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Solid state diffusion bonding of ZrC to Zr-based alloys

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Solid state diffusion bonding of ZrC to Zr-based alloys

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Introduction

Australian Government

ansto

Nuclear-based science benefiting all Australians

Institute of Materials Engineering

"A major centre of materials engineering and expertise in Australia with a multidisciplinary team of scientists and engineers"

Materials for Extreme Environments

Radiation, Elevated Temperature, High Stress and Corrosion
Fission and Fusion Applications

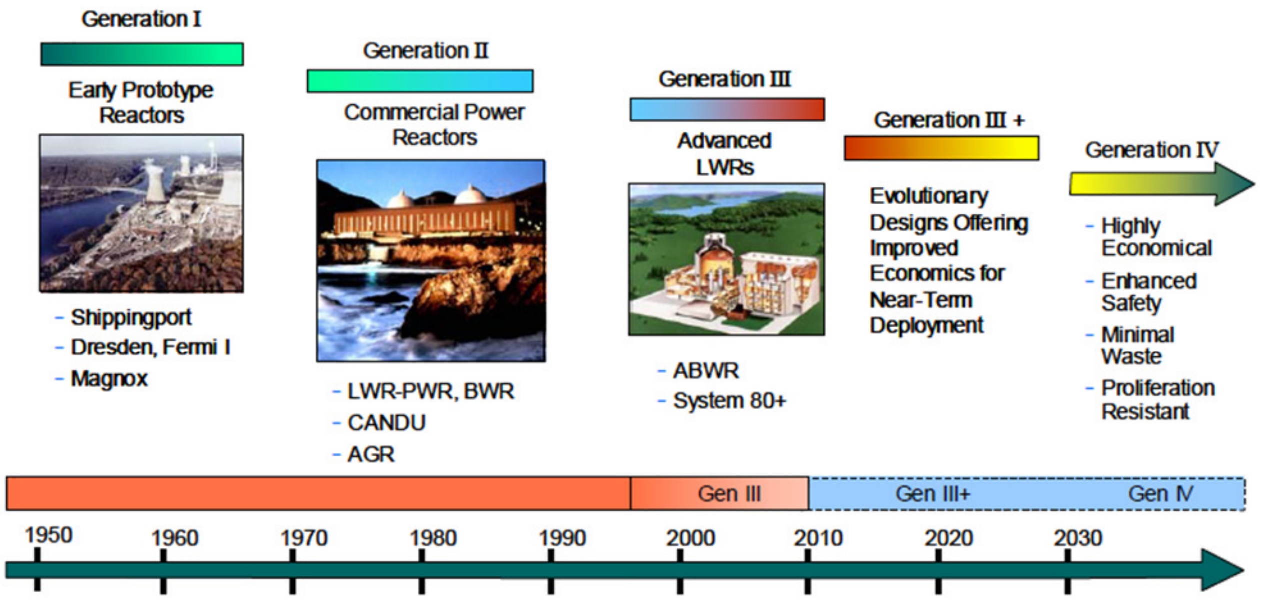
Introduction

- Nuclear Power will play a vital role in realising a clean energy future
  - Lowest cost, low emission technology for producing baseload power
  - Currently generates 30% of baseload power in EU
  - Two nuclear power plants to be built at Vogtle, GA (First in US in 35 years)
- Currently, worldwide, there are:
  - **433** nuclear reactors in **operation**
  - **63** nuclear reactors under **construction**
  - **160** nuclear reactors in **planning**

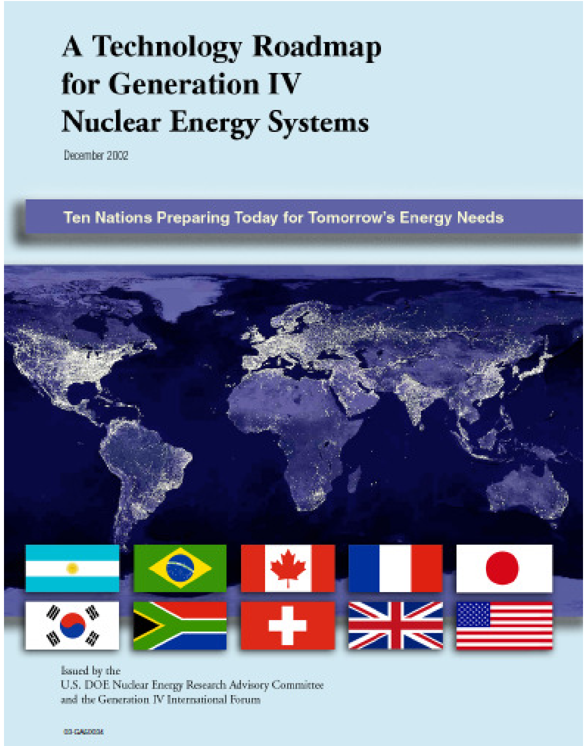
Source: http://www.world-nuclear.org/info/reactors.html

Olkiluoto-3, Finland Source: theage.com.au

Background



A Technology Roadmap for Generation IV Nuclear Energy Systems



A Technology Roadmap
for Generation IV
Nuclear Energy Systems

December 2002

Ten Nations Preparing Today for Tomorrow's Energy Needs

Issued by the
U.S. DOE Nuclear Energy Research Advisory Committee
and the Generation IV International Forum

- Generation IV initiative (GIF) lauched by US DOE in 2000
  - 10 Member Nations + Euratom
  - Aim: *Design and develop a new generation, of energy efficient, sustainable, safe and reliable, proliferation resistant nuclear reactors by 2030*

