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Fuel Overview

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Mar 19, 2012

Acknowledgements

- This overview presentation is based on several:
  - NFIR Projects; an international collaborative program led by EPRI
  - Coauthored open literature papers
- R&D described here involved several organizations:
  - CEA, CRIEPI, EPRI, Halden, ITU, NFD, Windscale
- Contributions from and collaboration with several colleagues is thankfully acknowledged

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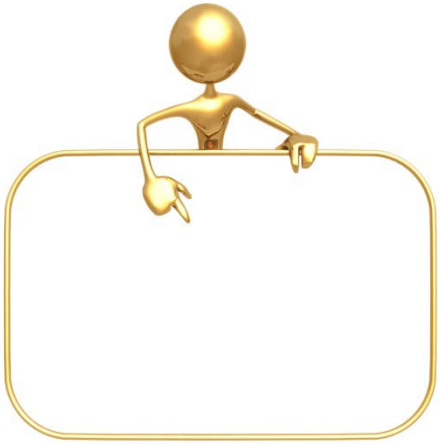
Presentation Outline



- Fuel Data Needs
- Disc Irradiation Approach
- Typical Results
- High Burn-up Structure
- Thermal Diffusivity Recovery
- Microstructure Studies
- Fuel Fragmentation
- MOX Fuel
- Future R&D Needs
- Conclusions

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Fuel Data Needs

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Fuel (UO_2) Data Needs

- Fuel properties of prime interest:
  - Densification and swelling
  - Formation of HBS at the pellet rim
  - Thermal conductivity of pellet materials
  - Fission gas <u>behavior</u> in steady and transient conditions, including precipitation, localization, and release
  - Creep rate and mechanical properties
- These are indispensable inputs to fuel performance and design codes

→ Safe, reliable, and economical use of LWR Fuel

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Specific Project Objectives (Examples)

- Thermal conductivity as a function of burn-up; the effect of grain size and fuel variants (including MOX);
- Determination of density and changes in porosity;
- Fuel melting point and specific heat measurements;
- Evolution of fuel microstructure with burn-up
  - Onset of HBS formation and its dependence on grain size, additives, temperature and/or hydrostatic restraint;
  - Implications of HBS formation on FGR and fuel fragmentation; and
- Evolution of the mechanical properties including thermal creep measurements; the mechanisms involved in visco-plastic deformation behavior of the pellet material

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Disc Irradiation Approach

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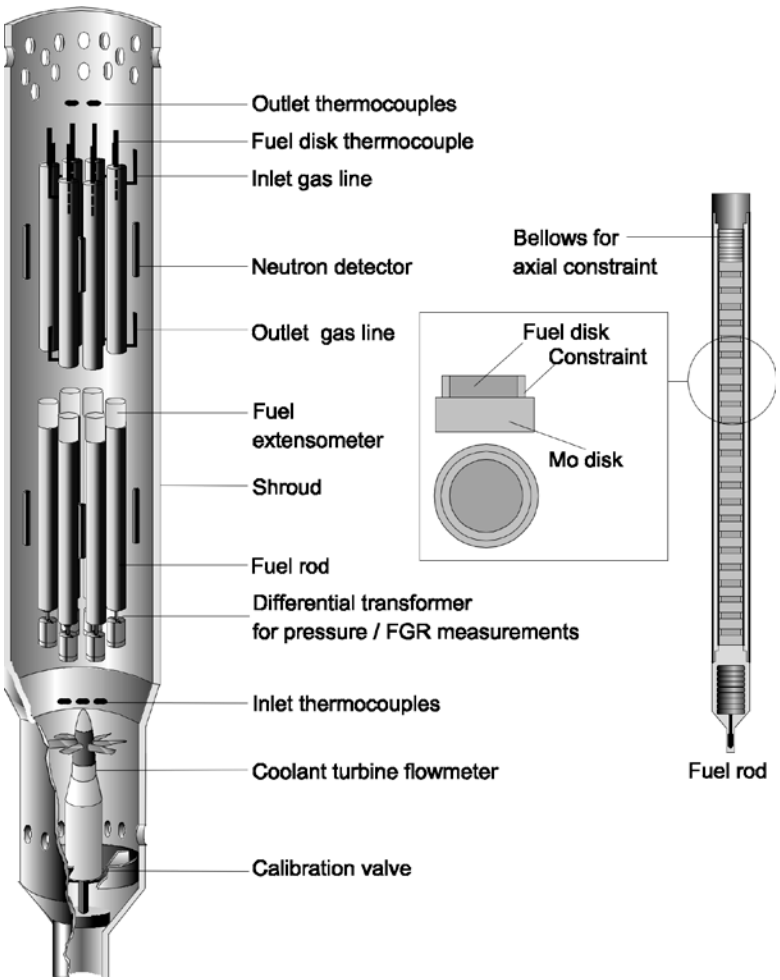
Disc Irradiation Concept

- Nearly isothermal irradiation
  - Fuel discs sandwiched between two Mo disc
    - With or without restrain
  - Each rodlet containing several fuel discs
- Several rodlets irradiated in HBWR in IFAs equipped with
  - ND, neutron flux detector
  - TF, thermocouples
  - PF, pressure transducer
  - EF, elongation detector
  - GF, gas sweep with external γ-spectroscopy



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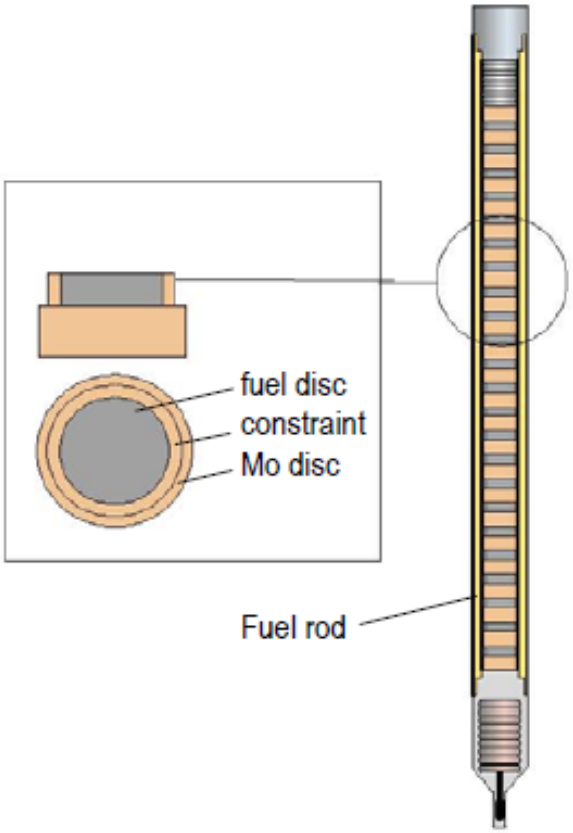
Experimental design

- Mo disc thickness and gap from the cladding aimed at discharge temperatures of approximately 500 and 700°C
- Mo rings around fuel discs provided restraint and prevented fuel discs turning into unusable fragments for PIE purposes



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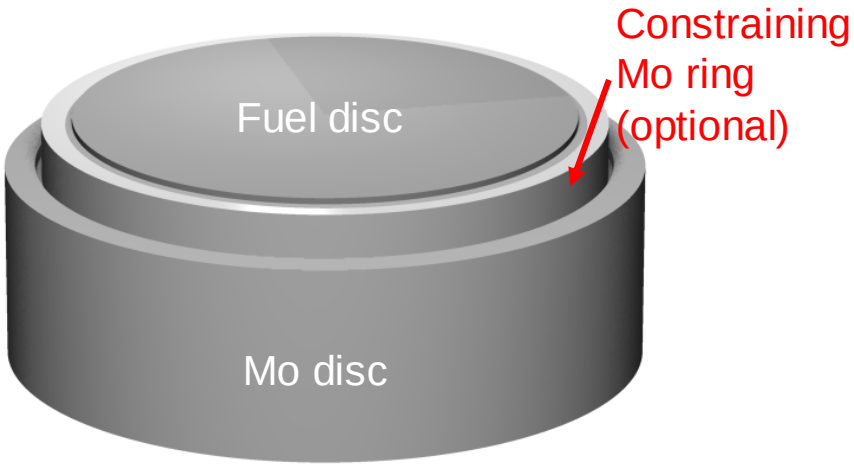
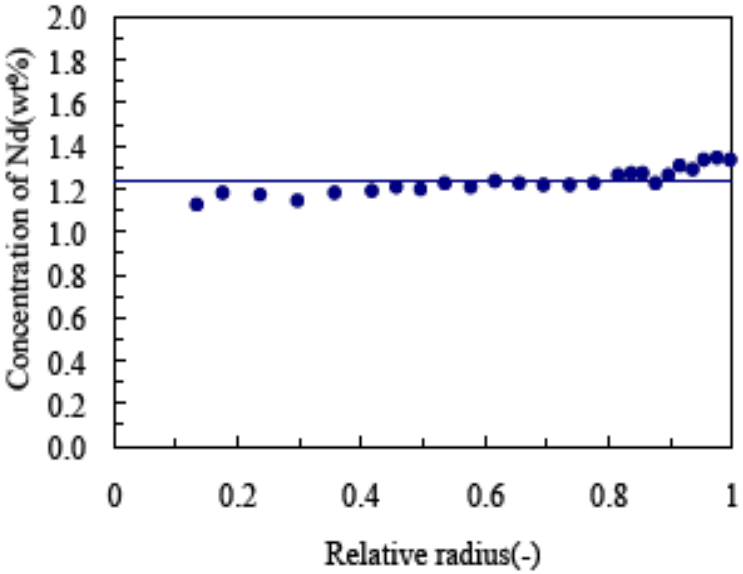
Disc Irradiation Concept (cont'd)

- A wide range of burn-up up > 100 GWd/t achieved in HBWR
- "As-irradiated" fuel samples ideal for advance PIE and properties measurements
  - Minimum sample size
  - Free of strong gradients in burn-up or irradiation temperature
- Applied successfully
  - Five irradiations since 1989





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Collaborative Disc Irradiation Programs

IFA# (Program)	Time period	Fuel Temperature (°C)	Max burn-up (MWd/kg)
IFA-569 (NFIR)	1989-1991	600-800	77
IFA-563 (HPG)	1991-1996	900-1200-1400	36
IFA-601 (HBRP)	1993-1996	500-700-900-1200	103
IFA-649 (NFIR)	2001-2006	550-700	103
IFA-655 (HPG)	2001-2006	500-700	125

