Better Materials for Nuclear Energy

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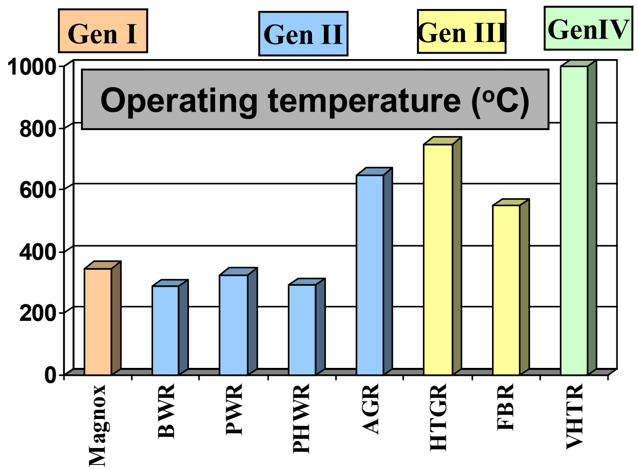
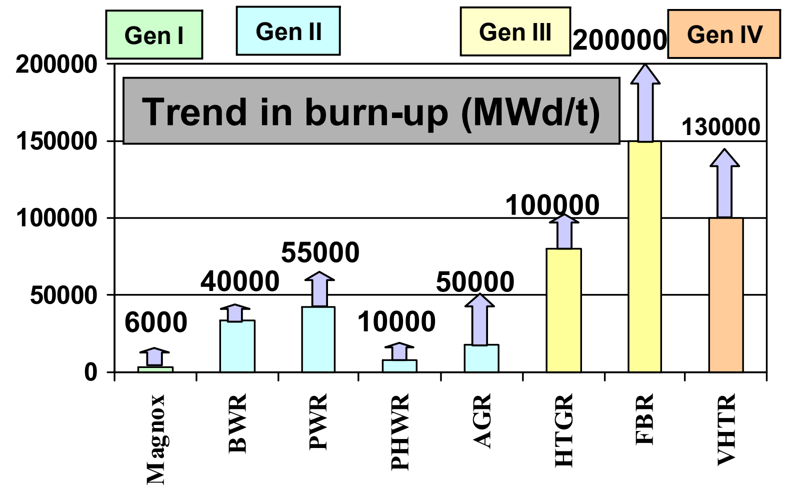
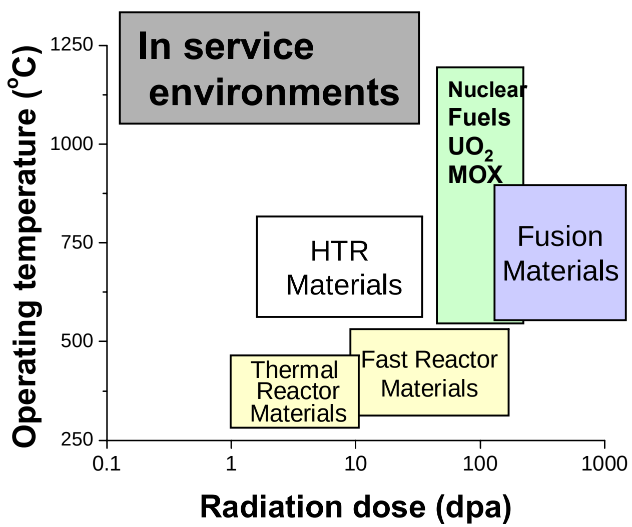
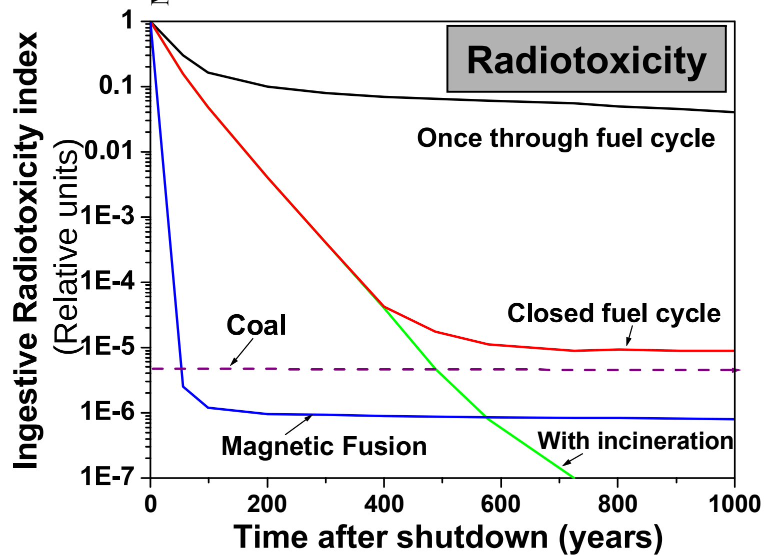
Increasing demands on materials









Pushing the burn-up

- Innovative fuel cycle for utilizing larger nuclear energy potential
- Increase in burn-up in a single cycle

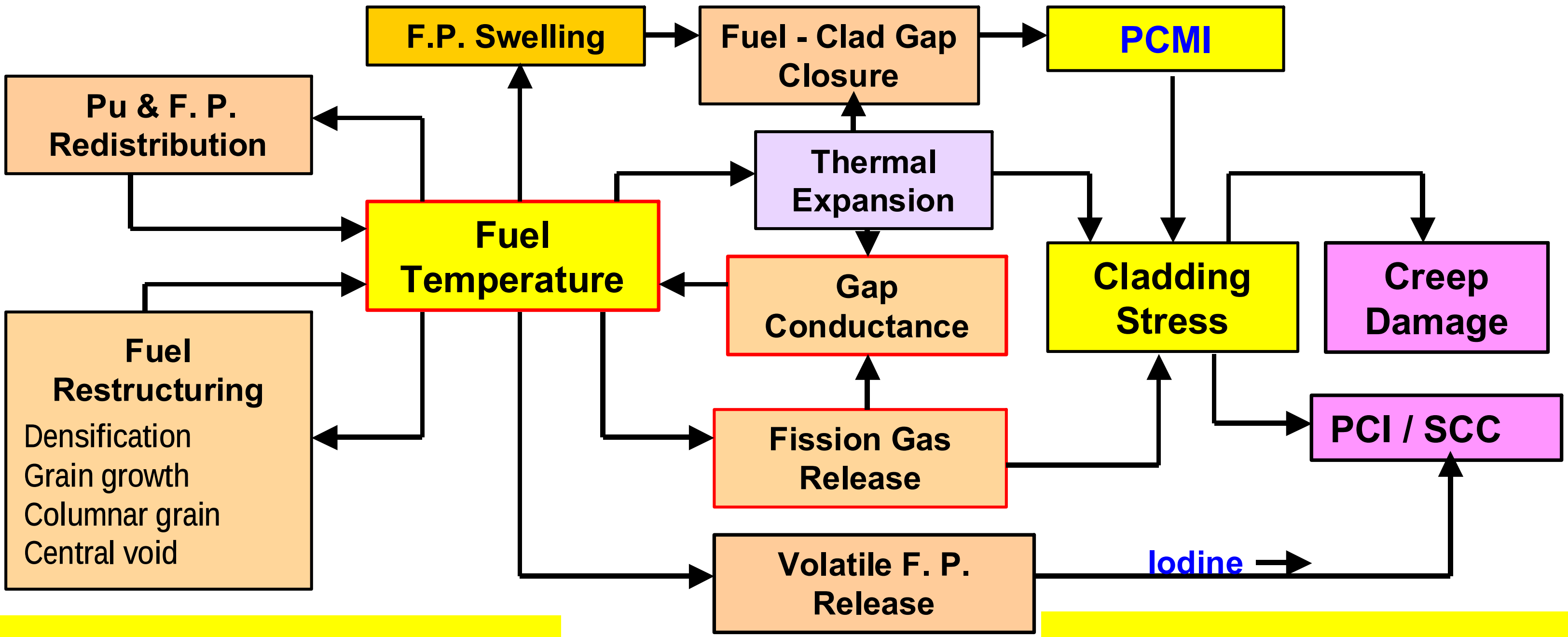
Fuel

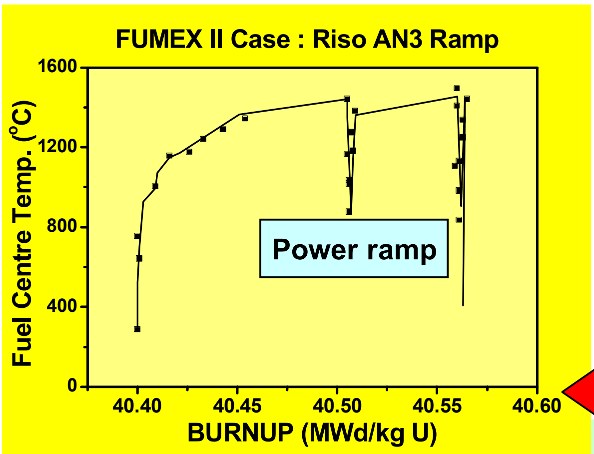
- Fuel restructuring
- Fission gas release
- Fuel clad interactions
- Reactivity Control

Structural materials

- Radiation damage
  - Dimensional stability
  - Property degradation
- Clad - coolant compatibility

