SAND2015-8817C

Fatigue Behavior and Notch Sensitivity in Nanocrystalline Metals

Tim Furnish, Brad Boyce, John Sharon, Christian Arrington, Jamin Pillars, Blythe Clark

Sandia National Laboratories
Albuquerque, NM 87123

October 7, 2015

MS&T 2015 Materials Science & Technology Conference and Exhibition, Columbus OH

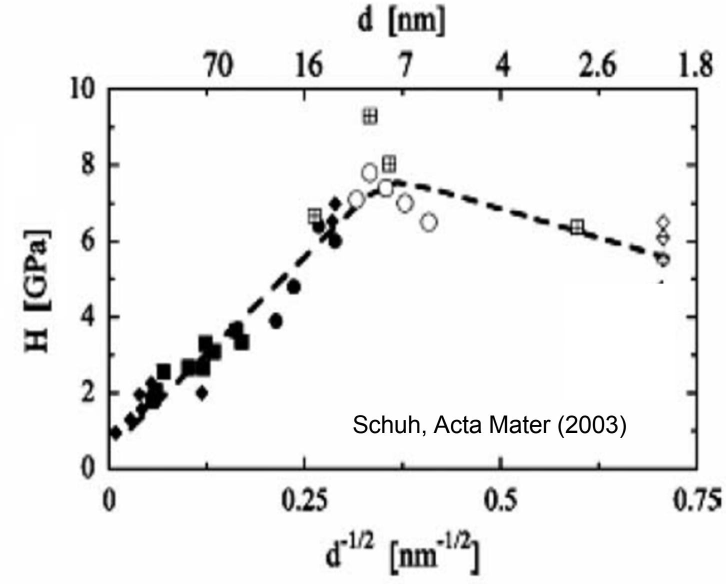
U.S. DEPARTMENT OF ENERGY | Office of Science

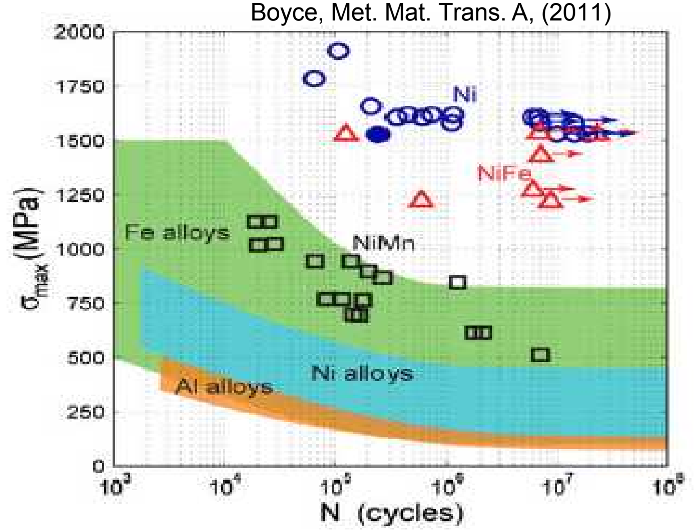
NNSA National Nuclear Security Administration

Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000. SAND2015-4425 C.

Nanocrystalline metals show unique mechanical behaviors and the potential for tunable properties

- **Nanocrystalline (NC)** materials are generally defined by having an average grain size of < 100nm
- When grains are at the nanoscale, **unique mechanical properties** and plastic responses are observed
- Typical NC metals show:
  - ultra-high strength & hardness (up to 10+ times that of CG counterparts!)
  - high wear-resistance
  - high fatigue strengths