Background

Proposed Gen-IV Reactor Designs

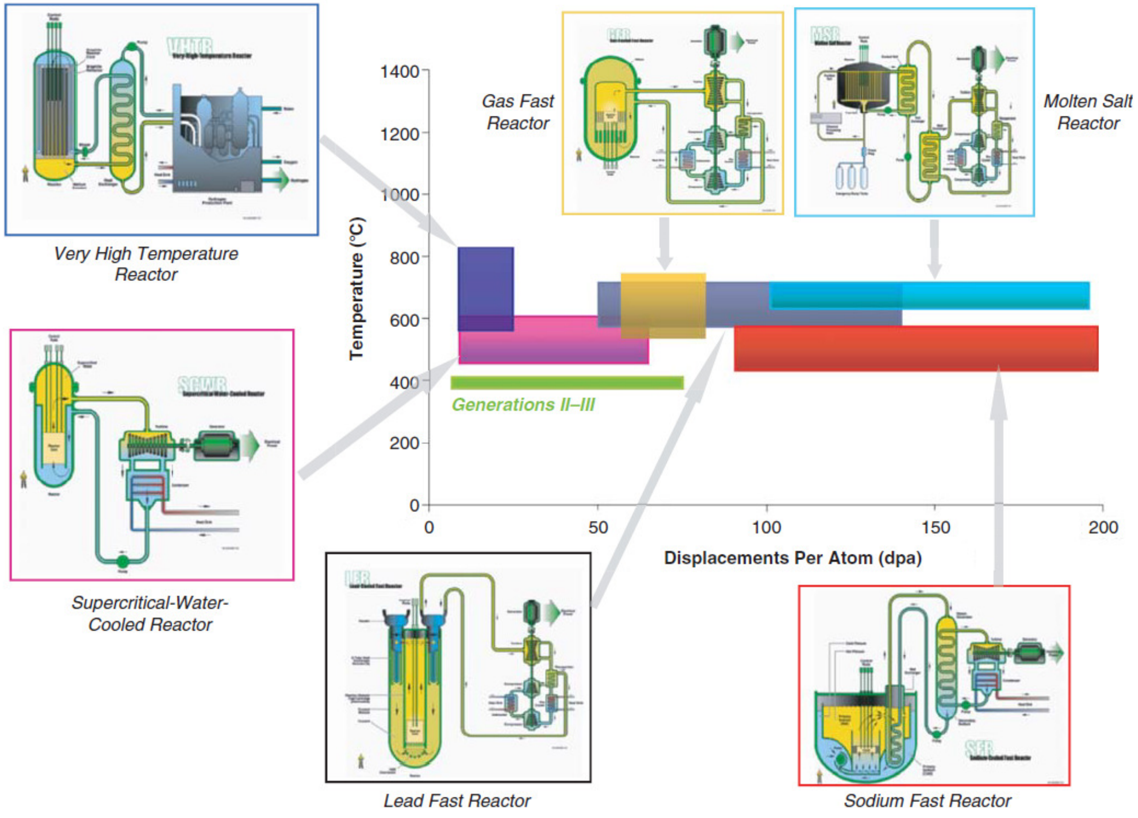
Reactor System	Coolant	Neutron Spectrum	Core Outlet Temperature (ºC)	Pressure (High = 7-15MPa)
Very High Temperature Reactor (VHTR)	Gas (eg. He)	Thermal	>900	High
Gas Cooled Fast Reactor	Gas (eg. He)	Fast	~850	High
Sodium Cooled Fast Reactor (SFR)	Molten Salt (fluoride salts)	Fast	700-800	Low
Lead Cooled Gas Reactor (LFR)	Liquid Metal (eg. Pb, Pb-Bi)	Fast	550-800	Low
Molten Salt Reactor (MSR)	Liquid Metal (Na)	Thermal	~550	Low
Supercritical water-cooled reactor (SCWR)	Water	Thermal/Fast	350-620	Very High

Source: Murty *et al.*, 2008

Background

Proposed Gen-IV Reactor Designs

- Materials exposed to
    - Higher temperatures
    - Higher neutron doses
    - High stresses
    - Extremely corrosive environments
- Additional radiation-induced material effects
    - 1D/2D and 3D Defects
    - Segregation
    - Creep
    - Diffusion
    - Precipitation
    - Volumetric Swelling



Source: Guerin *et al.*, 2009

Background

Proposed Gen-IV Reactor Designs

- Reactor Technology Challenges
  - **Availability of durable, radiation tolerant materials**
  - Sustainability & waste disposal
  - Economics
  - Safety and reliability
  - Proliferation resistance & physical protection

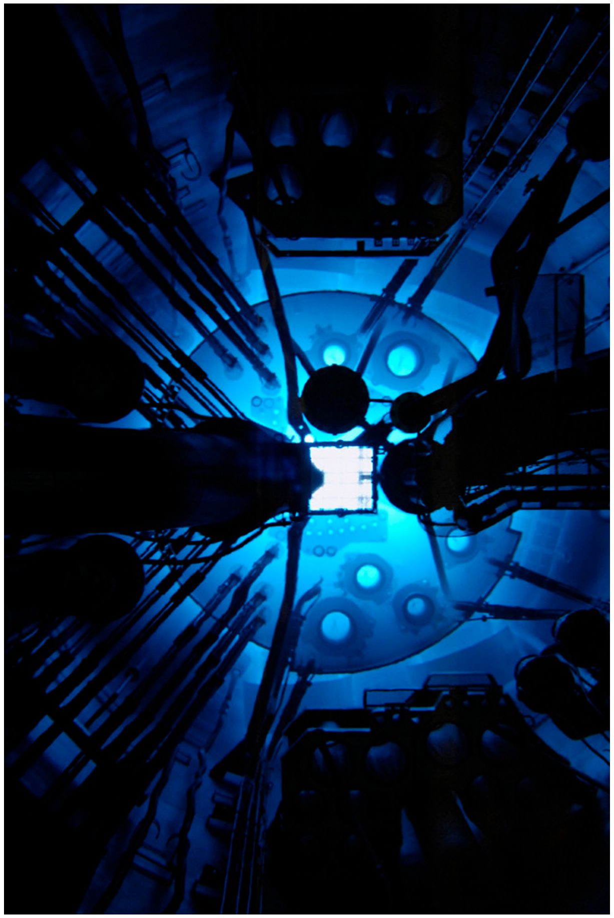
- Radiation Tolerant Materials
  - Dimensional stability under irradiation
  - Large interfacial area
  - Low defect mobility
  - Good recovery from irradiation induced defects

Displacement Cascade

(○ -vacancies; ● interstitials)