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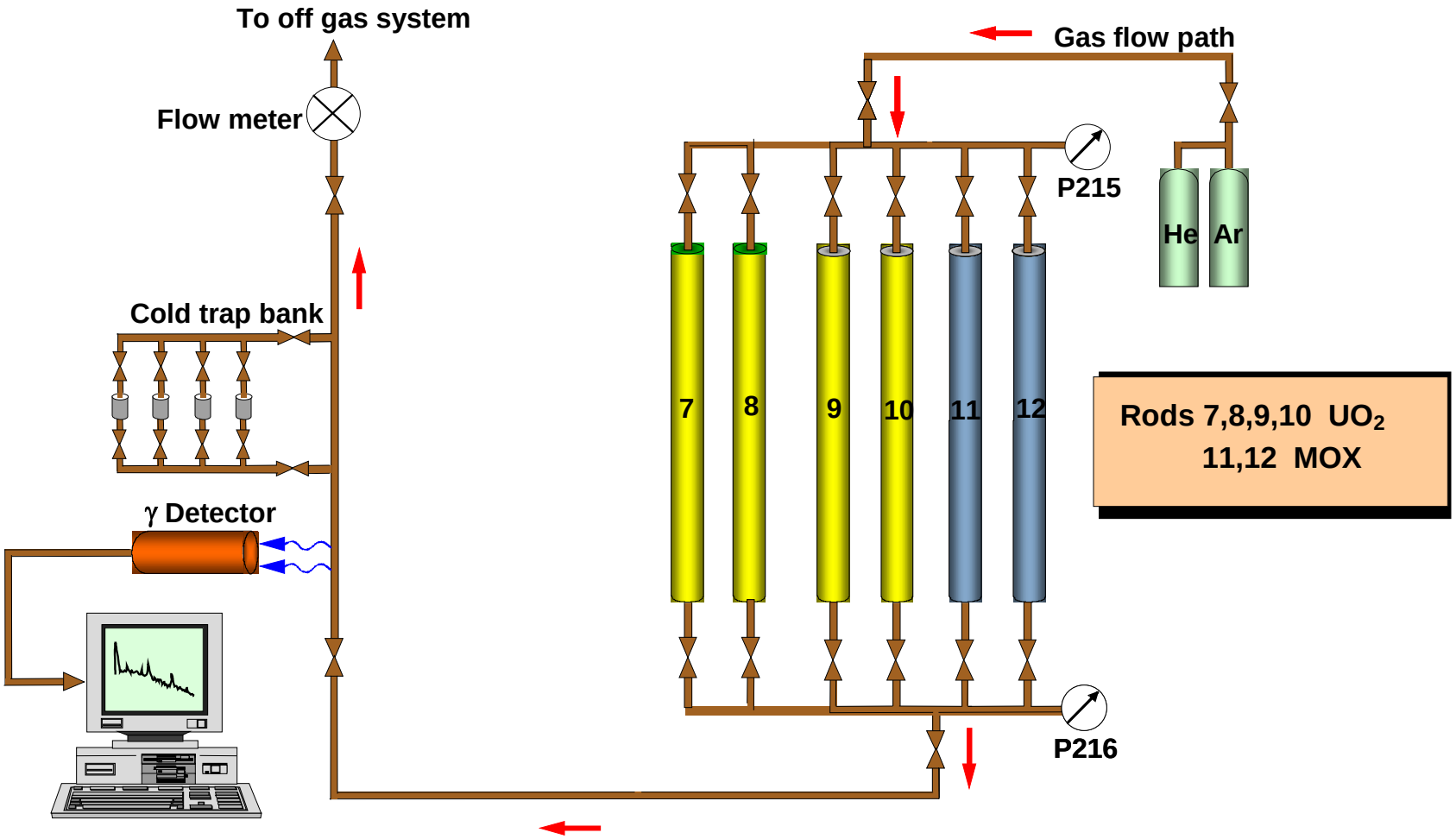
Gas flow system: IFA-655



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Gas flow analysis

Assuming Booth model from short-lived fission products for spherical grains:

Isotopic release rate from γ-spectrometry

Recoil release to birth rate

$$\frac{R}{B} = \frac{S}{V}\sqrt{D}\sqrt{\frac{\alpha}{\lambda}} + \frac{R}{B}_{recoil}$$

Isotopic birth rate from fission yield calculations

Square root of precursor enhancement factor / decay constant

Surface area to volume ratio * square root of diffusion coefficient

In start-up diffusivity analysis S/V fixed

Through-life analysis D fixed

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Typical Results

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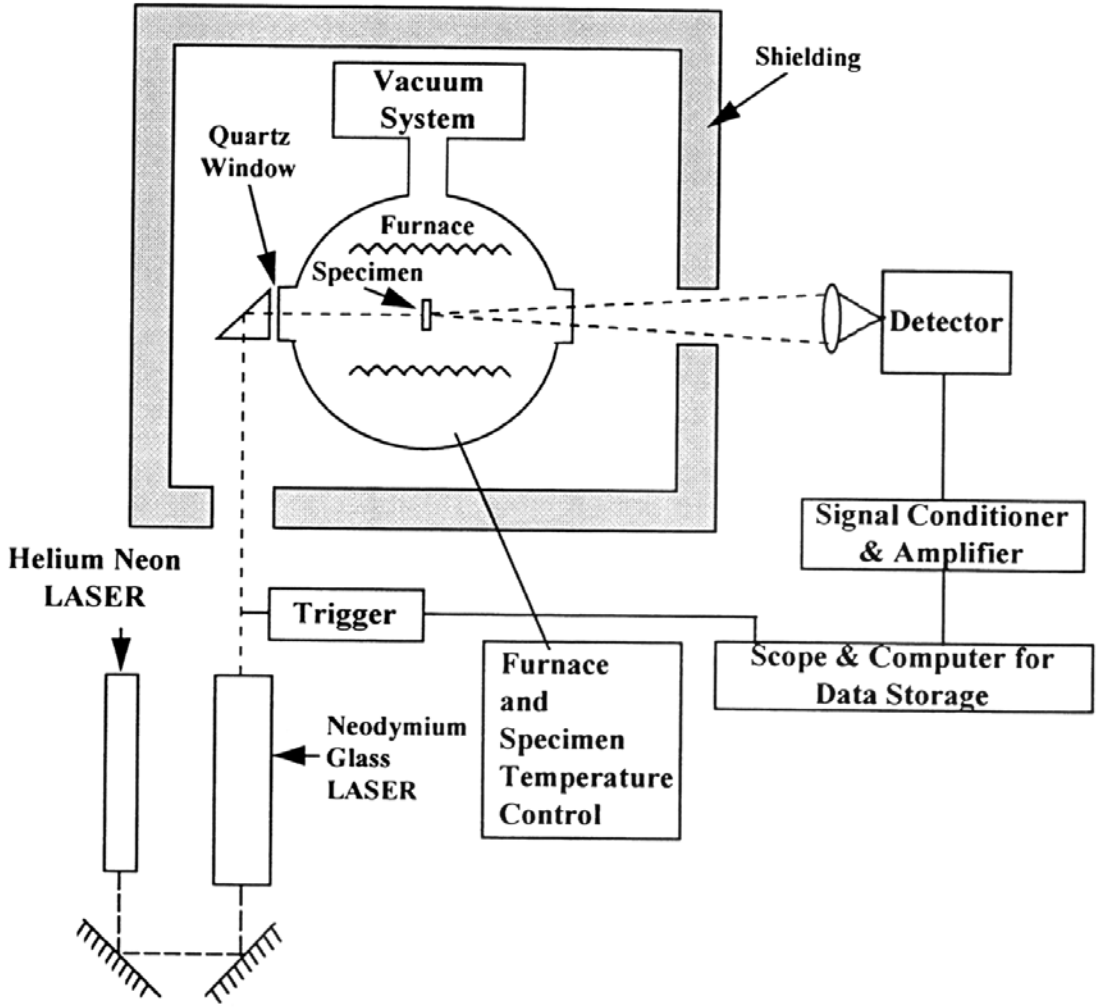
Laser Flash Method



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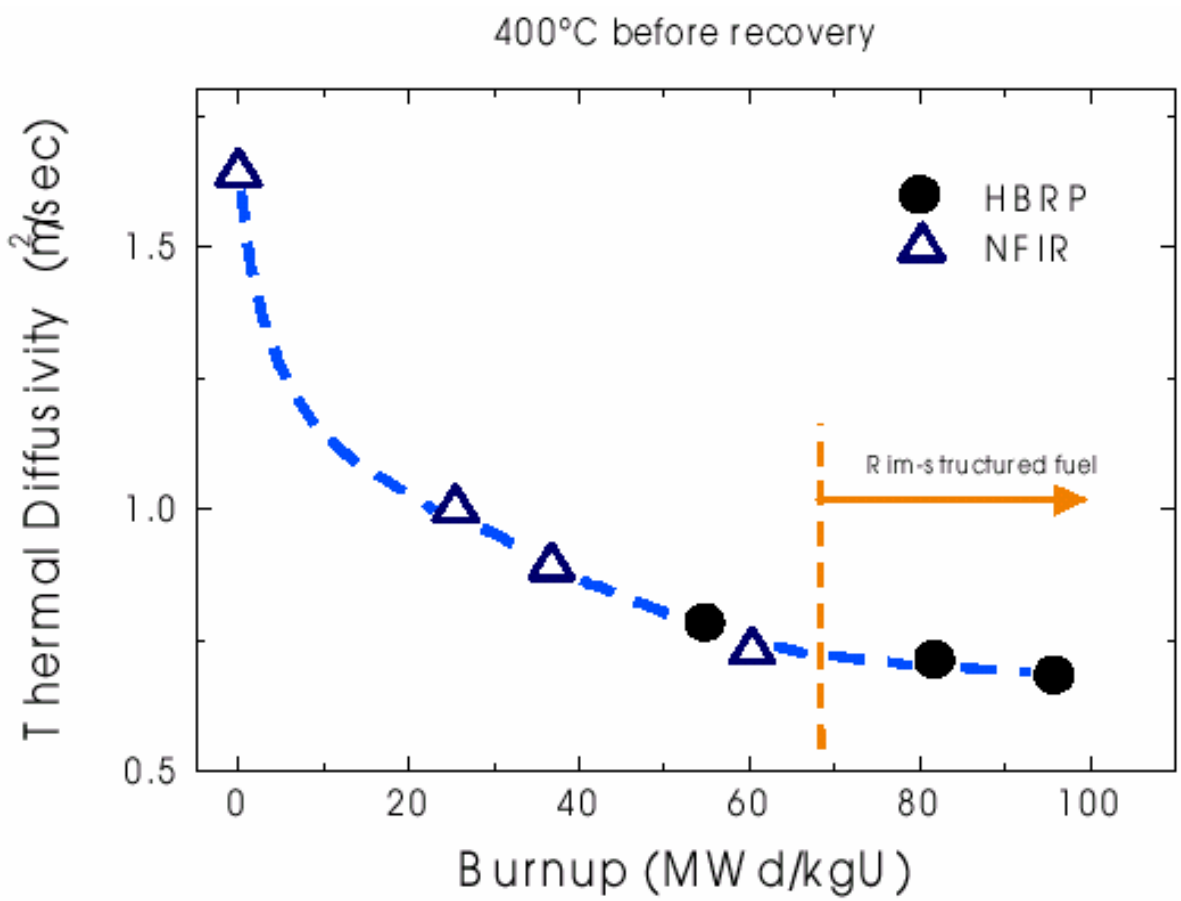
IFA-601& IFA-569 PIE Data Compared--TD



