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Impact of Fuel Assembly Transportation on Zirconium Alloys: toward a Mechanistic Understanding

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Impact of Fuel Assembly Transportation on Zirconium Alloys: toward a Mechanistic Understanding

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Zirconium alloys are commonly used in pressurized water reactor as fuel rod cladding tubes. After irradiation and cooling in pool, the spent nuclear fuel assemblies are transported for wet storage to a devoted site. During dry transportation, at temperatures around 400°C, the cladding experiences a creep deformation under the hoop stress induced by the internal pressure of the fuel rod. A recovery of the radiation damage can occur during transportation that can affect the subsequent mechanical properties [1].

The recovery of the radiation damage during heat treatments has been investigated using micro-hardness tests at room temperature on neutron irradiated cladding materials made of fully recrystallized Zr-1%Nb alloy. Transmission electron microscopy (TEM) observations performed on irradiated thin foils have also shown that, simultaneously with the recovery of the hardness, the dislocation loop density, induced by irradiation, falls while the loop size increases (Fig. 1). Moreover, the TEM analysis has revealed that only vacancy loops are present in the material after long-term annealing, the interstitial loops having entirely disappeared. A numerical cluster dynamic modeling [2] has been used in order to reproduce the material recovery for various annealing conditions (Fig. 1).

Furthermore, the mechanical behavior of the cladding after post-irradiation creep test has been investigated. Creep tests under internal pressure were conducted at 400 and 420°C on the neutron irradiated recrystallized Zr-1%Nb alloy. After depressurization and cooling, ring tensile tests were carried out at room temperature. In addition, transmission electron microscopy observations have been performed after testing [3]. The post-creep mechanical response exhibited a decrease of the strength compared to the as-irradiated material. This decrease is associated with a significant recovery of the ductility, which becomes close to the ductility of the unirradiated material (Fig. 2).

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The transmission electron microscopy examinations, conducted on ring samples, revealed that the radiation defects have been annealed (Fig. 3). It was also observed that as for the unirradiated material, the deformation occurred homogeneously throughout the grains. No dislocation channeling was observed contrary to the as-irradiated material [4]. These observations explain the recovery of the strength and of the ductility after post-irradiation creep that may also occur during dry transportation.

![Stress-strain curve](image1)

**Figure 2:** Stress-strain curve obtained during ring tensile test on Zr-1%Nb alloy at room temperature.

![Dislocation microstructure](image2)

**Figure 3:** Dislocation microstructure after ring tensile test subsequent to a post-irradiation creep test at 420°C.

**References**


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F. Onimus, J. Ribis, B. Bourdiliau, C. Cappelaere
INDUSTRIAL BACKGROUND: END OF LIFE OF THE FUEL ASSEMBLY

Spent Nuclear Fuel Assembly

- Temperature from 400°C up to 450°C

In-reactor use → Dry transportation → In-pool storage

Fuel assembly

- Neutron irradiation of the Zr alloy cladding

- Post-irradiation creep of the Zr alloy cladding and radiation damage recovery

- Room temperature mechanical properties at retrieval?
Recrystallized Zr-1%Nb alloy

Chemical composition (% wt)

<table>
<thead>
<tr>
<th>%wt</th>
<th>O</th>
<th>Sn</th>
<th>Fe</th>
<th>Cr</th>
<th>Nb</th>
<th>Zr</th>
</tr>
</thead>
<tbody>
<tr>
<td>RXA Zr-1%Nb</td>
<td>0.12</td>
<td>0</td>
<td>0.02</td>
<td>0</td>
<td>1</td>
<td>Bal.</td>
</tr>
</tbody>
</table>

Cladding in Zr alloy

Displacement cascade

Dislocation loop

→ creation of a high density of small loops
After heat treatment at 400°C during 250 h
\[
<\rho_b> = 1,2 \times 10^{22} \text{ m}^{-3} \quad \text{<d> = 14 nm}
\]
After heat treatment at 450°C during 960 h
\[
<\rho_b> = 2,5 \times 10^{21} \text{ m}^{-3} \quad <\text{d}> = 18 \text{ nm}
\]
\[
<\rho_b> = 1,2 \times 10^{20} \text{ m}^{-3} \quad <\text{d}> = 176 \text{ nm}
\]
The loop size increases while the density decreases
Radiation damage recovery
Loop nature using inside / outside contrast method

<table>
<thead>
<tr>
<th>Annealing</th>
<th>Vacancy loops</th>
</tr>
</thead>
<tbody>
<tr>
<td>As-irradiated</td>
<td>50%</td>
</tr>
<tr>
<td>350°C 250 h</td>
<td>50%</td>
</tr>
<tr>
<td>350°C 500 h</td>
<td>50%</td>
</tr>
<tr>
<td>400°C 250 h</td>
<td>65%</td>
</tr>
<tr>
<td>400°C 500 h</td>
<td>71%</td>
</tr>
<tr>
<td>450°C 960 h</td>
<td>100%</td>
</tr>
</tbody>
</table>

