Electron Microscopy for Microstructural Studies on Fast Reactor Structural Materials

C. N. Venkiteswaran, V. Karthik, Divakar Ramachandran, B. K. Ojha, V. V. Jayaraj, C. Padmaprabu, V. Anandraj, P. Visweswaran, Ranvijay Kumar, Shaji Kurien, T. Johny, Jojo Joseph, S. Venugopal and T. Jayakumar

Metallurgy & Materials Group
Indira Gandhi Centre for Atomic Research
Kalpakkam, TN 603102, India

E-mail: divakar@igcar.gov.in
Contents

• Introduction
  – PIE facilities at IGCAR
  – Projects
• Electron Microscopy – Importance
• Fractography studies
• TEM of irradiated structural materials
  – Ion irradiation
  – Neutron irradiation
• Summary and Conclusions
PIE Facility at RML-IGCAR

Seven \( \alpha/\beta/\gamma \) inert atmosphere hot-cells for PIE of FBTR and experimental sub-assemblies
PIE at RML-IGCAR

- Fuel receipt
- Sodium Removal
- Visual Examination
- Profilometer
- Dismantling

NDE:
- ECT / XR / GS / NR

- FG Analysis
- Metallography
- Hi-T Tensile test
- Small Specimen Test
- Microscopy

- Performance assessment of irradiated Fuels & Structural materials
- Feedback to Designers, Fabricators & Operating Personnel
- Inputs for development of Advanced Fuels and Structural Materials
PIE Projects at RML-IGCAR

- 25+ years of experience O&M of α, β, γ Hot Cells
  - Development and refurbishment of in-cell equipments
- Progressive life extension of FBTR
  - PIE of fuel sub-assemblies from 25 to 165 Gwd/t
  - PIE of Control rod assembly
  - PIE of Nickel reflector subassembly
  - PIE of grid plate material
- Evaluation and validation of PFBR design
  - PIE of experimental MOX fuel pin – BOL studies
  - PIE of MOX test fuel sub-assembly after 112 Gwd/t burn-up
  - Irradiation of structural materials
- FBTR as an irradiation facility
  - Production of Radio Isotopes for medical use
  - In-reactor creep rate studies on Zr alloy used in PHWRs
  - PIE of experimental FBTR fuel pins
- Collaborative Projects for other sectors
Irradiation Induced Defects

Macroscopic consequences -
Cracks, Fracture, Swelling, Embrittlement...
Irradiation Effects in Fast Reactor Structural Materials – Radiation Damage

- High neutron fluence > $10^{15} \text{n}^2/\text{cm}^2\cdot\text{s}$ at 523 – 973K leads to void swelling and irradiation creep
- Dimensional changes limit burn-up
- 20%CW 316 at 530 C under $1.5 \cdot 10^{23} \text{n}/\text{cm}^2$ (75 dpa) undergoes 10% linear expansion
- (i) lattice atoms displaced from equilibrium positions, and (ii) production of helium by $(n,\alpha)$ reactions
- Long-range diffusion and clustering of defects can give rise to microstructural changes – dislocation loops, SFT, precipitation, voids, solute segregation

Incubation period – dislocation density and cavity density reach steady-state values; radius of the cavities reach critical value for growth
Linear swelling regime - cavities grow steadily as a result of the biased absorption of interstitial atoms by other sinks
Why Electron Microscopy for PIE?