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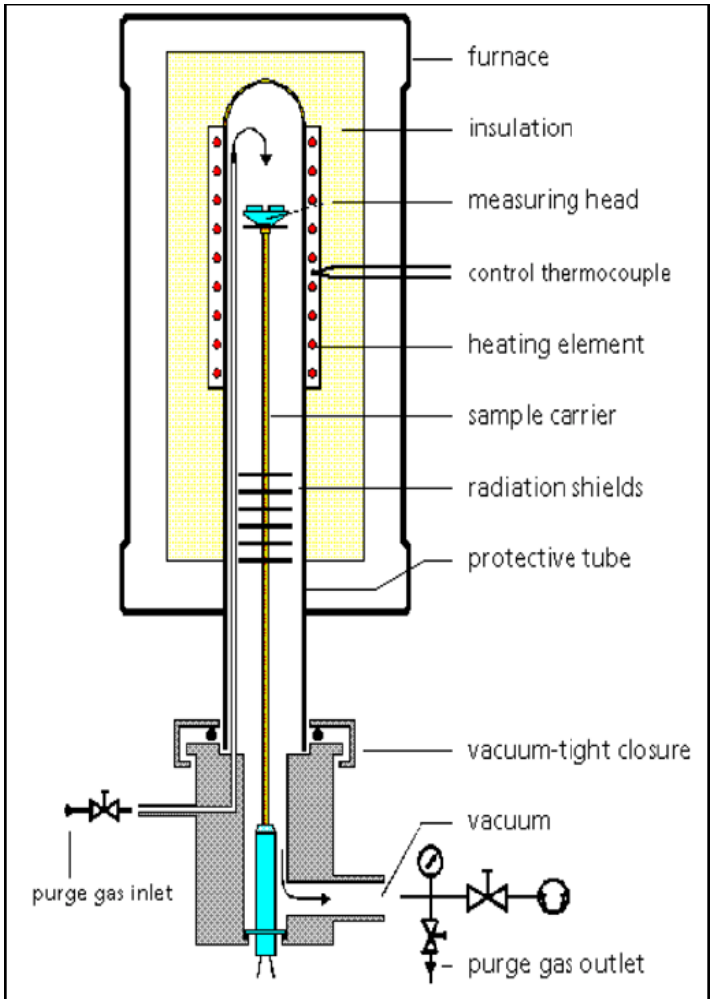
DSC method of C_p determination



- Differential scanning calorimetry (DSC) adapted for remote operation in hot cell
- Two identical crucibles under the same external heating or cooling conditions
  - In flowing high purity Ar in a furnace
- Procedure adapted for ‘sample’ or ‘reference standard’ in one crucible, while other is empty

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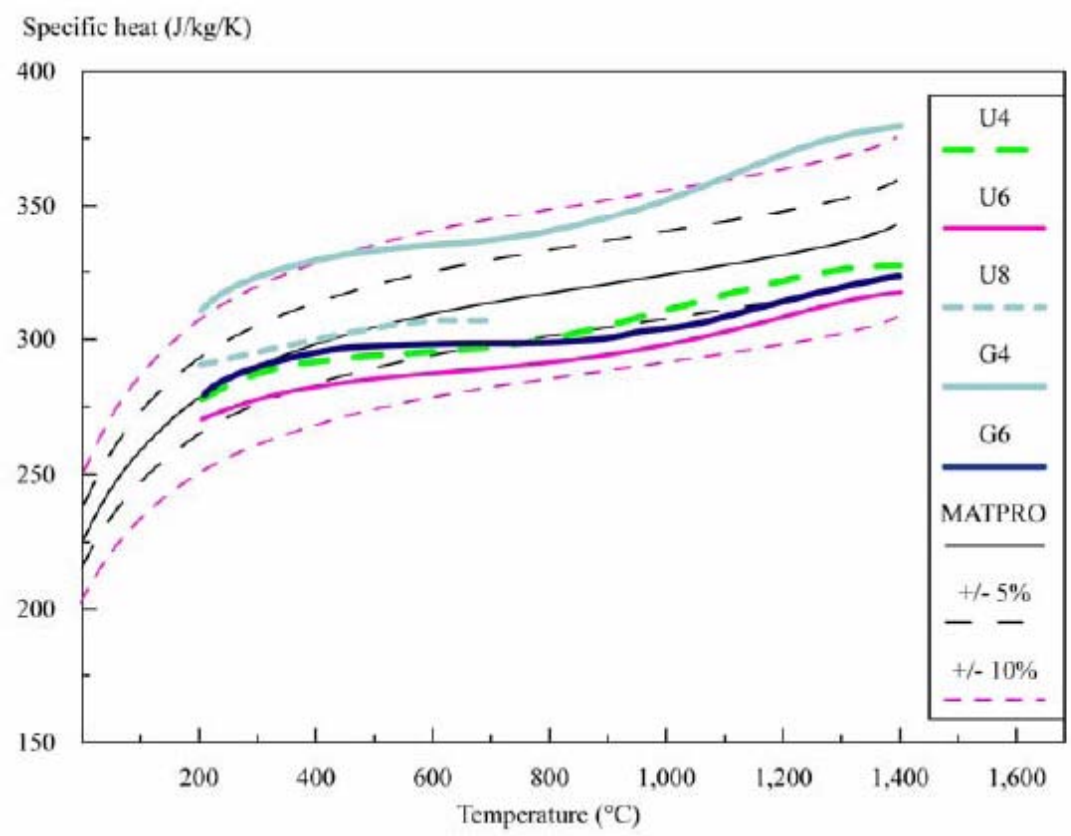
C_p data: comparable to MATPRO within 10%



- MATPRO does not account for irradiation and doping

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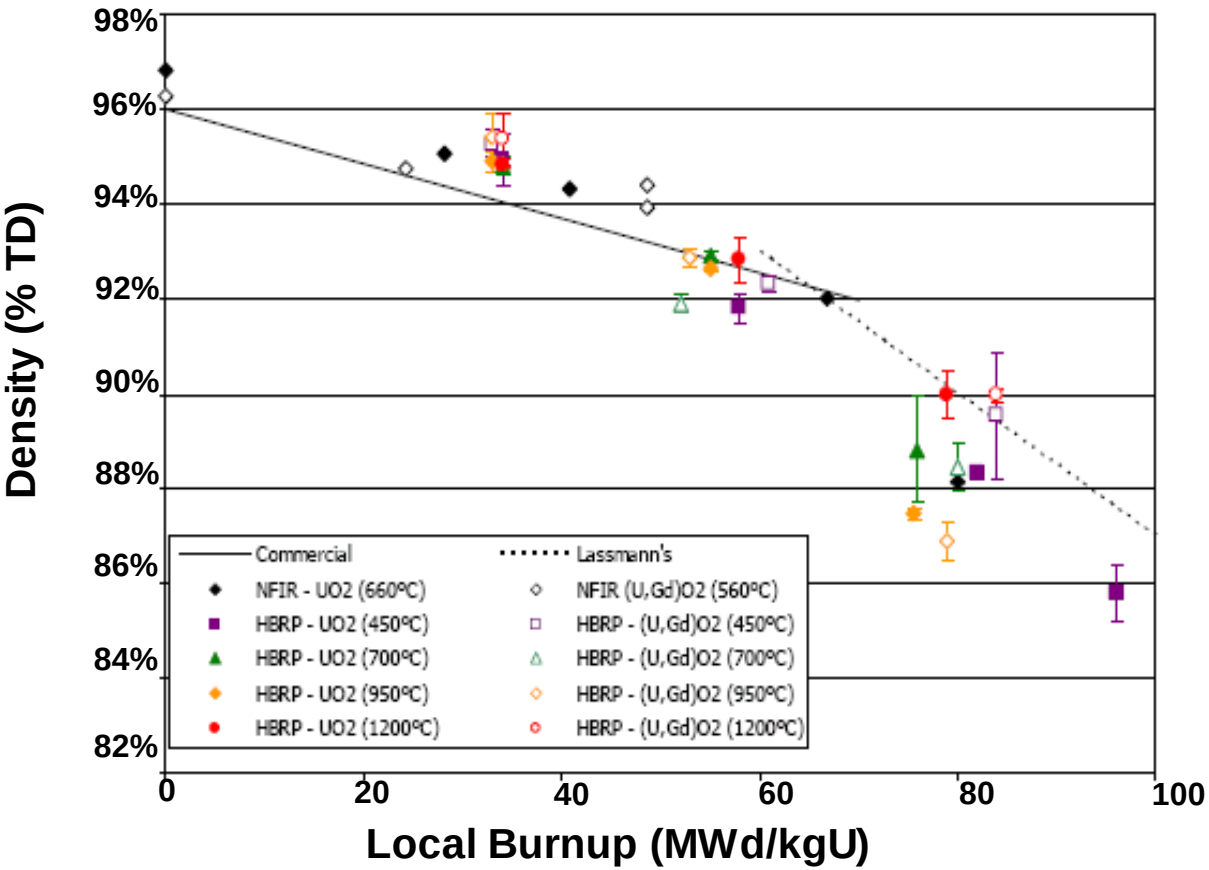
IFA-601& IFA-569 PIE Data Compared--Density



- **Dotted line is based on swelling data**
  - Converting to density neglects fission products in the lattice; hence expected to be higher

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High Burn-up Structure (HBS)

