Neutron Diffraction Studies of Micromechanics of Material Deformation

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Outline

- Introduction and motivation
- New engineering diffractometers: SMARTS and ENGIN-X
- Metallic glass composites
- In-situ-reinforced Si₃N₄
- Polycrystalline ferroelectrics
- Future directions: DANSE
Fracture of a Fiber Composite under Tension

- **Aim**: prediction of strength and lifetime
- **Need**: “realistic” constitutive laws

**Complications**
- Fabrication processes
- Inhomogeneous dislocation densities
- Changes in grain size
- Geometrical constraints
- Interface introduced with different properties
- Residual stresses
Motivation and Approach

- Little information about deformation and \textit{in-situ} constitutive behavior of materials.
- Need to link experimental data with rigorous micromechanics modeling.
- **Approach**: Use neutron diffraction to investigate deformation in materials and complement it with modeling.
- **Critical issues**:
  - Need for model specimens
  - “High selectivity” of diffraction
  - Only elastic lattice strains are measured with diffraction
  - Lack of “realistic” constitutive laws to calculate stress and interpret diffraction data
Advantages of ND

- Non-destructive.
- Ability to distinguish different phases.
- Can measure elastic strain and texture.
- Multi-scale: \textit{nm} to \textit{cm}.
- Deep penetration.
- \textit{In-situ} experiment capability.

⇒ Determination of \textit{in-situ} constitutive behavior

Bragg’s law:

$$\lambda = 2dsin\theta$$

Differences in lattice spacing

⇒ Elastic lattice strain

$$\varepsilon_{hkl}^{el} = \frac{d_{hkl} - d_{hkl}^0}{d_{hkl}^0} = \frac{d_{hkl}}{d_{hkl}^0} - 1$$

\begin{align*}
\bullet & \quad \bullet \\
\bullet & \quad \bullet \\
\bullet & \quad \bullet \\
\bullet & \quad \bullet \\
n & \quad n
\end{align*}
Neutron Powder Diffraction: Data Analysis

Rietveld Method*

- Least-squares-based fitting method.
- Requires a priori phase information.
- Fits the whole diffraction pattern.
- Phase discrete.
- Can distinguish superimposed reflections.
- Yields detailed crystallographic information: lattice constants, texture, site occupancies, phase fractions, thermal parameters, etc.

Fitting Parameter

“Weighted Pattern” Residual:

$$ R_{wp} = \left[ \frac{\sum_i w_i (I_{io} - I_{ic})^2}{\sum_i w_i (I_{io})^2} \right]^{1/2} $$

Third generation neutron powder diffractometer.

- 10-30 fold performance improvement over NPD.

First dedicated engineering diffractometer.

- Optimized for engineering stress/strain studies.

State-of-the-art ancillary equipment:

- 250 kN (60,000 lb) load frame.
- Controlled atmosphere furnace ($T_{\text{max}} = 1500^\circ \text{C}$ under load, $2000^\circ \text{C}$ stand alone).

- Radial collimators for 1 mm$^3$ spatial resolution.

- Rapid and accurate specimen handling capability.

- “Expert System” for experiment design and real time monitoring.
SMARTS: Spectrometer (for) MAterials Research (at) Temperature (and) Stress

Cave cutaway schematic

Cave with load frame & furnace installed on translator

Los Alamos Neutron Science Center, Los Alamos National Laboratory
**SMARTS Load Frame**

- Spectrometer for MAterials Research at Temperature and Stress (SMARTS)
- Schematic setup for *in-situ* compression loading
- Measurement time is about 10-20 minutes per load level
- Measure elastic strains in two directions simultaneously
- Bulk measurement contrary to conventional X-ray measurements

**Bragg’s law:** \( \lambda = 2dsin\theta \)
SMARTS Furnace

- $T_{\text{max}} = 1500^\circ\text{C}$ under load
- $T_{\text{max}} = 2000^\circ\text{C}$ stand alone
- Vacuum or inert atmosphere
ENGIN-X Diffractometer

ISIS Neutron Scattering Facility
Rutherford Appleton Laboratory (Didcot, UK)
Main problem with BMGs: catastrophic failure under unconstrained loading.

Main deformation mechanism is via shear bands (at room T).

Addition of reinforcements has been shown to increase damage tolerance and toughness.

Critical questions:

» What is the *in-situ* mechanical behavior of reinforcements?

» How do reinforcements interact with shear bands?
W-Fiber / BMG-Matrix Composites: Compressive Loading Behavior

- Vitreloy 1 matrix: $Zr_{41.2} Ti_{13.8} Cu_{12.5} Ni_{10} Be_{22.5}$
- Tungsten fiber composites:
  - Same ultimate stress as monolithic Vit.1
  - Large increase in ductility
  - Knee in stress strain curve as tungsten fibers yield

- In-situ deformation of W?
- What happens to BMG?

W-Fiber / BMG-Matrix Composites: Thermal Residual Stresses

- CTE mismatch: $\alpha_W (4.5 \times 10^{-6} \text{ 1/K}) < \alpha_{BMG} (10 \times 10^{-6} \text{ 1/K})$

- Measured residual strain in W fibers using neutron diffraction

- Calculated thermal residual stresses in both phases using FEM

- Residual stresses are generated just below $T_g$

W-Fiber / BMG-Matrix Composites: Finite Element Model

- Full 3-D model due to loading along fibers
  - Unit cell model
  - Plane strain along \( z \)
- Hexagonal stacking in all models to accommodate high volume fractions
- Thermal residual stresses: no relaxation below \( T_g \)*
- Constitutive laws:
  - \( W \): deduced \textit{in-situ} behavior
  - BMG: von Mises or Mohr-Coulomb**
    \[ \tau_c = 946 - 0.04 \sigma_n \text{ [MPa]} \]

W-Fiber / BMG-Matrix Composites: Compressive Loading Behavior

- Important to account for *in-situ* deformation and residual stresses
- W yields at -1300 MPa, BMG yields at -1900 MPa
- Composite yielding at -360 MPa (20% W-BMG), -1060 MPa (80% W-BMG)
- Model struggles at high stresses (multiple shear bands in BMG?)