Source: Buschow et. al, 2001

Research Motivation



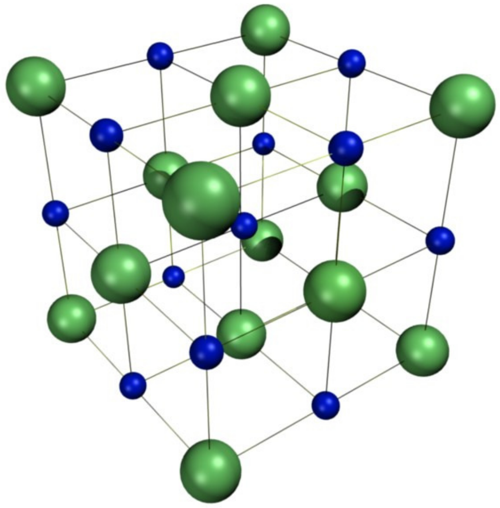
ANSTO OPAL Reactor Core

Source: ansto.gov.au

- Many reactor components in proposed Generation IV systems will require high-strength interfacial bonding for maximised performance
- Several refractory non-oxide ceramics are proposed for application in Gen-IV reactors
  - SiC, TiC, **ZrC**
  - Applications: heat exchangers, thermal insulations, core-reactor components and high thermal load components
- A UHTC joined to a metallic material
  - exhibits **high refractoriness**
  - **retains structural integrity** of the underlying metallic substrate

Zirconium Carbide

- A promising UHTC Gen-IV material
  - Diffusion barrier layer on TRISO fuel in earlier Generation GFRs
  - Proposed as a fuel matrix component in Gen-IV GFR
  - Other potential applications in elevated temperature nuclear environments
- Key Nuclear Properties
  - Low neutron absorption cross section
  - High melting point (~3420ºC)
  - Recovery from irradiation induced defects
  - Resistance to attack from fissile products
- Crystal Structure
  - Interstitial carbide
  - NaCl, B1 FCC structure
  - Stoichiometry: $ZrC_{0.55}$ to $ZrC_{0.98}$
- **Major limitation: low fracture toughness**



ZrC NaCl Structure

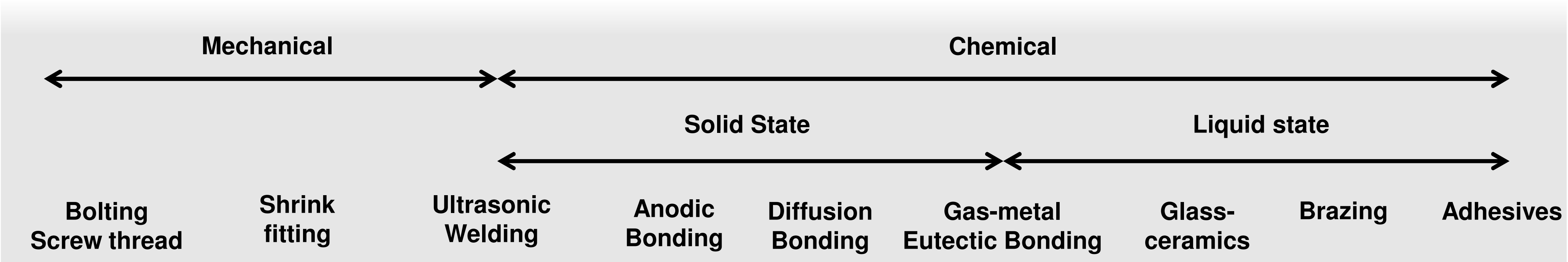
Zirconium Alloys

- Zircaloy-4, Zircaloy-2 (Zr+Sn), Zr-Nb
- Nuclear Applications
  - Cladding, pressure tubes and structural components
- Key Nuclear properties
  - Low neutron absorption cross section
  - Excellent corrosion resistance
  - Good mechanical properties
- HCP Crystal Structure (@RT)
  - Strong anisotropy
  - $\alpha \rightarrow \beta$ transformation ~850° C

- **Limited to operation at ~400° C in nuclear applications due to creep/embrittlement**

Zircaloy Cladded Nuclear Reactor Fuel
Source: cameco.com

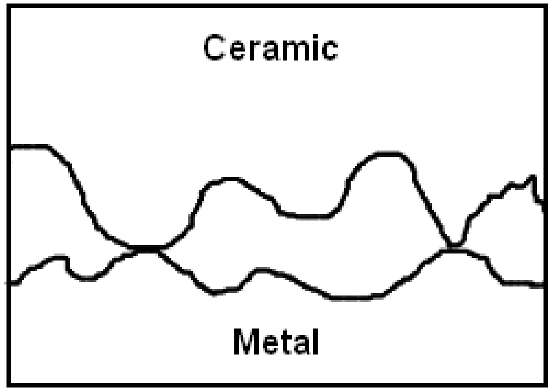
Joining ZrC/Zircaloy-4



Source: Fernie *et. al* . 2009

Method: Solid State Diffusion Bonding

- Usually carried out in a HUP or HIP
- Structure, CTE mismatch and surface roughness important
- Zircaloy-4/ZrC CTE mismatch is relatively low
- Key Steps
    - (a) asperity contact
    - (b) yielding under large localised stresses
    - (c) deformation and diffusion mass transfer
    - (d) removal of interfacial voids and bond formation



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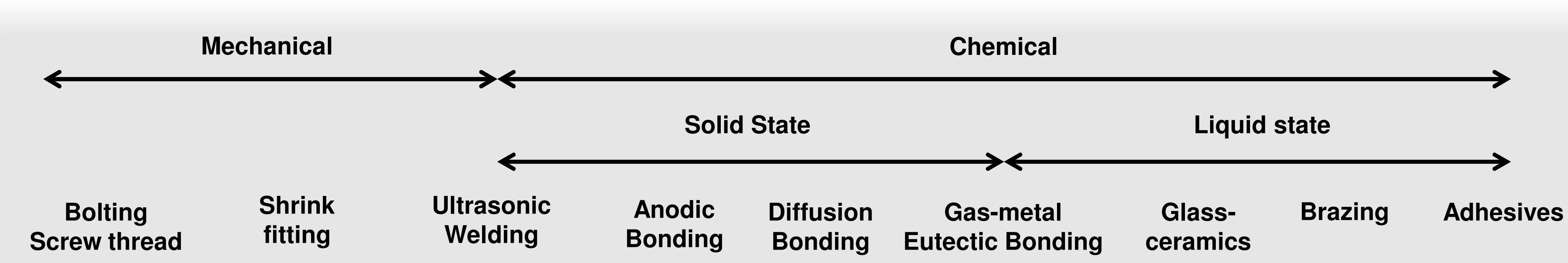