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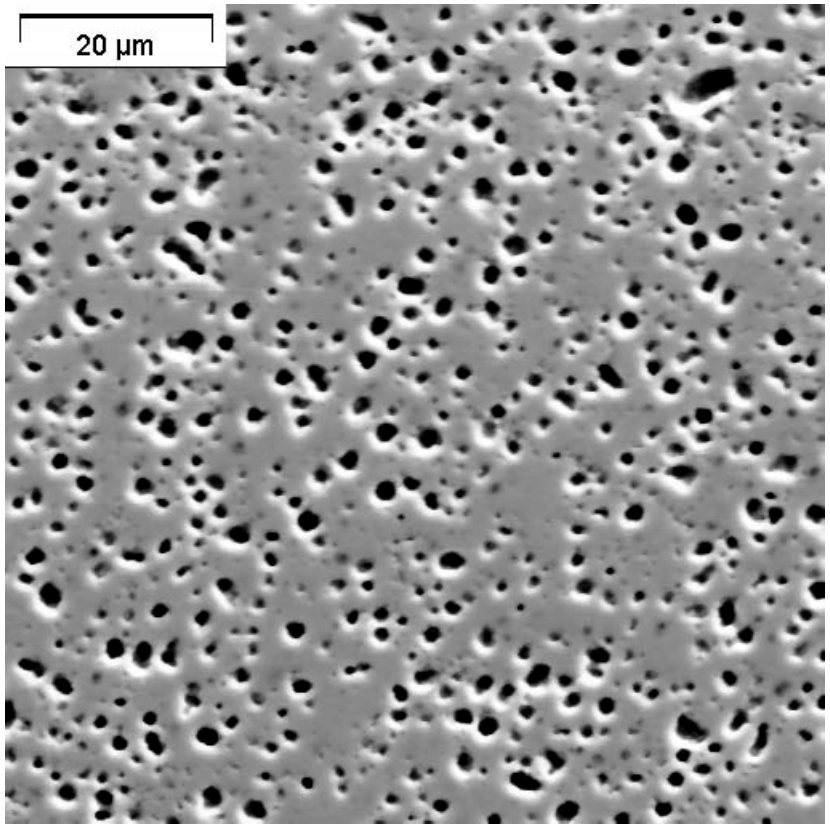
High Burn-up Structure (HBS)



- Highly porous;
- Amorphous structure;
- Fission gas trapped in small sub-micron bubbles
- Grains sub-divided
- Tendency to swell and crack upon heating
- A high concentration of over-pressurized gas bubbles

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IFA-601

- One of the objective: Threshold temperature for HBS formation
- Design: Fuel discs irradiated to 4 different burn-ups at 4 different temperatures
- Extensive PIE performed: Key results described in previous publications

Rod #	Fuel Temperature (°C)	Max burn-up (MWd/kg U)
1	600	55, 70, 85, 100
2	800	55, 70, 85, 100
3	1000	55, 70, 85, 100
4	1250	55, 70, 85, 100

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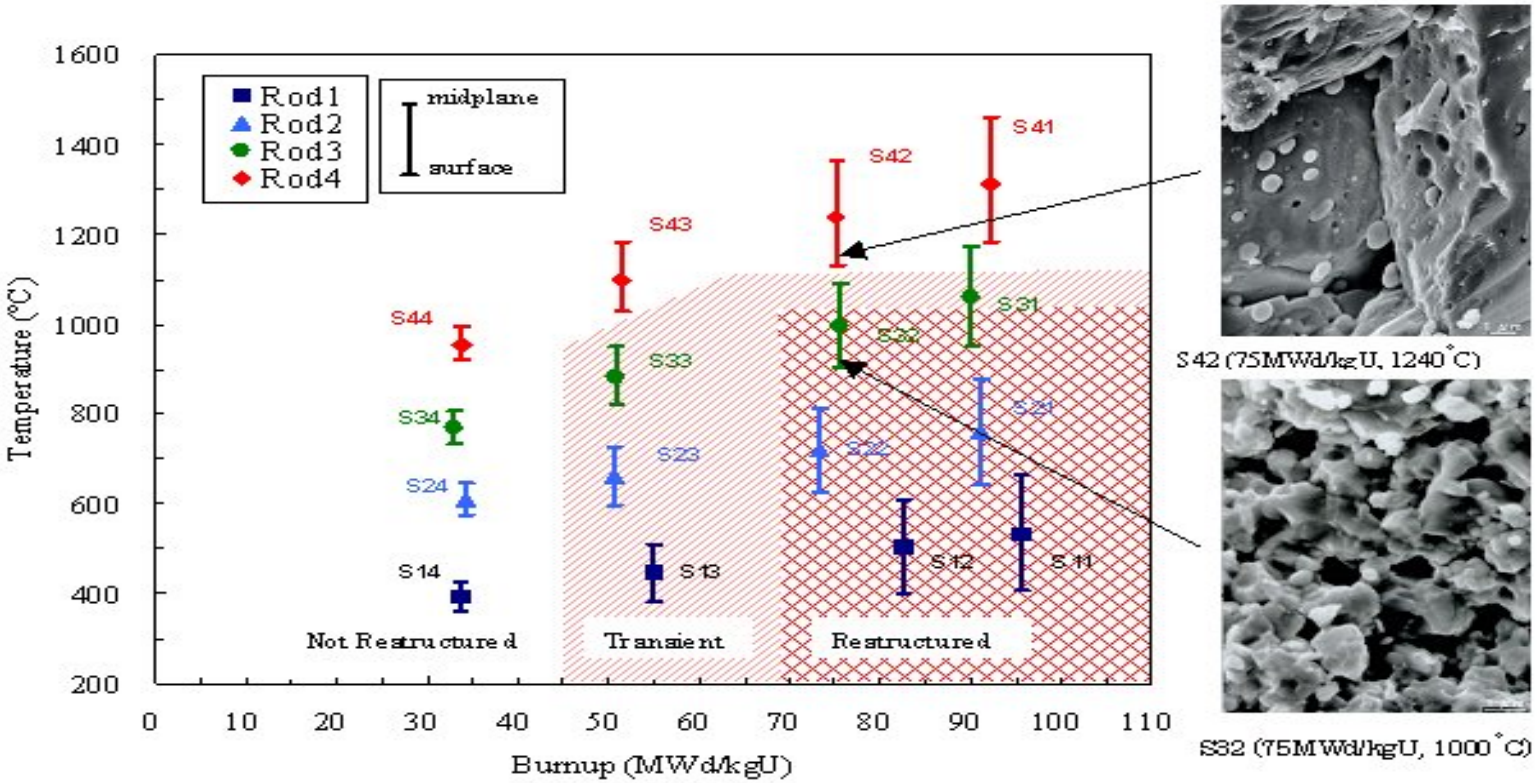
Onset of HBS Formation as a Function of Burn-up



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Thermal Diffusivity (TD) Recovery

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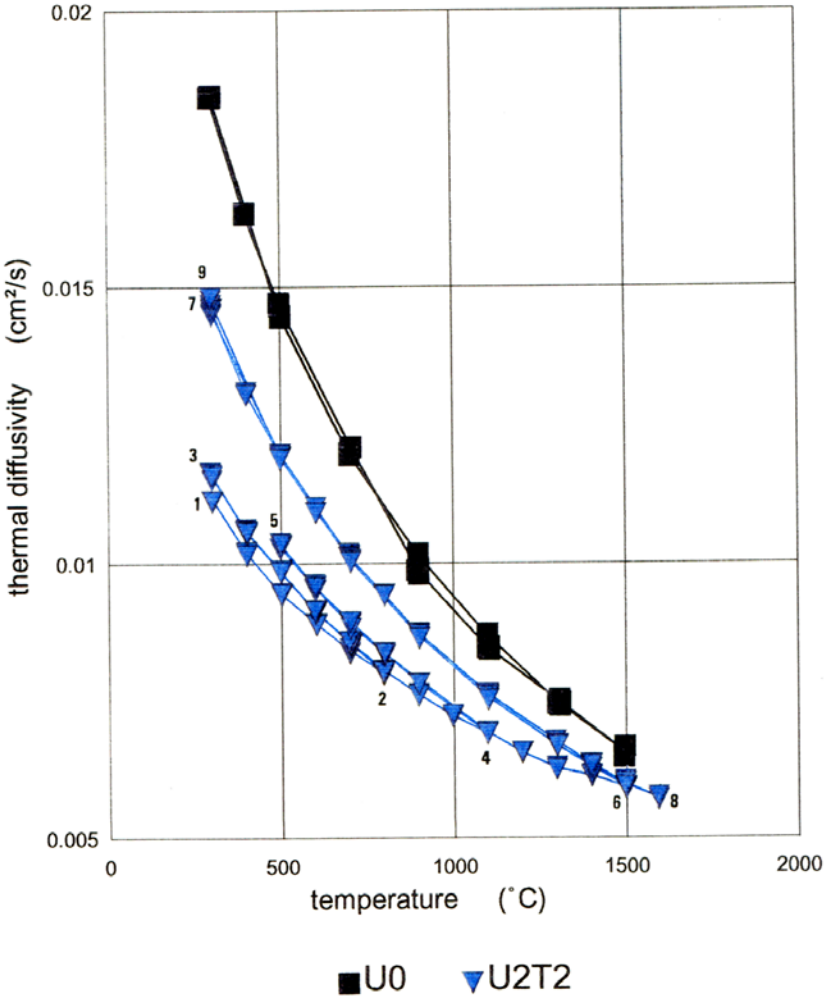
TD-Recovery Phenomenon



$$\%\,\mathrm{Recovery} = \frac{TD_{fully-anneald} - TD_{as-received}}{TD_{as-received}} \times 100$$

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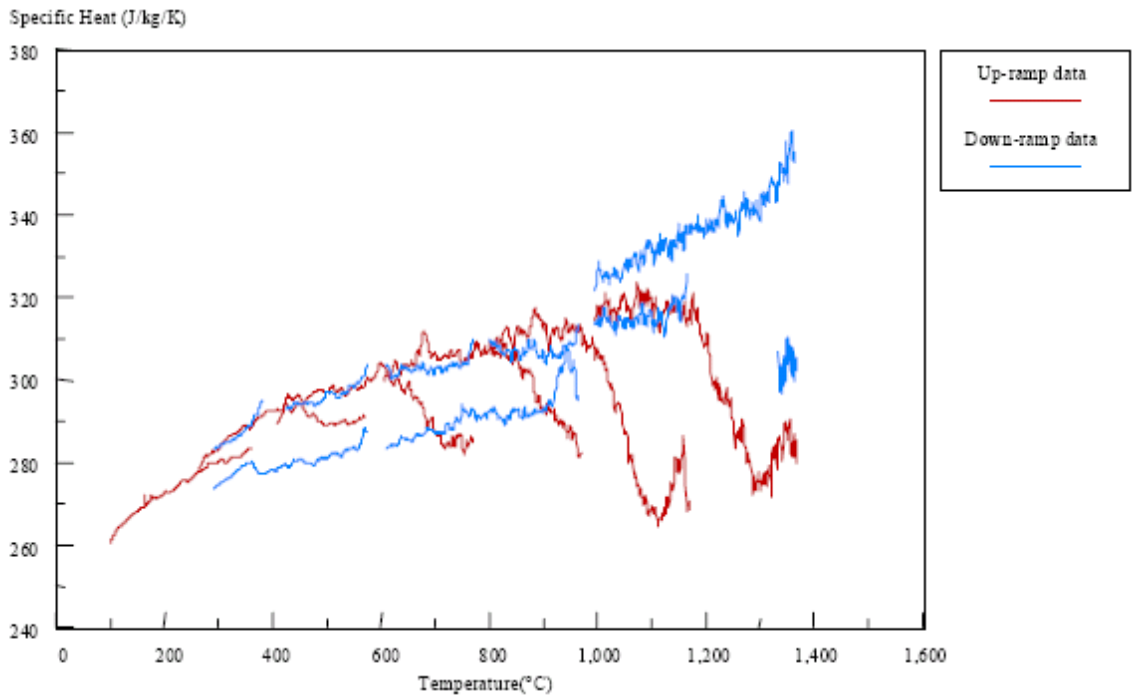
Irradiated UO_2: Up and down ramps showed different DSC response