Interactive Phenomena Operating in Fuel during Irradiation

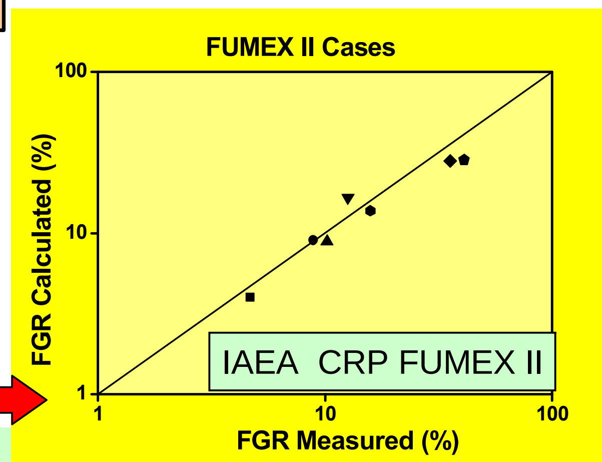




Fuel temperature validation

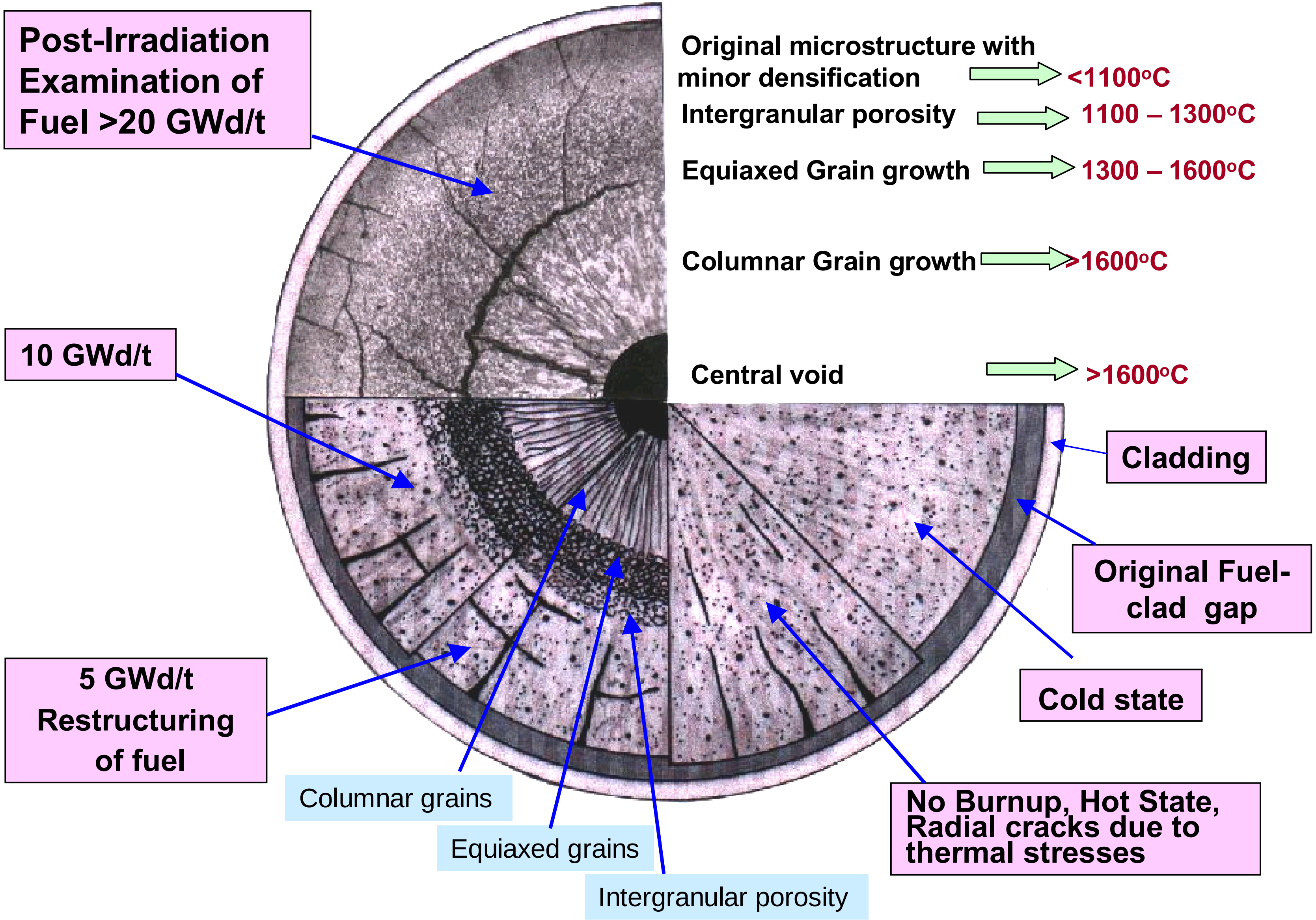
PROFESS

BARC Code Developed & Validated

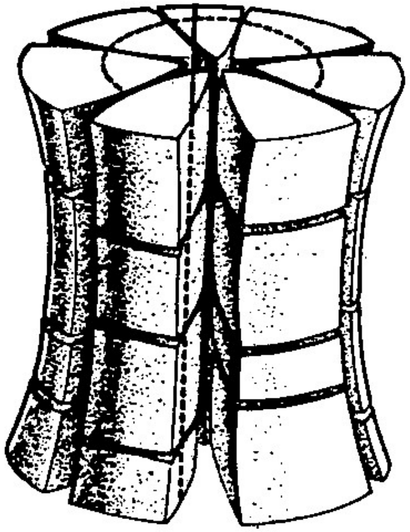


Fission gas release validation

Microstructural Evolution in Oxide Fuel

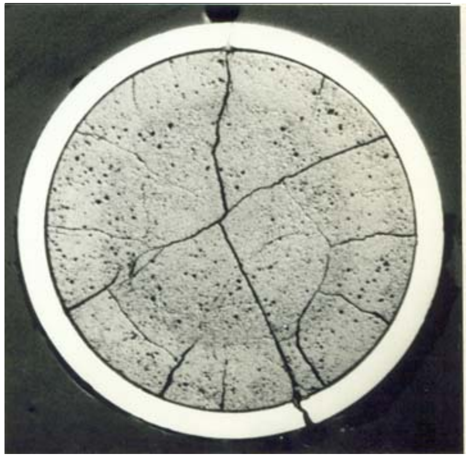


Pellet Clad Interaction at High Burnup

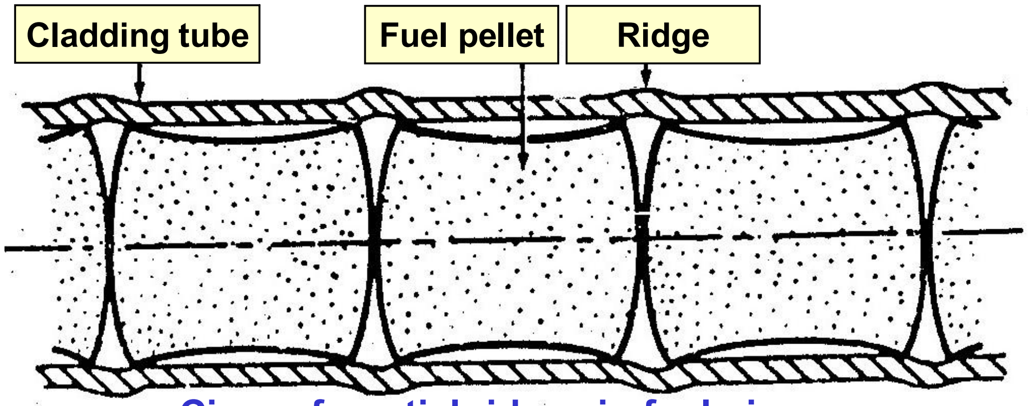


Hour-glassing of fuel pellet due to radial thermal gradient

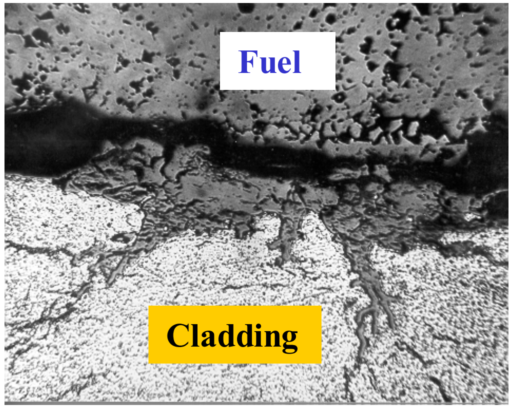
Protection against PCI/SCC failure is required for pushing the fuel burnup.



PCI/SCC Failure



Circumferential ridges in fuel pins



Incipient PCI/SCC cracks

Fission Gas Release

Athermal Release

- From pellet surface by recoil and knock out

Diffusional Release

(Equivalent Sphere Model with re-solution)

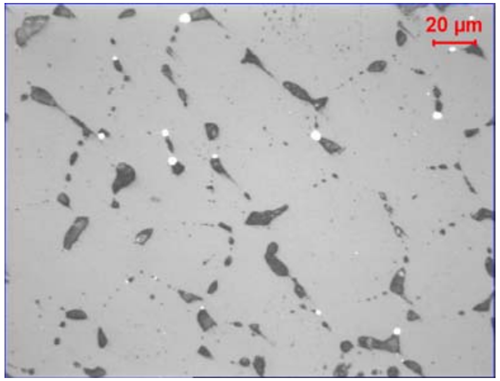
$$(\partial c/\partial t) \quad = \quad D\nabla^2 c \quad - \quad gc \quad + \quad bm \quad + \quad \beta$$

Gas conc. in grain	Diffusion to grain boundaries	Capture by traps.	Re-solution from bubbles	Generation by fission

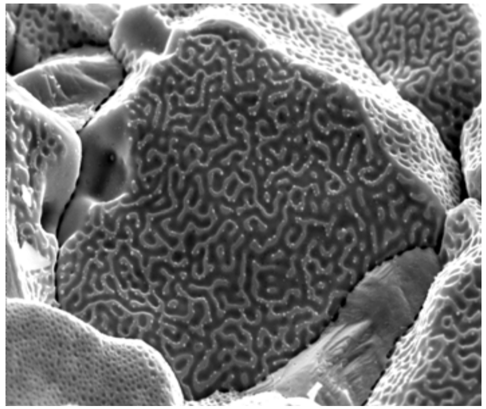
- Gas atom migration to grain boundaries

Gas atom collection at grain boundaries

- Grain boundary sweeping accumulates fission gas to reach early saturation



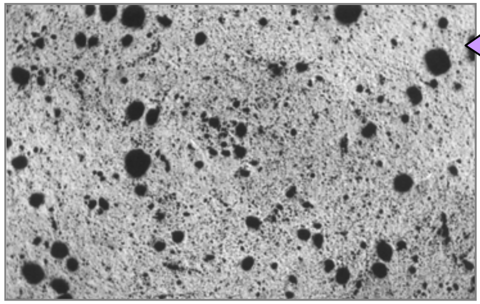
Nucleation and growth of gas bubbles on grain boundaries



Interconnected channels of gas bubbles at grain faces

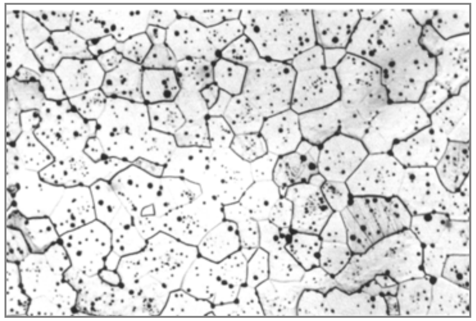
Strategies for Improving Fuel Performance

- **Large grain size:** reduces the fission gas release
- **Stable porosity structure:** avoids densification
- **Improved fuel design:** reduces the heat rating
- **Fuel-clad barrier layer:** pure Zr, graphite

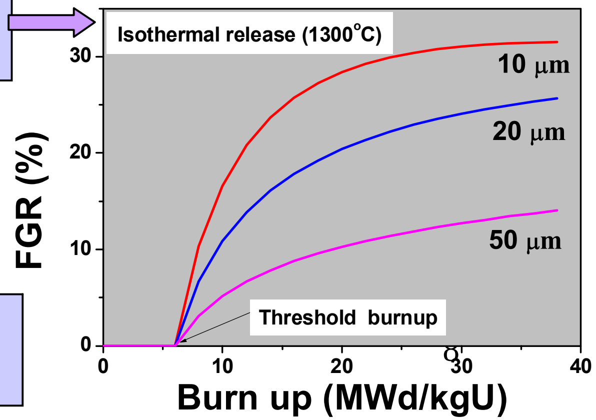


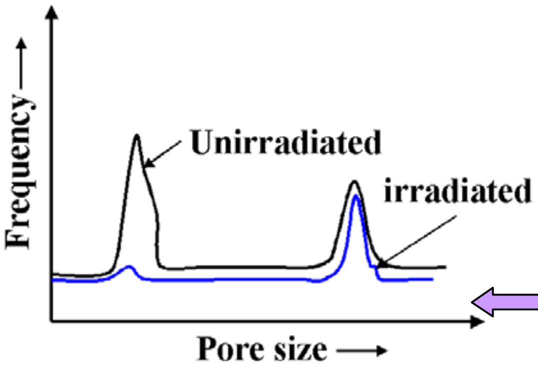
Controlled porosity pellet for better thermal performance

Large grain size pellet to reduce fission gas release



Effect of grain size on fission gas release





Pore size distribution in UO_2 before and after irradiation

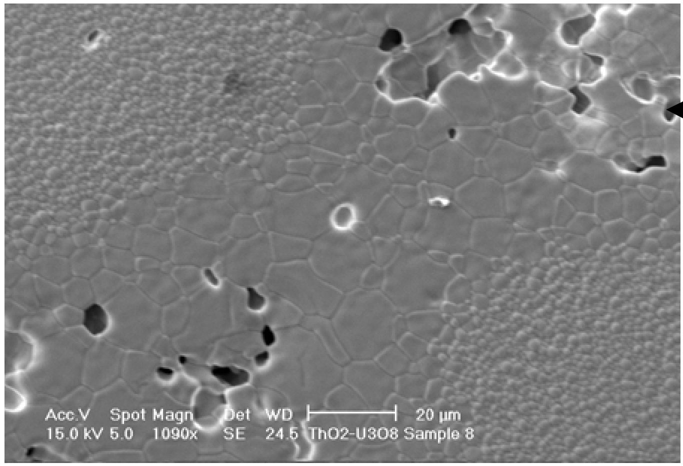
High Performance MOX Fuel Microstructure