Mechanical

Chemical

Solid State

Liquid state

Bolting Screw thread

Shrink fitting

Ultrasonic Welding

Anodic Bonding

Diffusion Bonding

Gas-metal Eutectic Bonding

Glass-ceramics

Brazing

Adhesives

Source: Fernie *et. al* . 2009

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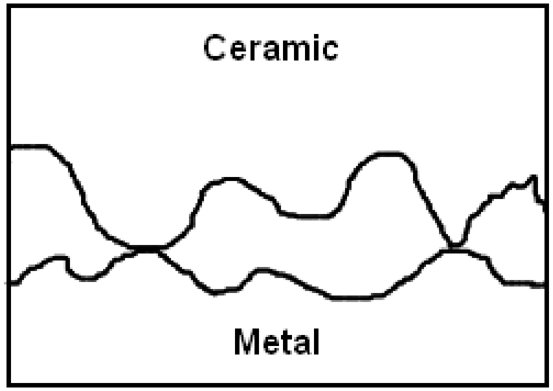

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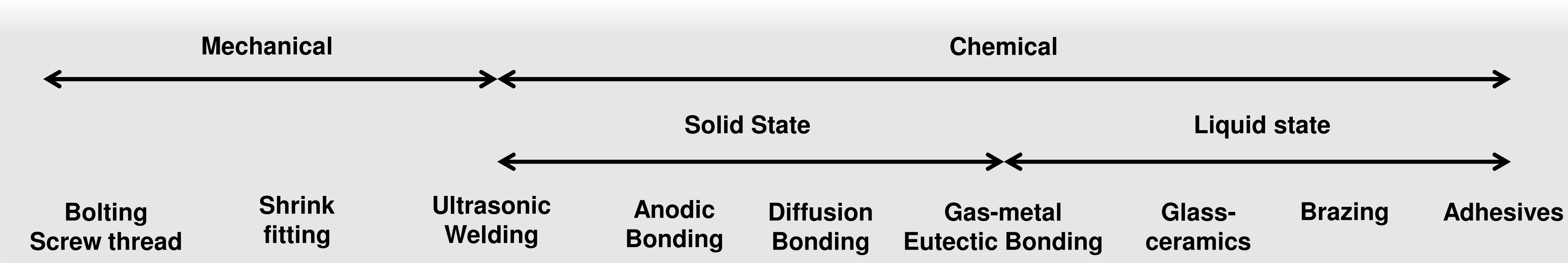

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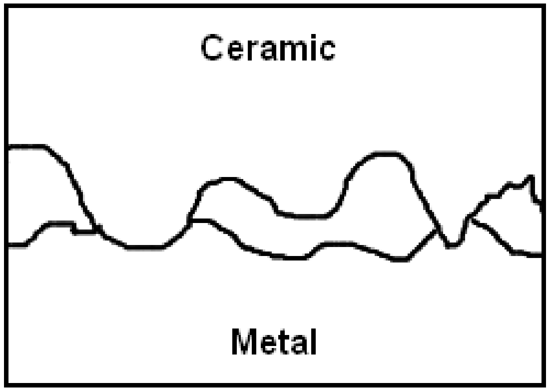

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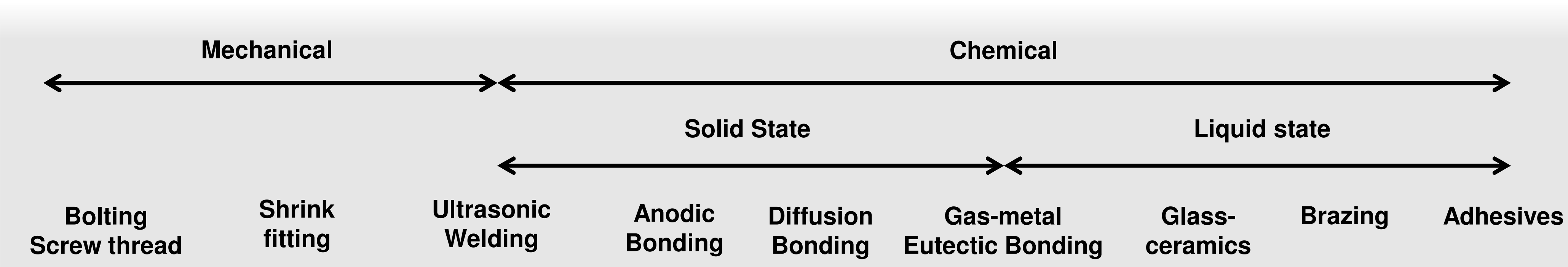

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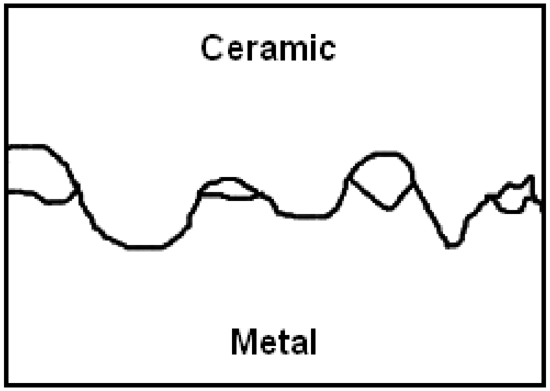