U6 sample



- Exothermic heat release with annealing
- The perturbations not seen in U0

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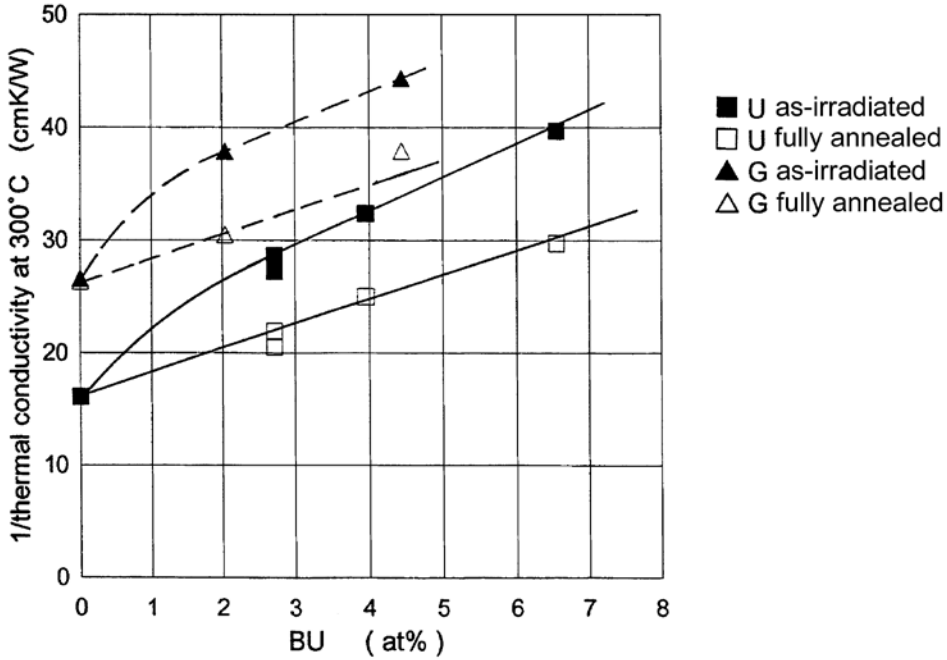
Results



- Inverse thermal conductivity (resistivity) of "as-irradiated" samples:
  - Is higher than that of "fully-annealed" samples
  - Does not extrapolated to zero burn-up (unirradiated) samples
- Points to a non-linear behavior as burn-up approaches to zero

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DISCUSSION (1)

eq. 1 $k = (\rho\ C_p).TD$

eq. 2 $k = f\ \ k_M$

eq. 3 $k_M = (a + b.T + c.\beta)^{-1}$

- At a microscopic level:
  - Radiation damage (point defects, dislocations etc.)
  - Tiny solid and gas clusters of fission products (small enough in size to be phonon scattering centers)

also impact the conductivity directly [additional terms in Eq. (3) are needed]

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DISCUSSION (2)

- These microscopic phonon scattering centers grow with temperature anneals and improve the conductivity.
  - Bubbles 5 to 7 nm → ~ 90 nm in annealing associated with TD measurement cycles
- Available data also indicate
  - Recovery at lower temperature is due to radiation damage annealing but it is due fission atom annealing at higher temperatures
  - Kinetics of radiation damage annealing is rapid out-of-pile but may be significant in-pile (dynamic defect balance)

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Microstructure Studies

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Fuel Microstructure and Gas Localization Studies

- Evolution with burn-up and T(t) history "seen" by the specimen
- Has a direct relevance to:
  - Fuel behavior, in general
  - FGR, in particular
- Two fuel types examined
  - Isothermally irradiated fuel discs (IFA-569)
  - LWR irradiated fuel specimens
- Optical microscopy and quantitative SEM and TEM

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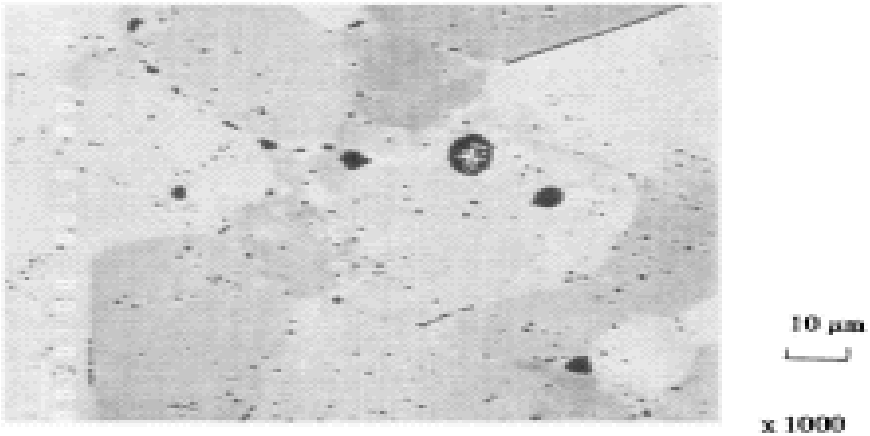
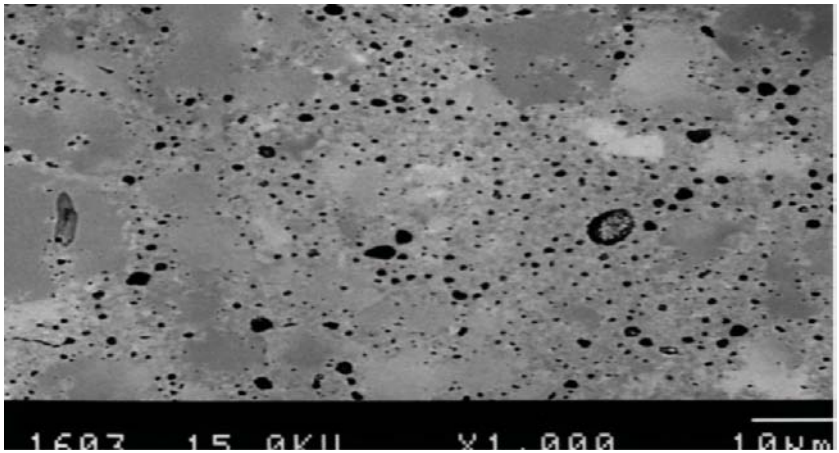
As-irradiated Fuel Discs

- Optical
  - With increase in burn-up some inter-granular porosity
- SEM
  - Grain boundaries essentially free of bubbles
  - Sub-micron porosity, mainly intra-granular, ~ 1 μm bubbles
- TEM
  - Maximum ~7 nm bubbles and ~ 5 nm precipitates in U2
  - Similar observations but with increasing number densities in U4 – U6
  - U8 had fully developed HBS, especially near outer rim of the disc

SEM of U6 at 0.6 mm from disc rim



SEM of U8 at 1.44 mm from disc rim



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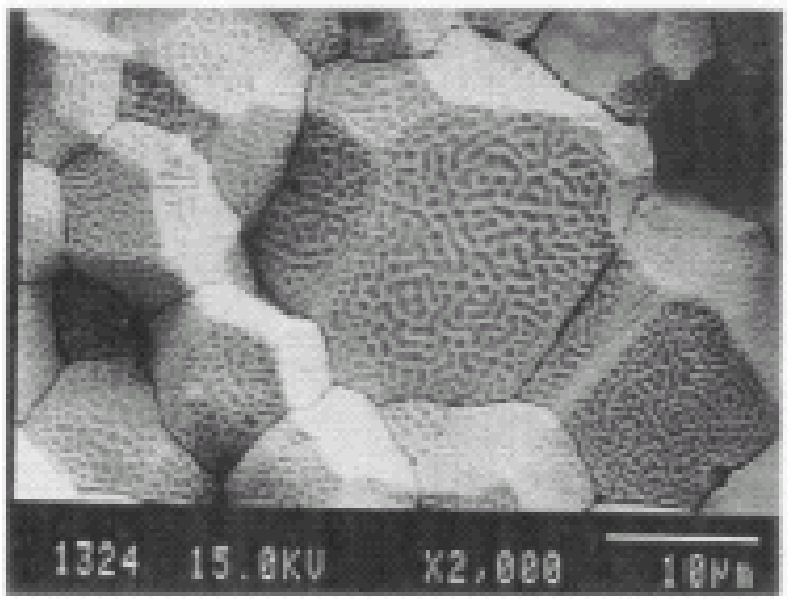
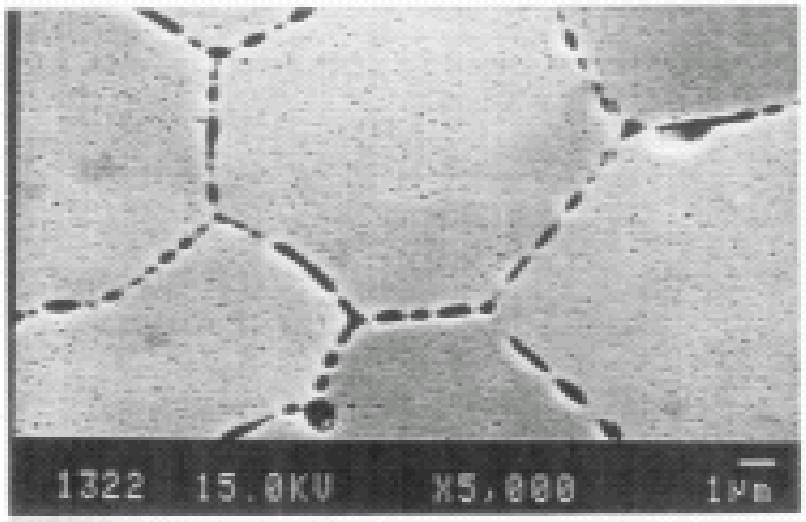
Post-annealing — Medium Burn-up Fuel Discs

