IAEA 60 Years
Atoms for Peace and Development

Cladding embrittlement, swelling and creep

Workshop on radiation effects in nuclear waste forms and their consequences for storage and disposal, 12-16 September 2016, Trieste, Italy

Scope

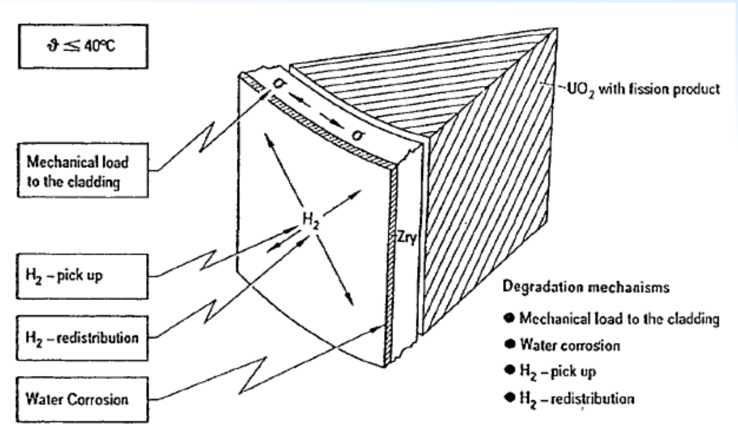
- Spent fuel, the behaviour of zirconium clad fuels from power reactors
  - Pre-disposal behaviour
- Beginning of storage conditions
- Focus on the following degradation mechanisms
  - Cladding embrittlement (behaviour of hydrogen)
  - Cladding creep
  - Swelling (oxidation, evolution of spent fuel in storage)
  - Why is it important?
- Degradation mechanisms will be looked at in terms of phases of the BEFC:
  - Storage
    - Wet
    - Dry
  - Transitions
    - Wet to dry, retrieval/transport of spent fuel after storage

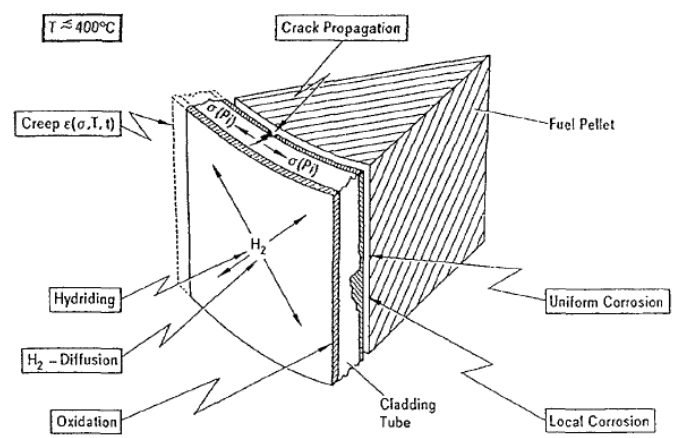
Beginning of Storage Conditions

- At the beginning of storage, the spent fuel has undergone a number of changes as a result of reactor service
- Examples include:
  - Irradiation which causes modifications to clad (neutron damage) and fuel pellet microstructures;
  - fission gas production leading to the fuel rods being pressurized;
  - internal attack of the fuel clad by fission gas products;
  - thermal changes;
  - impacts from mechanical interactions;
  - corrosion/pick-up of chemical elements from the reactor coolant.
- These conditions can impact on spent fuel performance during storage, transport and subsequent fuel handling operations
  - Area of topical interest is the behaviour of hydrogen in zirconium alloy clad material

Degradation Mechanisms Impacting Zirconium Clad Fuel in Wet and Dry Storage

- Essentially the same degradation mechanisms in wet and dry storage. Differ in significance; for example temperature related mechanisms are not significant in wet storage
  - Oxidation
  - Thermal Creep
  - Stress Corrosion Cracking (SCC)
  - Delayed Hydride Cracking (DHC)
  - Hydride Reorientation
  - Hydrogen Migration and Re-distribution

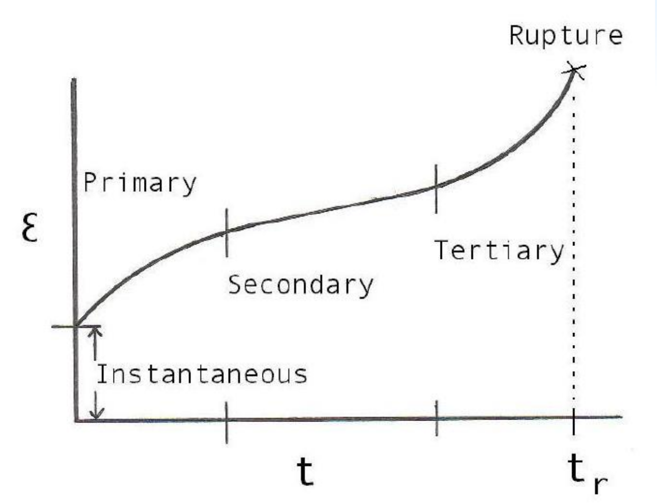




References 1 and PEEHS. M., FLEISCH. J., J. Nucl. Mater., 137 (1985) 190-202.

Thermal Creep – Fuel Cladding

- Thermal creep is a time dependent deformation under an applied load
  - For fuel rods this is normally associated with an increase in fuel rod length primarily as a result of the internal rod pressure (RIP)
  - Generally only occurs at high temperature
    - Insignificant creep at pool storage temperatures
  - Greater the creep the thinner the fuel clad wall becomes
  - Greater the potential for the fuel clad to rupture
- Three creep stages
- Safety considerations
  - Containment (IAEA-SF-1)
  - Goal is to prevent conditions which would lead to fuel clad rupture



Creep Stages

Primary – Initially rapid and slows with time

Secondary – Uniform rate

Tertiary- Accelerated rate l leading to rupture

Thermal Creep –Dry Storage

- *‘Considered’* as one of the most limiting degradation mechanism in spent fuel dry storage

- To protect against the possibility of fuel clad rupture during dry storage a 1% creep strain limit was originally introduced in dry storage licensing
  - Bounding condition based upon post-pile burst tests (up to 40GWd/t(U))
    - More realistic creep burst test introduced for high burnup fuel
  - Minimum 1% uniform strain was reached by all fuel cladding tested before tertiary creep starts

- Understanding has been improved over the years through increasing the experimental dataset, development of creep models etc.