Performance Requirement for High Burn-up Fuels:

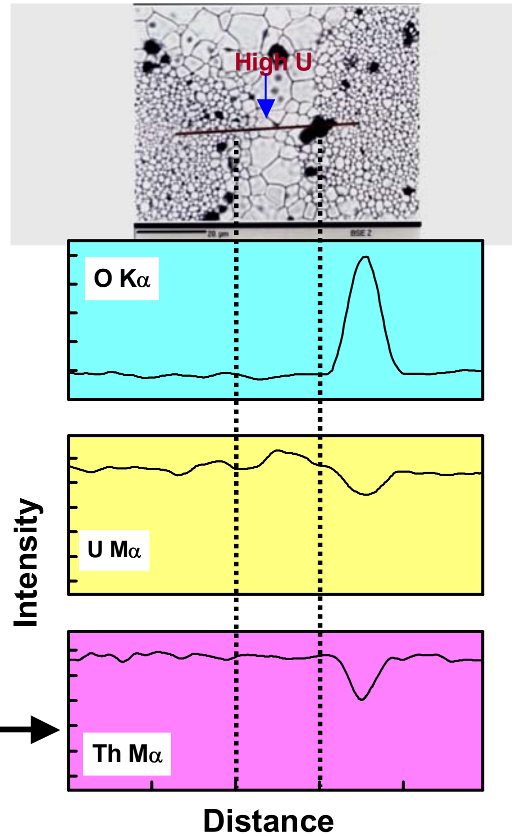
1. **"Soft pellets" – To reduce PCMI.**
2. **Large grain size - To reduce FGR.**

Suggested Microstructure - Rock in Sand

A hybrid of islands of fine grains (Fertile rich) to give plasticity and large grains (Fissile rich) to reduce FGR. CAP process being developed for (Th-U^{233}) MOX Appears to achieve this.



Microstructure of $(Th,U)O_2$



Elemental scan for U Mα, Th Mα, and O Kα for $(Th-U)O_2$ pellet

In-service Degradation of Structural Materials

Radiation Induced	Environment Induced
• Mechanical Prop: Hardening, Embrittelement	• Chemical Interaction with coolant & fission products
• Dimensional Changes: Creep, Growth	• Hydrogen Damage
• Segregation: Precipitation, Agglomeration	• Introduction of New Corrosion Modes (Irradiation Assisted Stress Corrosion Cracking)
• Transmutation: He Embrittelement (n,α)	

Hydrogen Damage in PHWR Pressure Tubes

Hydrogen Embrittelement

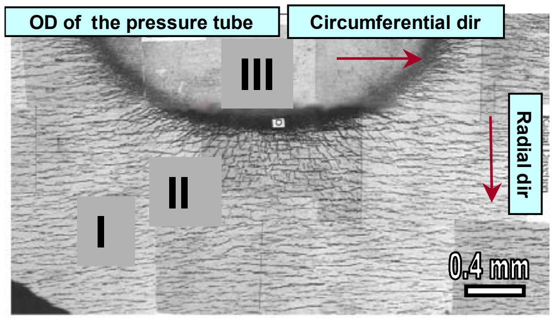
- Radial-Axial Hydride Lowers Frac. Toughness
- Restriction on Cold Pressurization
- Remedy: Hydrogen Getters (Yttrium, Bulk Met Glass)

Delayed Hydride Cracking

- Hydrogen migration under stress gradient
- Zero Clearance Rolled Joint
- Optimization of microstructure & texture for increasing K_{IH} & reducing DHC Velocity

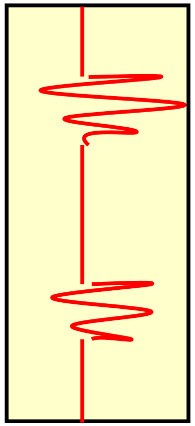
Hydride Blistering

- Hydrogen migration under temperature gradient
- Avoid PT-CT contact

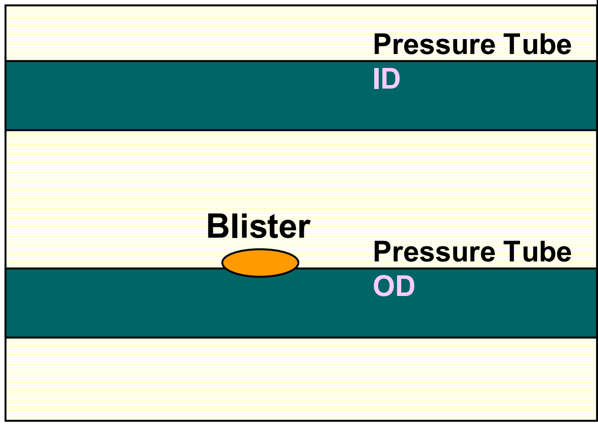


Hydride Blister

A-scan



B-scan Image



INSIGHT Jl., 1998

Modulus and Density difference between α_{Zr} & δ-hydride leads to Longitudinal (V^L) & Shear (V^S) velocity differences

$V^L_{\alpha\text{-}Zr}$ = 4750 m/s
$V^L_{\delta\text{-hydride}}$ = 5400 m/s
$V^S_{\alpha\text{-}Zr}$ = 2350 m/s
$V^S_{\delta\text{-hydride}}$ = 1900 m/s

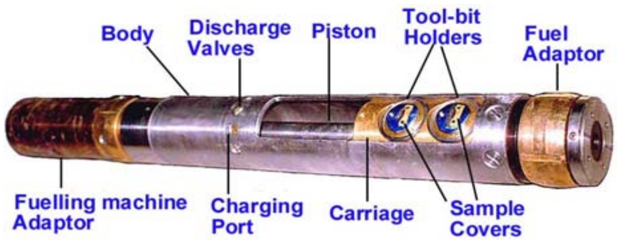
Life Management of PHWR Pressure Tubes

In-Service Inspection | **Scraping Tool** | **Material Surveillance**

BARC Channel Inspection System (BARCIS)

3 Ultrasonic Testing Probes: Flaw Detection in PT
PT Wall Thickness

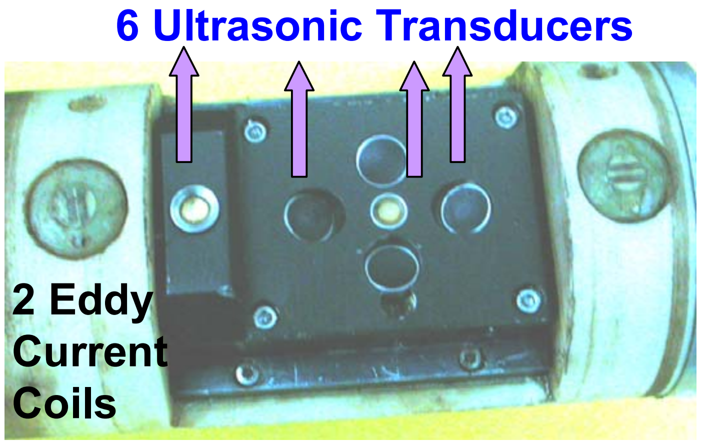
3 Eddy Current Coils: ID Flaw, GS Location, PT-CT Gap



BARC Tool Sliver Sample for H Analysis

Periodic removal of pressure tube to determine
- **Fracture toughness**
- **DHC velocity**
- **Deuterium Pick-up**

Non-Destructive Examination for Structural Integrity Assessment of PHWR Pressure Tube

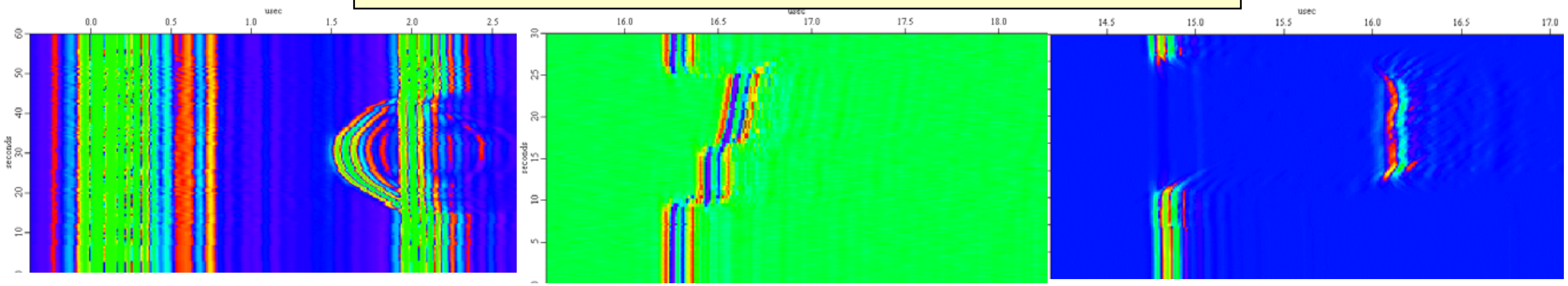


Inspection Head

- Validation of Existing NDE Techniques
- Sizing and imaging of flaws by ultrasonic time-of-flight technique

IAEA CRP on Pressure Tube Inspection & Diagnostics

Ultrasonic Images of simulated flaws



Delayed Hydride Cracking

Bearing Pad Fret

Debris Fret

Improvements in Fabrication Flow Sheet

Pressure Tube

- **Quadruple Melting** → Control on H (<5ppm), Cl (<0.5 ppm) & P (<10 ppm) to improve fracture toughness
- **Pilgering** → Better Microstructure Control, Surface Finish & Dimensional Control

Calandria Tube

- Seamless for homogenous microstructure, texture, properties & avoiding weld defects
- Seamless tubes qualified for irradiation growth

Specimen	Growth Strain (10^{-4})
Seamless Longitudinal	4.70
Seam welded Long.	4.78
Seamless Transverse	2.78
Seam Welded Transverse	3.89

Improvements in Fabrication Flow Sheet

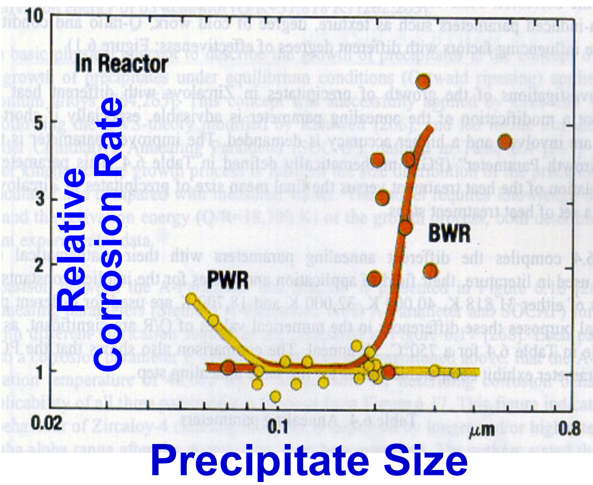
Fuel Tube

Spot Welding of Bearing Pad & Spacers instead of Brazing

- Minimize HAZ
- Avoid Be Handling

Control of Intermetallic Precipitate Size

- Achieved by controlling Cumulative Annealing Parameter (CAP)