Intergranular porosity in post-TD U4



Grain boundary interlinkage in post-TD U4



Centre

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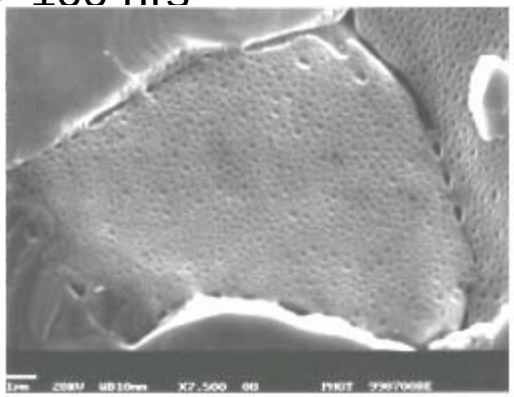
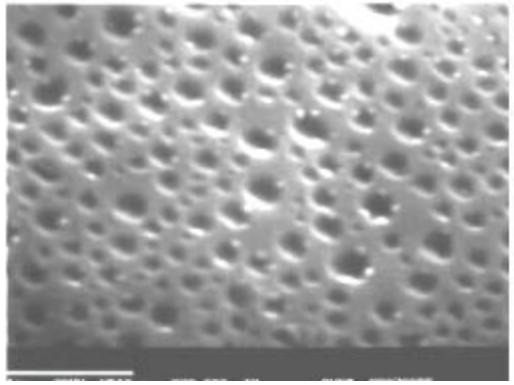
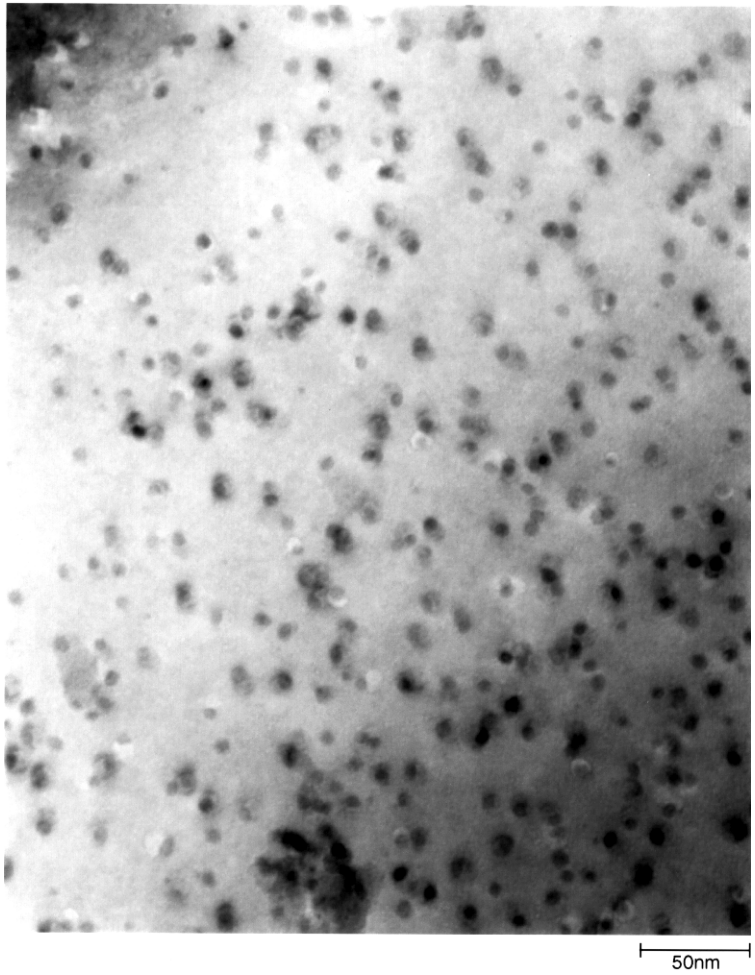
Medium Burn-up Fuel — Time Holds above 1200ºC

- IT7→ 4 hrs (similar to)
- IT8→ very short duration
- IT9→ 100 hrs







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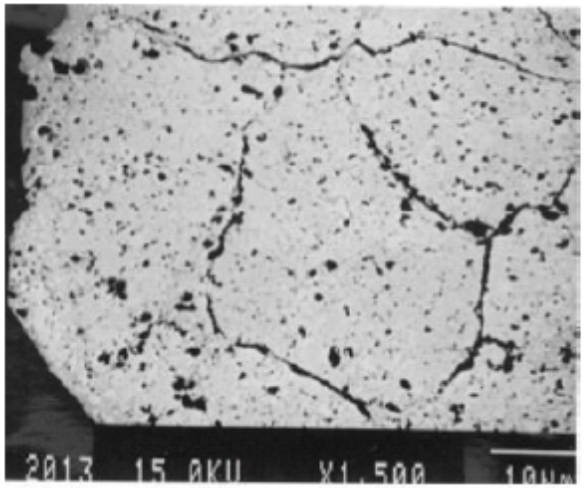
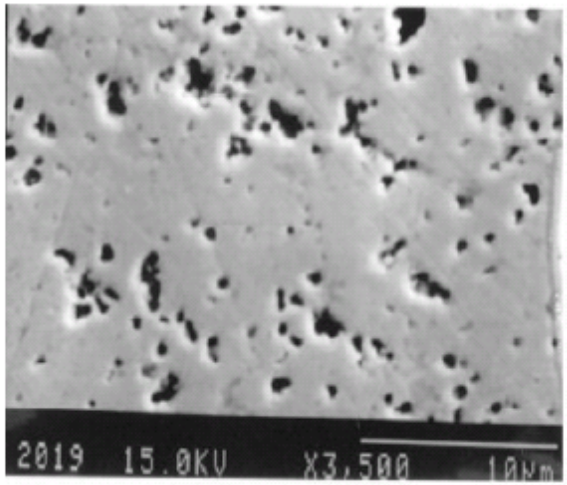
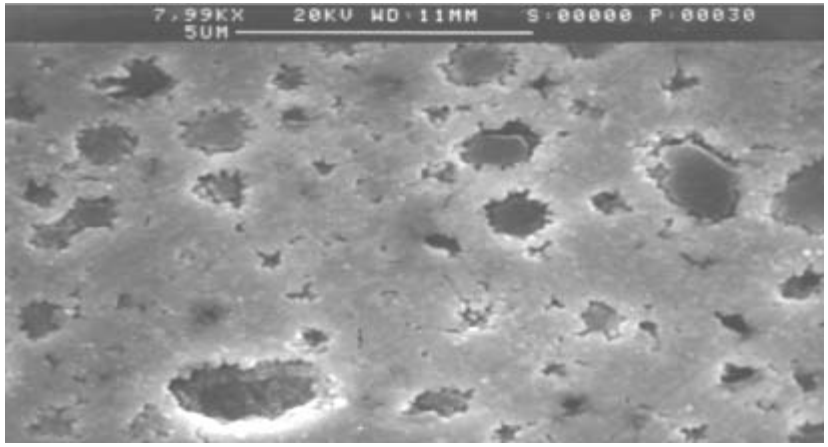
Highest Burn-up Disc Fuel

As-irradiated





Heated to 800ºC



- When heated as unrestrained sample:
  - Tended to fragment/disperse when heated above 800ºC
  - Fragment size ranged from several hundred microns to a few microns
- Sub-micron bubbles in HBS expanding and fracturing the surrounding fuel

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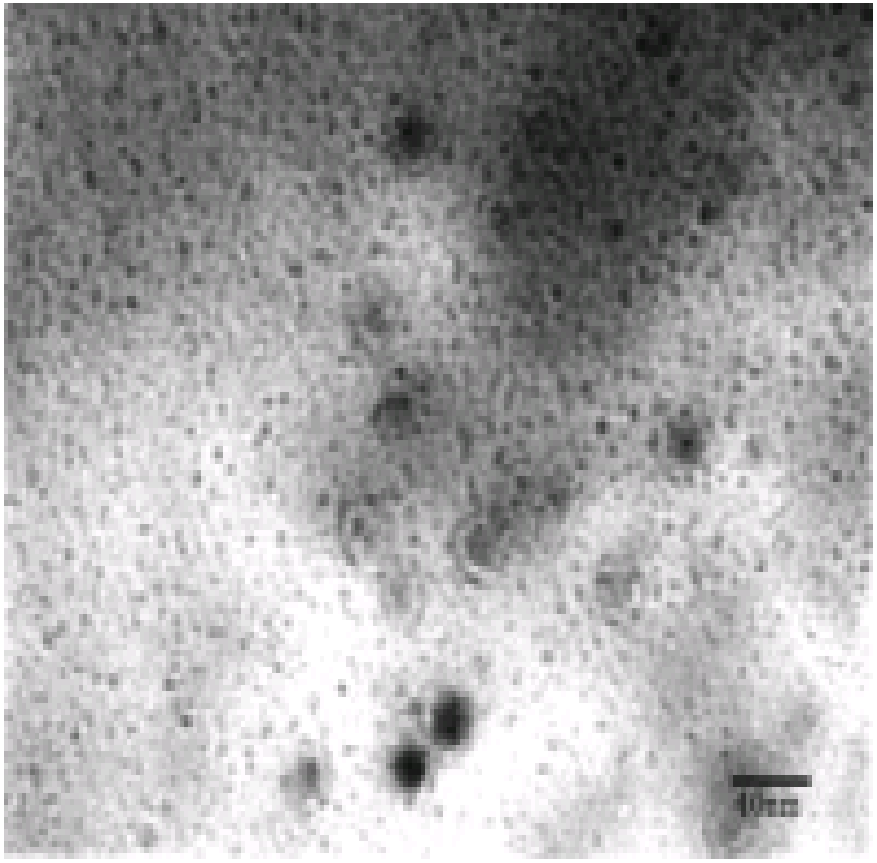
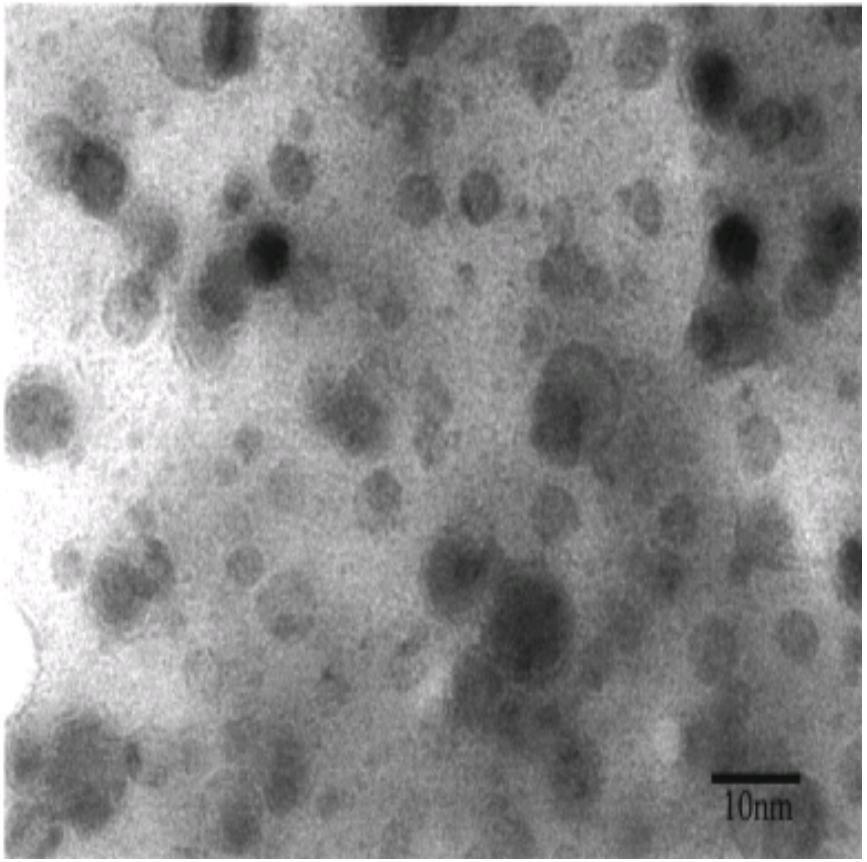
TEM of LWR Fuel at ~ 60 GWd/t Burn-up



