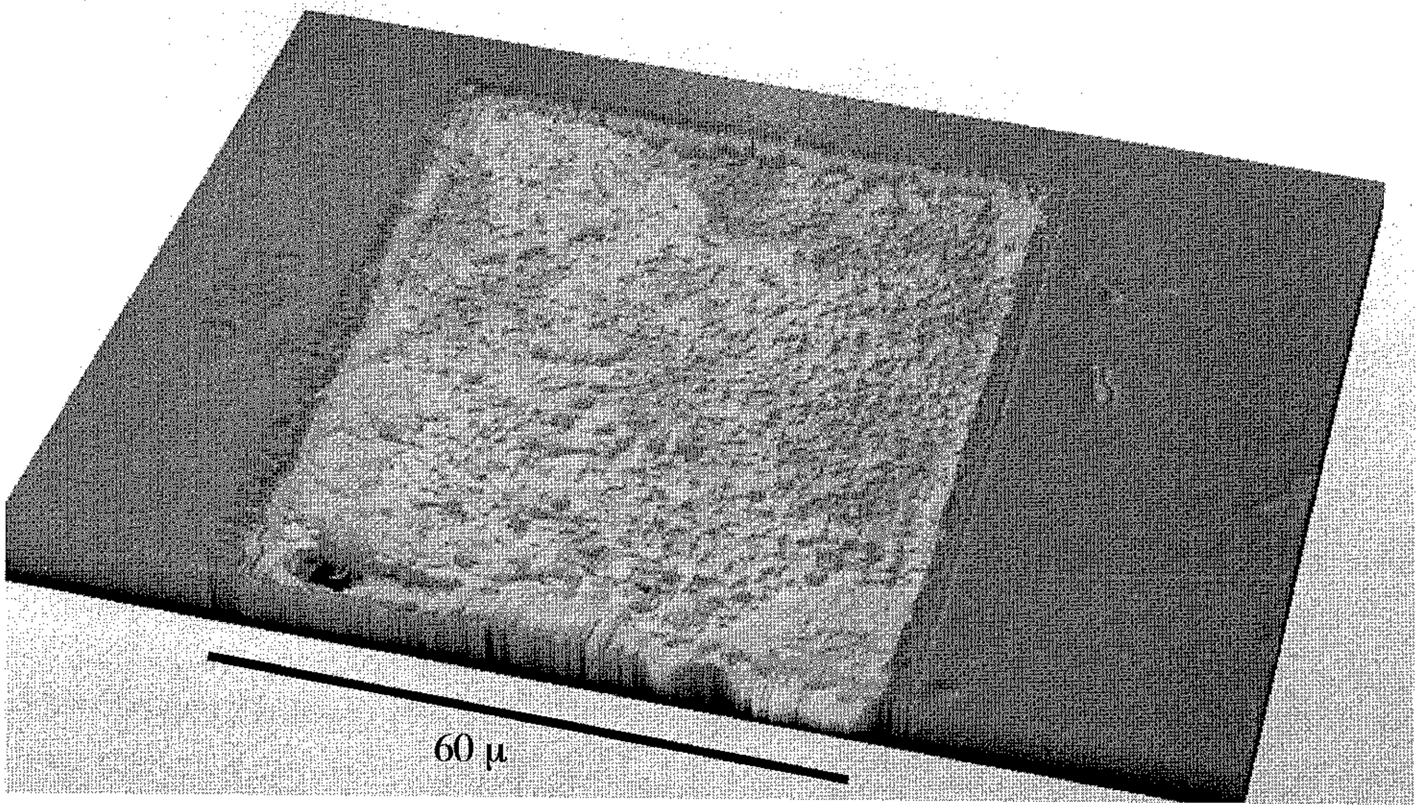
- Optical Profilimetry of Crater in Uranium