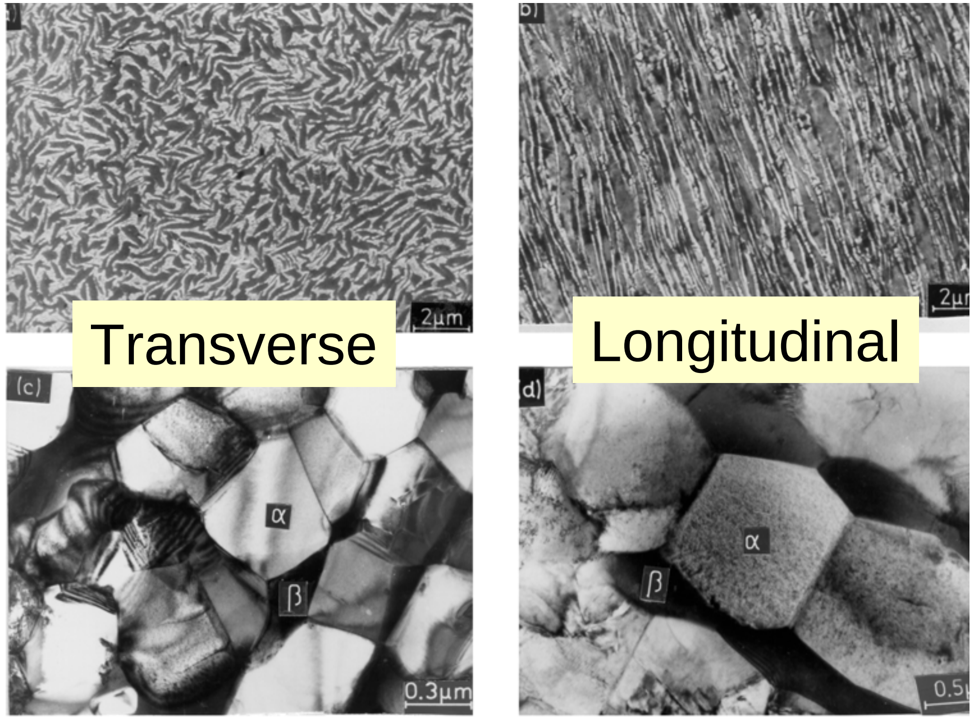
Reactor Type	Corrosion Concern	Desired Particle Size	CAP (Hours)
PWR/PHWR Reducing environ.	Uniform	> 0.1 µm	2 $\times 10^{-18}$ – 5×10^{-17}
BWR Oxidizing environ	Nodular	< 0.15 µm	$\leq 10^{-18}$

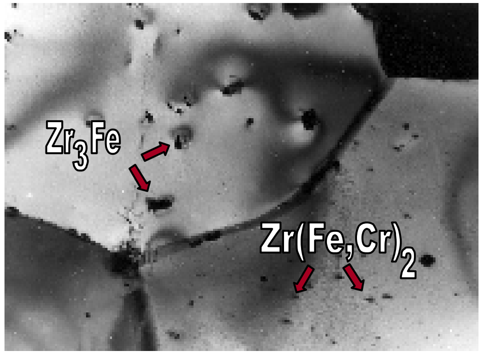
Microstructure of PHWR Components

Pressure Tube

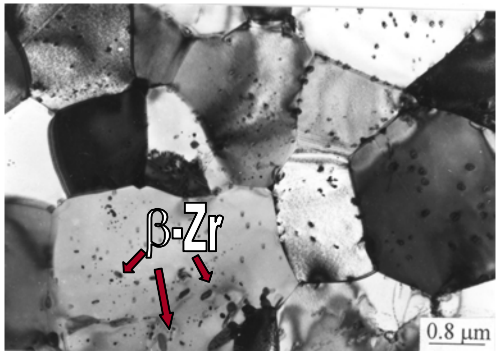
Zr-2.5Nb extruded

Cladding tube





Zircaloy 4



Zr-1Nb

Two phase (α–matrix + β-Zr stringers)

Future Directions in Zr Alloy Development

Approach

Optimization of Existing Alloys

- Chemical Composition
- Microstructure
- Texture

New Alloy Development

- Zr- Sn – Nb Alloys
- BCC Zr Alloys
- Isotopically Denatured Zr

Requirements

Clad Tubes

- Improved Corrosion resistance & residual ductility for higher Burn-up & partial boiling
- Increased hydrogen tolerance

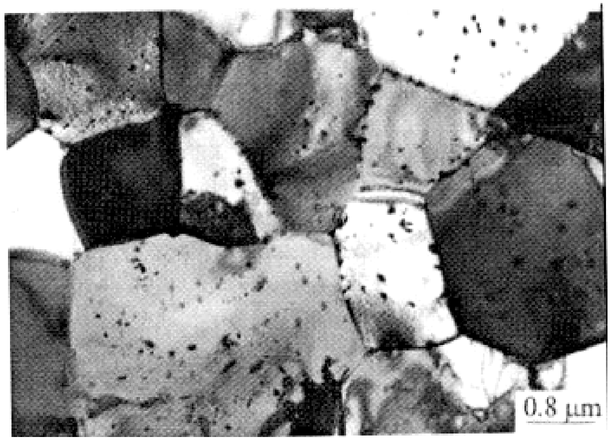
Pressure tubes

- Increased flaw tolerance
- Hydrogen immunity
- Improved creep resistance

Development of Zr - Sn - Nb - Fe Alloys

Primarily for Higher Fuel Burn-Up

- Higher Coolant Temperature
- Permits Higher Li Addition
- Lower Irradiation Creep & Growth
- Higher Corrosion Resistance
- Lower Hydrogen Pick-up



Trans IIM, 2004

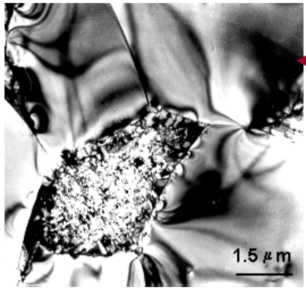
- ZIRLO (Westinghouse) : Zr – 1 Sn – 1 Nb – 0.1 Fe
- Alloy 635 (Russian alloy) : Zr – 1.2 Sn – 1 Nb – 0.4 Fe

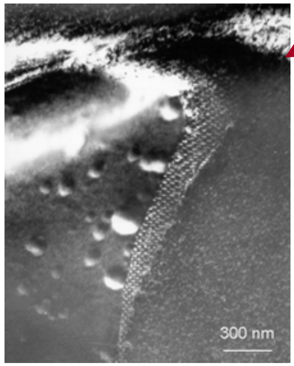
PIE after 70,000 MWD/Te : Good Performance of ZIRLO Clad

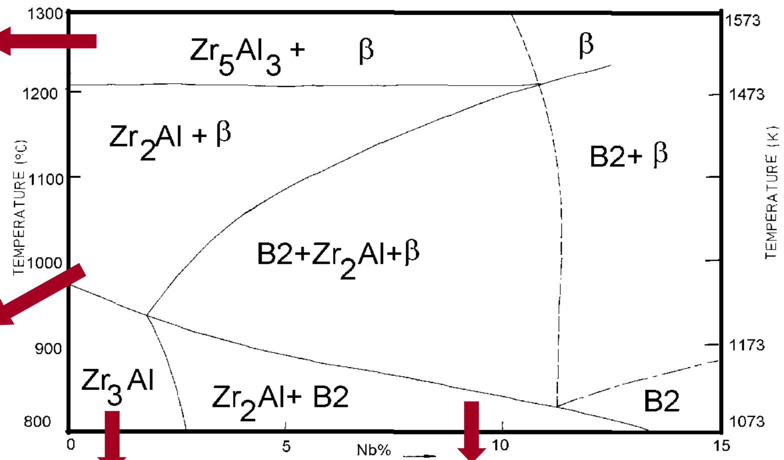
Zr alloys with bcc crystal structure

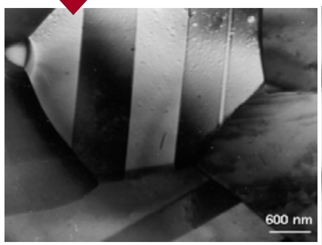
HCP has problems related with
- Anisotropy
- Growth
- Hydride

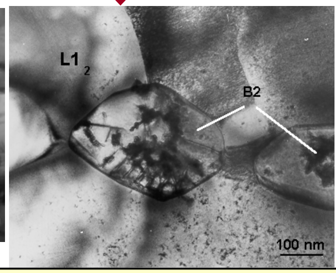
BCC Zr alloys:
- Isotropic
- Higher hydrogen solubility
- Omega embrittlement, Higher σ_a

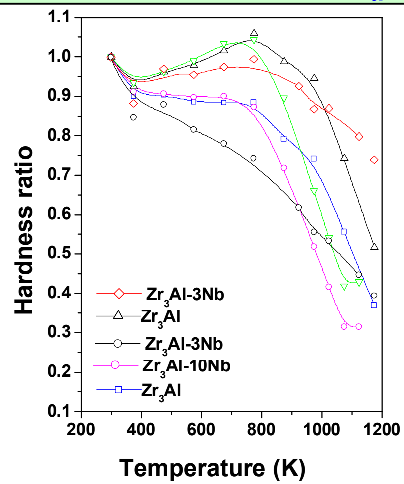


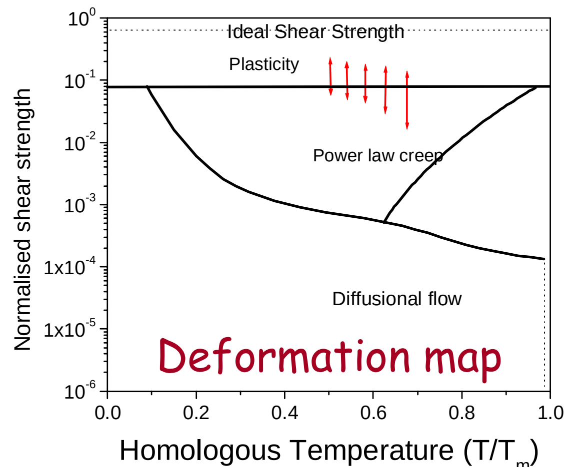










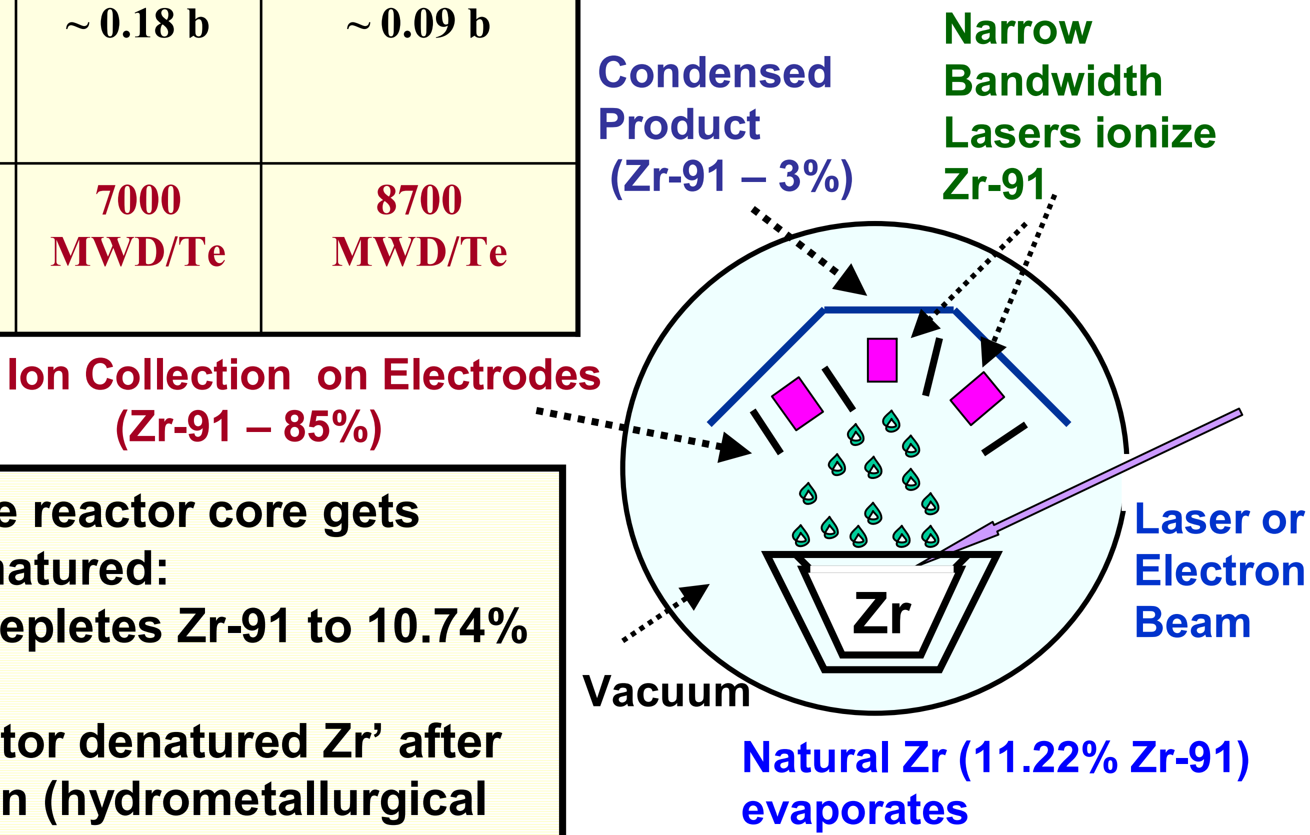


Oxidation properties comparable
Zircaloy-2: 6.8 mdd Zr_3Al-Nb: 8.9 mdd

Isotopically Denatured Zirconium

Composition of Zr	Natural Zr Zr-91 11.22%	Denatured Zr Zr-91 3%
Absorption Cross-section $\sigma_{Zr\text{-}91} \sim 1.24$ b	~ 0.18 b	~ 0.09 b
Burn-up in PHWR 23 Te Coolant Channel	7000 MWD/Te	8700 MWD/Te

