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MECHANICAL BEHAVIOR OF UO₂ UNDER IRRADIATION: A MOLECULAR DYNAMICS STUDY

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FUEL DESIGN, AN ENGINEERING CHALLENGE

**Macrograph of a fuel pellet after nuclear reactor**

**In reactors**
- Temperature, $\nabla T$
- Stress, $\nabla \sigma$
- Burn-up

**Higher burn-up**

**In interim storage or long-term disposal**
- Swelling $\leftarrow$ He bubbles

**Mechanical properties?**
- Elasticity, yield stress, hardness, toughness…

**Method:** atomistic simulations with molecular dynamics

**Goal:**
- Understanding phenomena
- Input parameters for mesoscale models
I. Mechanical behavior in Bulk
II. Mechanical behavior with Point Defects
III. Mechanical behavior with He Bubbles
IV. Crack propagation
V. Summary
**MECHANICAL PROPERTIES CALCULATION**

Fluorite structure 
*(Fm$^3$m)*

Output

- Onset of crack localization
- Yield stress & strain
- Energy release rate, Toughness
- Crack propagation

Interatomic potentials:

- Yakub: good elastic constants
- Morelon: good phase transitions

Energy release rate:

$$ G_c = L_Z \int_0^{\varepsilon_c} d\varepsilon \, \sigma(\varepsilon) $$
I. Mechanical behavior in Bulk

II. Mechanical behavior with Point Defects

III. Mechanical behavior with He Bubbles

IV. Crack propagation

V. Summary
MECHANICAL BEHAVIOR - IN BULK

- Weakest crystallographic direction: <111>
- Difference with Griffith criteria ↔ phase transformation

Strain orientation | $G_C$ (J/m$^2$) |
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>&lt;100&gt;</td>
<td>12.73</td>
</tr>
<tr>
<td>&lt;110&gt;</td>
<td>9.54</td>
</tr>
<tr>
<td>&lt;111&gt;</td>
<td>9.51</td>
</tr>
<tr>
<td>Griffith</td>
<td>3.4 – 7.5</td>
</tr>
</tbody>
</table>

$G_C$ (Griffith) = $2\gamma_S$

$\gamma_S$ : surface energy

Phase transition: fluorite $\rightarrow$ PbO$_2$

Y. Zhang JNM 430 (2012) 96
P. Fossati PRB 88 (2013) 214112
I. Mechanical behavior in Bulk

II. Mechanical behavior with Point Defects

III. Mechanical behavior with He Bubbles

IV. Crack propagation

V. Summary
After NPT relaxation (300 K) → Formation of small clusters (mainly int.)

For systems > 2% of defects, clusters are small dislocation loops
General decrease of the toughness with increase of defects

Threshold at ~1.8% defects: plasticity, no phase transformation
Crack initiation: at cluster of point defects

“Plasticity” → due to nanodomain creation
I. Mechanical behavior in Bulk

II. Mechanical behavior with Point Defects

III. Mechanical behavior with He Bubbles

IV. Crack propagation

V. Summary
Experiments

$UO_2$ irradiated $6 \times 10^{12}$ Au.cm$^{-2}$, 600°C

- Bubble density saturates at: $4 \times 10^{23}$ m$^{-3}$
- Bubble size: $1 - 2$ nm

HRTEM: C. Sabathier NIMB 266 (2008) 3027

To fit experimental observations:

Bubble: $\varnothing$ 2 nm, density of bubbles = $10^{24}$ m$^{-3}$, He density in bubble = $4 \times 10^{-2}$ mol/cm$^3$, $\rightarrow$ Pressure inside bubble = 500 MPa

MD simulations

Density in bubble

- 3$\times$10$^{-2}$ mol/cm$^3$
- 8$\times$10$^{-2}$ mol/cm$^3$

He resolved in int.

He resolution for density $> 4 \times 10^{-2}$ mol/cm$^3$
bubble: Ø 2 nm, He density in bubble = $4 \times 10^{-2}$ mol/cm$^3$, Pressure = 500 MPa

- Phase transition at the onset of the crack
- Crack initiates systematically at the bubble surface
Strain orientation | \( G_C \) (J/m\(^2\))
<table>
<thead>
<tr>
<th></th>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Bulk</td>
<td>Cavity</td>
<td>He bubble</td>
</tr>
<tr>
<td>&lt;100&gt;</td>
<td>12.73</td>
<td>7.21</td>
</tr>
<tr>
<td>&lt;110&gt;</td>
<td>9.54</td>
<td>5.96</td>
</tr>
<tr>
<td>&lt;111&gt;</td>
<td>9.51</td>
<td>5.62</td>
</tr>
</tbody>
</table>

- \( \varepsilon_c, \sigma_c, \) and \( G_C \) decrease with cavity and He bubble
- Easy fracture initiation
- Decrease of phase transformation
- \( G_C \sim G_c(\text{Griffith}) \)
- \( G_C \) bubble \( \geq G_C \) cavity
I. Mechanical behavior in Bulk

II. Mechanical behavior with Point Defects

III. Mechanical behavior with He Bubbles

IV. Crack propagation

V. Summary
CRACK PROPAGATION - METHOD

- Strip geometry
- Create initial notch
- Relaxation NPT 300 K, 20 ps
- Impose deformation along <111> direction
- Strain-rate $10^9$/s