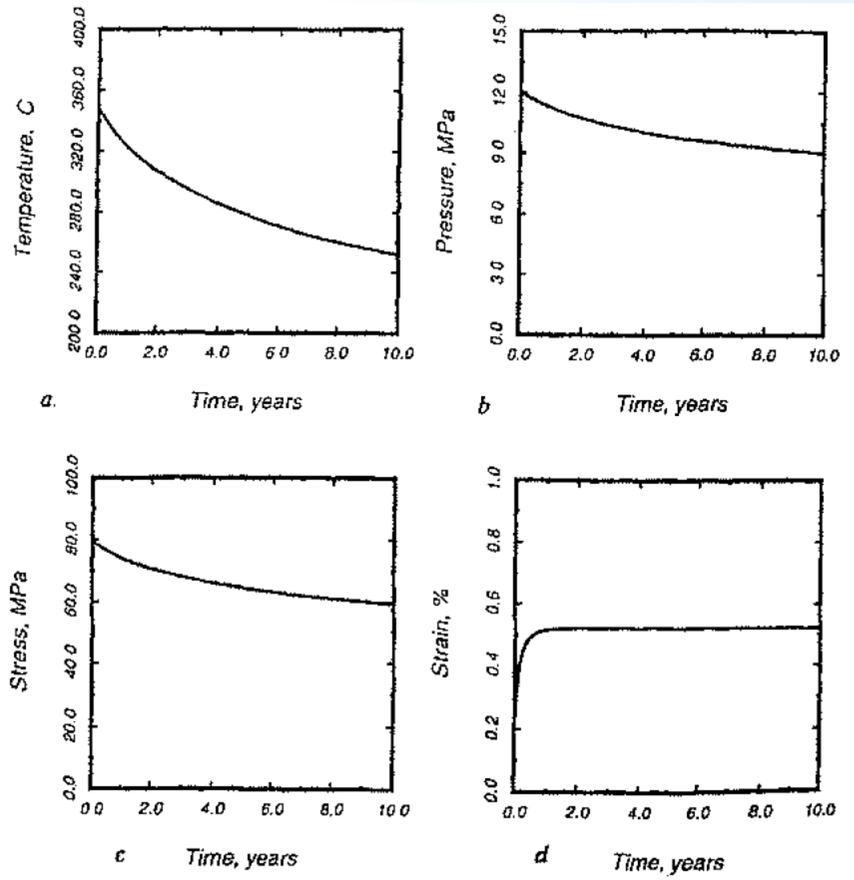
- 2003 US NRC concluded for fuel <40GWd/t(U) no gross rupture would occur provided the clad temperature does not exceed 400°C.

Thermal Creep – Dry Storage

- Thermal modes considered in dry storage:
  - Mode I -Rapid decrease in Temperature (range up to 400ºC to T= 300ºC)
  - Mode II – Medium decrease in Temperature (Range 300ºC- 200ºC)
    - All creep deformations considered to be terminated
  - Mode III Low to no decrease in Temperature (Range <200ºC)
    - associated >40+ years storage

- Temperature decreases with time, leads to a reduction in internal rod pressure and Stress
- Increasing fuel burnup increases clad corrosion and internal rod pressure
  - As hoop stress is a function of clad thickness and internal rod pressure then hoop stress will also increase
- MOX fuel has the highest stresses and strains in dry storage c.f. the equivalent UO_2 fuel

- Manufacturers responded by introducing new clad materials to reduce corrosion
  - For example MDA, M5, Zirlo



See References 2 & 3

Effect of hydrogen on creep deformation

- As fuel burnup is increased so does the hydrogen content in fuel clad
- Japanese studies (Reference 4) at various hydrogen contents, temperatures and hoop stresses
- For unirradiated clad
  - Hydrogen content below solubility limit: For BWR Zry-2 cladding, creep showed a slight acceleration by charged hydrogen, while no significant tendencies were observed for PWR Zry-4 cladding;
  - Hydrogen content above solubility limit: Creep suppressive effects increased with increased hydrogen content in PWR cladding, while, for BWR cladding, a slight accelerating effect was observed in low stress region, and suppressive effects were observed in the high stress region;
  - No effects on creep rate due to hydrides reoriented in the radial direction were observed.
- Are unirradiated tests bounding?
  - Creep strain of irradiated cladding was smaller than unirradiated cladding for all tests
  - Creep suppressive effects in irradiated cladding

PWR Creep Tests on HBU Zirlo

Spanish studies – ENRESA (& partners) on Development of ZIRLO creep laws and dry storage behaviour

- ZIRLO irradiated cladding up to 68 GWd/t(U) average rod

TEST MATRIX

Samples	oxide (μm)	ºC	MPa
2C-III	119	400	220
2C-II	111	400	190
2C-I	102	400	160
1C-I	49	360	220
1C-IV	66	380	220
1C-II*	60	400	190

Cool-down slower than 2ºC/min until 150ºC where H in solution considered negligible and then free cooldown.

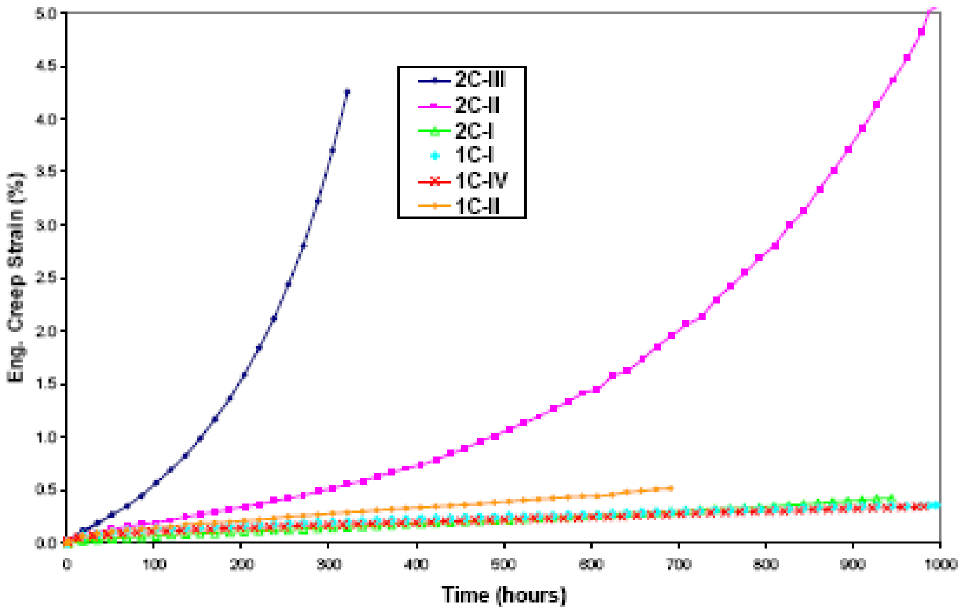
* With controlled cooling up to 300º C. Afterwards, keeping that temperature and 130 MPa hoop stress during 48 h

Other participants:

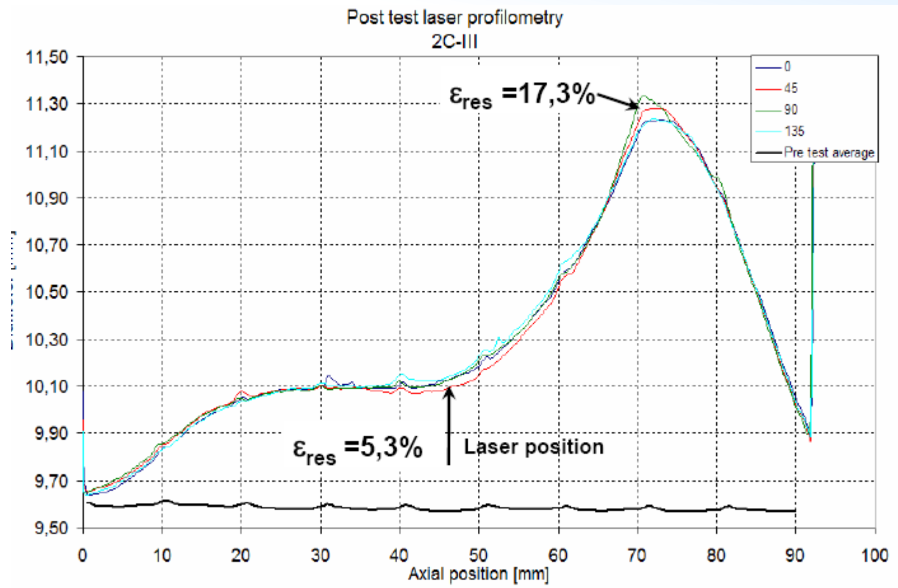
CSN CONSEJO DE SEGURIDAD NUCLEAR

ENUSA INDUSTRIAS AVANZADAS, S.A. GRUPO ENUSA

PWR Creep Tests on HBU Zirlo



Tertiary creep only in Samples 2C-III and 2C-II



Sample 2C-III: Failed

Detailed results reported to - Seoul 2008 Water Reactor Fuel Performance Meeting: Results of thermal creep tests on highly irradiated Zirlo.   Lloret (Enusa), Quecedo (Enusa), Conde (CSN), Gago (ENRESA), Fernández (ENRESA)