Laser Separation



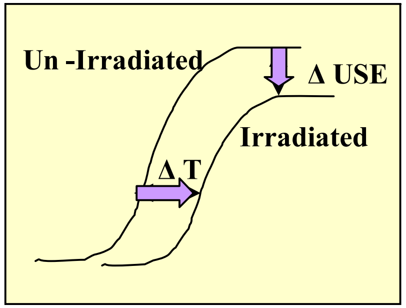
- Zirconium in the reactor core gets isotopically denatured:
  6×10^{21} n/cm^2 depletes Zr-91 to 10.74%

- Recycling 'reactor denatured Zr' after decontamination (hydrometallurgical process) will drastically reduce radioactive waste

Reactor Pressure Vessel Steel Embrittlement

Radiation Embrittlement

Energy

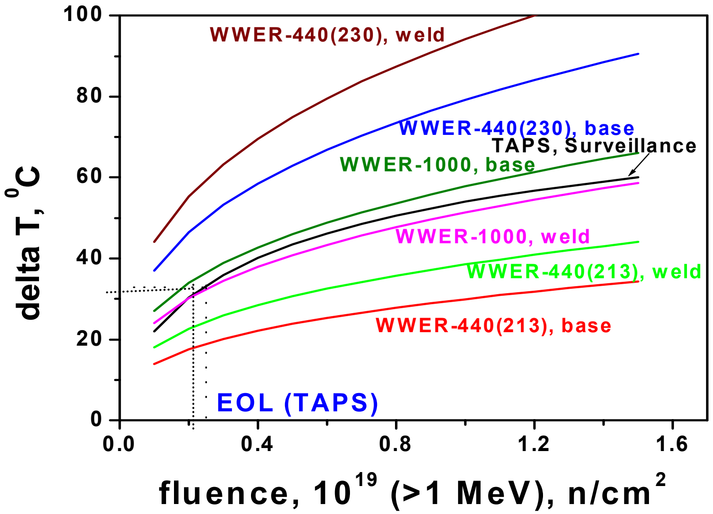


Temperature

Embrittlement Mechanisms	Remarks
Matrix damage	Lattice defects due to neutron bombardment
Precipitation effect	Ni, Mn, Cu, Si-enriched precipitates
Segregation effect	P,Ni, Si to dislocations, P at grain boundary

Major characteristics	Western	WWER 1000
Alloying elements for optimizing strength, toughness, weldability & hardenability	Mn, Mo	Cr, Mo, V, Ni
Elements causing irradiation embrittlement	Cu,P, Ni	Cu, P, Ni, Mn, Si
Predictive equation used for irradiation embrittlement $\Delta T = CF \times FF$ Compositional Fluence	CF = f(Cu,Ni) $FF = F^{0.28-0.01\log F}$	CF = f(Cu,P) = 20 for weld = 23 for base $FF = F^{0.33}$

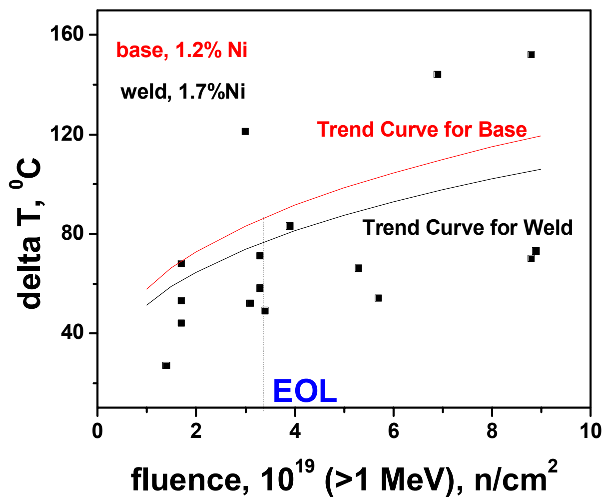
Intercomparison of Embrittelement Trends of RPVs of Different Origins



For TAPS, ΔT_{EOL} = 33°C (Surveillance)

= 73°C (Design Limit)

Intercomparison of Embrittelement Trends in WWER-1000 RPV



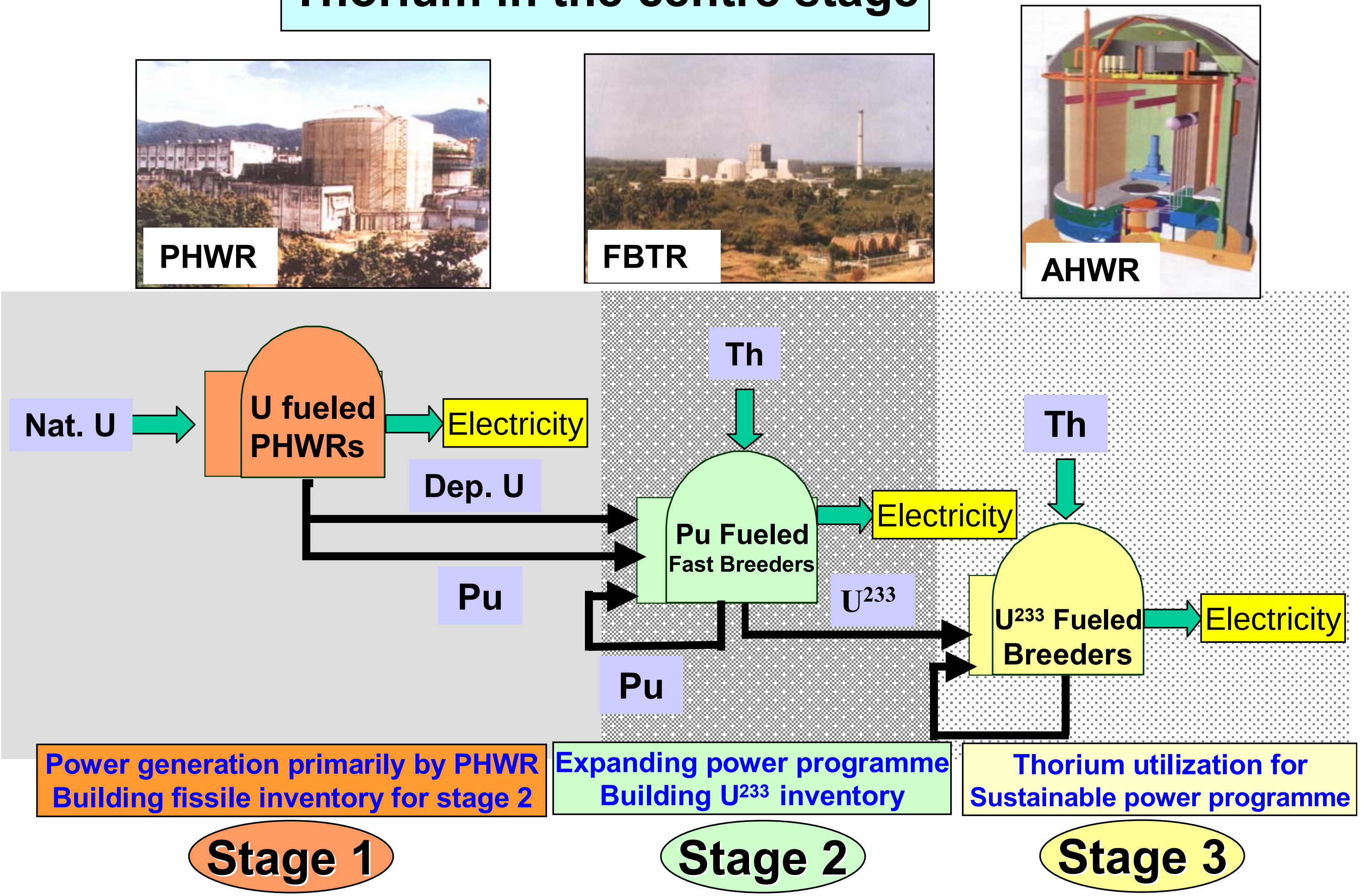
ΔT_{EOL} = 92°C (Design Limit)

Approach to Extend RPV Life

- Reduction in Ni
- Controlling Dual Presence of Mn & Ni
- Reduction of P, Cu, Si
- Lowering Non-Metallic Inclusions

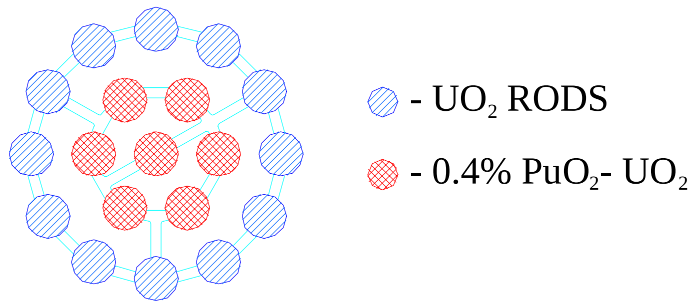
Three Stage Indian Nuclear Programme

Thorium in the centre stage



MOX fuel for PHWRs

- Uranium conservation and demonstration of high burn-up fuels for PHWRs.
- MOX fuel design and fabrication capability.
- Natural uranium savings (~40%)
- Lower volume of spent fuel storage/ reprocessing.
- 11,000 MWd/T achieved in MOX bundles



MOX Fuel Bundle for PHWR

Fuel subassembly inside glove box

Indian Fast Breeder Reactor Programme

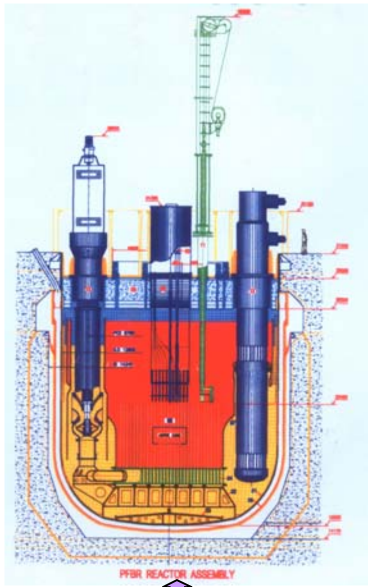
FBTR- 40 MW_{th} (loop type) with indigeneously developed mixed carbide fuel is in operation since 1985.

Construction of FBR- 500 MW_e (pool type) with MOX fuel has started in Oct. 2004.