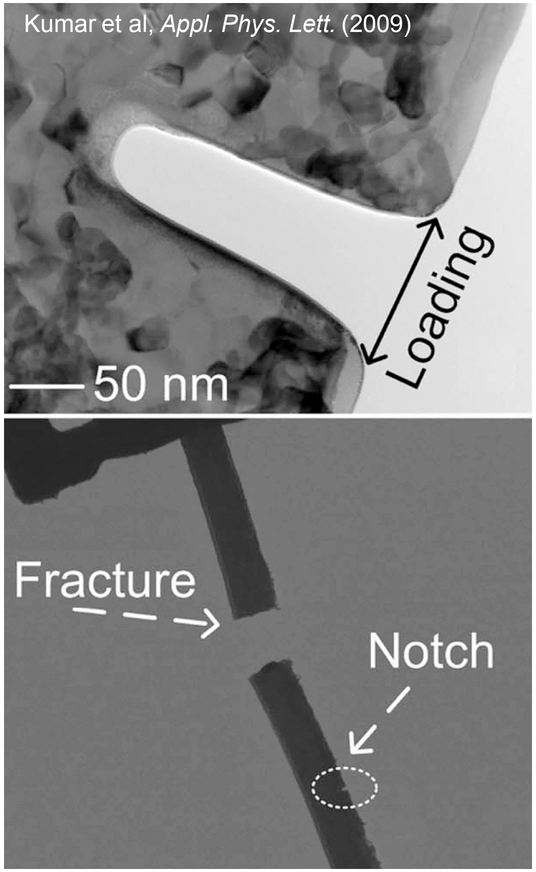




The potential for ultrahigh strength, hardness, and fatigue resistance make NC metals good candidates for advanced applications – but we are only beginning to understand their unique deformation behaviors.

Some reports have suggested that "stress concentration may not be relevant at the nanoscale…" Is this true?

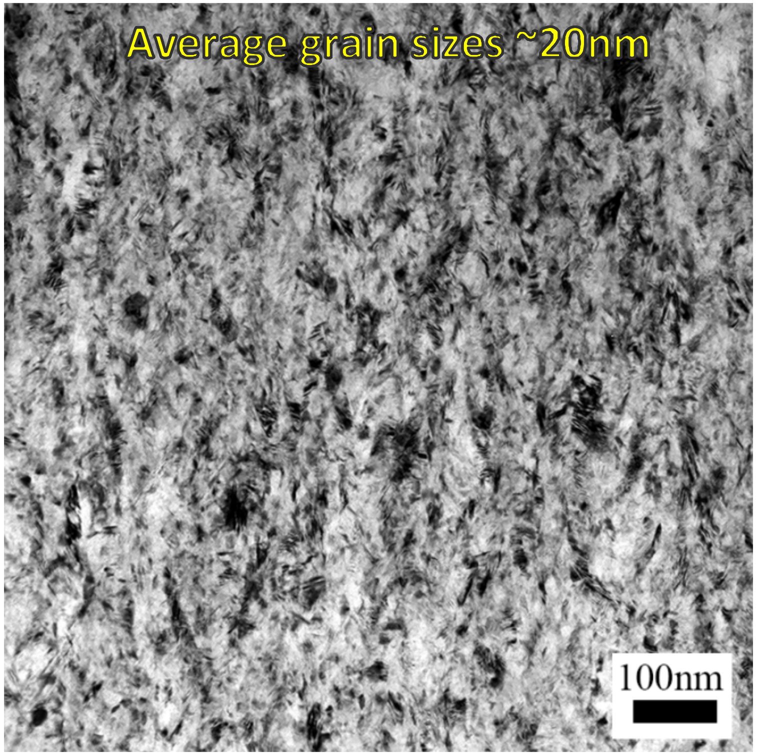
- Stress concentrations and their potential effects on material behavior are "old" problems. But…
- Stress concentration effects are
  - Complicated during *plasticity*
  - Even more convoluted during *cyclic* loading
  - Unknown for NC metals!
- Some reports have shown insensitivity to nano-notches in NC materials during monotonic loading