Summary - Creep

- The original 1% creep strain in storage criteria used in licensing spent fuel dry is bounding and considered to be very conservative

- Thermal creep does not appear to be a critical threat for spent fuel integrity as it is a self limiting phenomenon

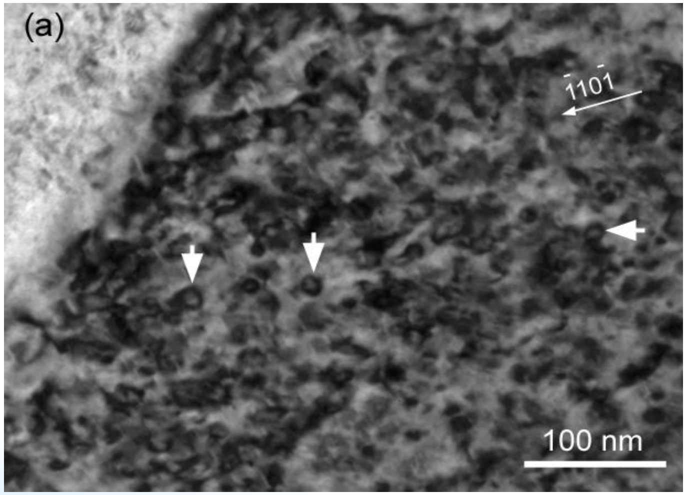
Cladding Embrittlement

- Fuel clad embrittlement is caused by
  - Neutron damage
  - Hydrogen adsorption and precipitation

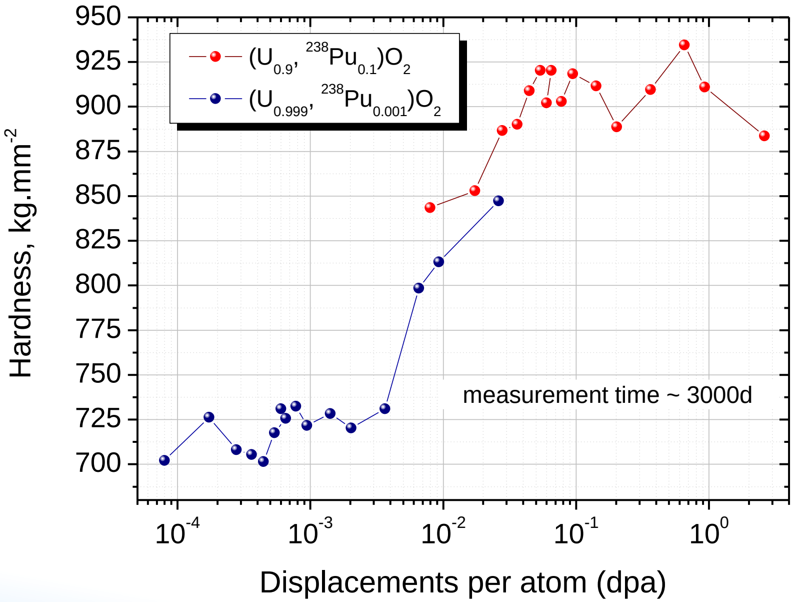
- Impact
  - Results in hardening of the material and loss of material ductility
  - The greater the loss in clad ductility the greater the potential for pin fracture during subsequent fuel handling

Neutron Damage

- Neutron damage
  - Results in the formation of point defects clusters (dislocation loops and voids)
  - Clusters act as a dislocation restricting their ability to glide under stress
  - Saturation of the effect is reached at ~5GWd/t(HM)
  - At elevated temperatures the damage is annealed



Example of neutron damage
Arrows show <a> dislocation loops



Hydrogen Behaviour in Zirconium Alloys

- Zirconium alloys absorb hydrogen through reaction with and radiolysis of the coolant
- The amount absorbed is alloy dependent. For 50 GWd/t(U), average hydrogen values for Zy-4 low tin is 600ppm, for M5® 100 ppm max.

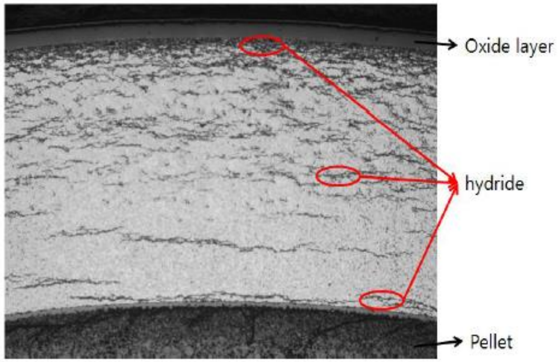
$$Zr + 2H_2O \longrightarrow ZrO_2 + 2H_2 \quad \text{- corrosion}$$

$$H_2O + \gamma_{energy} \Longrightarrow O_2 + H_2 \quad \text{- radiolysis.}$$

- When the fuel is initially cooled to the pool temperature, hydrogen in solid solution in the alloy precipitates in the form of hydride platelets primarily oriented in the circumferential direction

$$Zr + H_2 \longrightarrow \underbrace{ZrH_2}_{\text{ppt}^n\text{ hydride}} + \underbrace{H_2}_{\text{Solid Solution}} + \underbrace{H_2}_{\text{Trapped}}.$$