Outputs: microstructure evolution, local energies, local stresses

Strip geometry: $241 \times 65 \times 4$ nm
Initial notch: Ellipse $40 \times 8$ nm

Applied strain

Initial notch
\[ \varepsilon = 0.036 \]
\[ \varepsilon = 0.042 \]
$\varepsilon = 0.047$
\[ \epsilon = 0.052 \]
\[ \varepsilon = 0.058 \]

\[ \text{surface} \]

\[ \text{PbO}_2 \]

\[ \text{fluorite} \]

\[ \text{epot (eV)} \]

\[ -7.501 \]

\[ -10.54 \]
\[ \varepsilon = 0.064 \]
$\varepsilon = 0.069$

\[ \text{epot (eV)} \]

-7.501

-10.54

surface

PbO$_2$

fluorite
$\varepsilon = 0.075$
ε = 0.081

\[ \text{epot (eV)} \]

\[ \text{PbO}_2 \text{ structure} \]
\[ \varepsilon = 0.087 \]
\[ \varepsilon = 0.093 \]
$\varepsilon = 0.098$

epot (eV)

-7.501 surface

PbO$_2$

fluorite

-10.54
$\varepsilon = 0.104$

epot (eV)

- Surface
- PbO$_2$
- Fluorite

CRACK PROPAGATION - IN BLUK
$\varepsilon = 0.111$
\[ \varepsilon = 0.117 \]
$\varepsilon = 0.123$
\[ \varepsilon = 0.129 \]
Crack propagates by cleavage in the PbO$_2$ structure at the crack tip.

Sub-cracks appear, grow and coalesce in secondary phase.

\[ \varepsilon = 0.135 \]
ε = 0.036

1.8% Frenkel pairs randomly distributed
\[ \varepsilon = 0.047 \]
\[ \varepsilon = 0.052 \]
$\epsilon = 0.058$

epot (eV)

surface

PbO$_2$

fluorite

-11.82

-6.329
\[ \varepsilon = 0.064 \]
$\varepsilon = 0.069$
$\varepsilon = 0.075$

epot (eV)

- Surface
- PbO$_2$
- Fluorite
CRACK PROPAGATION - WITH POINT DEFECTS

\[ \varepsilon = 0.081 \]
\[ \varepsilon = 0.087 \]
\[ \varepsilon = 0.111 \]
\[ \varepsilon = 0.135 \]

No phase transition

Crack propagates by growth and coalescence of close sub-cracks
\[ \varepsilon = 0.036 \]

4×\(10^{23}/m^3\) He bubbles, 2 nm, with 4×\(10^{-2}\) mol/cm\(^3\) He
\[ \epsilon = 0.042 \]
\[ \epsilon = 0.052 \]
CRACK PROPAGATION - WITH HE BUBBLES

\[ \varepsilon = 0.064 \]
$\epsilon = 0.069$
ε = 0.075

CRACK PROPAGATION - WITH HE BUBBLES

epot (eV)

-5.046

surface

PbO₂

fluorite

-8.28
\[ \varepsilon = 0.081 \]
\( \varepsilon = 0.087 \)
$\varepsilon = 0.098$

diagram showing crack propagation with HE bubbles in a material with a fluorite or PbO$_2$ surface and epot in eV.
$\epsilon = 0.104$
\[ \varepsilon = 0.111 \]

**Same crack propagation than in bulk**

**Sub-cracks initiated on the He bubbles**
Crack propagation via coalescence of sub-cracks

Decrease of the toughness with the presence of defects

Sub-cracks initiated at the surfaces of the He bubbles
SUMMARY

Mechanical behavior of UO$_2$ with MD simulations

In Bulk
- Most fragile crystallographic orientation: $<111>$
- Difference with Griffith: due to phase transition

With point defects – 0.04 to 7%
- Decrease of yield stress and toughness
- Threshold at 1.8% point defects → plasticity

With He bubbles – 2 nm, $10^{24}$/m$^3$
- Decrease of yield stress and toughness
- Initiation of crack at the bubble

Crack propagation
- Propagation with coalescence of sub-crack
- Sub-cracks initiated at defects
Thank you for listening

Questions?

Acknowledgments

JP Crocombette
P Fossati

Challenge project – C097073
PHASE TRANSITIONS UNDER COMPRESSION

- Cotunnite ($Pnma$)
- Fluorite ($Fm\overline{3}m$)
- Rutile ($P4_2/mnm$)
- Scrutinyite ($Pbcn$)

Oxygen is represented by red spheres, uranium by grey spheres.

Compression and tension directions indicated by arrows.
All potentials yield to the same phase transitions

Morelon potential is the closest to the reference values
Temperature increase → elasticity loss

Disappearance of phase transition with the increase of the temperature
Goal: study damage created by He bubble in UO$_2$ (defects, pressure, …)
Method: He Bubble in UO$_2$ (≠ density, size, temperature)
Main results:

- No influence of the temperature (300 – 1500 K)
- He partially soluble in UO$_2$ (→ interstitial)
- Density threshold ≈ 0.04 mol/cm$^3$ (= 1 He/Schottky)
- Max pressure in bubble 10 GPa, He state equation ≠ VdW
- Pressure at 0.04 mol/cm$^3$ = 500 MPa
300 K – nanobubble 0.22 mol/cm³

- **Gaz structure** → amorphous with increasing density
- ≠ Xénon (fcc structure for high density)
General no phase transformation

Threshold at \(~1.8\%\) defects