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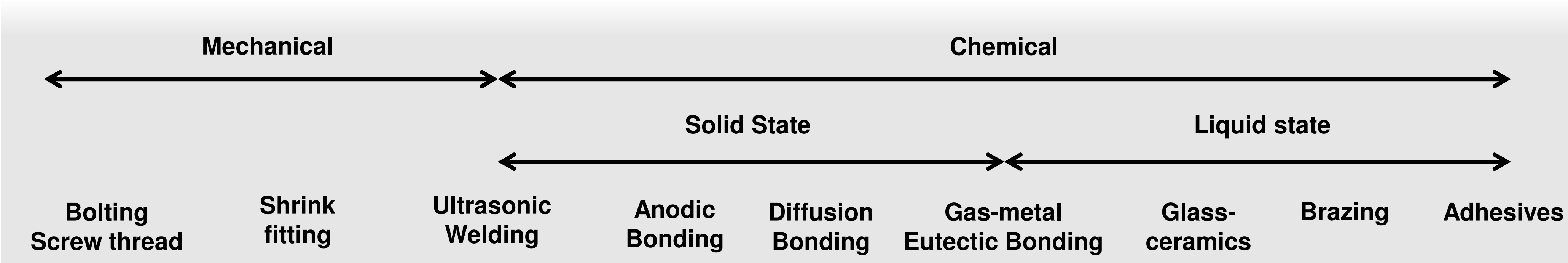

Mechanical

Chemical

Solid State

Liquid state

Bolting
Screw thread

Shrink
fitting

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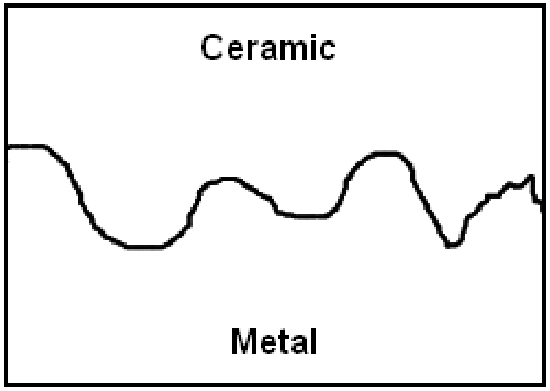


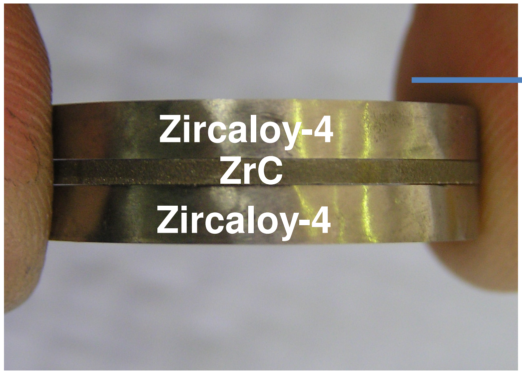
Joining ZrC/Zircaloy-4

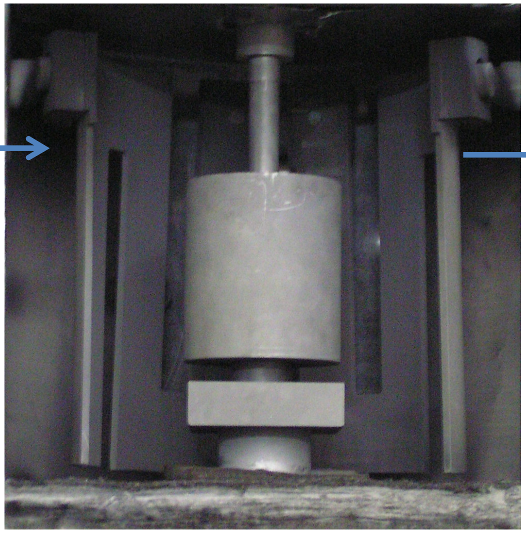
- **Preparation**
  - Zirconium Carbide
    - ~2% Hf, 3-5 micron APS
    - Cold Uniaxially Pressed, HUP, HIP
    - No sintering aids
    - Ø25mm size, 2mm thick
    - Grinding/polishing steps to 1µm diamond surface finish
  - Zircaloy-4
    - Zr-1.56Sn
    - Hot rolled, annealed, blasted & pickled
    - Ø25mm size, 3mm thick
    - Grinding polishing steps to 1µm surface diamond finish

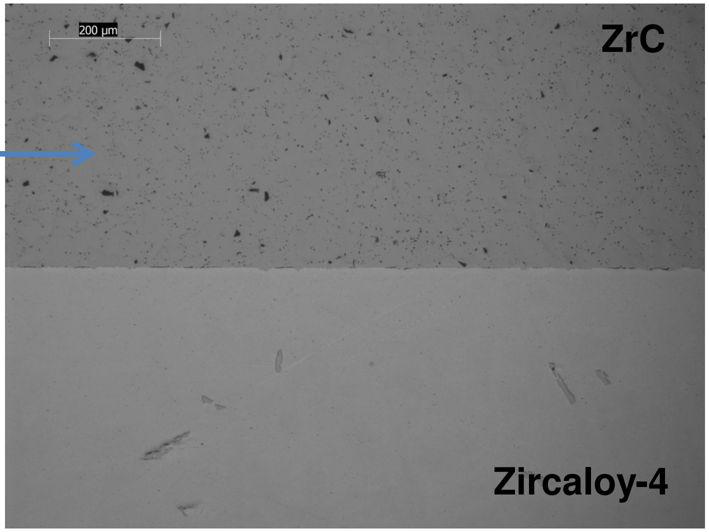
ZrC Processing Steps

	Stage 1	**Stage 2**
Process	HUPing	HIPing
Temperature	2000ºC	2000ºC
Pressure	20MPa	100MPa
Bulk Density after Processing (ASTM C20)	~96.5	~99%