- Resolution
- Electron – Matter Interactions
- Depth of Field / Focus
- Diffraction

- Limitations
  - Sampling
  - Image artefacts
  - Electron beam damage
  - Specimen preparation

- Atomic-scale events --> Microstructural evolution --> macroscopic effects
- Radiation damage mechanisms are usually very complicated with interactions between structural and microchemical changes
- Nanometre-sized point defect clusters – vacancy / interstitial dislocation loops, stacking fault tetrahedra, bubbles and voids; high dislocation densities; modified precipitation behaviour; solute segregation
- Electron microscopy affords routine high-magnification direct imaging of irradiation features along with micro-chemical information
- Special case of HV-TEMs where *in situ* irradiation is possible
Electron Microscopy of FR Structural Materials

- Mechanical behaviour of irradiated fuel cladding
  - Fractography studies on tensile test fracture surfaces
- Microstructural study on Irradiated fuel cladding and wrapper
  - Ion irradiation
  - Neutron irradiation
Irradiated FBTR clad – Remote Tensile Tests

<table>
<thead>
<tr>
<th>Condition</th>
<th>Test Temp K</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ambient</td>
<td>298K</td>
</tr>
<tr>
<td>Fuel Handling</td>
<td>453 K</td>
</tr>
<tr>
<td>In-Reactor</td>
<td>703 – 783 K</td>
</tr>
</tbody>
</table>

70 mm length of clad tube fitted with Mandrels, fixed in a gripper and loaded on a screw-driven tensile test machine with a furnace in hot cell.
Tensile tests on Irradiated SS 316 Clad

Effect of damage level (dpa)

Effect of Test Temperature

Test Temperature = Irradiation Temperature
Trends in Strength and Ductility

- Decreasing Strength Levels in high temperature and RT tests beyond 60 dpa
- Declining UTS with increasing void swelling
- Elevated temperature and room temperature ductility of 2 – 4% at high dpa

RT - Room temperature, $T_{test}$ – Test temperature, $T_{irrad}$ – Irradiation temperature (430C – 500C)
Carbide fuel pins have seen a peak displacement damage of 83 dpa in FBTR fuel sub-assemblies that have seen a burn-up of up to 165 GWd/t. Tensile tests on clads at various stages have been carried out indicating a successive reduction in residual strength and ductility. Fractography of these clad specimen have been carried out in a SEM.

Tensile tests were carried out on clad tube sections after fuel dissolution. Care was taken to protect fracture surfaces after the test. An in-cell cutting machine was used to extract sections including the fracture surface. These were decontaminated and sputter coated with gold to minimise and trap transferable contamination. Specimens were then transferred to the SEM facility in a shielded pot.
Ductile fracture mode at low damage level
Change in fracture mode at higher damage

56 dpa – Mixed mode of fracture with ductile dimple voids & cleavage facets

81 dpa – Cleavage surfaces suggesting flow localization; dimples and microvoids are less abundant
Fracture mode vs dose (dpa)

- 13 dpa – ductile fracture
- 56 dpa – mixed mode
- 83 dpa – cleavage facets
Specimen extraction from irradiated wrapper

- Hollow end-mill cutter, Shear punch tool
- Uniform Temperature and fluence within specimen
- Multiple examinations like swelling, Testing & Microscopy
- Easy handling due to reduced dose
Microstructural changes in irradiated SS316 Wrapper

- Extensive Void formation beyond 30 dpa, of size 8 – 30 nm
- Microstructure with Precipitates in addition to dislocation loops
- Retention of CW at 83 dpa
- Presence of Ni and Si rich M6C precipitates - η Phase
- Radiation induced Precipitates – G Phase - M6(Ni,Co)16Si7
Removal of Ni and Si from solid-solution in the austenitic matrix has an effect on supersaturation of vacancies that leads to void nucleation. Thus radiation induces segregation and precipitation which leads to onset of increased swelling.
- 30 dpa: voids and dislocation loops
- 40 dpa: radiation enhanced Ni and Si enriched \( \eta \) phase precipitation observed
- 83 dpa: radiation induced G phase \( \text{M}_6(\text{Ni,Co})_{16}\text{Si}_7 \) formation
- Voids associated with precipitates and matrix voids
Alloy D9 – Ti-modified 316 grade, 20%CW

0.05C 0.7Si 1.5Mn 15Cr 2Mo 15Ni 0.3Ti Bal Fe

Fine scale precipitation at the scale of ~ 10 nm is associated with dislocation networks

