The Effect of Microstructure on Delayed Hydride Cracking Behaviour in Zircaloy-4 Fuel Cladding – An IAEA Coordinated Research Programme

DHC CRP-II

The objective:

to evaluate the effect of microstructure on DHC velocity in Zircaloy-4 cladding; the experimental methods have been transferred so that consistent procedures are used to obtain reproducible measurements.
Cladding materials – Zircaloy-4

- LWR
  - Cold-worked (CW)
  - PWR Stress-relieved, 480 °C, 3.5 h (CWSR)
  - BWR Recrystallized, 565 °C, 1.5 h (RXA)
- CANDU
  - Two fabricators, 500 °C, 8 h
  - Atucha, 510 °C, 8.5 h
## Cladding materials - dimensions

<table>
<thead>
<tr>
<th></th>
<th>Outside diameter mm</th>
<th>Wall thickness mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>LWR</td>
<td>9.5</td>
<td>0.6</td>
</tr>
<tr>
<td>CANDU</td>
<td>13.1</td>
<td>0.39</td>
</tr>
<tr>
<td>Atucha</td>
<td>11.9</td>
<td>0.55</td>
</tr>
</tbody>
</table>
# Microstructure

<table>
<thead>
<tr>
<th>reactor</th>
<th>Grain size µm</th>
<th>Grain shape</th>
<th>Texture $F_R$</th>
</tr>
</thead>
<tbody>
<tr>
<td>LWR CW</td>
<td></td>
<td>Elongated</td>
<td>0.65</td>
</tr>
<tr>
<td>PWR</td>
<td></td>
<td>Elongated</td>
<td>0.64</td>
</tr>
<tr>
<td>BWR</td>
<td>3</td>
<td>Equiaxed</td>
<td>0.60</td>
</tr>
<tr>
<td>CANDU Zircatec</td>
<td>4</td>
<td>Elongated/Equiaxed</td>
<td>0.65</td>
</tr>
<tr>
<td>CANDU Sandvik</td>
<td>6</td>
<td>Elongated/Equiaxed</td>
<td></td>
</tr>
<tr>
<td>Atucha</td>
<td>2 to 3</td>
<td>Elongated/Equiaxed</td>
<td>0.56</td>
</tr>
</tbody>
</table>
## Strength

<table>
<thead>
<tr>
<th></th>
<th>UTS (250 °C) MPa</th>
</tr>
</thead>
<tbody>
<tr>
<td>LWR CW</td>
<td>532</td>
</tr>
<tr>
<td>PWR CWSR</td>
<td>510</td>
</tr>
<tr>
<td>BWR RXA</td>
<td>270</td>
</tr>
<tr>
<td>CANDU Zircatec</td>
<td>392</td>
</tr>
<tr>
<td>CANDU Sandvik</td>
<td>424</td>
</tr>
<tr>
<td>Atucha</td>
<td>367</td>
</tr>
</tbody>
</table>
Specimen preparation & testing

- 200 ppm hydrogen added
- Circumferential hydrides
- 13 mm long section
- Two notches extended 1.5 mm by fatigue
- Crack detection by potential drop or crack opening
- Testing in temperature range 150 to 300 °C
Pin-Loading Tension Specimen, Fixture and Assembly (Studsvik Nuclear)
Test history  $K_i \ 15 \text{ MPa}\sqrt{\text{m}}$
Fracture history
DHC velocity in PWR cladding
DHC in BWR cladding (Zircaloy-4)
DHC in LWR cladding
DHC in non-LWR cladding

![Graph showing DHC in non-LWR cladding](image)
Comparison of claddings

![Graph comparing claddings with different materials and temperatures.](image-url)
Fatigue to DHC transition

Crack Direction

Fatigue

DHC
DHC to ductile fracture transition
Striations

Zr-2.5Nb

Zircaloy-4
Summary of observations

- Zircaloy-4 fuel cladding can fracture by DHC
- Velocity passes through maximum value of slightly $>10^{-7}$ m/s (8.6 mm/day) at 280 °C
- Between 150 and 280 °C Arrhenius behaviour: $V=A\exp(-Q/RT)$; $Q\approx55$ kJ/mol
- $V$ scattered in recrystallized material
- No striations on fracture surface of Zircaloy-4
Some potential explanations

- Recrystallized material: $K_{IH}$
- Importance of texture and strength
- High temperature behaviour: $K_{IH}$
Definition of $K_{IH}$
Possible explanation for behaviour of RXA material

![Graph showing the relationship between V (m/s) and Ki (MPa√m) at 250 °C. The graph includes data points for RXA, CW, and CWSR.]
Possible explanation for behaviour of RXA material

![Graph showing possible explanation for behaviour of RXA material at 200 °C. The graph plots V m/s against KI MPa √m, with data points for RXA and CW & CWSR.](Image)
Comparison with CW Zircaloy-2 pressure tubes

![Graph showing comparison of V (m/s) vs 1/T (1/K) with various data points for different temperatures and experimental setups.]