Joining ZrC/Zircaloy-4



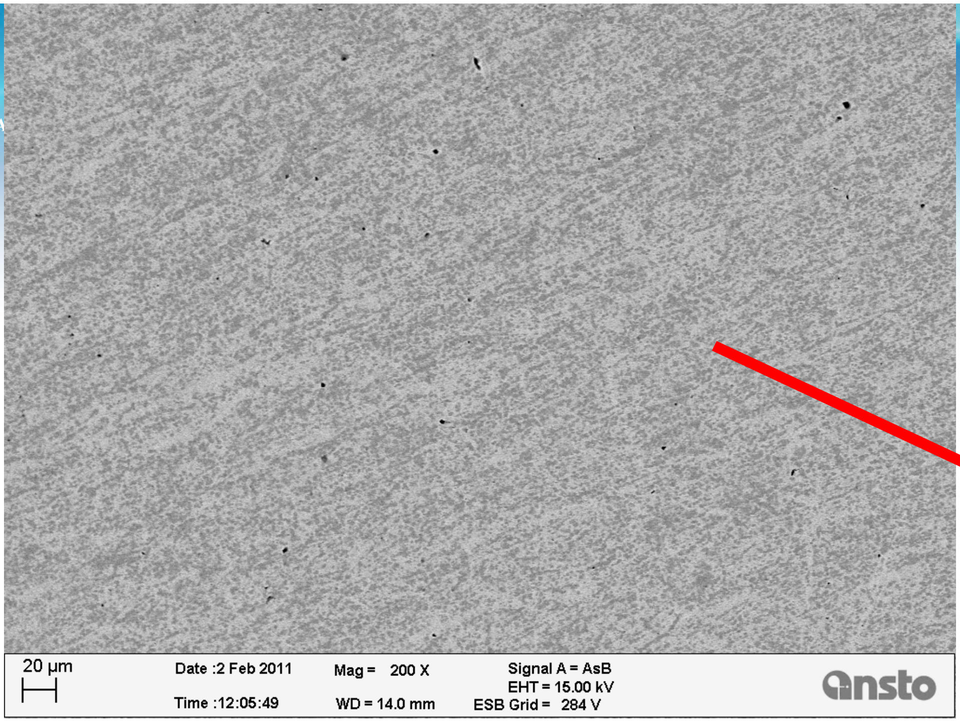


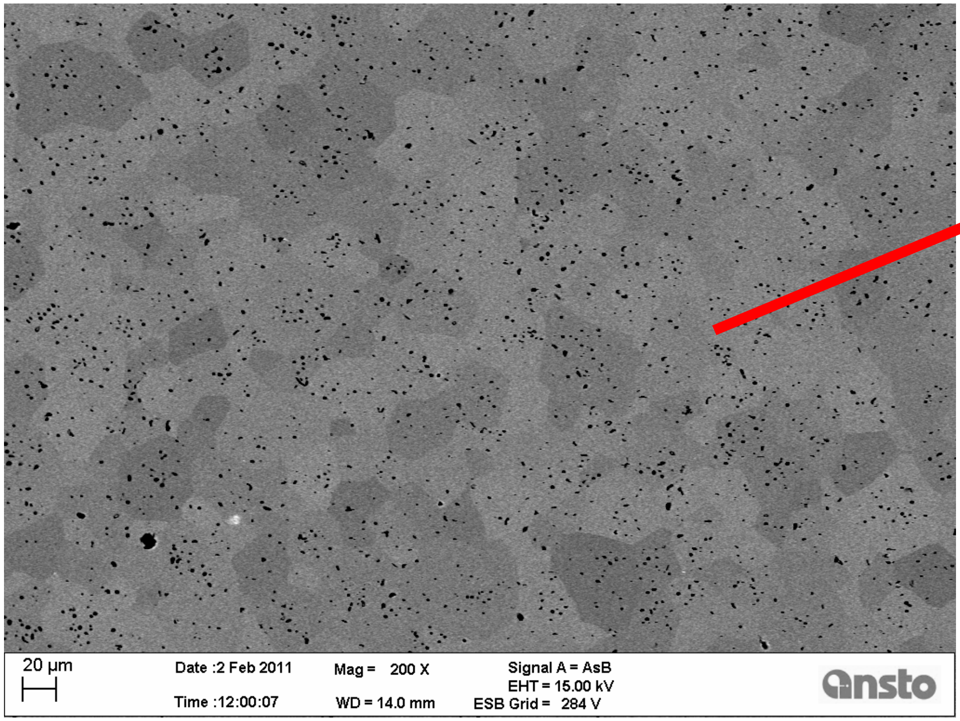


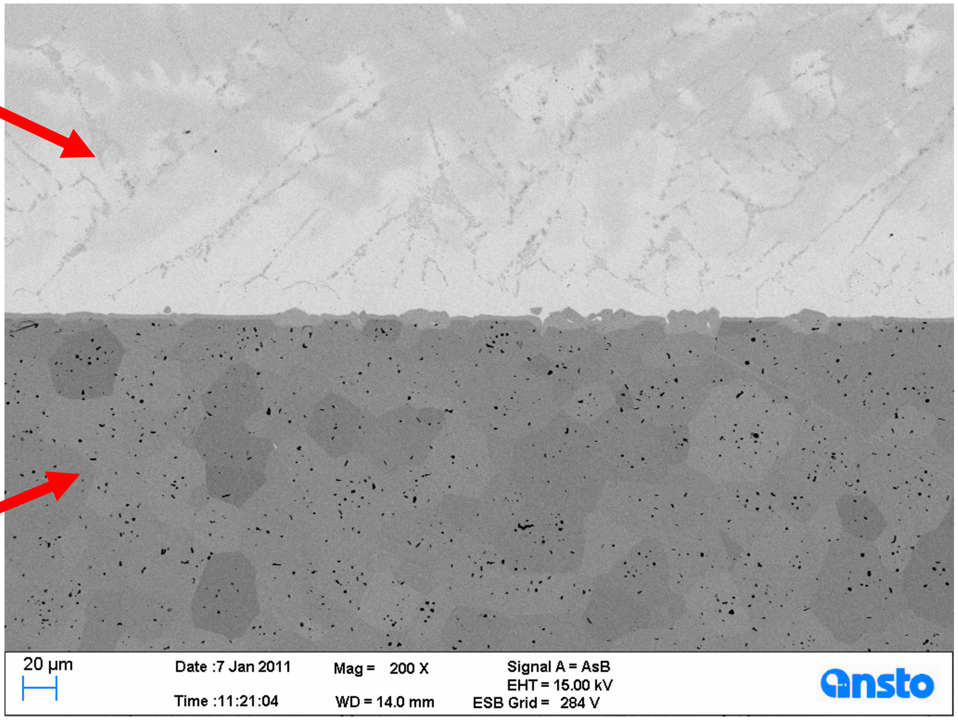
- Samples joined in hot uniaxial press, 0.01Pa vacuum, no interlayer
- Processing parameters: 1300ºC, 40MPa
- Void-free solid-state diffusion bond produced
- Macro-deformation of Zircaloy-4 during joining process