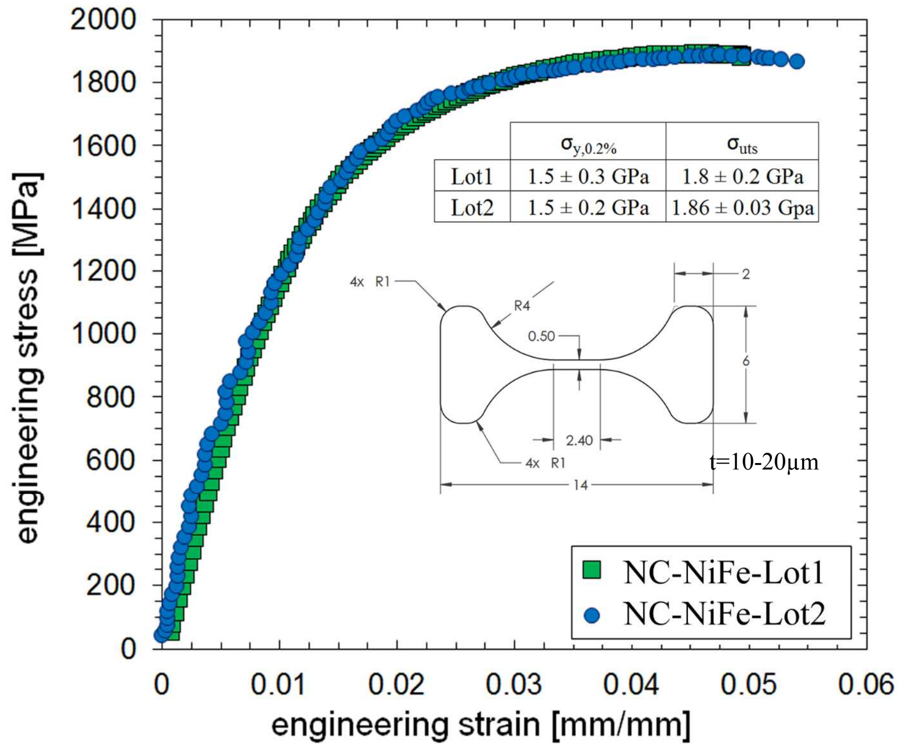


- Is any notch sensitivity observed during *fatigue* in NC metals?
- Are there potential size dependencies (e.g. grain size-to-notch size ratio) that may be playing a role?
- If fatigue stress concentrations are prevalent, how do they affect the deformation and fracture behaviors?

Understanding stress concentrations and flaw sensitivity will be paramount for the reliable and predictable performance of NC metals for advanced applications.

Two sets of NC NiFe specimens were pulse-electrodeposited at different current densities – grain size and tensile properties were identical

