Modeling HIC and Failure of Ti Alloy Drip Shields within the Yucca Mountain Nuclear Waste Repository

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Engineered Barrier System (EBS)
The primary function of DS is to protect WP from seepage drips that form high ionic strength aqueous solutions at high temperatures, which could lead to crevice corrosion (CC).

The failure of DS, on its own, does not constitute a breach of EBS.
DS Criteria to Protect WP from CC

- CC initiates only after the establishment of aqueous conditions ($T < T_{aq}$), and when $T > T_{cc}$, the CC threshold temperature
- DS is required to provide protection for 2000 years on realistic conditions to 6000 years on more aggressive, but much less likely, conditions
Titanium Grade 7 Alloy (Ti-7)

<table>
<thead>
<tr>
<th>Element</th>
<th>Ti</th>
<th>Pd</th>
<th>Fe</th>
<th>C, O, N, Ni, Mo</th>
</tr>
</thead>
<tbody>
<tr>
<td>%wt</td>
<td>Balance</td>
<td>0.12 - 0.25</td>
<td>0.08 - 0.18</td>
<td>trace</td>
</tr>
</tbody>
</table>

- High strength to weight ratio (~393 kg m² s⁻²)
- Exceptional corrosion resistance (25~40 nm/yr)
- Potentially susceptible to hydrogen induced cracking (HIC)

From AECL report 11608

From AECL report 11284
Corrosion and Hydrogen Absorption Processes

Water Reduction

H₂O → H₂

Oxide Growth

Ti⁴⁺ → TiO₂

O₂⁻

Metal Oxidation

Ti → TiO₂

Hydride Formation

TiHₓ

Aqueous Environment

Passive Oxide

Oxide Transformation

TiOOH

Metal

Hydrogen Reduction

H⁺ → H₂
Failure Model for Ti-7 Drip Shields

- Degradation modes: general corrosion (GC) and HIC
- The two processes are inextricably linked since the rate of hydrogen absorption is controlled by the GC rate
- Corrosion occurs only when aqueous conditions are established:
  \[ T < T_{aq} \]
- Source of absorbable hydrogen:
  \[ \text{Ti} + 2 \text{H}_2\text{O} \rightarrow \text{TiO}_2 + 4 \text{H} \]
- Only a small fraction of the hydrogen produced will absorb into the metal:
  \[ \gamma = 4 \frac{f_H}{M_{Ti}} \]
- The absorbed hydrogen is uniformly distributed in the metal:
  \[ C_H = \frac{m_H}{\rho_{Ti} A (h_0 - h_c)} \]
- Hydrogen produced by the corrosion process absorbs into Ti. On the other hand, the hydrogen already absorbed will be released as the metal containing the hydrogen converts to oxide. There is also possibility of re-distribution of hydrogen between the metal and the oxide.

\[
dm_H = \left[ \gamma \rho_{Ti} A - (1 + \gamma - \eta) \frac{m_H}{h_0 - h_c} \right] dh_c
\]

- Sufficiently high stress intensity exists causing instant brittle failure once the absorbed hydrogen concentration exceeds a critical value.

\[
C_H(t_{HIC}) = C_{HIC}
\]

- Failures by GC if corrosion penetrates the wall allowance.

\[
\int_{0}^{t_{GC}} R_{GC} \, dt = h_0
\]
Schematic of the Model

- Failure by HIC prior to failure by GC, i.e. $t_{\text{HIC}} < t_{\text{GC}}$
Hydrogen Concentration $C_H$

\[
C_H(t) = \frac{\gamma}{\gamma - \eta} \left\{ 1 - \left[ 1 - \frac{h_c(t)}{h_0} \right]^{\gamma - \eta} \right\}
\]

No release ($\eta = 1$)

\[
C_H(t) = \frac{\gamma}{1 - \gamma} \left\{ \left[ 1 - \frac{h_c(t)}{h_0} \right]^{\gamma - 1} - 1 \right\}
\]

No re-distribution ($\eta = 0$)

\[
C_H(t) = 1 - \left[ 1 - \frac{h_c(t)}{h_0} \right]^{\gamma}
\]

$\eta = \gamma$

\[
C_H(t) = -\gamma \ln \left[ 1 - \frac{h_c(t)}{h_0} \right]
\]
Comparisons

$\gamma = 1/600$

($f_H = 0.02$)