Results

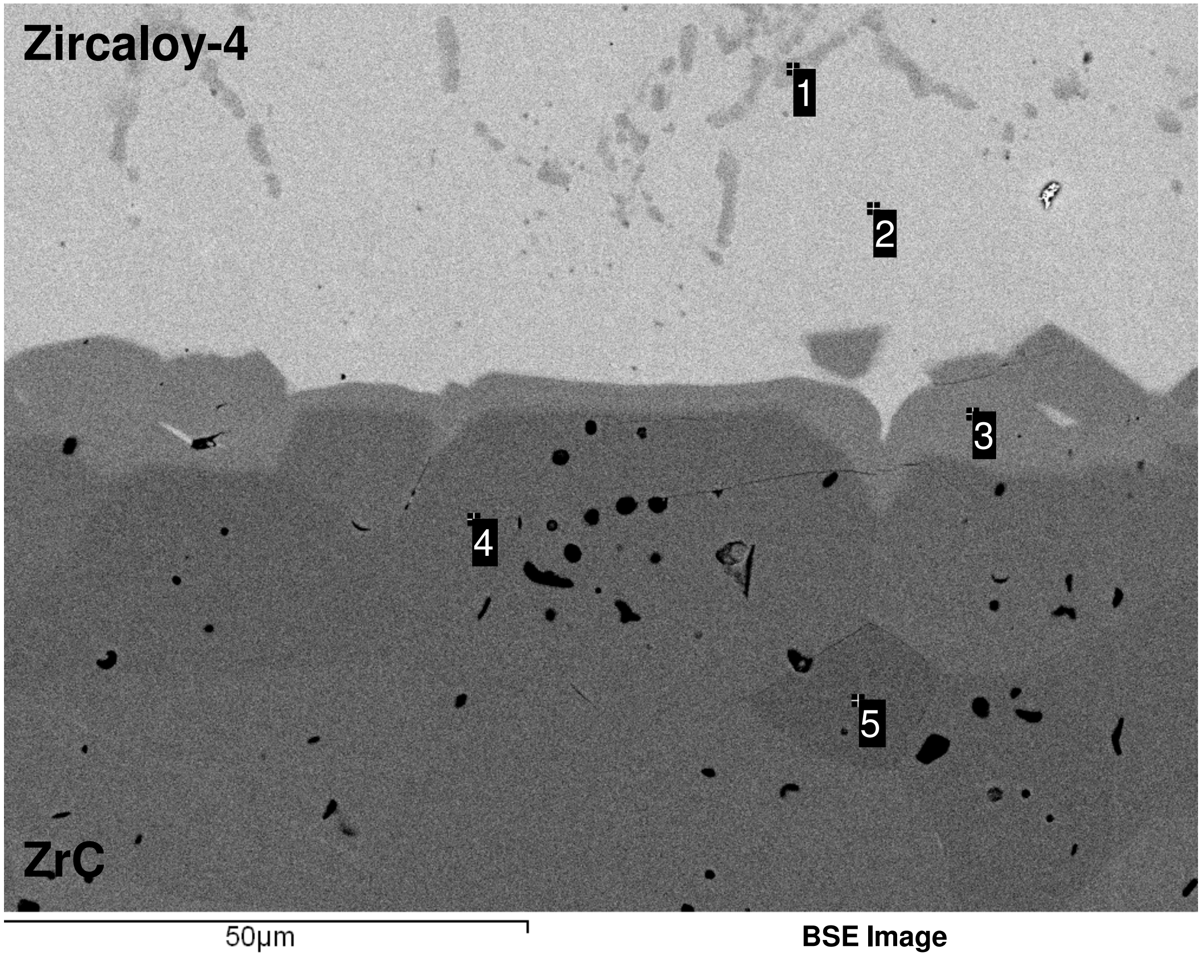
- **BSE SEM Microscopy**







Results



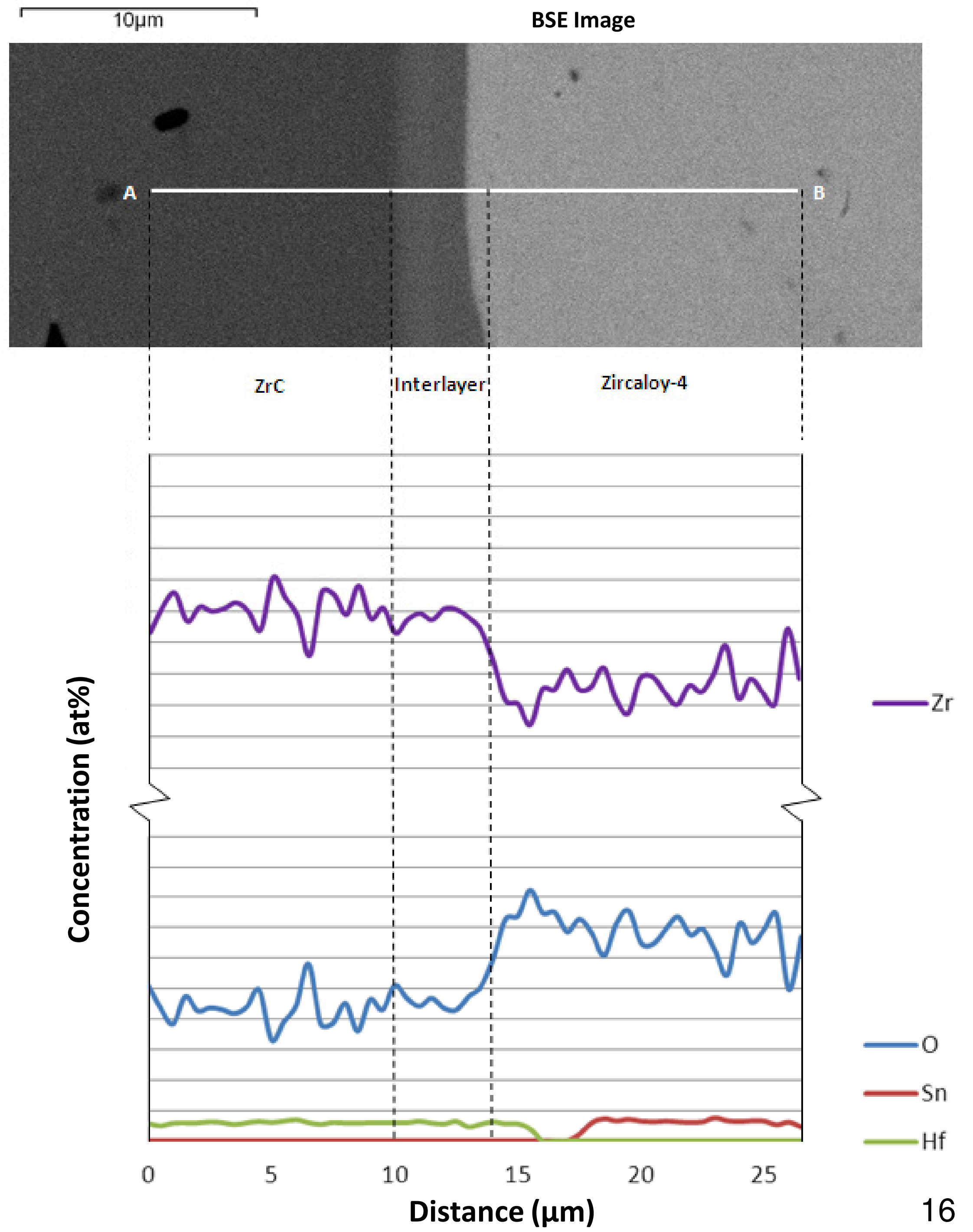
BSE Image

EDS: Minor Elements

ID	Elemental wt%		
	O	Sn	Hf
1	1.99	0.75	
2	2.17	1.81	
3	3.07		
4	1.36		2.37
5	1.18		2.36

Results

- **Observations: EDS Linescan**
  - Higher [O], [Sn] in Zircaloy-4
  - Evidence of Hf diffusion into Zircaloy-4
  - Marked changes in [Sn], [Hf] near interlayer



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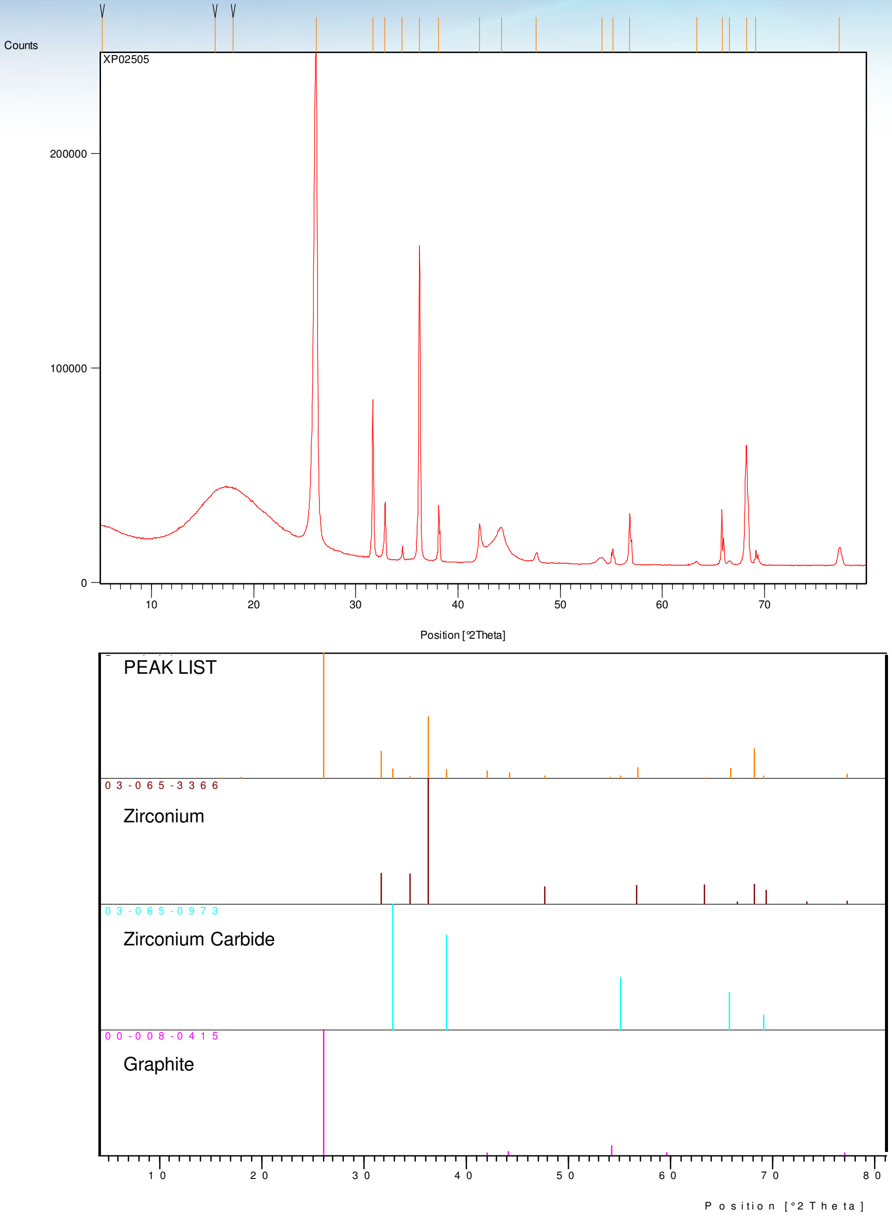

- **X-ray Diffraction**
  - Zr and ZrC presence confirmed
  - Free carbon also detected
  - Need smaller beam size for more accurate resolution of interface structure

Conclusions & Future Work

- **Conclusions**
  - Void-free ZrC/Zircaloy-4 solid-state diffusion bond achieved
  - Observations
    - Higher relative [O] in Zircaloy-4
    - Evidence of surface oxidation
    - Evidence of Sn segregation at interface
    - Evidence of Hf diffusion into Zircaloy-4
    - Macro-deformation of Zircaloy-4 during joining process

- **Future Work**
  - Optimise process temperature and pressure to minimise residual stresses and macro-deformation
  - C detection & mapping (eg. Raman)
  - Mechanical testing of diffusion couples
  - Irradiation of samples

Acknowledgements

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