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

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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.
- 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 Volume Swelling

- Depending on Irradiation History bubbles form:
  - Circular Lenticular Bubbles
  - Extended Lenticular Bubbles
  - Multi-lobed Lenticular Bubbles
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 ($V_{ii}$)
Intergranular Volume Swelling
In-House Software Developed for Image Analysis

\[ P = 2L + (n\pi + 4 - 2n)r_p, \]

\[ A = \frac{n}{2} \pi r_p^2 + 2Lr_p, \]

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

- Volume of porosity obtained
- \( L, r_p \) and \( n \) allow bubble morphology to be examined
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
BNFL Magnox Generation

Fuel TEM

AGR Fuel Pin
Slice of fuel pin cut
Segment of fuel examined in SEM

TEM sample examined in SSEM to locate analysis position

Segment of fuel electropolished and examined in TEM
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.
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.
(AGR Fuel ramped in Halden)
Berkeley SIMS

- Vacuum Generators (VG) MT500 SIMS
- FEI Ga\(^+\) Liquid Metal Ion Gun (LMIG) (Resolution 100nm)
- VG EX05 Ar\(^+\) Ion Gun (Resolution 30\(\mu\)m)
Advantages for Fuel Analysis:

- Isotopic sensitivity e.g. can map/profile $^{239}\text{Pu}$ or $^{240}\text{Pu}$ in MOX
  (Does not have the same peak overlap problems associated with EDX/WDX)
- Increased spatial resolution, 0.1\(\mu\)m relative to large 1\(\mu\)m 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
SIMS

SEI and Isotopic maps using Ga\(^+\) LMIG

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

Aim to apply mapping to Pu isotope distributions in irradiated and unirradiated MOX fuel.
Isotopic Analysis of Fuel - Used to generate isotopic radial profiles
Depth Profiles - Iodine Diffusion in Uranium

- Ga\(^+\) ion gun crater
- 60\(\mu\)m x 60\(\mu\)m
- 6.6\(\mu\)m deep
- Crater surface roughness 0.5\(\mu\)m
BNFL
Magnox Generation

SIMS

- Optical Profilimetry of Crater in Uranium
- Iodine concentration measured as a function of depth

- Use Fick's Diffusion Laws to obtain Diffusion Coefficient

\[ c(z, t) = c_0 \exp \left( \frac{-z^2}{4Dt} \right) \]

\[ \ln[c(z,t)] \text{ vs } z^2 \]

\[ \text{Gradient} = \frac{-1}{4Dt} \]
$D_1(T) \times 10^{-15} \text{cm}^2\text{s}^{-1}$

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
Summary

- Quantitative Fission Gas analysis has been applied extensively to AGR UO₂ 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.