**β-Si₃N₄: Neutron Diffraction Experiments**

- Single phase sample ($\beta$-Si₃N₄) – AS800 or GS44 from Honeywell
- Multiple reflections used in elastic constant and CTE determination
- Si₃N₄ fitting parameters:
  - Space group $P6_3/m$ – hexagonal; $a = 7.608\,\text{Å}$, $c = 2.911\,\text{Å}$
  - 6-term background function, absorption, Debye-Waller (thermal) parameter
\[ \beta-\text{Si}_3\text{N}_4: \text{ Coefficient of Thermal Expansion} \]

- Diffraction data used directly in CTE calculation*.
- Multiple reflections employed; higher precision.
- Least squares analysis of redundant data.
- Result for CTE tensor of AS800 \( \beta-\text{Si}_3\text{N}_4 \):

\[
\alpha_{ij} = \begin{bmatrix}
3.50 & 0 & 0 \\
0 & 3.50 & 0 \\
0 & 0 & 4.06 \\
\end{bmatrix} \times 10^{-6} \text{ } 1/K
\]

- Polycrystalline value:

\[ \alpha = 3.69 \times 10^{-6} \text{ } 1/K \]

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Self-Consistent Model (SCM)

- **Model Assumptions:**
  - Eshelby inclusion theory
  - Stresses and strains within an ellipsoidal inclusion are uniform
  - Homogeneous equivalent medium (HEM)

- **Output:**
  - Direct comparison with neutron diffraction measurements
  - Averages over grain sets representing reflections
  - Information about material behavior on a microscopic scale
  - *hkl* dependent behavior
  - Accurate description of texture
Elastic Constants of **AS800 β-Si₃N₄ at 1375°C**

- Employed self-consistent modeling (EPSC)
- Least square fitting of \( hkl \)-dependent elastic strains in both longitudinal and transverse directions
- Polycrystalline average:
  \[ E = 310 \text{ GPa}, \ \nu = 0.31 \]

  manufactured values at 1200°C:
  \[ E = 293 \text{ GPa}, \ \nu = 0.28 \]

\[
C_{ij} = \begin{bmatrix}
460 & 160 & 240 & 0 & 0 & 0 \\
160 & 460 & 240 & 0 & 0 & 0 \\
240 & 240 & 310 & 0 & 0 & 0 \\
0 & 0 & 0 & 140 & 0 & 0 \\
0 & 0 & 0 & 0 & 140 & 0 \\
0 & 0 & 0 & 0 & 0 & 150 \\
\end{bmatrix} \quad \text{(GPa)}
\]

Literature values (at room T):
\[ C_{11} = 430, \ C_{33} = 570, \ C_{12} = 190, \ C_{13} = 130, \ C_{44} = 110 \text{ (GPa)} \]

Creep Mechanisms of ISR Si$_3$N$_4$

- Creep of ISR Si$_3$N$_4$ described by formation and growth of cavities in grain boundary phase**.
- Si$_3$N$_4$ grains remain elastic.
- Results in curvature of semi-log plot of creep rate vs. stress.
- Leads to the following creep equation:

\[
\varepsilon_s = B \sigma \exp\left(\frac{-\Delta H}{RT}\right) \frac{f^3}{(1-f)^2} \exp(\alpha \sigma)
\]

where \( \alpha \approx \frac{2\sigma_c}{9\sigma^2} \) and \( f : \text{vol. frac. of g.b. phase} \)

\[
\& = A' \sigma \exp(\alpha \sigma) \quad A'(\sigma) = 0.08 \left( \frac{\sigma - \sigma_c}{25} \right)
\]

Creep of **GS-44** at 1200°C: *Constant Stress Test*
Constitutive Behavior of Ferroelectric Materials

- Ferroelectric and piezoelectric materials couple electrical signals to mechanical displacements.

- Ideal for applications in vibration control, sensors, transducers, and micromechanical devices.
How Does Ferroelectricity Work?

PbTiO₃

Six equivalent $<001>_{\text{cubic}}$ directions give six equivalent states at room temperature

High temperature (non-polar cubic)

Room temperature ($<001>$ polarized tetragonal)

$\sigma / a = 1.065$

180° Switching

$T < T_c$

E, $\sigma = 0$

apply large electric field or stress

90° Switching

ferroelectricity switching induced by electric field

ferroelasticity switching induced by stress

180° Switching

E

$90°$ Switching

$\sigma$
Microscopic Effects

- Regions of organized unit cell polarizations are separated by twin boundaries called domain walls.

- Application of stress or electric field induces motion of domain walls, changing polarization and strain in the crystal.
Meso-/Macroscopic Effects

- Grains within a polycrystal possess randomly oriented domains
- Electrical poling is used to align a significant number of domains and produce a technologically viable ceramic material
- Domain motion may be constrained by grain orientation and local boundary conditions
Compression of Single Phase Tetragonal PZT

- Strain gauge data indicate linear elastic behavior
- March coefficient results suggest minor 90° domain switching
- Lattice strains are approximately linear
Comparison of Various PZTs

Tetragonal

Rhombohedral