$\eta = \gamma$

$\eta = 0$

$\eta = 1$

$\eta = \gamma$ is used in Monte Carlo simulations
Monte Carlo Program --- EBSPA

Monte Carlo simulation for performance assessment of the engineered barriers system proposed for Yucca mountain high-level radioactive waste repository

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Shoesmith Lab, U. of Western Ontario

File: C:\My Programs\Visual Basic\EBSPA
- Parameters
- WP shell flaws
- Outer lid flaws
- Inner lid flaws

Sample size 1000

Save to file
- Input summary
- Simulation results
- Both

Note

Save
Exit
Flow Chart of Monte Carlo Simulation

\[ T < T_{aq} \]

- **GC**
  \[ h_c = \Sigma R_{GC}(t) \Delta t \]
  \[ h_c(t_{GC}) \geq h_0 \]
  - **No**
    - GC failure
  - **Yes**
    - \( t_{HIC} < t_{GC} \)

- **HIC**
  \[ C_H(t) = \Sigma \Delta m_H(t) / [\rho T A(h_0 - h_c(t))] \]
  \[ C_H(t_{HIC}) \geq C_{HIC} \]
  - **Yes**
    - HIC failure
  - **No**
Two Scenarios in Simulations

- **Realistic**: conditions thought most likely prevail in the repository
- **Worst-case**: impact of the degradation processes overstated

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Distribution</th>
<th>Realistic</th>
<th>Worst-case</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_{aq}$ (°C)</td>
<td>N/A</td>
<td>120</td>
<td>160</td>
</tr>
<tr>
<td>$R_{GC}$ (scale factor)</td>
<td>Weibull</td>
<td>22.4566</td>
<td>37.4155</td>
</tr>
<tr>
<td>$R_{GC}$ (shape factor)</td>
<td>Weibull</td>
<td>0.8057</td>
<td>0.8846</td>
</tr>
<tr>
<td>$C_{HIC}$ (ppmw)</td>
<td>Uniform</td>
<td>800 – 1200</td>
<td>400 – 1000</td>
</tr>
<tr>
<td>$f_H$ ($T &lt; 100°C$)</td>
<td>Uniform</td>
<td>0 – 0.02</td>
<td>0.02 – 0.04</td>
</tr>
<tr>
<td>$f_H$ (100 &lt; T &lt; 120°C)</td>
<td>Uniform</td>
<td>0.02 – 0.04</td>
<td>0.04 – 0.06</td>
</tr>
<tr>
<td>$f_H$ (T &gt; 120°C)</td>
<td>Uniform</td>
<td>–</td>
<td>0.06 – 0.08</td>
</tr>
</tbody>
</table>
➢ Effective initial failure: CPF = 0.0001
➢ DS should not fail until 29,000 years on realistic conditions
➢ Even for the Worst-case, the DS would likely survive for more than 6000 years
Conclusions

- Probabilistic model based on anticipated repository environment and available data on the corrosion of titanium and its alloys

- Considers general aqueous corrosion and HIC as the degradation modes; the model inevitably predicts failure by HIC prior to failure by general corrosion

- The use of Monte Carlo simulations to predict the lifetime of DS on the realistic and worst-case scenarios

- The DS should not fail until 29,000 years on realistic conditions, and even for the worst-case scenario, the DS would likely survive for more than the required 6000 years
Further Improvements

- Hydrogen transport in titanium. The coupled absorption and diffusion equations could be solved by the finite element method.
- Influence of the enrichment of hydrogen near the DS surface, which then tends to be released as corrosion proceeds.
- A decrease in hydrogen absorption efficiency as the oxide film, a transport barrier to hydrogen, thickens.
- A shift in support of GC of titanium from water reduction to oxygen reduction, a process that does not produce hydrogen.
- Assessment of galvanic coupling between the titanium surface and carbon steel structure components, a process that would be expected to accelerate the rate of hydrogen production and absorption.
Electrochemistry and Corrosion Studies at Western

Group leader: Professor D. W. Shoesmith
(NSERC/OPG industry chair in nuclear fuel disposal chemistry)
Website: http://publish.uwo.ca/~ecsweb/

Thank You