Phenomenon of (irradiation assisted) stress corrosion cracking for internals of PWR & BWR systems

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http://www.jrc.ec.europa.eu

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CONTENT

- Ageing
- Corrosion
- SCC – PW SCC
- Radiation effects
- IASCC
- CONCLUSIONS
- Effort to plan life management of ageing NPPs
- Reliability of in-core in-vessel structural materials
- Control of the degradation of the structural materials
- Guiding replacement campaigns

- Rising issues for alloys in LWR conditions:
  - Corrosion and
  - Stress Corrosion Cracking (SCC PW SCC)
  - IASCC
Industry’s Top Ten R&D Priorities (from MRP-205)

1. Inspection & Evaluation (I&E) Guidelines: Reactor Internals
2. NDE Technology: Dissimilar Metal (DM) Butt Welds
3. PWSCC Mitigation: Environmental Controls
4. I&E Guidelines: Bottom Mounted Nozzles
5. Vibration Fatigue: Small Bore Piping
7. NDE Qualification Program: Ni-Alloy Penetrations
8. NDE Accessibility: Reactor Internals
9. PWSCCC Mitigation: Stress Improvement (SI) of Butt Welds
10. Thermal & Irradiation Embrittlement: Synergistic Effects on CASS & SS Welds - Internals
Essential service water system pipe degradation
Stress Corrosion Cracking

- **Attachment welds**: SCC significant at welded pad/bracket locations in the vessel shell
- **Nozzles**: SCC of nozzles is a significant issue
- **Closure studs**
- **Penetrations**: SCC of CRD stub tubes (high residual stresses in sensitized weld material)
- **Safe ends**: Observed at several plants. SCC is a potentially significant degradation mechanism for safe ends
## Main Degradation Mechanisms

<table>
<thead>
<tr>
<th>Component</th>
<th>Irr. Emb.</th>
<th>Fatigue</th>
<th>Thermal Ageing</th>
<th>Wear</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reactor pressure vessel</td>
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<tr>
<td>Control rod drive mechanisms</td>
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<tr>
<td>Internals structures</td>
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<td>Reactor coolant pump easing</td>
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<tr>
<td>Piping and safe ends</td>
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<tr>
<td>Pressurizer</td>
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<tr>
<td>Surge and Spray lines</td>
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<tr>
<td>Steam generator tubing</td>
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<tr>
<td>Steam generator shell and nozzles</td>
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</tbody>
</table>
Lovisa
PWSCC Experience in Vessel and Other Components

Plants with leaks:

Plants with cracks/indications:
- Ringhals 3, 4 – axial, reactor vessel nozzle (2000)
Corrosion & radiation

- Effects of irradiation on materials are well investigated
  - Defects due to irradiation
  - Mechanical properties evolution with dose

- For LWRs, passive materials used
- Corrosion enhanced by water radiolysis
  - oxidant species (OH\(^-\), H\(_2\)O\(_2\), etc.)
  - together with reducing species (H, etc)
  - influence free corrosion potential
- Radiation affects also semi-conductive properties of the oxides, particularly the behaviour of the passive layer
Irradiation hardening increases with \((\text{dose})^{1/2}\)
Radiolysis of Water by n- and $\gamma$ Radiation

\[ H_2O + (\gamma, n) \rightarrow H_2, H_2O_2, HO_2^*, OH^*, H^*, e_{aq}^-, H^+ \]

- **Excitation**
  - $H_2O^*$

- **Primary products formation**
  - $H_2, H_2O_2, HO_2^*, OH^*, H^*, e_{aq}^-$

- **Molecules formation**
  - $H_2, H_2O_2, O_2$

**Radiolysis**
- mainly in the reactor core region
- mainly by n-irradiation
- $\gamma$ is important for recombination

**Redox-potential and ECP corrosion behaviour**

- **PWR**: $H_2$-addition (in VCT) → recombination with $O_2 + H_2O_2$ to $H_2O$ → $H_2 \gg O_2 + H_2O_2 < 5$ ppb
  → low redox-potential → ECP = -800 to -500 mV$_{SHE}$