→ Only vacancy loops at the end of the recovery
→ Interstitial loops recover faster than vacancy loops

What is the loop recovery mechanism?
As-irradiated microstructure

Shrinking of the vacancy and interstitial loops

Shrinking of the smallest loop at the expense of the biggest loop

Annealed microstructure
Predictive modelling approach: cluster dynamics

Single-vacancy evolution:

\[
\frac{dC_v(1)}{dt} = \text{production} - \text{annihilation}
\]

No creation of point defects during annealing

Cluster evolution:

\[
\frac{dC(n)}{dt} = a_{n-1} C(n+1) - b_n C(n) + c_{n-1} C(n-1)
\]

Mean-field modeling

Sinks: clusters, dislocation lines, grain boundaries / free surfaces

Vacancy emission

Vacancy absorption

Vacancy cluster
Cluster dynamic modelling $\rightarrow$ computation of loop size and density

$\rightarrow$ Correct agreement between experiment and modeling
Evolution of vacancy and interstitial loop size distribution at 400°C

Evolution of the proportion of vacancy loops during heat treatment

<table>
<thead>
<tr>
<th>Annealing temperature (°C)</th>
<th>Annealing time (h)</th>
<th>Experiment Number of analysed loops</th>
<th>Percentage of vacancy loops (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>As-irradiated</td>
<td>As-irradiated</td>
<td>-</td>
<td>50 [23]</td>
</tr>
<tr>
<td>350</td>
<td>250</td>
<td>19</td>
<td>50</td>
</tr>
<tr>
<td>350</td>
<td>500</td>
<td>24</td>
<td>50</td>
</tr>
<tr>
<td>400</td>
<td>250</td>
<td>37</td>
<td>65</td>
</tr>
<tr>
<td>400</td>
<td>500</td>
<td>38</td>
<td>71</td>
</tr>
<tr>
<td>450</td>
<td>960</td>
<td>13</td>
<td>100</td>
</tr>
<tr>
<td>450</td>
<td>960</td>
<td>13</td>
<td>100</td>
</tr>
</tbody>
</table>

→ Correct agreement between experiment and modeling
High density of small loops
-> pinning of dislocations
-> radiation hardening

\[ \Delta \tau_c = \alpha \mu b \sqrt{Nd} \]

Hardness as a function of the loop density & size

→ Radiation hardening recovery during heat treatment
→ Correct prediction of radiation hardening recovery
After post-irradiation creep:
→ decrease of the loop density
( + increase of the loop size)
→ Radiation damage recovery during creep
→ Effect on the tensile mechanical behavior?
- Irradiation induced hardening (increase of the yield stress and ultimate tensile strength)

- Loss of macroscopic ductility (decrease of the uniform elongation), but the failure remains ductile (strong necking)

→ What are the deformation mechanisms?
DEFORMATION MECHANISM OF AS-IRRADIATED MATERIAL

RT tensile test on non irradiated material

Ring tensile test at room temperature after irradiation

→ homogeneous glide of dislocations in the prismatic planes mainly

Channels in the basal plane

Channels in the prismatic planes

- Heterogeneous deformation inside the grains (dislocation channeling)
- both basal and prismatic slip can be activated (depending on the grain orientation)
-> easier basal slip than before irradiation
High density of small loops
-> pinning of dislocations
-> radiation hardening

Clearing of loops by gliding dislocations
-> microscopic strain softening
-> early localization of the deformation at the specimen scale
-> decrease of the Uniform Elongation observed during ring tensile tests

--> Why is the basal slip more easily activated after irradiation than before irradiation?
Loop Burgers vector: $\mathbf{b} = \langle a \rangle = 1/3 \langle 1120 \rangle$

Dislocation Burgers vector: $\mathbf{b} = \langle a \rangle = 1/3 \langle 1120 \rangle$

B dislocation - loop

P dislocation - loop

Glissile junction

Sessile junction

→ Easy Basal channeling / difficult Prismatic channeling
- Recovery of the radiation induced hardening after creep test
- Recovery of the macroscopic ductility (uniform elongation)

→ What are the deformation mechanisms?
After ring tensile test following post-irradiation creep:
- very few remaining loops → lower radiation hardening
- homogeneous prismatic glide mainly, no channel
- recovery of the uniform elongation

Prismatic glide mainly
CONCLUSIONS

Radiation damage recovery:
- Occurs by exchange of vacancies between loops
  → Shrinking of Int loops and of small Vac loops at the expense to bigger Vac loops

Cluster dynamic modeling:
→ Correct prediction radiation damage recovery & radiation hardening recovery

Mechanical behavior after neutron irradiation:
- decrease of the uniform elongation due to the dislocation channeling
- failure occurs after a strong necking -> ductile failure
- evolution of activated slip systems explained by junction between dislocations and loops

Impact of transportation (post-irradiation creep):
- radiation hardening recovery due to the loop annealing during creep
- recovery of the uniform elongation due to homogeneous glide of dislocations

→ better knowledge and understanding of the effects of transportation on the mechanical properties of the fuel assembly at retrieval
Thank you!