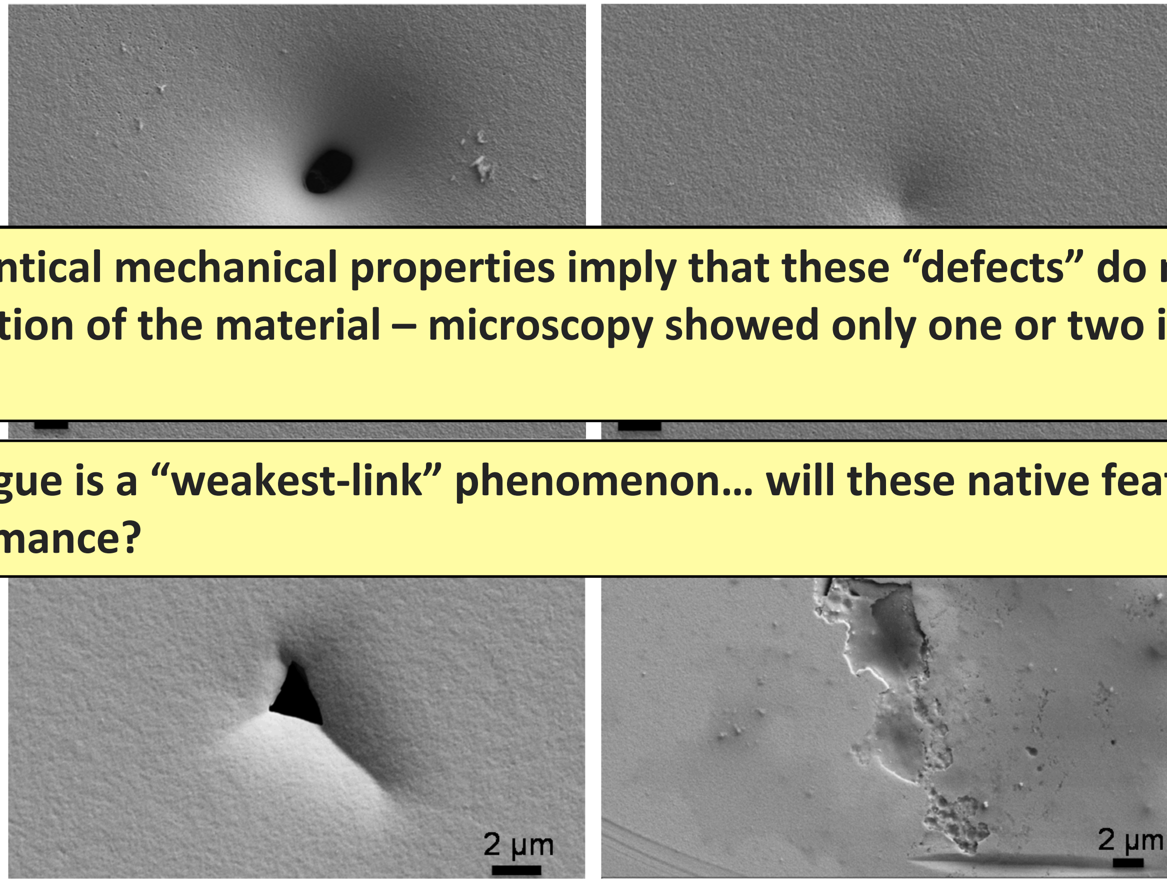




NC NiFe was selected as a representative NC material due to its ultrahigh strength & fatigue resistance – previous reports have also highlighted potentially unique fatigue deformation and crack initiation mechanisms, which are not well understood.

Electrodeposition can inherently lead to processing-related flaws. How might these native defects affect the fatigue properties?

“Lot1” and “Lot2” are nearly identical, except Lot1 consistently showed one or more 2-5 µm surface defects (not present in Lot2).

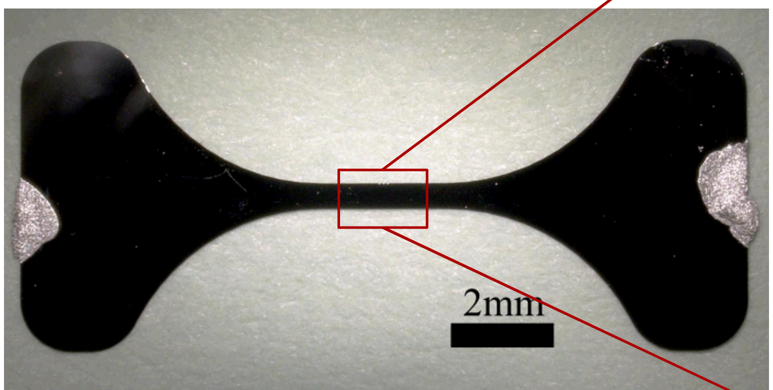


The nearly identical mechanical properties imply that these “defects” do not make up a significant portion of the material – microscopy showed only one or two in each Lot1 specimen.

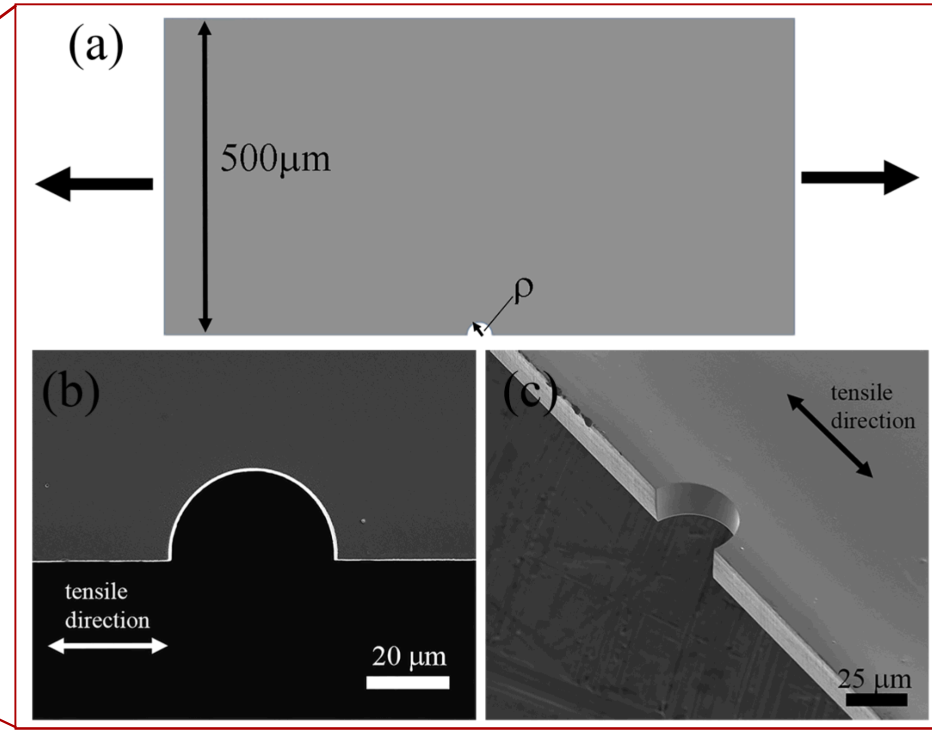
Generally, fatigue is a “weakest-link” phenomenon… will these native features affect the fatigue performance?