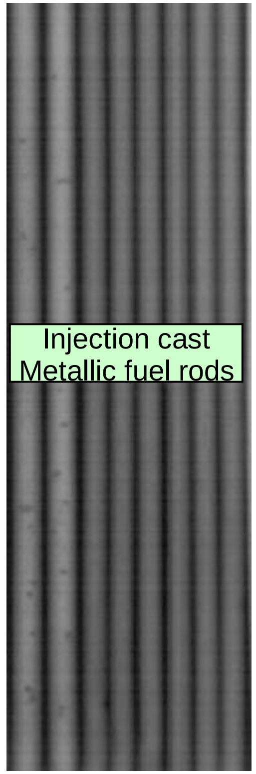
Metallic fuels with high breeding ratio are under Consideration for future fast reactors.

Fuel and core structural materials for fast reactors are new challenges for development.

Fast Reactor Fuel Fabrication Lab



Fast Breeder Reactor



Injection cast Metallic fuel rods

Properties of Reference FBR Fuels

Properties	$(U_{0.8}Pu_{0.2})O_2$	$(U_{0.8}\ Pu_{0.2})C$	$(U_{0.8}Pu_{0.2})N$	U-19Pu-10Zr
Heavy metal Density g/cc	9.78	12.96	13.50	14.30
Melting point ºK	3083	2750	3070	1400
Thermal conductivity (W/m ºK) 1000 K 2000 K	2.6 2.4	18.8 21.2	15.8 20.1	40
Crystal structure	Fluorite	NaCl	NaCl	bcc (γ)
Breeding ratio	1.1 - 1.15	1.2 – 1.25	1.2 - 1.25	1.35 - 1.4
Swelling	Moderate	High	Moderate	High
Handling	Easy	pyrophoric	Inert atmos	Inert atmos
Compatibility - clad coolant	average average	Carburisation good	good good	eutectics good
Dissolution & reprocessing amenability	Good	Demonstrated	risk of C^{14}	Pyro-reprocessing
Fabrication/Irradiation experience	Large Good	Good Indian Experience	very little	limited

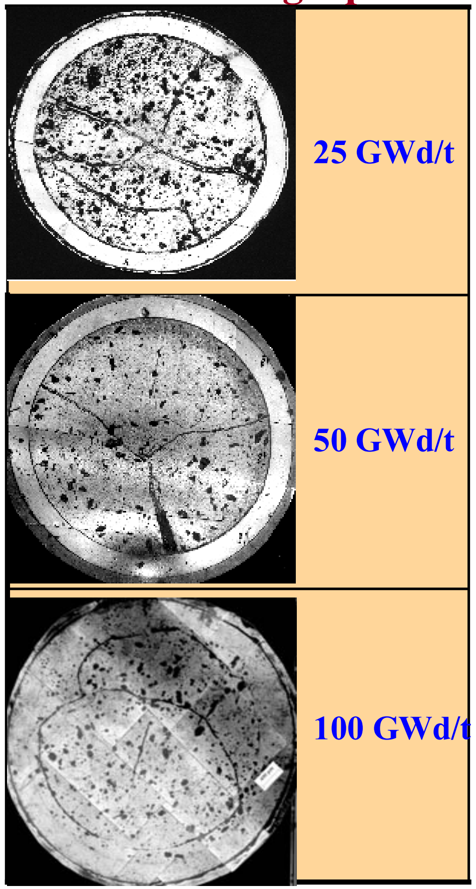
40 MW_{th} Fast Breeder Test Reactor (FBTR) Kalpakkam

$(U0_{0.3}, Pu_{0.7})C$ fuel in FBTR crossed a burn-up of 145 GWd/t.

PIE of FBTR fuel at 100 GWd/t

- No restructuring (low temperature)
- Diametral strain in cladding: 1.8%
- Fission gas release: 14%
- Fuel-clad gap closed
- No evidence of clad carburisation
- Residual ductility of clad 3%

Fuel Macrographs



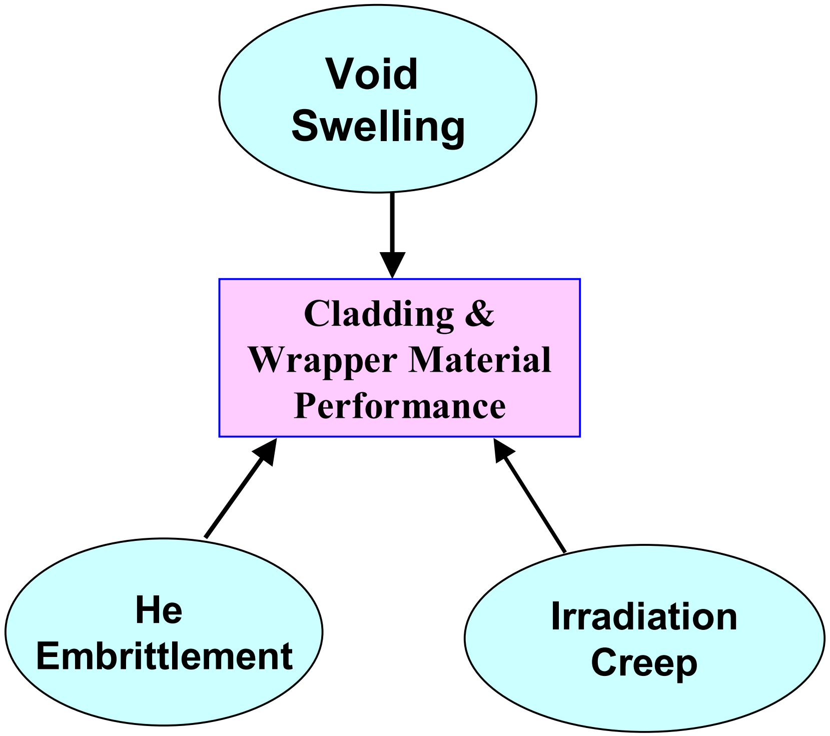
Life Limiting Processes in Core Structural Materials of FBR

Void Swelling

- Incubation period for swelling.
- Austenitic stainless steels (AISI 316) not resistant to swelling beyond 50 dpa
- Search for better materials, which can withstand exposure upto 150-200 dpa.

Void Swelling Resistance

- Enhancing vacancy-interstitial recombination
- Providing sites for recombination
- Optimisation of chemical composition
- Controlled cold work
- Coherent precipitate distribution



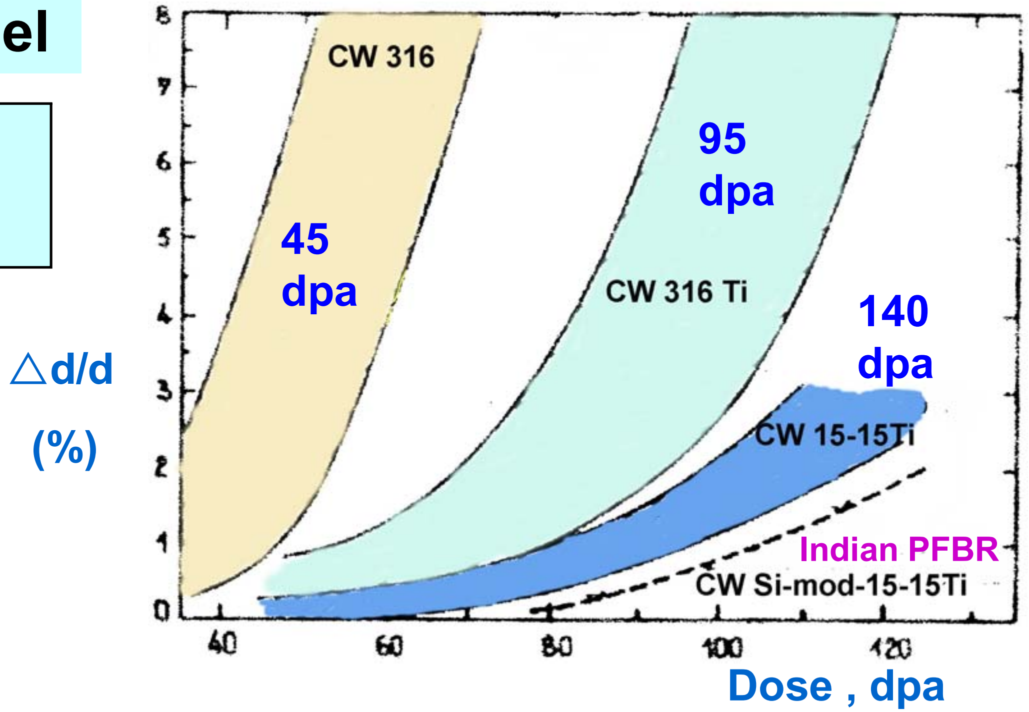
Candidate Materials: D9, PE16, 9Cr-1Mo, ODS steel

Stainless Steels for Fast Breeder Reactors

Austenitic Stainless Steel

- Good Creep Resistance
- Higher Void Swelling

D9: 15Cr, 15Ni, 2.5Mo
0.75Si, 0.04C,
5-7.5 x C: Ti



Ferritic Stainless Steel

Excellent resistance to Void Swelling
Poor Creep Strength
Rise in DBTT due to radiation

Not Suitable for Clad but feasible to use for Wrappers after Optimization of Chemical Composition & Microstructure to take care of DBTT Rise

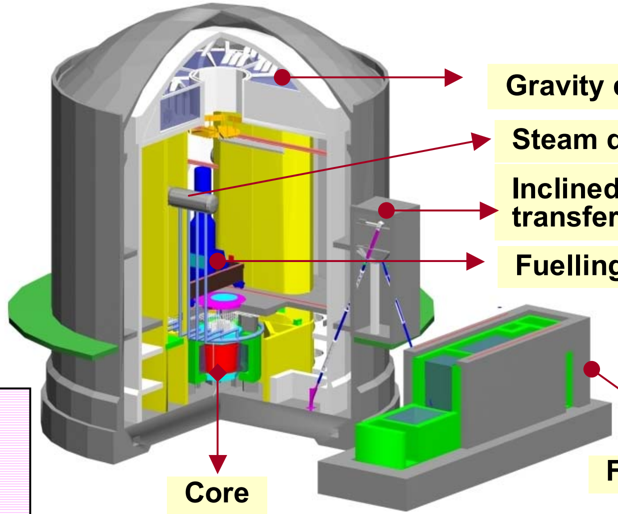
Future Nuclear Energy Sources & Systems

1. **Abundance of Resources**
   (large reserves to sustain requirement for a few generations)
2. **Resource consumption is matched by resource production.** (Neither breeding nor burning – just self-sustaining)
3 **Environmental friendly**
   (Low long lived radiotoxicity/ transmutation nuclide)
4. **Waste safety** (Fuel itself is a stable matrix for actinide and fission products, better than vitrified glass)
5. **Proliferation resistance**
   (U^{232} inherent presence in U^{233}/difficult to reprocess)

Thorium based Fuel cycle fits the bill

Advanced Heavy Water Reactor

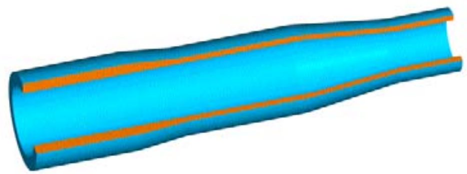
AHWR is a vertical pressure tube type, boiling light water cooled and heavy water moderated reactor using ^{233}U-Th MOX and Pu-Th MOX fuel.



Major Design Objectives

- **Power output – 300 MWe with 500 m³/d of desalinated water.**
- **A large fraction (65%) of power from thorium.**
- **Extensive deployment of passive safety features – 3 days grace period, and no need for planning off-site emergency measures.**
- **Design life of 100 years.**
- **Easily replaceable coolant channels.**

Salient Features of Pressure tube



- 120mm ID x 6300 mm length
- Replaceable through top end-fitting
- Unique shape by Pilgering route.
- - Thicker at one end, tapering at the other
- Controlled cold work to achieve required tensile properties.