Hydrogen in 3 forms



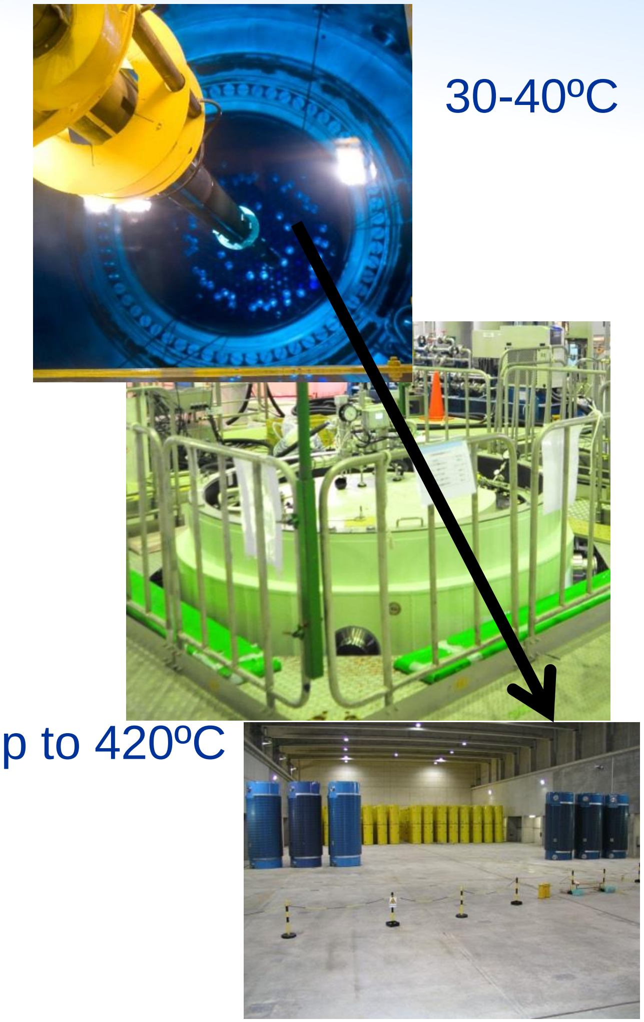
Cross section showing circumferential ZrH_2 precipitates (KAERI)

- Circumferential hydrides, in combination with hardening due to irradiation effects, decrease cladding ductility in response to axial and hoop loads, but the cladding retains enough residual ductility not to be impaired by fuel handling operations.

Transition from Wet to Dry Storage

- What is the issue?
- Potential for hydride reorientation
- Process:
  - As the fuel is transitioned from wet to dry the fuel clad temperature increases
  - Increasing the temperature causes ZrH_2 to dissolve
  - Results in an increase of the hydrogen in solid solution in the zirconium alloy matrix up to the hydrogen solubility limit.
  - The temperature increase also leads to an increase in the rod internal pressure (for example 4 to 9 MPa).
  - Subsequent cooling causes hydrogen in solid solution in the cladding materials to re-precipitate,
  - Possibility for hydrides to aligned in the radial and axial directions (or simply 'radial') due to the influence of the tensile hoop stress in the cladding; caused by the internal rod pressure that is no longer compensated by external means such as the coolant pressure in the reactor.
- Safety implication associated with subsequent fuel handling processes (accident conditions transport)

30-40ºC



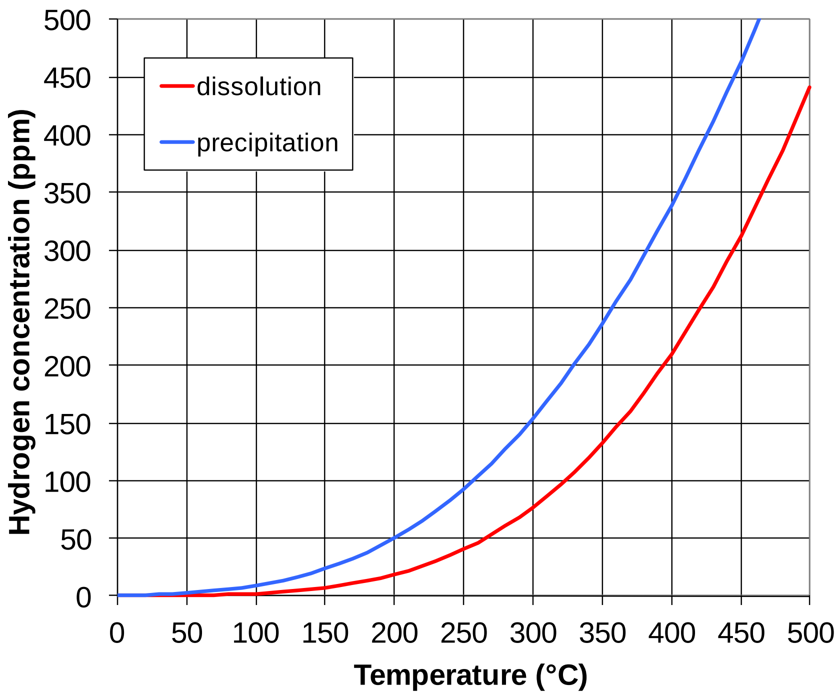
Up to 420ºC

Up to 400ºC

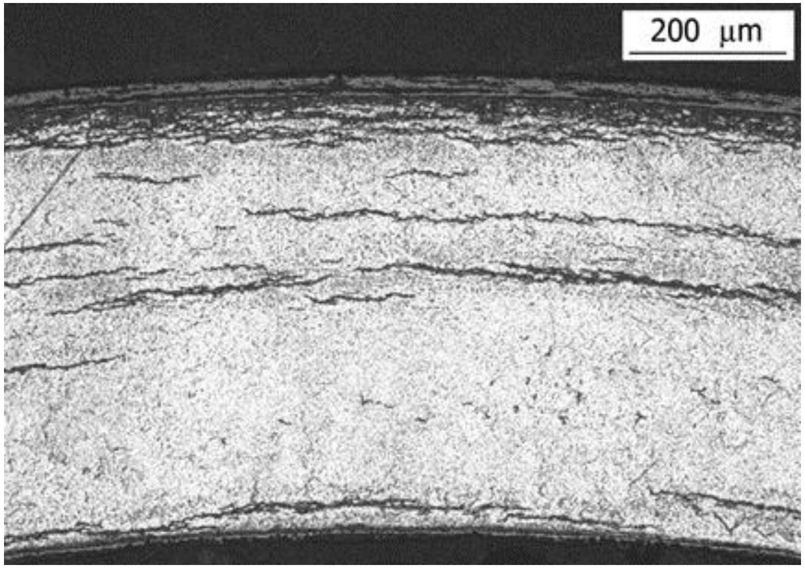
How much Hydrogen goes into solid solution

- For illustration purpose, the terminal solid solubility curves for dissolution and precipitation (TSSD and TSSP) established for non-irradiated alpha annealed zircaloy-4 are shown
- Heating from pool temperature to 420°C results in up to ~245 ppm of hydrogen going into solution in the alloy matrix (hydrogen content dependent)
- Upon cooling, hydride re-precipitation is initiated at ~335°C,
  - This is the expected behaviour when the material contains hydrogen at or near the solubility limit of 245 ppm.
- Assuming the alloy contained 600 ppm of hydrogen, then at 420°C the expectation is ~245 ppm of hydrogen will be in solution and the balance (~255 ppm) of hydrogen will remain as hydrides

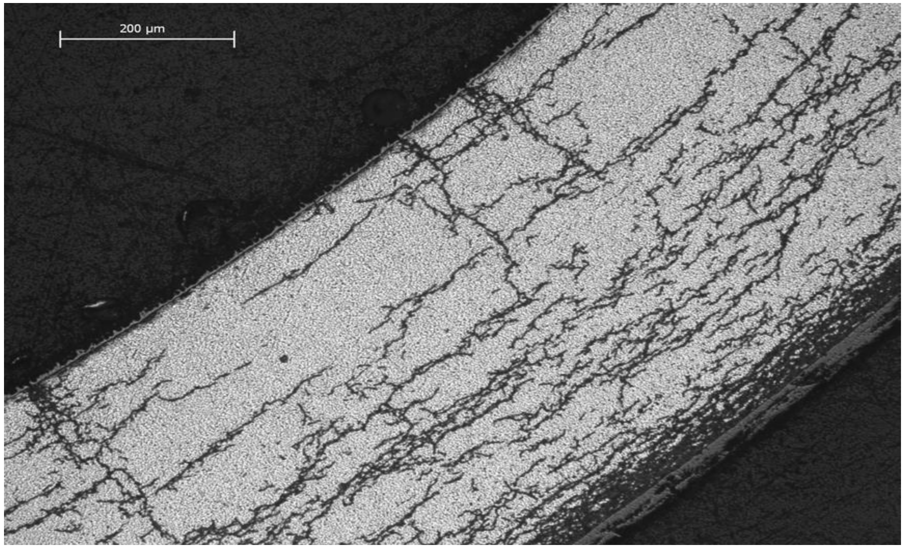
- Question is under what conditions does hydrogen in solid solution re-precipitate in the radial direction?