Examples of surface flaws observed in the Lot1 specimens – one or two features were found in each Lot1 specimen, none in the Lot2 material. Typical feature size in Lot1 is 2-5 µm.

Single-edged semi-circular notches with 10 and 20 μm radii were prepared using focused ion beam milling



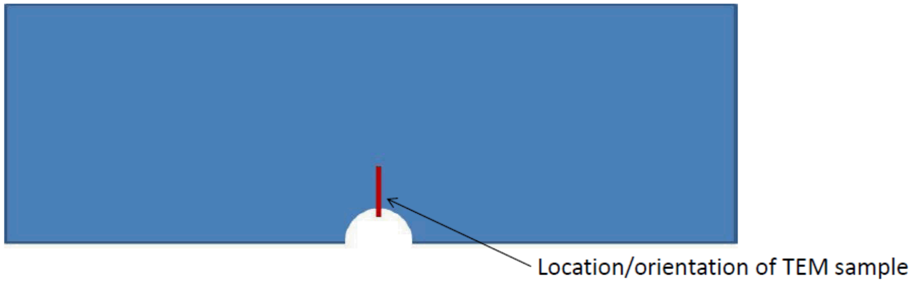
Example of the free-standing electrodeposited NC Ni-Fe tensile specimens used in this study. Gage width: 0.5 mm, gage length: 2.4 mm, thickness: 10-20 μm.