Indian AHWR Fuel Cycle

- **Thorium – CLOSED FUEL CYCLE**

Recycle both Fissile & Fertile Material

SHORT TERM:

⇒ **Based on (Th-Pu)MOX**

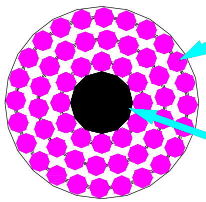
LONG TERM:

⇒ **Based on both (Th-Pu) and (Th-U^{233}) MOX.**

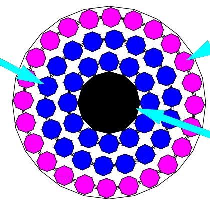
⇒ **Self – sustaining with respect to U^{233} and external Pu feeds from PHWR/FBR.**

⇒ **Recycle Th.**

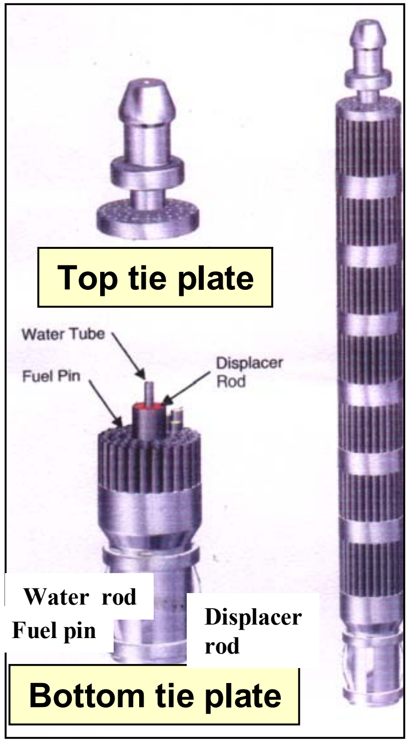
Fuel Cluster Cross section



Initial core

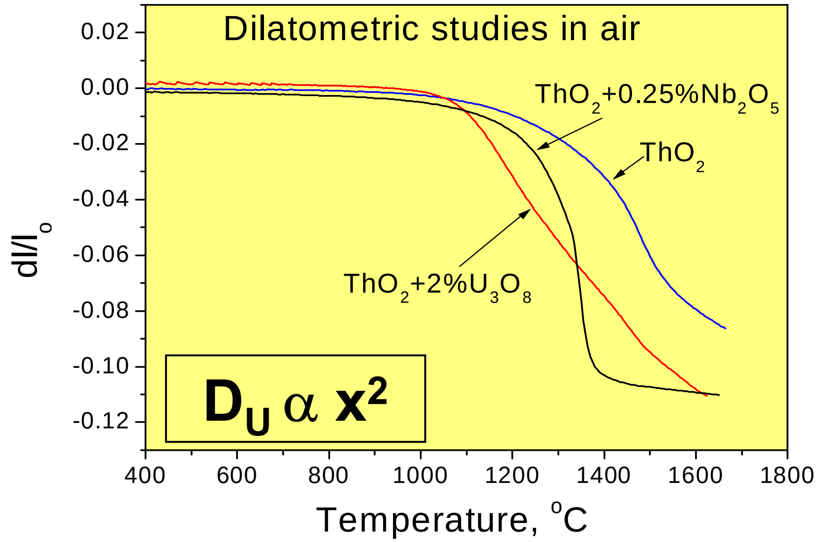


Equilibrium core

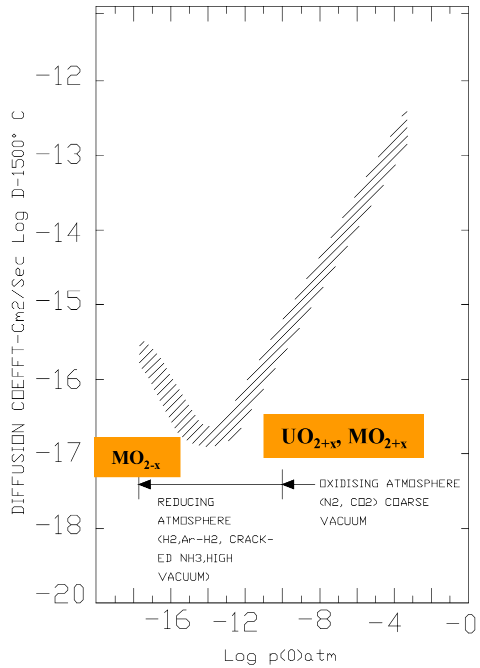


AHWR Fuel Cluster

Sintering behaviour of (Th-U)O_2 Pellets Made by CAP Process



Shrinkage behaviour of (Th-U)O_2 pellet



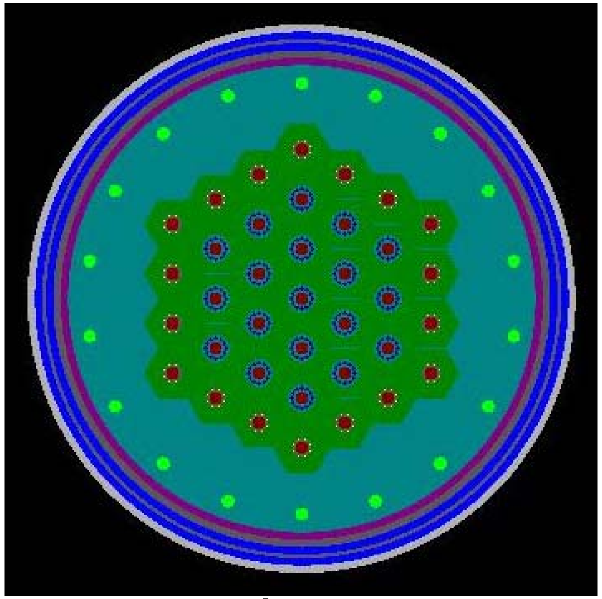
Oxygen potential Vs. diffusion Coefficient plot

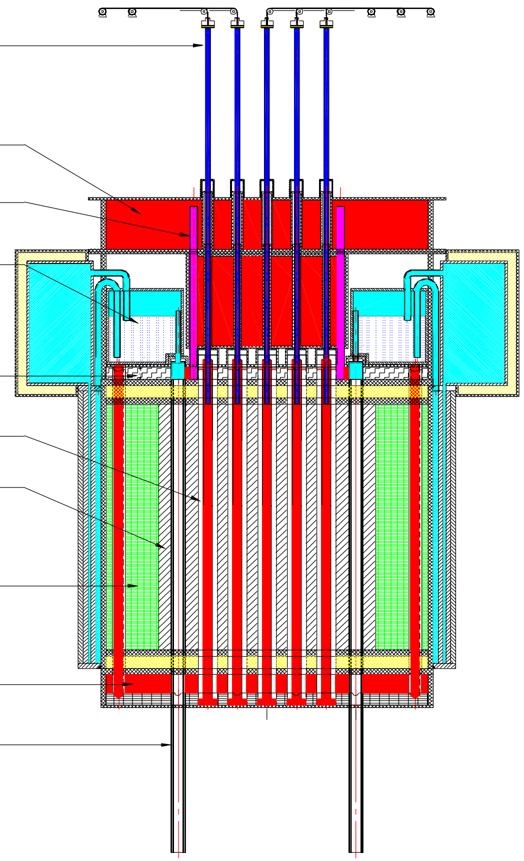
AHWR Fuel Pellets

Compact High Temperature Reactor

High temperature process heat for hydrogen production by water splitting

- Special materials
- Special fuel
- Passive systems for safe operation of the reactor





List of Materials for CHTR and their Selection considerations

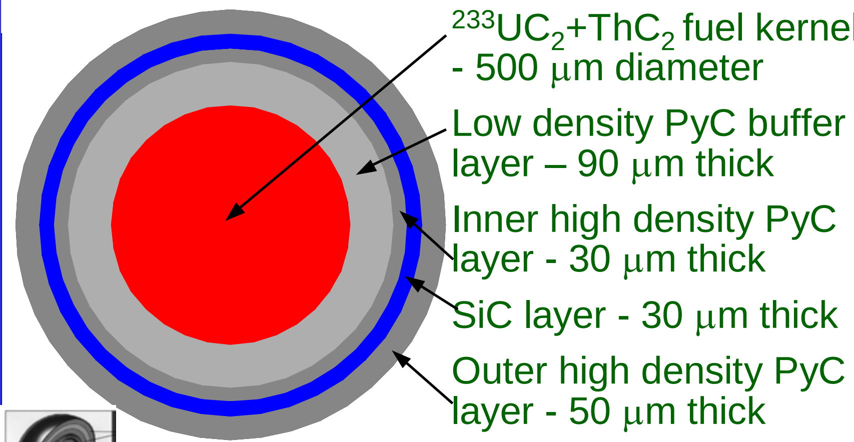
System	Material options	Reasons for selection
Fuel	UC_2, UO_2	Less Kernel migration
Moderator	BeO, Be, BeH_2	High temp. capability
Reflector	BeO, BeO+Graphite	Economical
Fuel Channel and Downcomer tubes	Coated Graphite	Low neutron abs. c/s, High temp. capability
Inner Reactor Vessel	Ceramic coated Mo-30%W, TZM, Mo-1% TiC, Nb-1% Zr, Ta	High resistance against Pb-Bi eutectic
Upper & Lower Plenums	Ceramic coated TZM (Ti -0.5%, Zr - 0.08-0.1%, C, Mo), Mo, Ta	Better corrosion resistanc against Pb/ Pb-Bi eutectic
Regulating System Driver & Control tubes	W, Niobium lined with PyC	Low neutron abs. c/s
Driving Fluid for Regulating system	Pb-Bi Eutectic, Gallium	Less neutron abs. c/s and less corrosive
Coolant	Pb/ Pb-Bi Eutectic	Low MP, High BP, Good safety features
Upper plenum Heat pipes	TZM/ Mo	Good corrosion resistance against Pb/ Pb-Bi eutectic

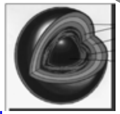
CHTR uses TRISO coated particle fuel which can withstand very high temperature (upto 1600 °C) facilitating high temperature operation

- **TRISO particles are embedded in a graphite matrix to form fuel compacts**
- **Number of TRISO particles per compact ≈ 3000**
- **Total number of TRISO particles in core ≈13.5 Million**
- **Burnup: 68 GWd/Te**

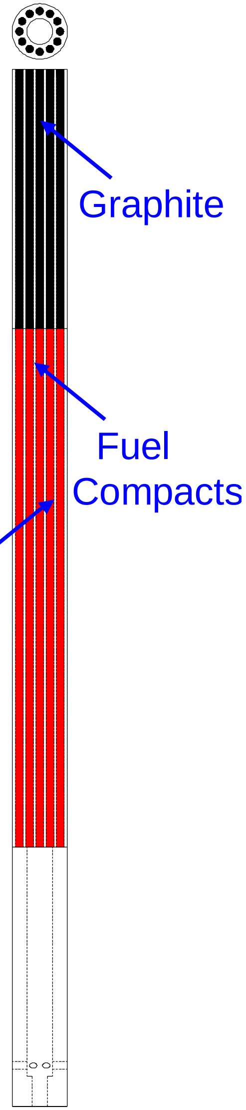
<u>Challenges:</u>

- **TRISO coatings by CVD process**
- **Achieve variable densities of coats**
- **Characterisation techniques**
- **Manufacture of fuel Compacts**



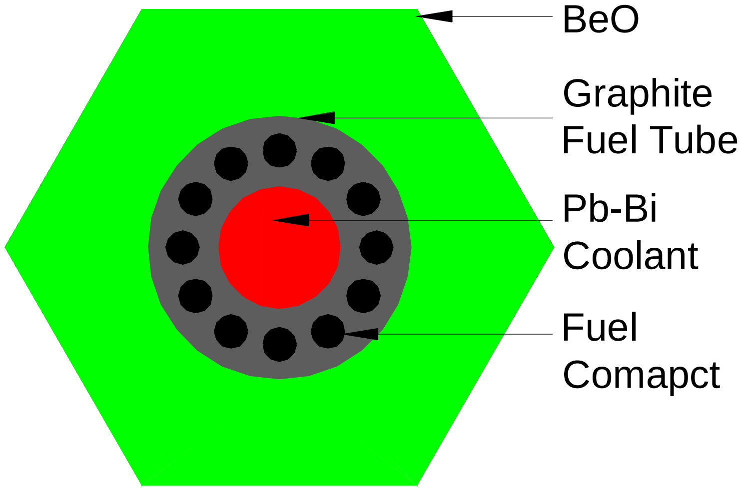


TRISO coated particle fuel-Schematic



Graphite fuel tube

Single fuel bed for CHTR



Compact High Temperature Reactor (CHTR) uses ^{233}U & Th based fuel, molten Pb-Bi coolant, BeO moderator, and (BeO+graphite) reflector material and has 1000 °C as coolant exit temperature

Severe operating conditions of CHTR poses many material related challenges

Materials	Reactor Components/ Systems
High density nuclear grade BeO	Moderator and reflector
High density, isotropic, nuclear grade graphite	Long fuel tube & down comer tube, large size reflector blocks, plenum flow guide blocks
Carbon-carbon composites	Heat pipes, alternate fuel tubes
Refractory metals/alloys e.g. TZM, Nb alloy, W etc.	Inner reactor shell, coolant plenums, heat utilisation vessels, Passive power regulation system, heat pipes, shutdown system
Oxidation and corrosion resistant Coatings	PyC, SiC, Silicides etc.