Example of Radial Hydride Treatment - High Burnup Zirlo™



As-irradiated with hydrogen content of 318±30 wppm



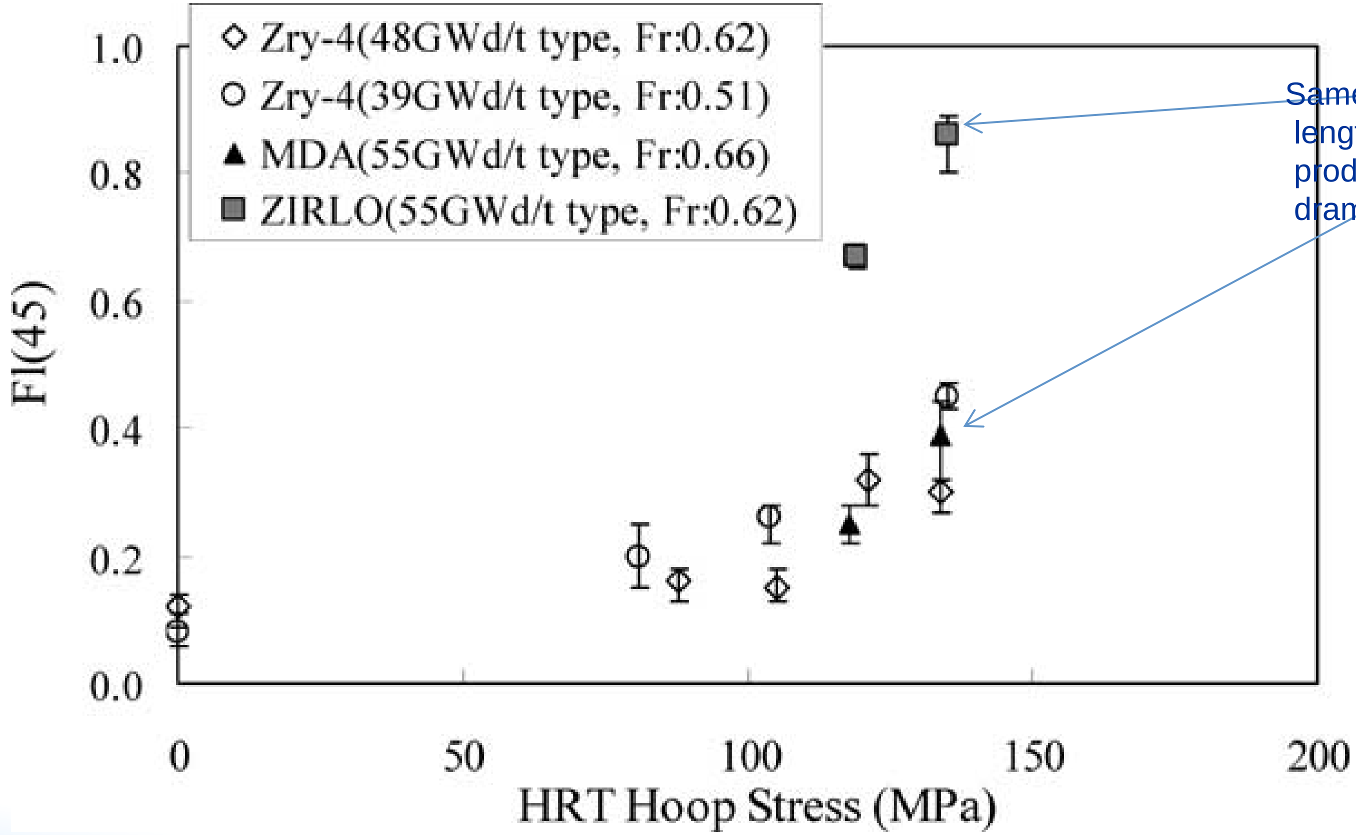
Clad sample subject to radial hydride treatment , 110 MPa hoop stress, heated to 400ºC, 24-h hold time, and then cooled at 5ºC/h
Hydrogen content - 325±72 wppm

Factors Which Can Affect Hydride Reorientation [5, 6]

- Material type
- Hydrogen content and distribution
- Fluence
- Temperature
- Applied stress
- Cooling rate
- Temperature history (e.g. cycling…)
- Impact of fuel pellet clad interaction