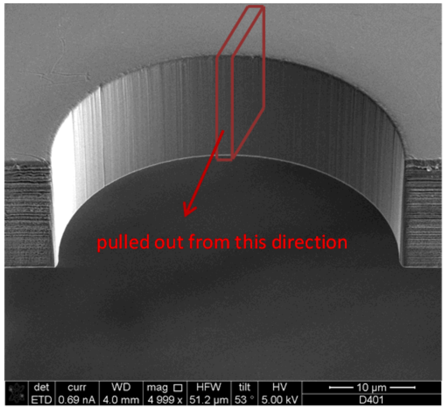


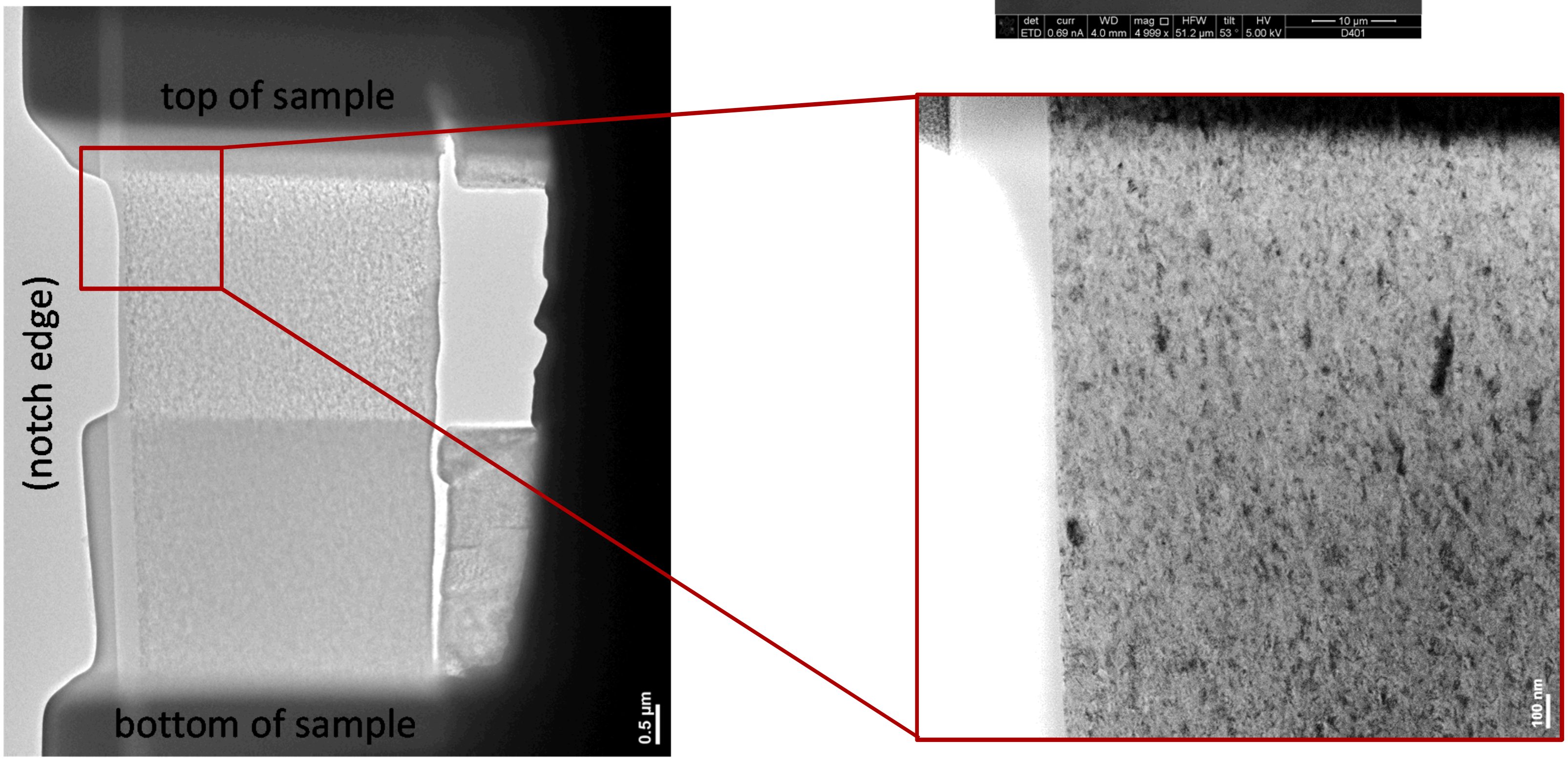
Schematic representation (a) and SEM micrographs (b,c) of the semi-circular notches FIB-milled at 30kV, 3nA

* Theoretical elastic stress concentration factors, Kt=2.89 and 2.43 for 10 and 20 μm notches, respectively

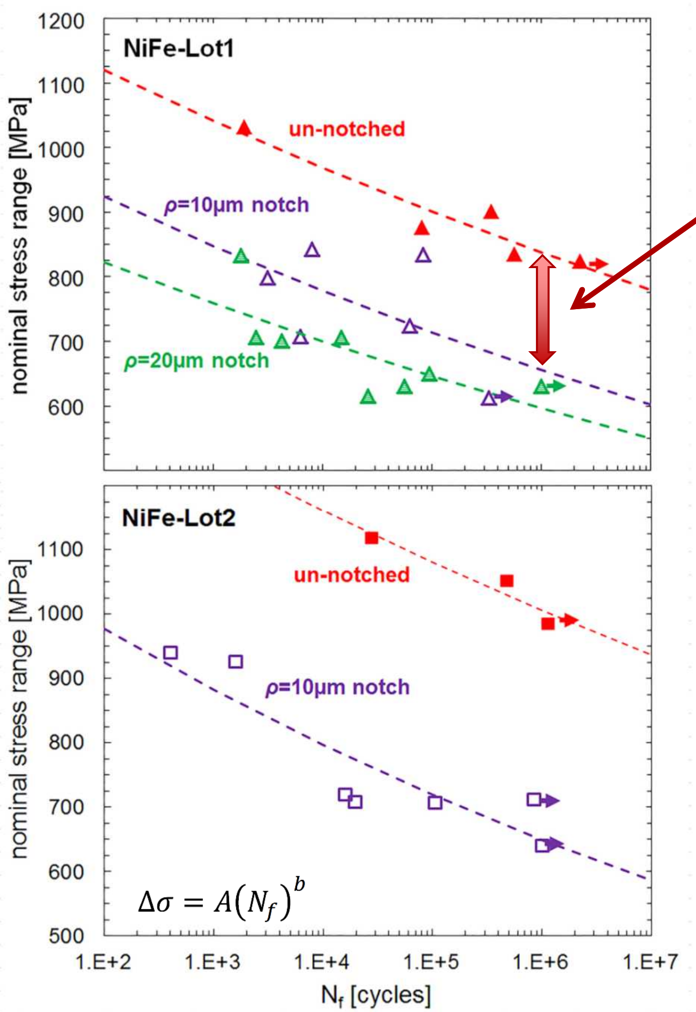
No obvious FIB-induced abnormal grain growth under notches