Graphite fuel tube

High density BeO prepared in BARC

Thermoelectric power generators for Compact High Temperature Nuclear Reactor (CHTR)

Developmental challenges

- Synthesis of n-type PbTe and p-type $(AgSbTe_2)_{0.15}(GeTe)_{0.85}$ alloys.
- Fabrication and characterization of Thermo-elements .
- Thin film metal contact deposition to thermo-elements.
- Metal strip interconnects with low contact resistance.
- Fabrication of devices.

Interface study using SEM & EDX

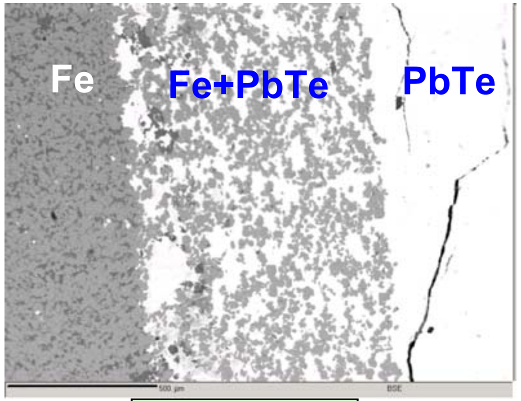
PbTe Thermoelement

No alloy formation at the PbTe/(PbTe+Fe)/Fe interfaces yielding clean and sharp interfaces.

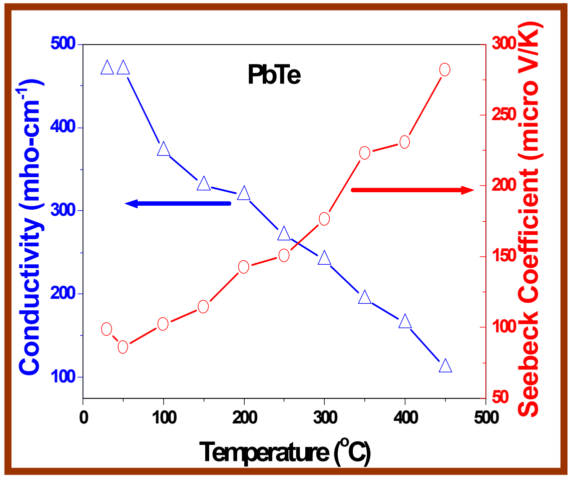
TAGS-85 Thermoelement

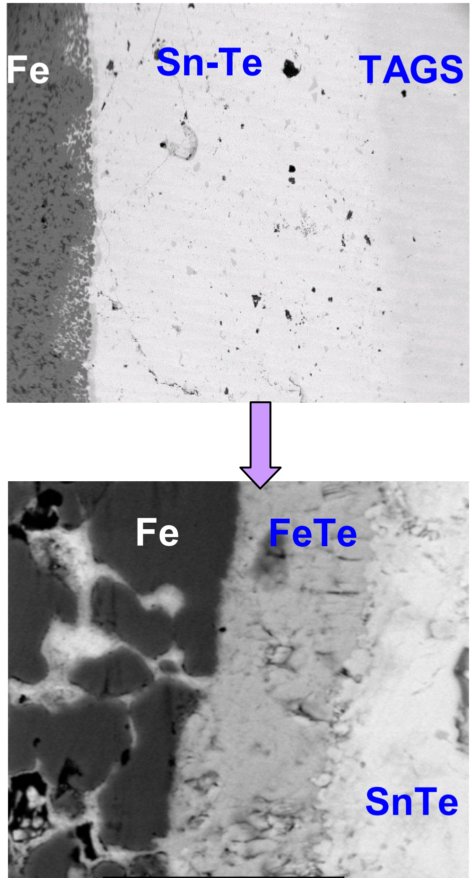
Reaction at Fe/SnTe interface results in the formation of FeTe phase.

No interaction observed at SnTe/TAGS-85 interface.



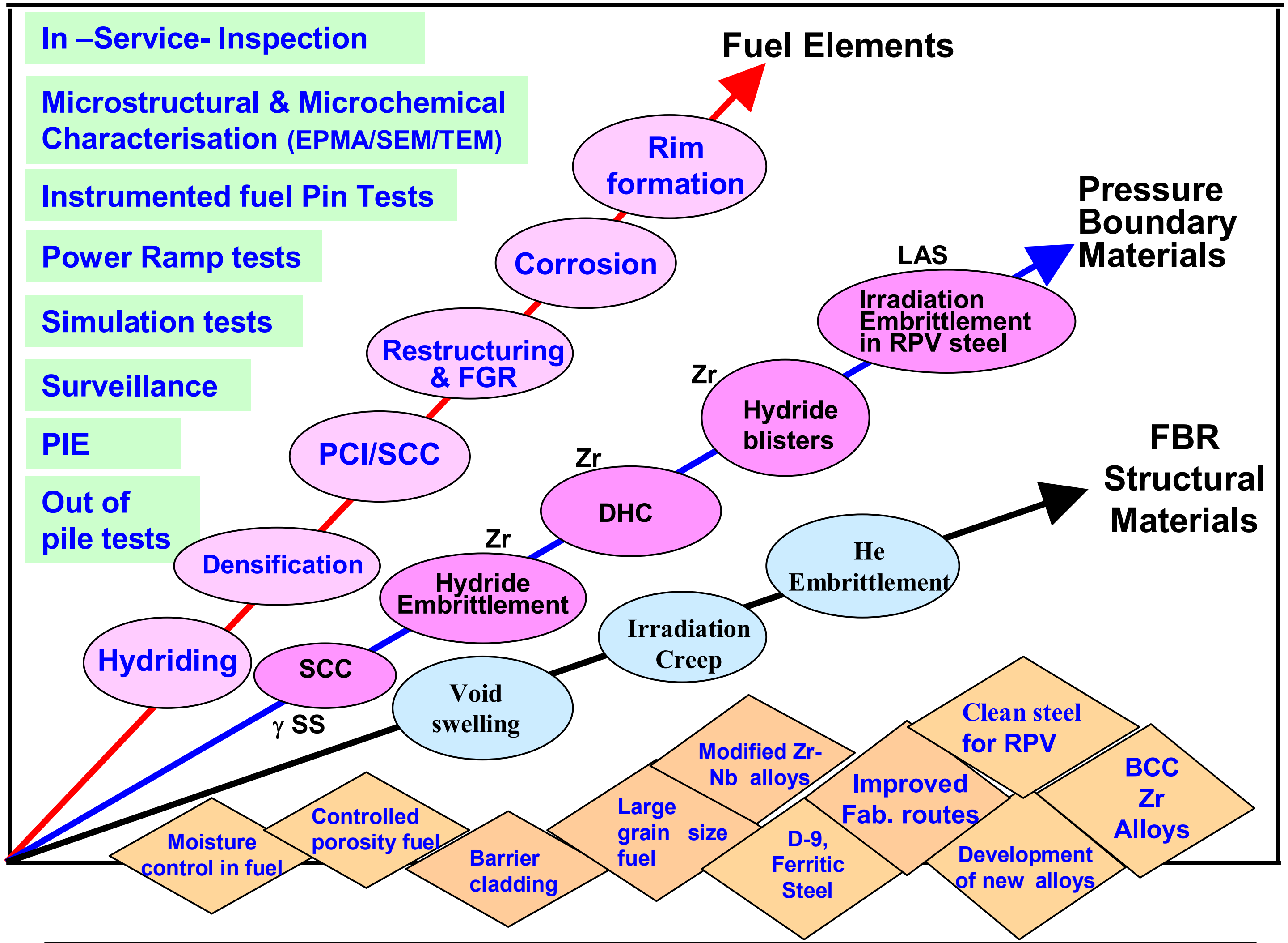
PbTe





TAGS-85

Summary



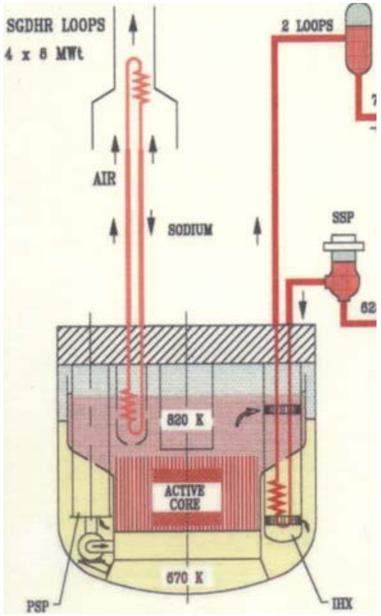
Solutions based on Physical Understanding & Modelling

Conclusions

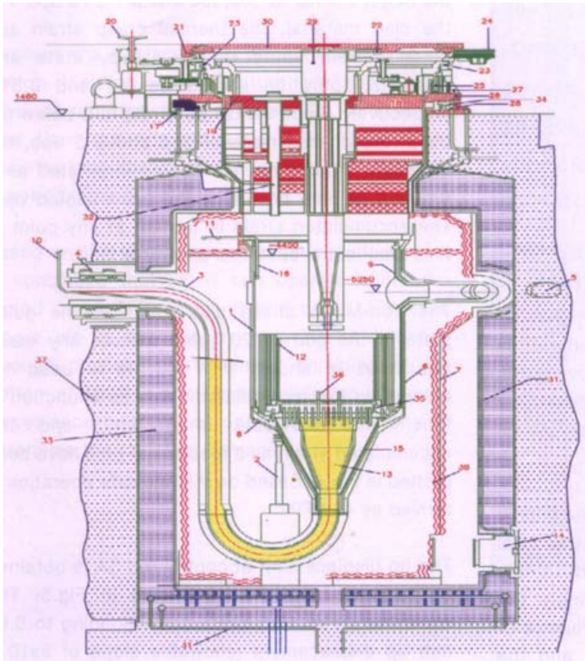
1. Increasing demands on materials with respect to higher operating temperature, higher fuel burnup, structural integrity at higher fluence and reduced radio-toxicity calls for optimization of presently used materials and /or development of new materials.

2. Inputs from R&D work in physical metallurgy and materials science towards optimization of manufacturing routes, identification and understanding of ageing degradation and establishing structure–property correlations are key to developing more forgiving materials and providing engineering solutions.

Thank You



PFBR Reactor Assembly
(Pool type)



FBTR Reactor Assembly
(Loop type)