- Mills & Huang
- Puls
- CW this paper
- CWSR this paper

Temperature: 0°C
Strength – a source of different values of V

![Graph showing the relationship between UTS (MPa) and V (m/s) at 250 °C. The graph includes data points for Zircaloy-4 cladding, RXA, and Zircaloy-2 pressure tubes.](image-url)
Reasons for drop-off at high temperature

- Insufficient hydrogen
  - **NO**: hydrides present up to 360 °C

- Temperature history
  - **NO**: cooled 50 to 75 °C to test temperature

- Applied $K_I$ approaching $K_{IH}$
  - **POSSIBLE**: analogy with Zr-2.5Nb
Comparison with Zr-2.5Nb pressure tube material
Demonstration of $K_{IH}$ change with $T$ in CW Zr-2.5Nb pressure tube

(Resta Levi)
Conclusions

• In Zircaloy-4 fuel cladding DHC velocity has two temperature regimes:
  • $< 280 \, ^\circ\text{C}$ follows Arrhenius behaviour
  • $> 280 \, ^\circ\text{C}$ $V$ drops off; possibly because $K_I \sim K_{IH}$
• $V$ depends on strength but not suppressed by radial texture
• In RXA material, $V = f(K_I)$; possible source of variation
• Striations not fundamental feature of DHC?
Plans for future

- New IAEA CRP being proposed under umbrella of Hydrogen Degradation.
- $K_{IH}$ is potential new focus
- Different materials – Zirlo, M5, other new materials
- Looking for volunteers for ideas and materials
Axial vs radial cracking (Sakamoto)

The diagram shows the relationship between temperature (in °C), V (m/s), and 1/T (1/K) for both axial and radial cracking. The data points are plotted for different temperatures, with logarithmic scales for V (m/s) and 1/T (1/K) to show the variation in cracking velocity with temperature.
Reproducibility (CWSR)

Temperature $^\circ$C

$1/T$ (1/K)

$V$ (m/s)

83786

86080
V vs crack length

CWSR 250 °C
Importance of Thermal History

Schematic illustration of DHCV temperature dependence

\[ V_{DHC} = V_0 \exp \left( \frac{-Q}{RT} \right) \]

\[ \log \text{DHC Velocity, } V_{DHC} \]

\[ V_{DHC} = 0 \]

\[ T_4, T_5, T_6, T_1, T_2, T_3 \]
Diffusion First Model

\[ \nu_c (T) = WD \left[ C(b) - C_P (T) \exp \left( \frac{\sigma_h V_H}{RT} \right) \right] \]

where

\[ \sigma_h = -2.4 \sigma_y \] Mode 1 plain-strain perfectly-plastic crack tip

\[ C(b) = C_t \] for \( T > T_{\text{TSSP}} \)

\[ C(b) = C_P (T_t) \] for \( T < T_{\text{TSSP}} \)
Diffusion first model

Crack velocity = constant x hydrogen diffusivity x 

\[ \text{[H concentration in solution in bulk –} \]

\[ \text{[TSSP x exp(-}\sigma_H V_H/RT)] \]

Just need D, TSSP, V_H, \sigma_Y (for \sigma_H)
Efficacy of DFM prediction

\[ \ln V = \frac{1}{T} \]

- Zircaloy CWSR measured
- Calculated Tpeak = Ttest + 50
- Calculated Tpeak = Ttest + 75

1/T
Distinguishing Experiments

- Diffraction
- Specimen thickness
- Incubation time
Pin-Loading Tension Specimen, Fixture and Assembly (Studsvik Nuclear)
Determination of $K_I$

$$K_I = \left[ \frac{P}{(2t\sqrt{W})} \right] f(a/W)$$

$f(a/W) = \text{geometry correction factor, determined experimentally from compliance measurements.}$

For LWR cladding:

$$f(a/W) = 92.203 - 468.73(a/W) + 787.15(a/W)^2 - 360.99(a/W)^3$$
Бороздки в изломах оболочечных труб

250 °C

Zircaloy-4
Zr-2,5Nb
Zr-1Nb
CRP “Hydrogen and Hydride Degradation of Mechanical and Physical Properties of Zirconium Alloys”

DHC-I “Delayed Hydride Cracking in Zr alloys in pressure tube nuclear reactors” 1998-2002

(http://www-pub.iaea.org/MTCD/publications/PDF/te_1410_web.pdf)
J ASTM International Paper ID JAI 101091

DHC-II “Delayed Hydride Cracking in Zr alloy fuel cladding” 2005 - 2009

J ASTM International Paper ID JAI 103008