As-irradiated; mid-radius



Ramped to 23 kW/m; Max temp. 1130ºC

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Fuel Fragmentation/Dispersal Studies

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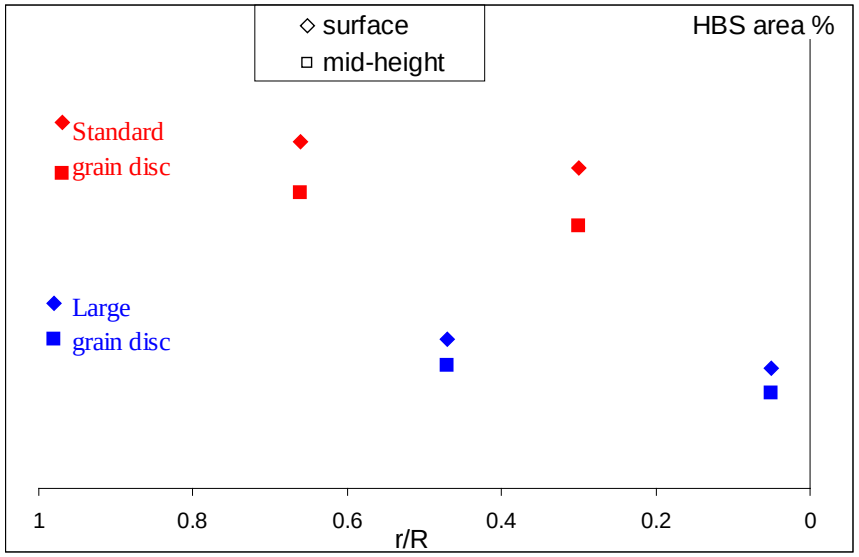
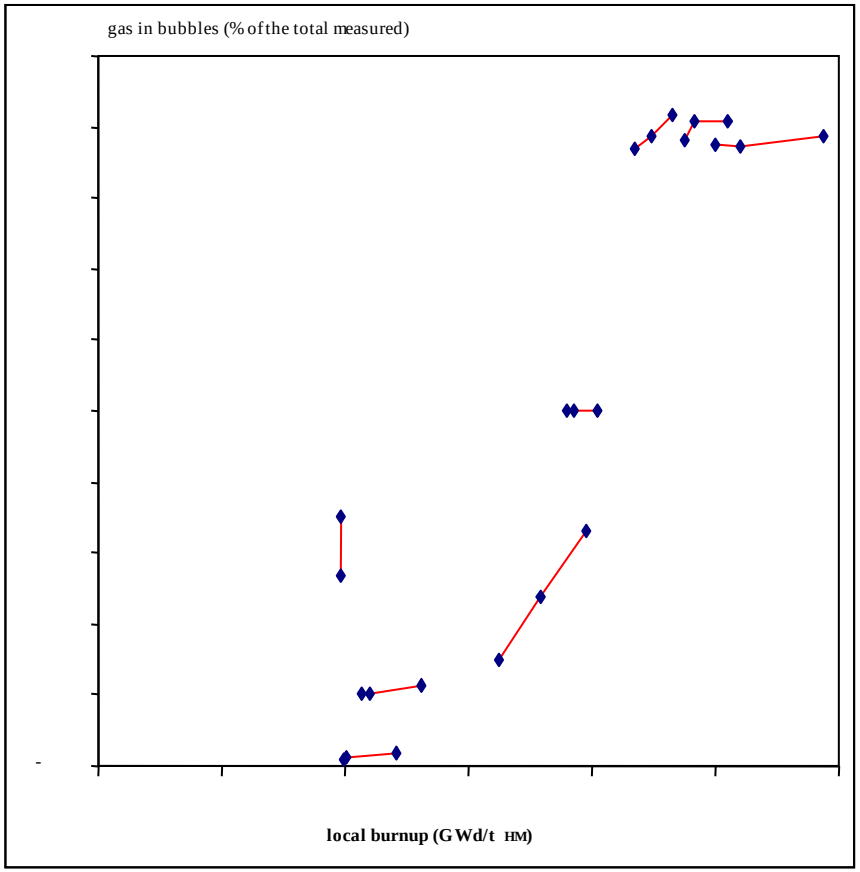
Detailed HBS Characterization

Grain size dependence



Fission gas content



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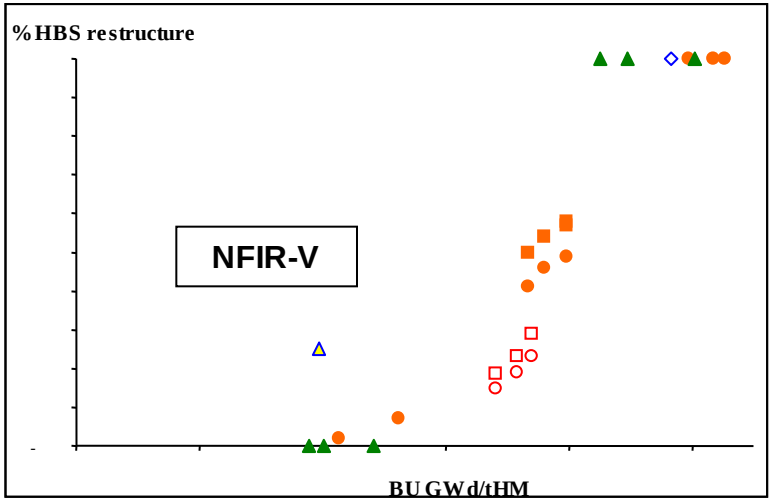
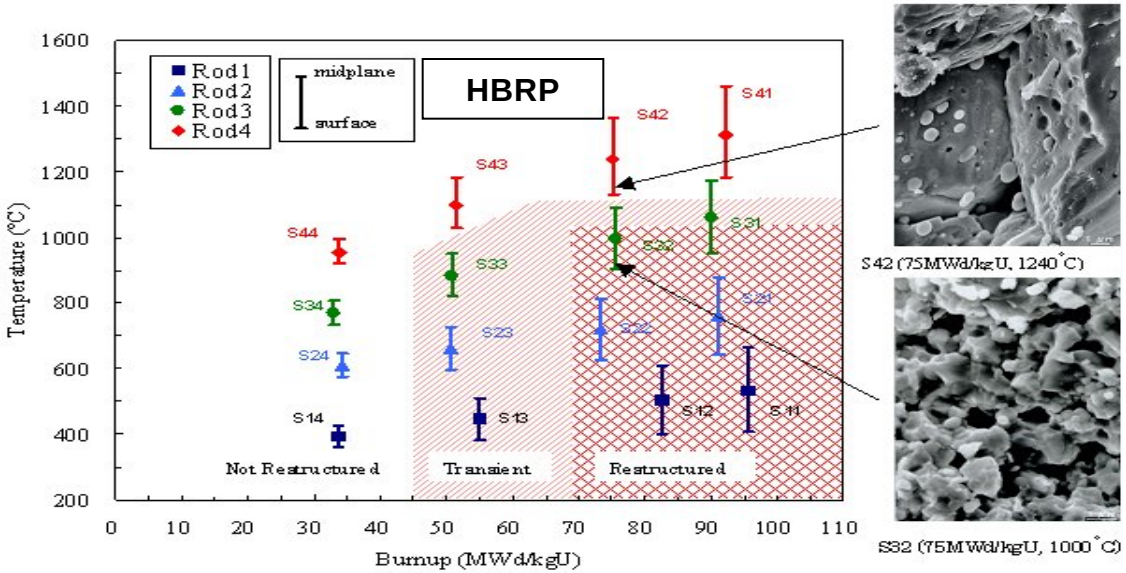
Onset of HBS Formation as a Function of Burn-up



- Fuel fragmentation is linked to the presence of HBS;
- A high concentration of over-pressurized gas bubbles in HBS tends to fragment the fuel in response to temperature transients (e.g., in LOCA and RIA)



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MOX Fuel

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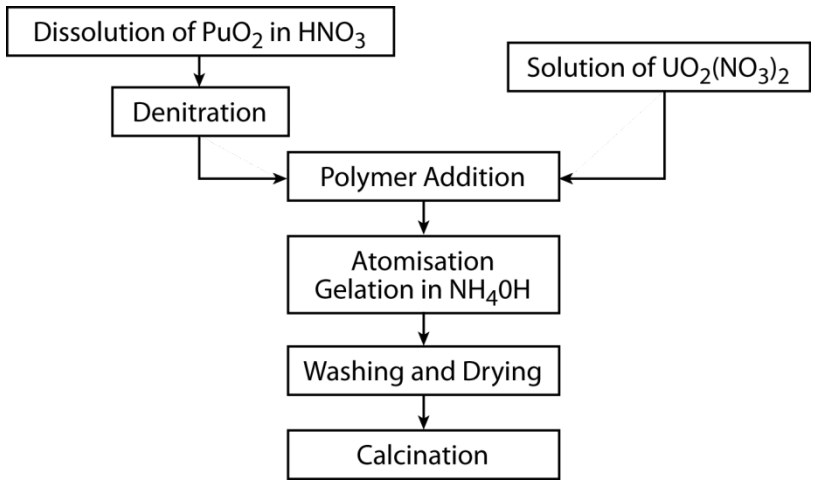
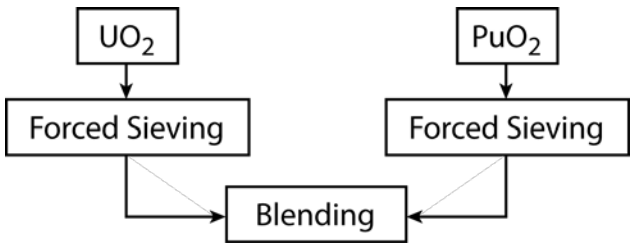
Fabrication of MOX variants