- **BWR/NWC**: $H_2O_2$ is non-volatile → $O_2 + \frac{1}{2} H_2O_2 > H_2$ → oxidizing → ECP = -50 to +200 mV$_{SHE}$

- **BWR/HWC or NMCA**: $H_2$-addition to feedwater → recombination with $O_2 + H_2O_2$ to $H_2O$ → $H_2 \gg O_2 + H_2O_2 < 5$ ppb → low redox-potential → ECP = -500 to -200 mV$_{SHE}$
- The integrity of fuel elements (zircaloy) may also be affected
- Localised corrosion phenomena are also affected by the free corrosion potential changes, particularly SCC phenomena
+ Radiation

Corrosion

IASCC

Stress

Radiolysis

Radiation corrosion

Material

Water

Stress Corrosion Cracking

Fracture Mechanics

Radiation creep

Radiation induced segregation
Present generation

LWR $\rightarrow$ BWR, PWR, VVER
HW $\rightarrow$ CANDU
GCR $\rightarrow$ MAGNOX, AGR
LM $\rightarrow$ FR
BWR - PWR

- Interactions corrosion behaviour and radiolysis
- Slightly diversified for the two main LWRs
  - Boiling water reactors (BWR) and
  - Pressurised water reactor (PWR)
# Water Chemistry Conditions in BWR and PWR

<table>
<thead>
<tr>
<th>BWR/NWC</th>
<th>BWR/HWC</th>
<th>PWR</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>T</strong> = 274 – 288 °C</td>
<td><strong>T</strong> = 274 – 288 °C</td>
<td><strong>T</strong> = 286 – 323 °C</td>
</tr>
<tr>
<td><strong>pH_T</strong> = 5.65</td>
<td><strong>pH_T</strong> = 5.65</td>
<td><strong>pH_T</strong> = 6.8 to 7.4</td>
</tr>
<tr>
<td>Neutral</td>
<td>Neutral</td>
<td>Slightly alkaline</td>
</tr>
<tr>
<td>High-purity water</td>
<td>High-purity water</td>
<td>Pure water</td>
</tr>
<tr>
<td>Impurities &lt; 1ppb</td>
<td>Impurities &lt; 1ppb</td>
<td>H₃BO₃/LiOH</td>
</tr>
<tr>
<td>O₂ ~ 200 ppb</td>
<td>O₂ &lt; 5 ppb</td>
<td>O₂ &lt; 5 ppb</td>
</tr>
<tr>
<td>H₂ ~ 15 ppb</td>
<td>H₂ ~ 50 to 300 ppb</td>
<td>H₂ ~ 2000 to 5000 ppb</td>
</tr>
<tr>
<td>ECP = -50 to +200 mV&lt;sub&gt;SHE&lt;/sub&gt;</td>
<td>ECP = -500 to -200 mV&lt;sub&gt;SHE&lt;/sub&gt;</td>
<td>ECP = -800 to -600 mV&lt;sub&gt;SHE&lt;/sub&gt;</td>
</tr>
</tbody>
</table>
**BWR - PWR**

**BWRs**: stainless steels (304 type) mainly used for core components
- exposed to water often up to 288 °C
- SCC is controlled by hydrogen conditioning (HWC: H water chemistry)
  - free corrosion potential (ECP) at low values

**PWRs**
- primary water chemistry based on B (added as boric acid)
- for neutronic purposes with Li addition (added as LiOH)
- increase pH to limit the general corrosion and activation of components
- large hydrogen concentration used to limit radiolysis effects
  - 25 to 35 ml of hydrogen per Kg water!
Boric Acid Corrosion

- Boric acid leakage is a consequence of Alloy 600 cracking
- This leakage can lead to boric acid corrosion of low-alloy steel
- Davis-Besse, March 2002