<table>
<thead>
<tr>
<th>(hkl)</th>
<th>dy (nm)</th>
<th>dTiC (nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>111</td>
<td>0.2070</td>
<td>0.2499</td>
</tr>
<tr>
<td>200</td>
<td>0.1793</td>
<td>0.2164</td>
</tr>
<tr>
<td>220</td>
<td>0.1268</td>
<td>0.1530</td>
</tr>
<tr>
<td>311</td>
<td>0.1081</td>
<td>0.1305</td>
</tr>
<tr>
<td>222</td>
<td>0.1035</td>
<td>0.1250</td>
</tr>
</tbody>
</table>
Ion vs. neutron irradiation – Temperature shift

The damage rate in Ni+ irradiation experiments is about three orders of magnitude higher than that typically experienced by materials under neutron irradiation in a fast reactor. The equivalence of the high rate damage to the much slower damage rate under neutron irradiation in a fast reactor can be expressed in terms of a temperature shift.

$$\Delta T = T_i - T_n = \frac{A T_i^2}{1 + A T_i}$$

$$A = \frac{R}{Q} \ln \left( \frac{G_i}{G_n} \right)$$

<table>
<thead>
<tr>
<th>$T_i$ (K)</th>
<th>$\Delta T$</th>
<th>$T_n$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>773</td>
<td>202</td>
<td>571</td>
</tr>
<tr>
<td>823</td>
<td>225</td>
<td>598</td>
</tr>
<tr>
<td>873</td>
<td>249</td>
<td>624</td>
</tr>
<tr>
<td>923</td>
<td>274</td>
<td>649</td>
</tr>
<tr>
<td>973</td>
<td>300</td>
<td>673</td>
</tr>
</tbody>
</table>


Low-energy Accelerators for Ion Irradiation

- Energy: 250 keV – 3400 keV
- Current: Several μA
- Damage rate: $7 \times 10^{-3}$ dpa/s

Irradiation temperature range
300 ºC – 750 ºC

Step height is used to estimate volumetric swelling
Ion irradiation – SS316L vs. Alloy D9

AISI 316 type SS pre-implanted with 15 appm He and irradiated with 5 MeV Ni+ ions to 100 dpa at 898 K

Alloy D9 pre-implanted with 100 appm He and irradiated with 5 MeV Ni+ ions to 100 dpa at 873 K
Ion irradiation performance of Alloy D9

- Under 5MeV Ni+ ion irradiation to 100 dpa,
- AISI 316 type SS shows a high density of voids of average size 2.7 nm
- 20 % CW Alloy D9 shows few voids
- Alloy D9 has a high density of microstructural traps for vacancies at titanium carbide precipitate – matrix interfaces and at defects in Alloy D9.
### PIE of PFBR MOX Test FSA in FBTR up to 112 GWd/t

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Specification</th>
</tr>
</thead>
<tbody>
<tr>
<td>No. of fuel pins</td>
<td>37 pins</td>
</tr>
<tr>
<td>Fuel stack length</td>
<td>240 mm</td>
</tr>
<tr>
<td>Pellet diameter OD &amp; ID</td>
<td>5.55 &amp; 1.75</td>
</tr>
<tr>
<td>Fuel pin OD &amp; ID</td>
<td>6.6 &amp; 5.7</td>
</tr>
<tr>
<td>Clad &amp; Wrapper material</td>
<td>20% CW D9</td>
</tr>
</tbody>
</table>

**Diametral profile of the D9 fuel pin clad**

**Reduced swelling of Alloy D9 clad**

**Retention of adequate strength and ductility**
PIE Techniques available at RML-IGCAR

- Visual examination
- Profilometer & laser dismantling
- Sodium removal
- X & Neutron radiography
- Gamma scanning
- Fission gas analysis
- Remote metallography
- High temperature tensile testing
- Small specimen testing
- Electron microscopy
- Fuel receipt
Acknowledgements

- Department of Atomic Energy
- Department of Science and Technology
- Centre for International Co-operation in Science