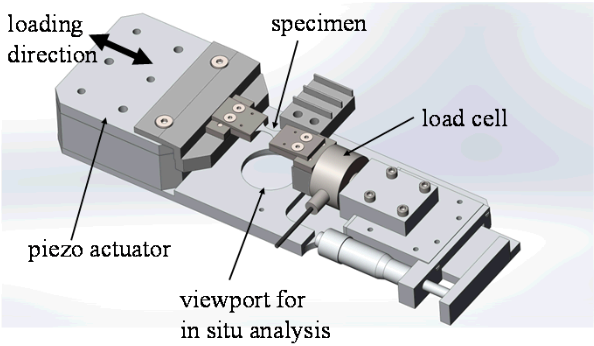
Notched fatigue tests revealed dramatic fatigue stress concentrations and high notch sensitivity!



$$K_f = \frac{\Delta\sigma_{unnotched,lim}}{\Delta\sigma_{notched,lim}}$$

$$q = \frac{(K_f - 1)}{(K_t - 1)}$$

q = 0 (totally notch insensitive)
q = 1 (fully notch sensitive, $K_f = K_t$



- Custom built piezo actuated *in situ* fatigue tester for thin foil tesing
- Constant-load, 4-6 Hz (sine) used in this study

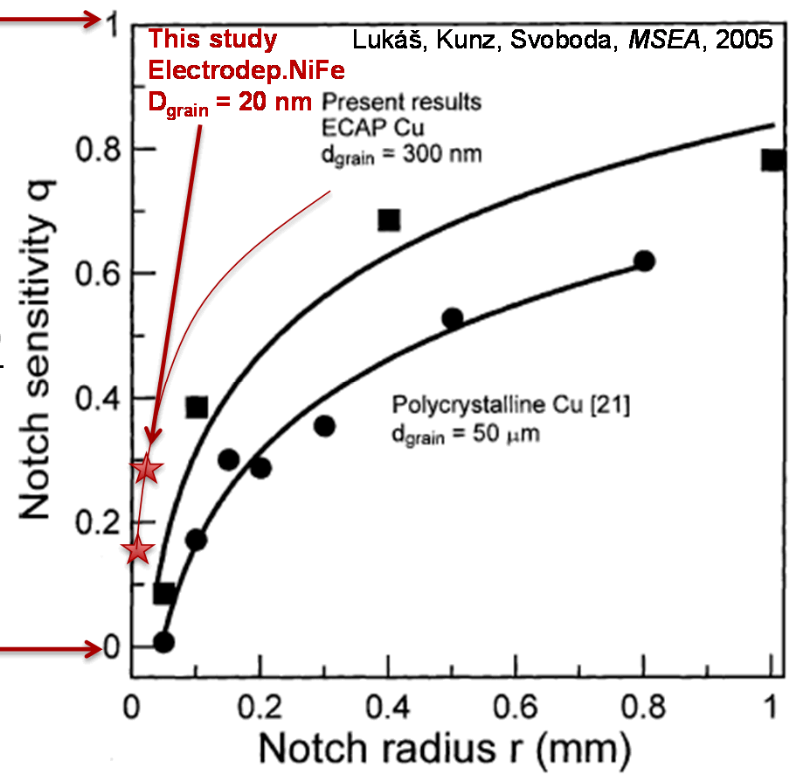
- Pronounced fatigue stress concentrations were observed due to the notches
- Notch sensitivity factors, q:
  - 0.24 (20µm notch, Lot1)
  - 0.15 (10µm notch, Lot1)
  - 0.29 (10µm, Lot2)

Not only is stress concentration *relevant,* but the notch sensitivity is an order of magnitude larger than for conventional metals!