- Order EA-03-009. Inspection requirements according to the parameter EDY

Davis-Besse
Davis-Besse
Plants with Replaced RPV Upper Head - USA

- 25 plants replaced RPV upper heads
  - 24 with Alloy 690 penetrations
  - 1 with Alloy 600 (Davis-Besse)
- 13 of the 21 remaining high and moderate susceptibility plants

**PWSCC. Vessel Head**

- The Alloy 600/82/182 has been changed by the Alloy 690/52/152 in the penetrations of the new vessel heads
- The inspection practices vary from country to country, reducing the inspection intervals for the vessel heads with Alloy 600
Stress Corrosion Cracking

- BWR internals susceptible to two forms of SCC:
  - Intergranular stress corrosion cracking (IGSCC)
  - Irradiation assisted stress corrosion cracking (IASCC)

- Degradation via IGSCC is potentially significant. Programmes to effectively manage this degradation mechanisms are required

- IASCC is a concern in BWR core internal components such as a portion of the core shroud and the top guide.
Degradation Incidents of RPVIs Safety Relevant

<table>
<thead>
<tr>
<th>Component</th>
<th>Degradation Mechanism</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Core Plate</td>
<td>IGSCC</td>
</tr>
<tr>
<td>2. Core Spray Internal Piping</td>
<td>IGSCC</td>
</tr>
<tr>
<td>3. Core Spray Sparger</td>
<td>IGSCC</td>
</tr>
<tr>
<td>4. CRD Guide Tube</td>
<td>No incidents of cracking reported</td>
</tr>
<tr>
<td>5. CRD Housing</td>
<td>No incidents of cracking reported</td>
</tr>
<tr>
<td>6. In-Core Housing</td>
<td>IGSCC</td>
</tr>
<tr>
<td>7. Jet Pump</td>
<td></td>
</tr>
<tr>
<td>- Diffuser</td>
<td>IGSCC</td>
</tr>
<tr>
<td>- Hold down beam</td>
<td>IGSCC</td>
</tr>
<tr>
<td>- Inlet mixer</td>
<td>Fatigue due to improper installation</td>
</tr>
<tr>
<td>- Riser</td>
<td>IGSCC</td>
</tr>
<tr>
<td>8. LPCI Coupling</td>
<td>No incidents of cracking reported</td>
</tr>
<tr>
<td>9. Orificed Fuel Support</td>
<td>No incidents of cracking reported</td>
</tr>
<tr>
<td>10. Core shroud</td>
<td>IGSCC/IASCC</td>
</tr>
<tr>
<td>11. Shroud Support</td>
<td>IGSCC</td>
</tr>
<tr>
<td>12. Top Guide</td>
<td>IGSCC/IASCC</td>
</tr>
</tbody>
</table>
Mitigation Technologies for SCC

Water chemistry control or surface treatment

- Hydrogen water chemistry
- Noble Metal Chemical Application (NMCA)
- Deposition of noble metals by plasma spray
- Surface melting/Solution annealing
+ Radiation

Corrosion

IASCC

Stress

Radiolysis

Radiation corrosion

Water

Stress Corrosion Cracking

Fracture Mechanics

Radiation creep

Radiation induced segregation

Material
Some Effects of Irradiation

- Radiolysis of reactor coolant by \( n \)- and \( \gamma \)-irradiation
  - change of oxidizing capacity of environment → corrosion, EAC

- Change of microstructure of reactor internals and RPV by \( n \)-irradiation
  - displacement damage
  - irradi.-induced hardening, embrittlement, segregation, creep, void swelling,…
  - IASCC of reactor internals, \( n \)-embrittlement of RPVs

- Transmutation and activation
  - activation of corrosion products, activity transport and built-up in piping system
  - dry well dose rates (Co-60, which is strong \( \gamma \)-emitter, is created by thermal neutron activation of natural Cobalt 59 (from wear-resistant Co-alloys))