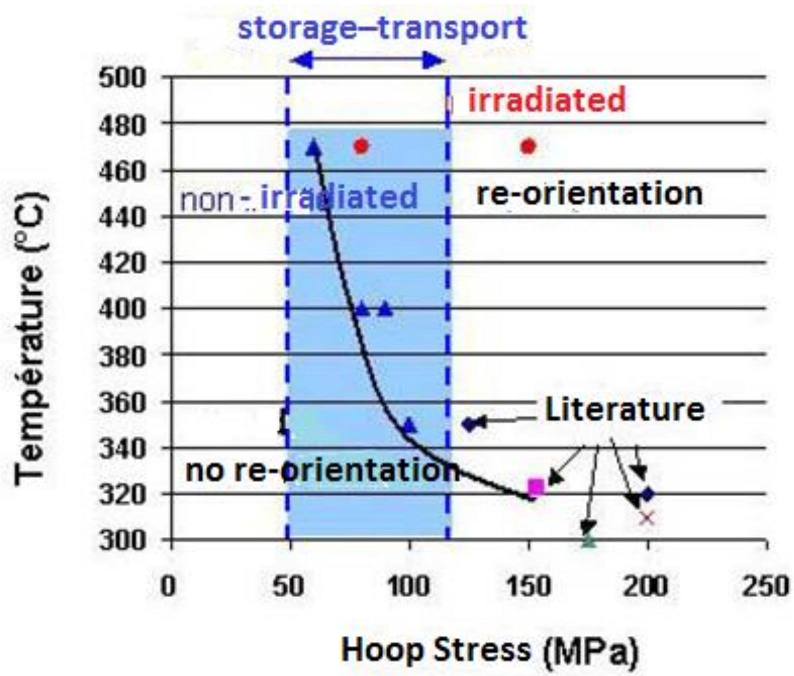
Cladding Material Studies

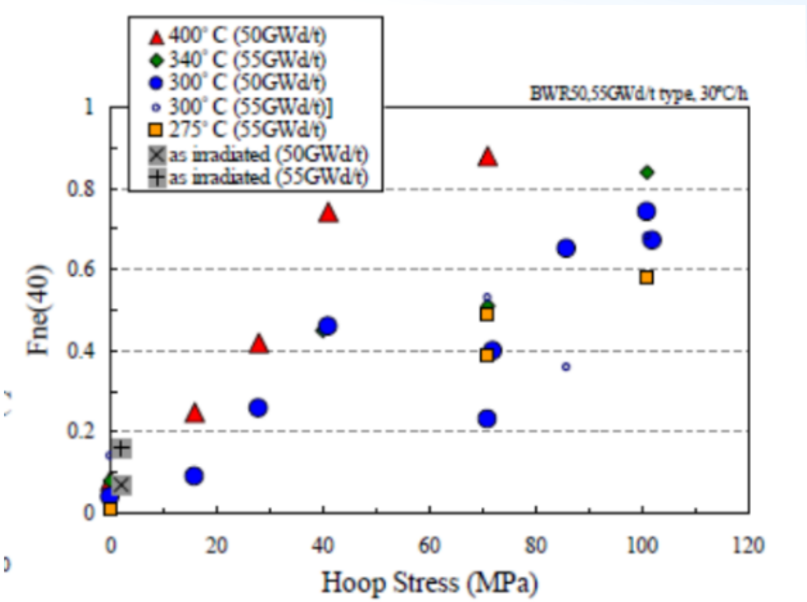
Japanese Studies



Same burnup & hoop stress, length of radial hydrides produced in the material dramatically different

Temperature Versus Applied Stress Studies



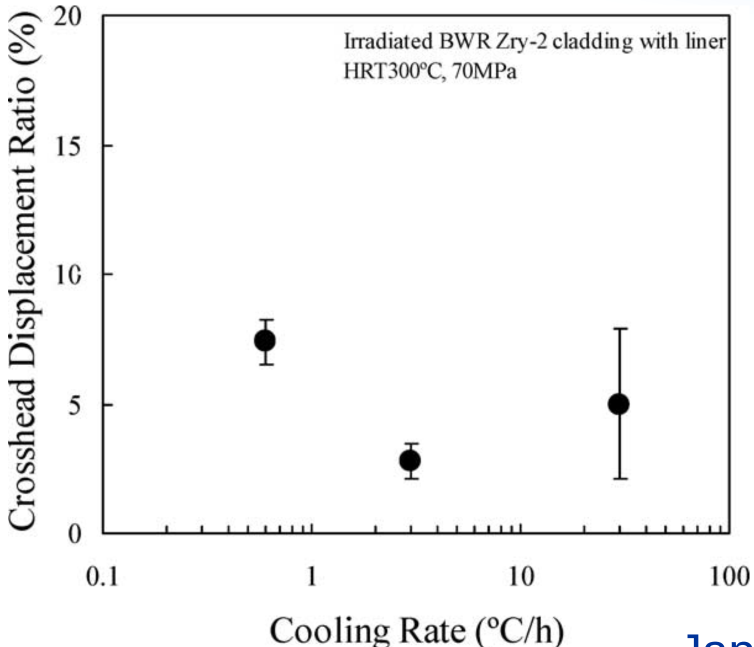


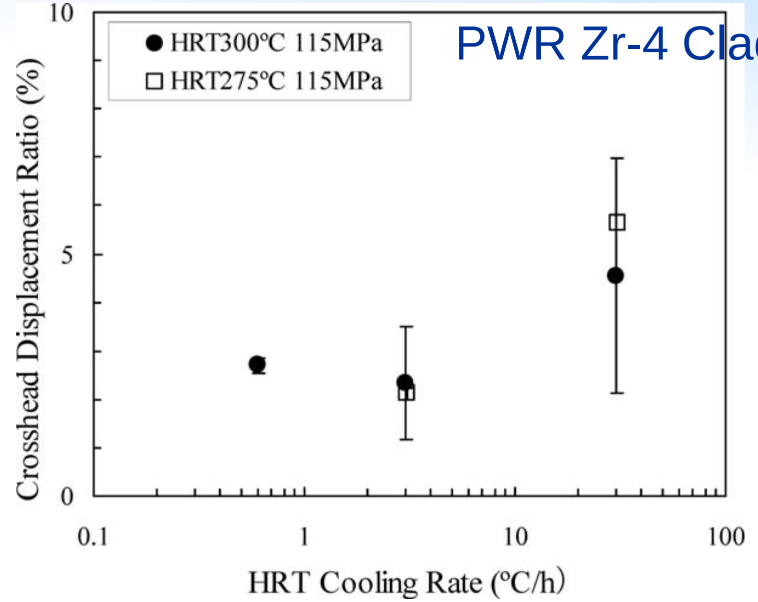
French investigations – AREVA TN sponsored

Japanese investigations – Zr-2 with liner

Higher the hoop stress the greater the potential for hydrogen to ppt^ in the radial direction

Cooling Rate Studies





PWR Zr-4 Cladding

Japanese studies

<u>Presence of a zirconium liner (BWR fuels) appears to be beneficial due to the difference in terminal solid solubility precipitation between the materials</u>

<u>Fuel that is rapidly cooled will be more ductile than fuel that is allowed to cool slowly</u>

Transport- Impact of fuel [5]

- Safety assessments which underpin the transport of fuel use assumptions on rod breakage and the amount of fuel released under accident conditions
  - Cask drop or external impact
  - <u>The amount of fuel released will impact the assumptions used in criticality assessments</u>

- With a move to high burnup fuel greater clad embrittlement and potential for hydride reorientation during fuel drying operations

- Need to establish whether existing assumptions are still bounding

JNES Studies Reported to IAEA SPAR III CRP

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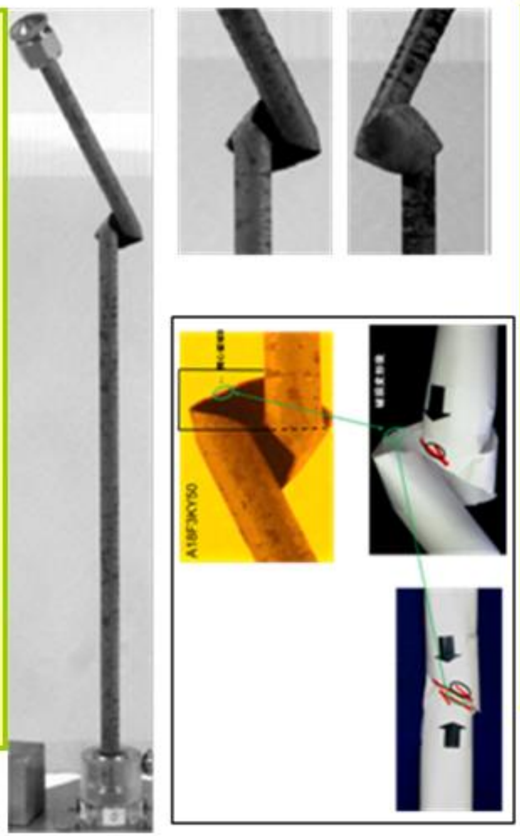
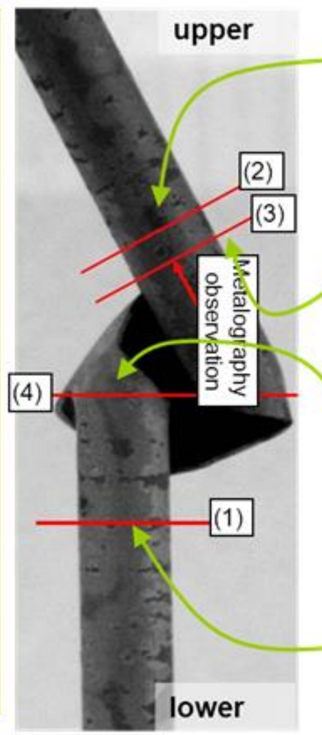
5.2-2 Breakage Morphology (Axial Dynamic Load Impact ; 3.5 kg-12 m/s)

