Evolution of the mechanical properties of aging erbium tritide films

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Metal tritide films

- Metal tritide films are essential for applications such as neutron generators, but the property changes as the T decays lead to problems.

- We have been following the mechanical properties of Erbium Tritide films as they age. We found that the films first strengthened, then softened.

- The two regimes can be explained by dispersion strengthening combined with a simple elastic softening due to the bubble growth.
Samples: ErT$_2$ layers on Mo/Si

Sample preparation

- 500 nm Er
- 95 nm Mo
- Silicon
- Hydrided with 100% Tritium
- Aged in vacuum

TEM cross-section
bright-field, ~{110} zone
62 days after hydriding.

Oxide forms during hydriding and upon air exposure.

Tritium decays into $^3$He, forming platelet-like bubbles on (111) planes.

$^3$He bubbles

6.4 nm bubbles

10 nm

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Tritium decay and $^3\text{He}$ content

**Calculated buildup of $^3\text{He}$ for 100% T-loaded ErT$_2$**

- Graph showing the $^3\text{He}$ content (at.%) over days after loading.
- Red line: T decay calculation.
- Red circles: IBA measurement.

**IBA of aged ErT$_2$ shows expected levels of T, $^3\text{He}$**

- Graph showing scattering yield vs. channel (energy).
- ErT$_2$ aged 1173 days.

Helium-3: 10.1 at.%
Tritium: 50.9 at.%
Nanoindentation of tritiated films

- Containment procedures allowed use of indenter outside T envelope.
- Only known nanoindentation of tritiated thin films.
- Finite-element modeling* is used to determine film yield strength and Young’s modulus separate from the substrate:
  - Properties of the indenter and underlying layers and substrate are fixed at known values.
  - Properties of the layer are varied until a good fit to experiment is obtained.
  - Tip yielding, stress, friction are all modeled.

Yield strength increased as $^3$He bubbles formed and grew. After an initial increase, the hardness of the films leveled off and then decreased as the films aged.
Young’s modulus initially stayed constant and then decreased at about the same time as the yield started to decrease.
Mechanical properties: two regions

- **Region 1**
  - increasing yield strength with bubble growth
  - nearly constant or decreasing elastic properties

- **Region 2**
  - decreasing yield strength
  - decreasing elastic properties

![Graph showing yield strength and Young's modulus over time for different materials](image)
Region 1 - dispersion hardening

- Bubbles pin dislocations
  - study in Ni(He) shows Orowan-type strengthening even though bubbles are shearable
  - implants with 1 to 10 at. %: spherical bubbles from 1 to 6.4 nm

- Calculation confirms pinning*
  Three effects provide binding:
  1) reduction in dislocation strain energy
  2) absence of core energy
  3) barrier to surface step formation in bubble

*calculation by S.M. Myers

Ni implanted with 1-10 at% $^4$He

**Bubble sizes**

- **TEM measurements**
  - sizes measured up 547 days: 15.7 nm
  - sizes consistent with no new nucleation after the first few weeks

- **Linear fit to volume**
  - constant thickness
  - assumes no nucleation after the first few weeks

![Graph showing TEM measurements and linear fit to volume.](image)
Bubble pressure

- Bubble pressure is needed to deduce number density
  - Since these are platelets we use Cowgill’s formula for $p_{\text{de}}$, calculated pressure for dipole expansion.\(^1\)

$$p_{\text{de}} = \left( \frac{2\gamma}{s} \right) \left[ \frac{(2r + b + s)}{(2r + b)} \right] + \frac{\mu d}{(2r + b)}$$

$d = \{111\}$ interplanar spacing  
$s = \text{platelet thickness} \ (2d<s<3d)$  
$r = \text{platelet radius}$  
$\gamma = \text{surface energy} \ (0.637 \ \text{J/m}^2)$  
$b = \text{burgher’s vector} \ (0.3623 \ \text{nm})$  
$\mu = \text{shear modulus} \ (70 \ \text{GPa})$

- The He equation of state by Kortbeek & Schouten\(^2\) then provides the He density in the bubbles, giving the number density.

\(^1\)Don Cowgill, HHIMC presentation, 2005.
Orowan-type strengthening for platelets

- For platelets the calculation of Critical Resolved Shear Stress $\tau$ is more complicated than for spheres or rods.
  - We use a formula computationally derived by Zhu and Starke$^1$:

$$\tau = 0.12 \mu \frac{b}{(D_p t_p)^{1/2}} \left[ f_v^{3/2} + 0.70 f_v (D_p t_p)^{1/2} + 0.12 f_v^{3/2} \frac{D_p}{t_p} \right] \ln\left( \frac{0.079 D_p}{r_0} \right)$$

$t_p = \text{platelet thickness}$
$D_p = \text{platelet diameter}$
$f_v = \text{volume fraction}$
$b = \text{burgher’s vector} \ (0.3623 \text{ nm})$
$\mu = \text{shear modulus} \ (70 \text{ GPa})$
$r_0 \sim \text{burgher’s vector (used } \frac{1}{2}b)\$

**Increase in yield strength**

**Yield strength** increases in proportion to \( \tau \) (CRSS)
- constant of proportionality is the Taylor Factor (typically 2-3)
- here we use 2

**Calculation of \( \tau \)** diverges when platelet diameters are equal to the average center-to-center spacing
- estimate of CRSS becomes invalid when bubbles nearly overlap.
Mechanical properties: two regions

- **Region 1**
  - increasing yield strength with bubble growth
  - nearly constant or decreasing elastic properties

- **Region 2**
  - decreasing yield strength
  - decreasing elastic properties
Region 2 - elastic softening

- Bubbles lower the average elasticity and strength
  - as the bubble volume fraction increases, elasticity and yield strength decrease.
  - FEM simulations using 2-12 vol.% bubbles, with varying sizes and shapes, quantify the effect.

Mises stress for nanoindentation of ErTₓ with 5 vol.% He

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Decrease in elasticity

**Elastic modulus** decreases as bubble volume density increases.
- might be expected from “rule-of-mixtures”
- FEM calculation is not scaled
Yield strength also decreases as bubble volume density increases.

- Not expected from simple “rule-of-mixtures” - FEM simulations required.
- Effect is clear in region 2, where dispersion hardening is no longer effective.
Summary

- **Region 1**
  - increasing yield strength with bubble growth
  - nearly constant or decreasing elastic properties
  - dispersion hardening

- **Region 2**
  - decreasing yield strength
  - decreasing elastic properties
  - elastic softening