- \( \gamma \)-heating
# IASCC Service Experience

<table>
<thead>
<tr>
<th>Component</th>
<th>Material</th>
<th>Reactor Type</th>
<th>Possible Sources of Stress</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fuel Cladding</td>
<td>304 SS</td>
<td>BWR</td>
<td>Fuel Swelling</td>
</tr>
<tr>
<td>Fuel Cladding</td>
<td>304 SS</td>
<td>PWR</td>
<td>Fuel Swelling</td>
</tr>
<tr>
<td>Fuel Cladding *</td>
<td>20%Cr/25%Ni/Nb</td>
<td>AGR</td>
<td>Fuel Swelling</td>
</tr>
<tr>
<td>Fuel Cladding Ferrules</td>
<td>20%Cr/25%Ni/Nb</td>
<td>SGHWR</td>
<td>Fabrication</td>
</tr>
<tr>
<td>Neutron Source Holders</td>
<td>304 SS</td>
<td>BWR</td>
<td>Welding &amp; Be Swelling</td>
</tr>
<tr>
<td>Instrument Dry Tubes</td>
<td>304 SS</td>
<td>BWR</td>
<td>Fabrication</td>
</tr>
<tr>
<td>Control Rod Absorber Tubes</td>
<td>304/304L/316L SS</td>
<td>BWR</td>
<td>B4C swelling</td>
</tr>
<tr>
<td>Fuel Bundle Cap Screws</td>
<td>304 SS</td>
<td>BWR</td>
<td>Fabrication</td>
</tr>
<tr>
<td>Control Rod Follower Rivets</td>
<td>304 SS</td>
<td>BWR</td>
<td>Fabrication</td>
</tr>
<tr>
<td>Control Blade Handle</td>
<td>304 SS</td>
<td>BWR</td>
<td>Low stress</td>
</tr>
<tr>
<td>Control Blade Sheath</td>
<td>304 SS</td>
<td>BWR</td>
<td>Low stress</td>
</tr>
<tr>
<td>Control Blades</td>
<td>304 SS</td>
<td>PWR</td>
<td>Low stress</td>
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<tr>
<td>Plate Type Control Blade</td>
<td>304 SS</td>
<td>BWR</td>
<td>Low stress</td>
</tr>
<tr>
<td>Various Bolts **</td>
<td>A-286</td>
<td>PWR &amp; BWR</td>
<td>Service</td>
</tr>
<tr>
<td>Steam Separator Dryer Bolts **</td>
<td>A-286</td>
<td>BWR</td>
<td>Service</td>
</tr>
<tr>
<td>Shroud Head Bolts **</td>
<td>600</td>
<td>BWR</td>
<td>Service</td>
</tr>
<tr>
<td>Various Bolts</td>
<td>X-750</td>
<td>BWR &amp; PWR</td>
<td>Service</td>
</tr>
<tr>
<td>Guide Tube Support Pins</td>
<td>X-750</td>
<td>PWR</td>
<td>Service</td>
</tr>
<tr>
<td>Jet Pump Beams</td>
<td>X-750</td>
<td>BWR</td>
<td>Service</td>
</tr>
<tr>
<td>Various Springs</td>
<td>X-750</td>
<td>BWR &amp; PWR</td>
<td>Service</td>
</tr>
<tr>
<td>Various Springs</td>
<td>718</td>
<td>PWR</td>
<td>Service</td>
</tr>
<tr>
<td>Baffle Former Bolts</td>
<td>316 SS Cold Work</td>
<td>PWR</td>
<td>Torque, differential swelling</td>
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<tr>
<td>Corc Shroud</td>
<td>304/316/347/L SS</td>
<td>BWR</td>
<td>Wcld residual stress</td>
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<tr>
<td>Top Guide</td>
<td>304 SS</td>
<td>BWR</td>
<td>Low stress (bending)</td>
</tr>
</tbody>
</table>

*Note: BWR = Boiling Water Reactor, PWR = Pressurized Water Reactor, AGR = Advanced Gas Reactor, SGHWR = Sodium Graphite High Temperature Reactor.*
Examples of IASCC: Cracked Baffle Former Bolts in a PWR
Lovisa core basket