Breakage position : 152 mm from upper end
Breakage length : about 40 mm

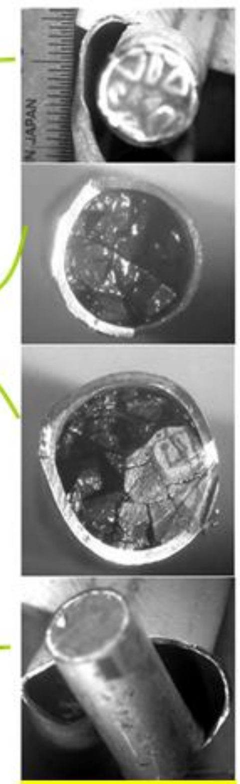
- ✓ S-shape deformation was not seen before the breakage at 0.1 – 0.2 ms after load impact.
- ✓ Sparks flied from the breakage part at 0.4 ms after load impact.
- ✓ Pushing displacement at spark was about 3 – 4 mm.



- ✓ Shearing breakage and spiral crack propagation occurred without buckling.
- ✓ It is thought that the influence on the pellet by load impact is restricted very close to breakage part.



(1) Rough cutting
(2) Rough cutting
(3) Metallography sample cutting
(4) SEM sample cutting

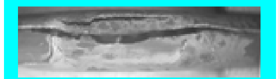


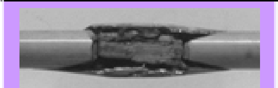
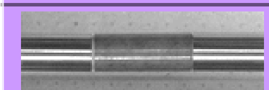
After-cutting Periscope Photograph

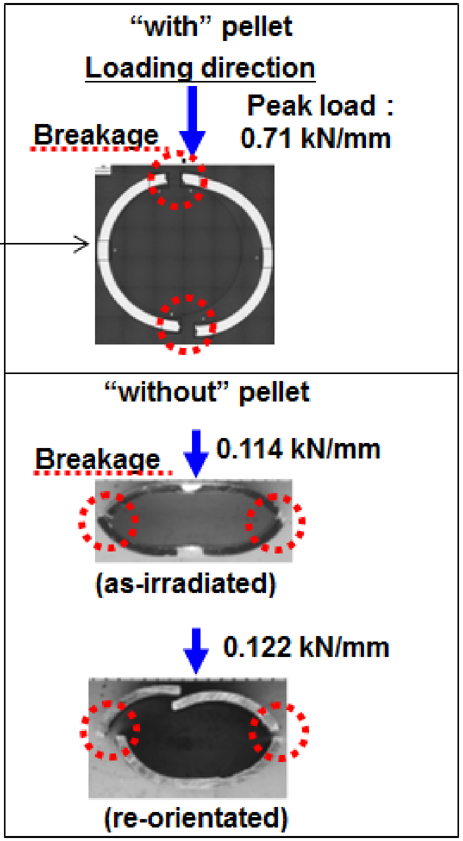
"Dynamic Load Test on SF" K.Kamimura, JNES (IAEA/SPAR-Ⅲ,Nov.4-8,2013, Busan Korea)

JNES Studies Reported to IAEA SPAR III CRP

Lateral Load Impact Test Result (BWR & PWR)

BWR fuel rod Post test	Weight mass	Impact speed	Peak load	Hydride re-oriented	Results
	2.60 kg	8.0 m/s	0.96 kN/mm	No	Failure Pellet dispersion
	2.60 kg	4.0 m/s	0.71 kN/mm	No	Failure
	0.63 kg	4.1 m/s	0.77 kN/mm	Yes *1	No-failure

PWR fuel rod Post test	Weight mass	Impact speed	Peak load	Hydride re-oriented	Results
	3 kg	12.9 m/s	1.5 kN/mm	No	Failure Pellet dispersion
	3 kg	11.0 m/s	1.1 kN/mm	No	Failure Pellet dispersion
	3 kg	10.9 m/s	1.4 kN/mm	Yes *2	No-failure*3



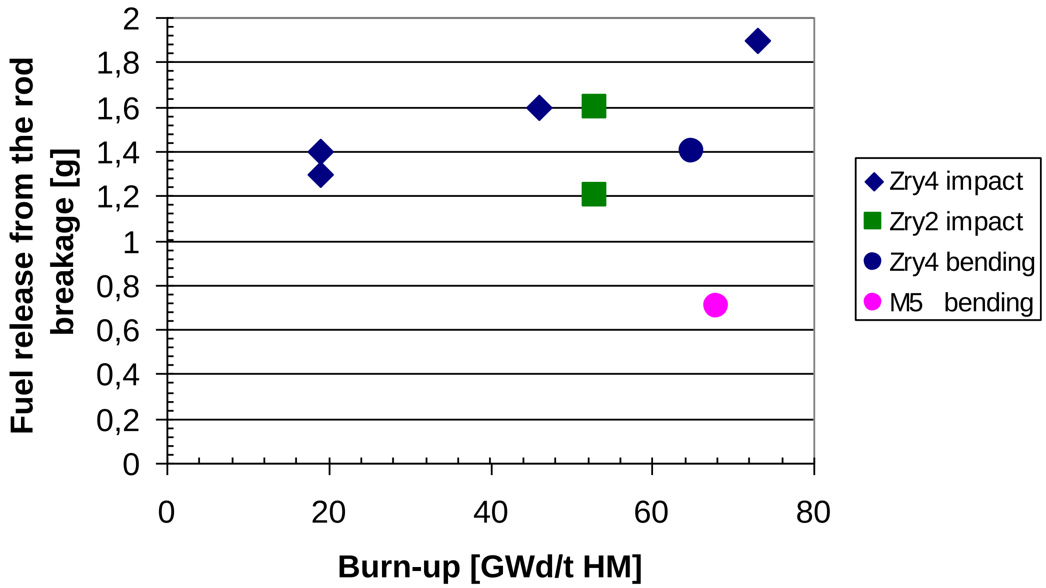
Hydride re-orientation treatment, *1 : 300°C_70MPa_30°C/h,
*2 : 300°C_115MPa_30°C/h

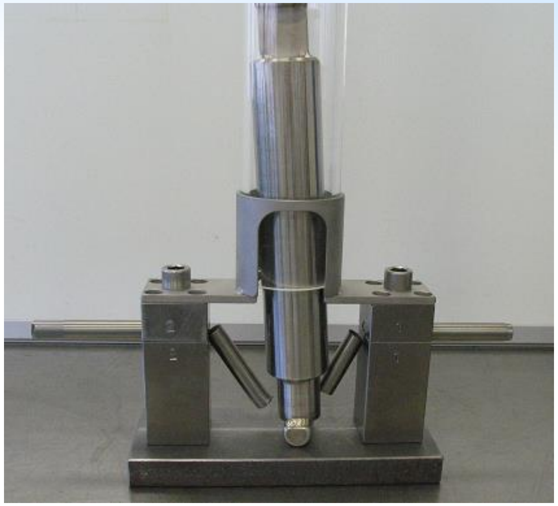
*3: This is due to support effect of dummy pellet which has higher strength than that of UO_2 pellet.