MOX-1



MOX-2



- MOX-1 by sol-gel process; MOX-2 by direct blending of powders
- Pu content in MOX-1 is 14.13 wt% and that in MOX-2 was 13.84 wt%.
- Pellets fabricated by standard pressing, sintering, and grinding steps; Thermally stable; 96% TD
- Cut into discs 8.2 mm dia; 1 mm thick

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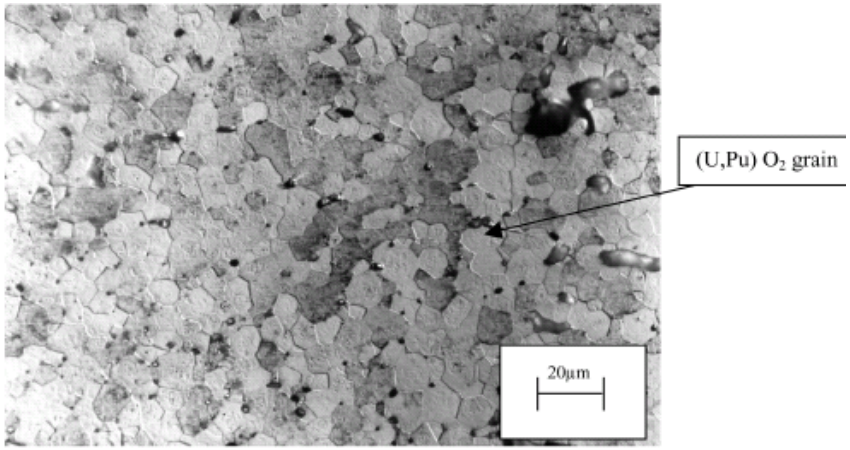
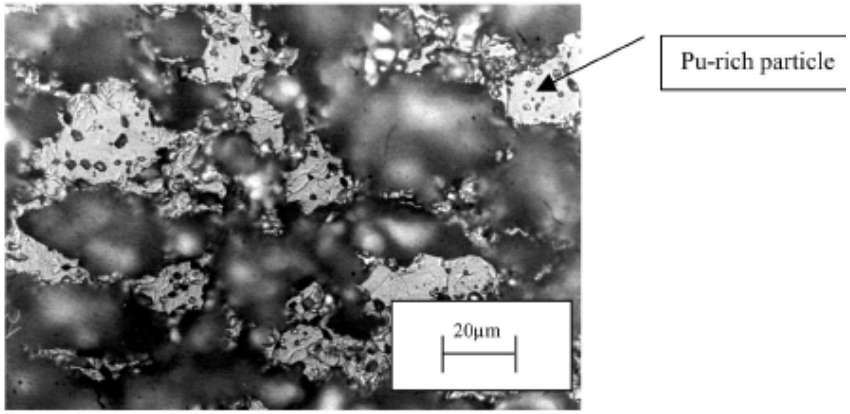
Microstructure

MOX-1



Pu-etched

MOX-2



Pu-etched

- MOX-1 grain size 10.9 µm.
- MOX-2 grain size: 5.2 µm (in U-rich phase) and 8.3 µm (in Pu-rich phase)
- Average size of Pu-rich agglomerates in MOX-2 is ~ 40 µm

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Homogeneous and Heterogeneous MOX Microstructure

- Since currently manufactured commercial MOX fuel is either MIMAS (micronized masterblend) or SBR (short binderless route):
  - Our experimental results do not *directly* apply to commercial MOX fuels but provide critical insights on MOX fuel performance
- Our results represent limiting MOX behavior and underscore the importance of initial microstructure, the plutonium agglomerate size, inter-agglomerate spacing, and fuel matrix plutonium content
- MOX-1 and MOX-2 are, in general, more homogeneous and less homogeneous, respectively, than the commercial MOX fuel. They were especially fabricated to:
  - Achieve the goals of this investigation
  - Continue further investigations through advanced fuel PIEs (NFIR) and further irradiations (HRP)

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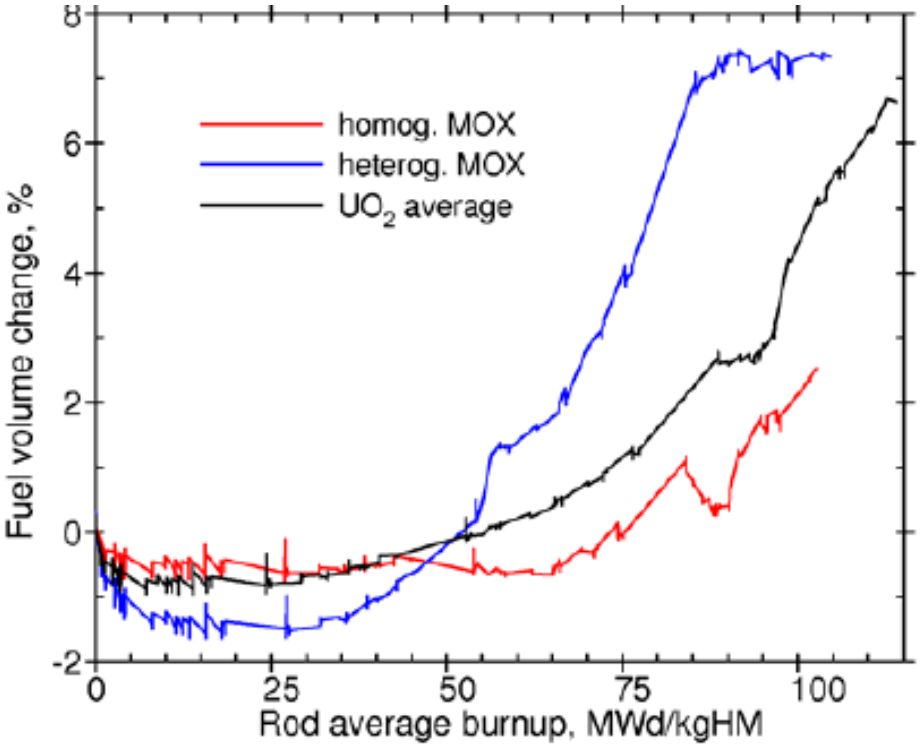
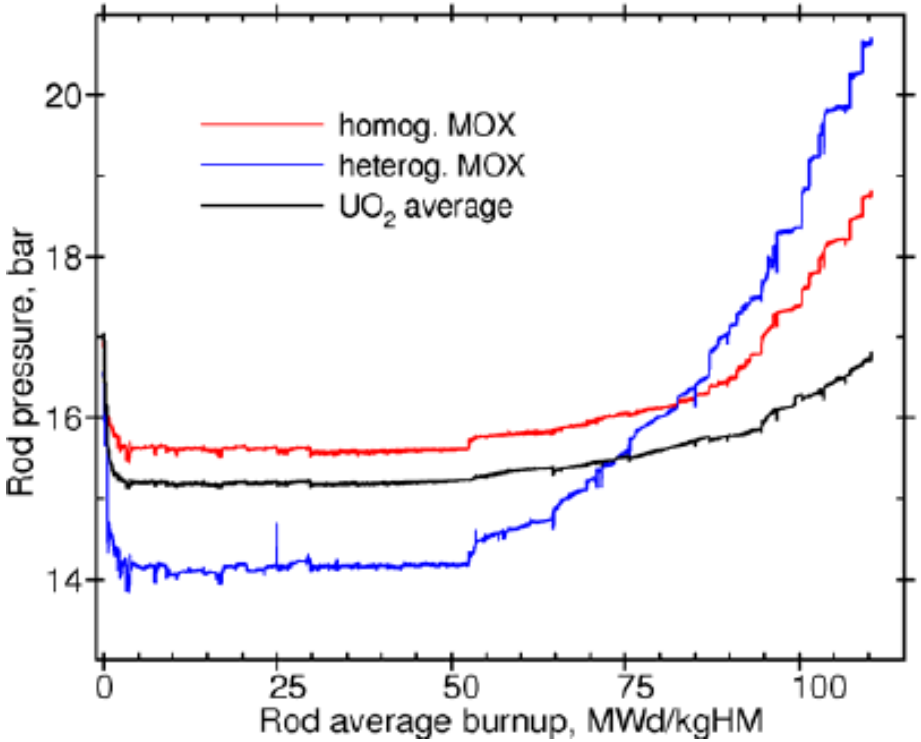
Results: Densification, swelling, and FGR in IFA-655





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Key observations on MOX

- Diffusion coefficient (tentatively) and FGR trend:

$$UO_2 \text{ fuel} < \text{MOX-1} < \text{MOX-2}$$

- Increased S/V at higher burnups in rods with gas lines, together with the corresponding increases in rod internal pressure in sealed rods
- These observations can be attributed to a combination of
  - More concentrated fissions and early formation of HBS in the large Pu oxide clusters where the locally high burnup occurs
  - Enhanced formation of open porosity within these clusters, due to interlinkage of HBS pores, or enhanced thermal or athermal diffusive release from small HBS grains

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Future R&D Needs

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Fuel Mechanical Properties and Creep

- Visco-plastic deformation behavior of fuel and the knowledge of concurrent mechanisms involved are the main needs PCMI behavior
- Out-of Pile approaches in various stages of development
  - Micro-indentation and acoustic methods
- Actual in-pile fuel mechanical behavior will need to be modeled:
  - Interaction of fractured ceramic (the pellet) in a metallic tube (the cladding) after strong PCMI has been established
- Underlying key question will be:
  - Relative significance of thermal and irradiation creep of the pellet

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Some Other Examples

- Threshold for fuel fragmentation/dispersal
  - Regulatory focus in RIA and LOCA events
  - Can one eliminate or delay HBS formation through fuel additives and grain size controls
- In-pile recovery kinetics
  - Role of radiation damage and fission gas bubble nucleation
  - Any concerns due to stored heat
- Fission gas localization and release
  - Large uncertainties in prediction yet important in fuel rod design margins
- Fuel mechanical behavior and interaction with cladding
- MD Modeling

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Conclusions

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Conclusions

- For fuel pellet behavior in a fuel rod temperature and microstructure are the co-pilots. They are highly dependent on one-another.
- The main approaches to licensing and burn-up extension issues in LWR fuel are:
  - PIE of as-irradiated fuel discs;
  - In-pile testing and measurements on instrumented fuel rodlets;
  - Experience-base from in-LWR fuel rod performance.
- Fuel (UO_2) R&D is often time consuming and expensive, with severely limited number of hot cell lab who can perform it. It is best take an international collaborative approach.
- Ultimate future goal: Full predictive capabilities validated to high burn-up UO_2 fuel system.

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