- Visual and ultrasonic inspection of all 312 locking bolts (Tecnatom)
- Removal of defective locking bolts (Westinghouse)
- Assembly of the new fixing system (Westinghouse)
- Internals TV-inspection of the core basket (Tecnatom)
Strong influence of radiation
Microstructure Changes can be Correlated to Irradiation Dose/Fluence

Note: \( \sim 15 \text{ dpa} = 10^{22} \text{ n/cm}^2 \ E \geq 1 \text{ MeV} \) (for PWR and BWR neutron spectra)

\( \sim 7 \text{ dpa} = 10^{22} \text{ n/cm}^2 \ E \geq 0.1 \text{ MeV} \) (for PWR and BWR neutron spectra)
Radiation Induced Segregation - RIS - occurs by preferential association between solute atoms and point defects.
Concentration Profiles for CP 304 SS after after neutron irradiation at 288°C to a fluence of $10^{22}$ n/cm$^2$ ($E > 1$ MeV)

- Cr, Mo deplete
- Ni, Si, P enrich
- Fe depends on alloy composition

\[ \text{Composition (wt\%)} \]

\[ \text{Position (nm)} \]

Grain boundary chromium depletion in irradiated austenitic iron base alloys
Grain boundary nickel enrichment in irradiated austenitic iron base alloys
Segregation of Minor Elements and Impurities

• Mo and Mn - strongly deplete with dose
• Si- very strong grain boundary segregation- as much as 10x. Segregation can lead to the formation of $\gamma'$ ($\text{Ni}_3\text{Si}$) in matrix or at grain boundary.
• P- thermally segregates and irradiation provides a small additional amount of segregation
• C, N, B- should also segregate, but there is little evidence due to difficulty in measurement
IASCC – Irradiation Assisted Stress Corrosion Cracking

- Terminology used to describe cracking of materials
- Exposed to nuclear reactor coolant and ionizing radiation
- Like all Stress Corrosion Cracking phenomena it requires critical combinations of applied stress or strain, environmental chemistry & metallurgical structure to occur

Major factors influencing Environmentally Assisted Cracking (EAC)

**Mechanical loading**
- Loading/strain rate \( dK/dt, d\varepsilon/dt, \nu \)
- Load/strain level \( K_i, \Delta K, \sigma, \varepsilon, R \)
- Residual stress

**Material**
- Composition
- Microstructure
- Heat treatment
- Surface condition
- Yield stress, hardness

**Environment**
- Composition
- pH, conductivity
- Temperature
- Redox, ECP
- Flow rate

**Critical conditions for SCC**
synergistic and interrelated parameters
IASCC added feature to EAC:
- atomic displacements
- neutron irradiation significantly alters metallurgical microstructure
- ionizing ($\alpha$, $\beta$ and $\gamma$) radiation modify the

Effects of irradiation on SCC:
- primary defects
- defects segregation
- dislocation interaction
- grain boundaries
- localized stress and strain
- environment
- stress relaxation by irradiation
- creep (beneficial factor for IASCC)
IASCC

- Radiation
- Stress
- Time
- Temperature
- Environment
Strong influence of radiation

$\mathrm{H}_2\mathrm{O} \rightarrow \mathrm{H}^+ + \mathrm{OH}^-$

$\rightarrow \mathrm{F}^+ + 2\mathrm{e}^- [\mathrm{y}, -K_{IC}]$

$\rightarrow \mathrm{H}_2\mathrm{O}_2$

$\mathrm{F}^+ + 2\mathrm{e}^- \rightarrow \mathrm{H}_2\mathrm{O}_2$

$\mathrm{Cr}^{3+} \rightarrow \mathrm{Cr}^{2+}$
SCC - RPV internals

**Displacement (mm)**

- Sample: 10 x 10 x 55 mm³

**Displacement (mm)**

- Sample: 3 x 4 x 27 mm³

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**Testing Environmentally Assisted Cracking (EAC) of Reactor Materials using Pneumatic Servo Controlled Fracture Mechanics (PSCFM),**


R. Novotny, F. Sevini, L. Debarberis, S. Petr, M. Kytka,
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- SCC
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- CONCLUSIONS
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