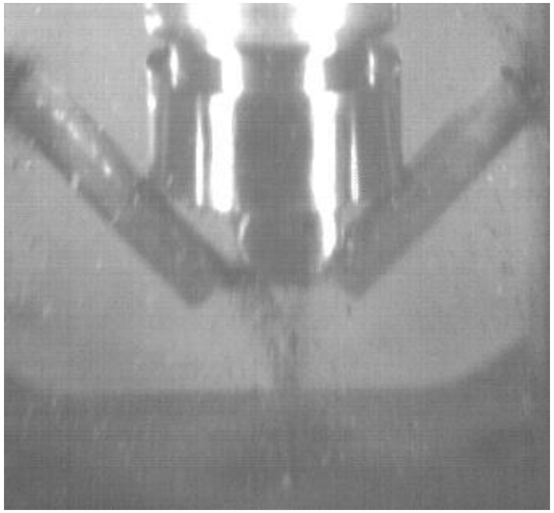
Failure mode and strength were different between "with" pellet (Lateral load impact test of fuel rod) and "without" pellet (Ring compression test of cladding).

GNS/ITU Reported to IAEA SPAR III CRP

Free fall hammer – test on HBU fuel rodlets







Summary - Embrittlement

Wet Storage

- Circumferential hydrides, in combination with hardening due to irradiation effects, decrease cladding ductility in response to axial and hoop loads, but the cladding retains enough residual ductility not to be impaired by fuel handling operations.

Transition Wet to Dry

- Studies to date have informed our understanding on how a number of parameters can influence hydrogen behaviour and the potential for hydrides to re-orientate
- The behaviour of fuel rods in real life may differ

Dry Storage

- Where hydrogen has gone into solid solution there is evidence to support that subsequent slow cooling in dry storage would increase the risk of the fuel clad being brittle

Transport

- Impact testing of HBU materials suggest that current safety assumptions on fuel debris accumulation under accident conditions are still bounding

Swelling

- Fuel pellet swelling can impact on fuel cladding stress and fuel pellet/clad interaction

- Post reactor operation the mechanisms which can lead to fuel pellet swelling (or volume increase) are:
  - Oxidation of exposed fuel
  - Helium accumulation/self irradiation

Fuel Oxidation

- <u>Only occurs where there is a breach in the fuel cladding</u>
  - normally associated with large defects >0.5mm

- In air it's a two stage process

$$UO_2 \rightarrow U_4O_9 \rightarrow U_3O_8$$

- Rate of conversion is temperature dependent
  - Below 250ºC the rate is very slow
- The formation of U_3O_8 results in a volume expansion >36%
- Where the defect is large this can result in axial or helical splitting of the fuel clad
- Where the defect is small the effected area is limited to the defect size and does not proceed beyond this

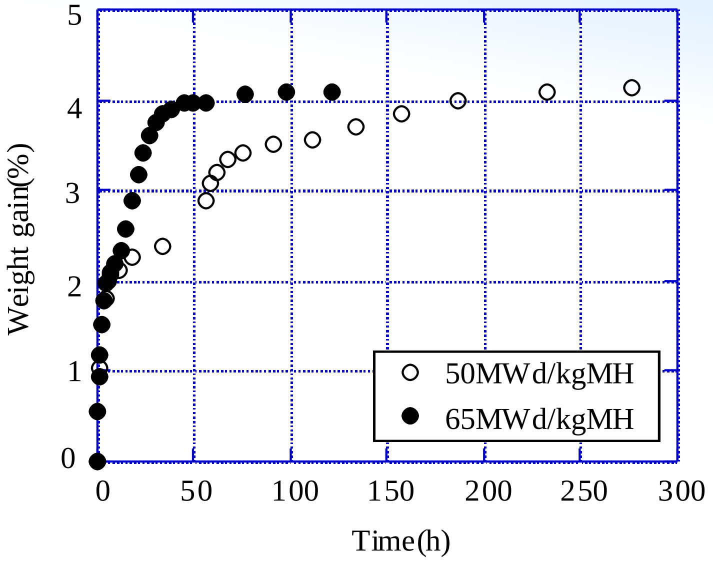
- Potential for conversion:
  - In oxygen rich pools
  - Failure of sealing arrangement dry storage
  - Drying of damaged fuel

Axial split

Helical split

Fuel Oxidation

- Safety consideration
  - Transfer of spent fuel from wet to dry or dry transport
  - What happens if damaged fuel is present
    - Potential to reach temperatures 420ºC during drying operations
    - Dry storage cask temperature limited to 400ºC



Fuel sample exposed to dry air at 350ºC

Self Irradiation – α-decay

- Isotopes which under go α-decay will lead to an accumulation of helium in the spent fuel matrix/fuel rod with time

$$ {}^{A}_{Z}X \xrightarrow[T_{1/2}.]{\alpha\text{-decay}} {}^{A-4}_{Z-2}Y + {}^{4}_{2}He\uparrow $$

$$ (\text{where } X = {}^{236}Pu,\ {}^{238}Pu,\ {}^{241}Pu\,(0.0023\%) \ldots\ldots) $$

- This could lead to an increase in fuel rod internal pressurisation and associated clad rupture or a failure of fuel rod end cap
- The implications of helium accumulation were considered by a number of institutions and programmes such as ‘STONE’ (Belgium) and ‘PRECCI’ (France) in the late 1990s/early 2000s

Self Irradiation – α-decay

- Is there any impact in interim storage?
- Studies of helium accumulation in 20 year stored BWR-MOX [7]
  - Calculated increase in helium after 20 years is twice the amount at discharge
  - Puncture gas analysis, for the period studied, implies that the gas is retained by the fuel pellet (see Table)
  - The potential for pellet swelling was also analysed by density measurements. Results were within the variance of literature values
- Self irradiation is considered in more detail in the lecture on "Radiation Effects in Spent Nuclear Fuel"

Fuel rod	W1 and A	W2 and D	W3 and B
He gas production[a] (cm^3/kgMOX)			
-Before storage	9.90	8.5	10.1
-After storage	20.1	17.9	20.4
He gas release[b] (cm^3/kgMOX)			
-Before storage	11.6	9.3	9.2
-After storage	9.7	7.9	9.8
He gas release (%)			
-Before storage	117.1	108.2	91.0
-After storage	48.3	44.2	48.1

[a]ORIGEN2/82 calculation; [b]measured.

Summary - Swelling

- Fuel pellet swelling can impact on fuel cladding stress and fuel pellet/clad interaction

- For spent fuel with large clad defects in the presence of an oxidative environment UO_2 will convert to U_3O_8 resulting in a significant volume increase which can lead to fuel clad splitting and the loss of fuel
  - This is the one of the reasons for damaged fuel to be conditioned for storage

- Although spent fuel self irradiation and helium accumulation does occur during interim storage, the evidence to date suggests that the helium is retained by the fuel pellet and no significant pellet swelling occurs for the time periods currently under consideration (i.e. <100 years)

References

1. INTERNATIONAL ATOMIC ENERGY AGENCY, Spent Fuel Performance Assessment and Research: Final Report of a Coordinated Research Project (SPAR-II), IAEA-TECDOC-1680, IAEA, Vienna, (March 2012) 49-76.
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Thank you!