Atom-Probe Tomographic Measurement of Trapped Hydrogen Isotopes

R. Karnesky (Sandia)
N. Teslich, M. Kumar (Lawrence Livermore)

TFG 2014
Local-Electrode Atom-Probe (LEAP) Tomography

Compositional and structural analysis at the atomic scale
- Pulse encodes \( z \)
- Area detector gives \((x, y)\)
- TOF encodes mass/charge

Instrument capability
- \(10^6-10^7\) nm\(^3\) analysis volume
- \(3 \times 10^{-11}-10^{-10}\) torr UHV
- 20–100 K specimen temp.
- 200 kHz electrical pulsing

Photo courtesy of R.P. Koll
First atomic imaging
Site-Specific Samples and Hydrogen Isotope Sensitivity

- Sc, ScH, ScD, ScHD, ScD₂
- Fe
- ScO
- Ga
- Ga
Hydrogen Segregates to Traps and Atom Probe can Image It
Engineering Needs Require Both High Strength and High Fracture Toughness

R. Rithie, Nat Mat 2011
Multiscale Experiment/Simulation Couplings are Important to Predict Strength and Fracture

“ICME@ Michigan” Framework
Foundational Engineering Problems & Cross-Cutting Pillars

MMM 2014
7th International Conference on Multiscale Materials Modelling
October 6-10, 2014 Berkeley, California
Science-Based Computational and Experimental Capabilities Advance Us Beyond the Empiricism of Current Methods

**Predictive modeling and simulation tools**

- **Atomistic simulations**
  - DFT/MD
  - GB mechanistic behavior

- **Grain-level framework**
  - MPM
  - Localized deformation coupled with microstructural evolution
  - Fracture processes

- **Fracture model**
  - Environmental fracture performance
  - Failure analysis

**Validation of atomic potentials**
- Deformation mechanisms
- Microstructure statistics
- Local deformation

**Experimental discovery and validation tools**

- HRTEM, LEAP tomography
  - GB structure / composition

- **In situ TEM, SEM**
  - Grain-level mechanical response
  - Microstructural evolution
  - GB statistics

- Macroscale mechanical testing
  - Intergranular fracture vs. H concentration, special GB fraction...
Intergranular Fracture Properties depend on Microstructure and Environment

Dislocations and twin boundaries within Ni
Angular Dependence of Inclined Twin Energy

- Calculated values (dots) at 0K versus a model assuming that boundary is made of (111) and (211) twin facets (line)
- Energetic contributions from junctions between CTB and LTB are neglected
Segregation in 35.26° Boundary

- Image made with $\mu_H = -2.35\text{eV}$
  - Corresponds to $\sim 2 \times 10^{-4}$ bulk concentration at 300K
- H segregates to GB and likely interacts with neighbors
- Adsorption Energy histogram is generated from map at 0K (lower right), it includes large peaks for bulk oct and tet sites.
Segregation Energy Histograms (0K)

$\Phi = 0^\circ$

$\Phi = 35.26^\circ$

$\Phi = 74.21^\circ$

$\Phi = 79.98^\circ$

$\Phi = 90.00^\circ$
Higher Angle Boundaries Show Greater S and D Segregation

4 at.% D, an enrichment of over ten from the bulk, consistent with Kirchheim, though higher than the results by Birnbaum.

Twin Boundary Has Some Sulfur Segregation, But No D Segregation
Summary

- Fracture prediction is impactful and multi-scale
- Current prediction of fracture is poor and will require strong coupling between experiments and simulations
- Atom-Probe Tomography provides a unique capability for 3-D characterization at the atomic scale
- It is sensitive to hydrogen isotopes (H environment greatly impact properties, but H is otherwise difficult to characterize)
- It can serve as input to sub-grain-level models
- It can validate atomic-scale models
Additional Acknowledgements

- ScD2 Project
  - N. Moody

- Fracture Project
  - B. Somerday, D. Medlin, R. Dingreville, B. Somerday, K. Hattar
  - W. Barrow, D. Spearot (University of Arkansas)

- Discussion of H Segregation and Trapping
  - D. Balch, N. Bartelt, C. San Marchi
  - D. Isheim (Northwestern University)
H Mapping Depends On (after T. Kelly)

- Overall H measurements (not localized) may strongly depend on:
  - Local Electrode and chamber cleanliness
  - Transfer time
  - Time in Analysis Chamber
  - Temperature during all of this
  - Pulse rate
  - Ion evaporation rate

- Localized H measurements (of phases) depend on
  - local field-evaporation field
    - causing changes in local tip shape, resulting in local magnification
  - Crystallographic faceting
  - Other defects (dislocations, grain boundaries)