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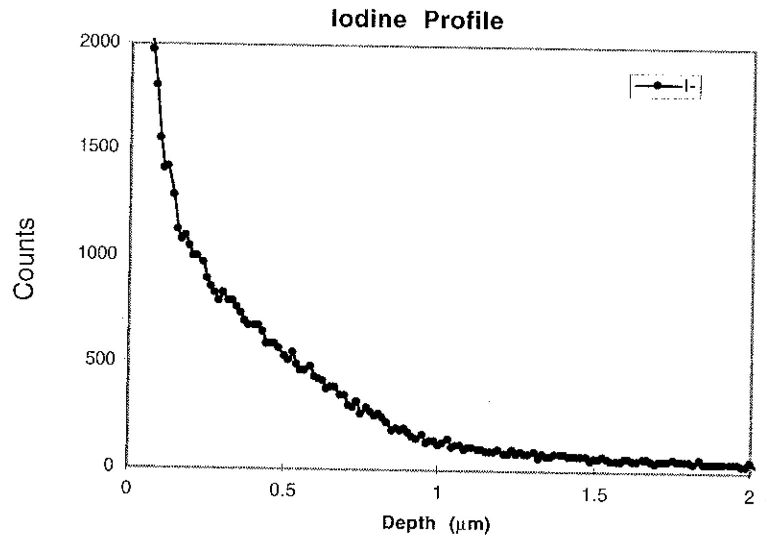


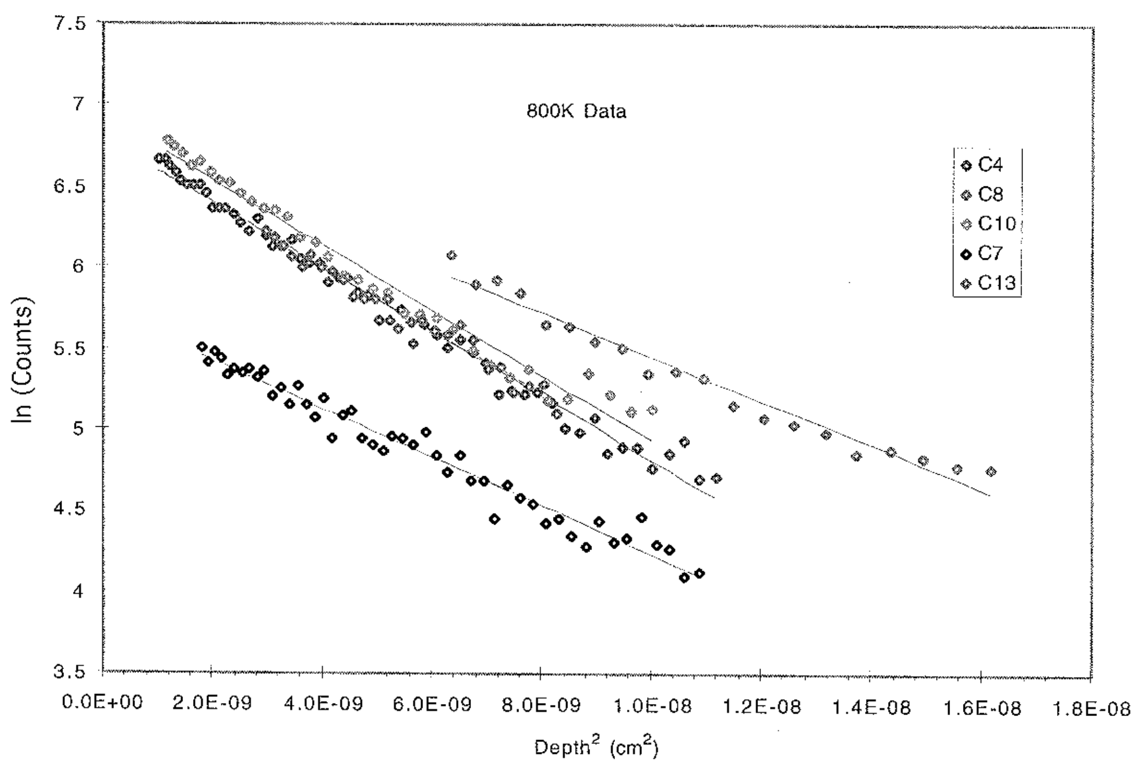
- Iodine concentration measured as a function of depth

- Use Fick's Diffusion Laws to obtain Diffusion Coefficient

$$c(z,t) = c_o \exp\left(\frac{-z^2}{4Dt}\right)$$

$$\ln[c(z,t)] \;\; vs \;\; z^2$$

$$Gradient = \frac{-1}{4Dt}$$



$D_I(T)$ x$10^{-15}cm^2s^{-1}$

800 K	0.73
	0.97
	1.07
	0.72
	0.76
Average	0.85
Stdev	0.16

Values agree with previous estimates from Xe release experiments

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Summary

- Quantitative Fission Gas analysis has been applied extensively to AGR UO_2 and now MOX Fuel.
- SIMS now being applied to MOX to:
  - Produce ^{239}Pu maps on unirradiated/irradiated fuel
  - Produce radial profiles on irradiated fuel.