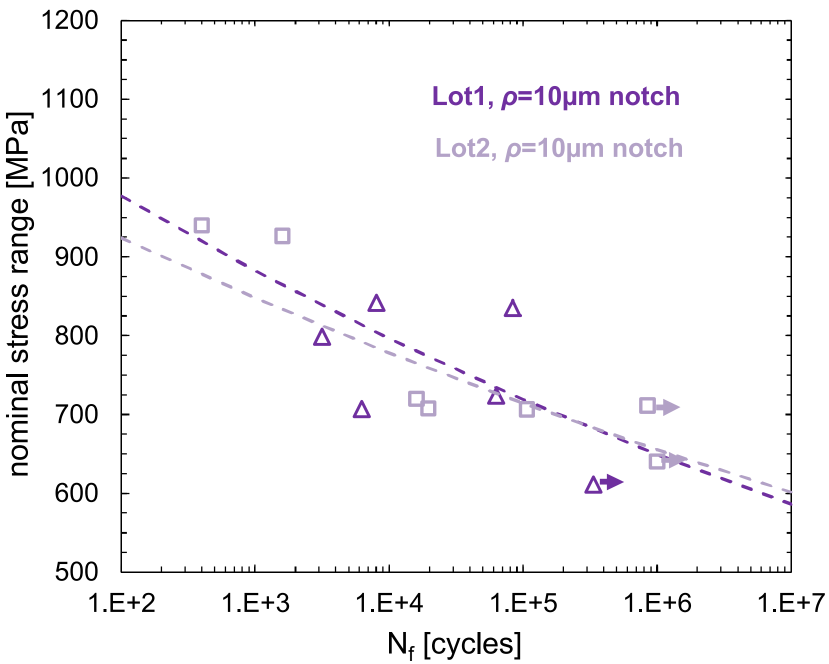
Notches obey elastic stress concentration factor

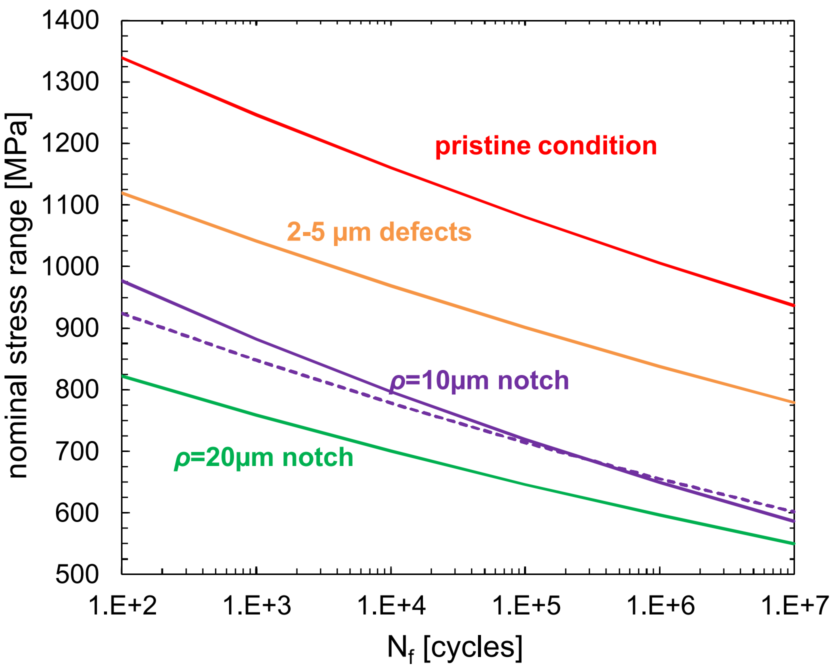
$$q = \frac{(K_f - 1)}{(K_t - 1)}$$

Notches are Irrelevant!!!



Primary vs. Secondary defects (critical flaw)

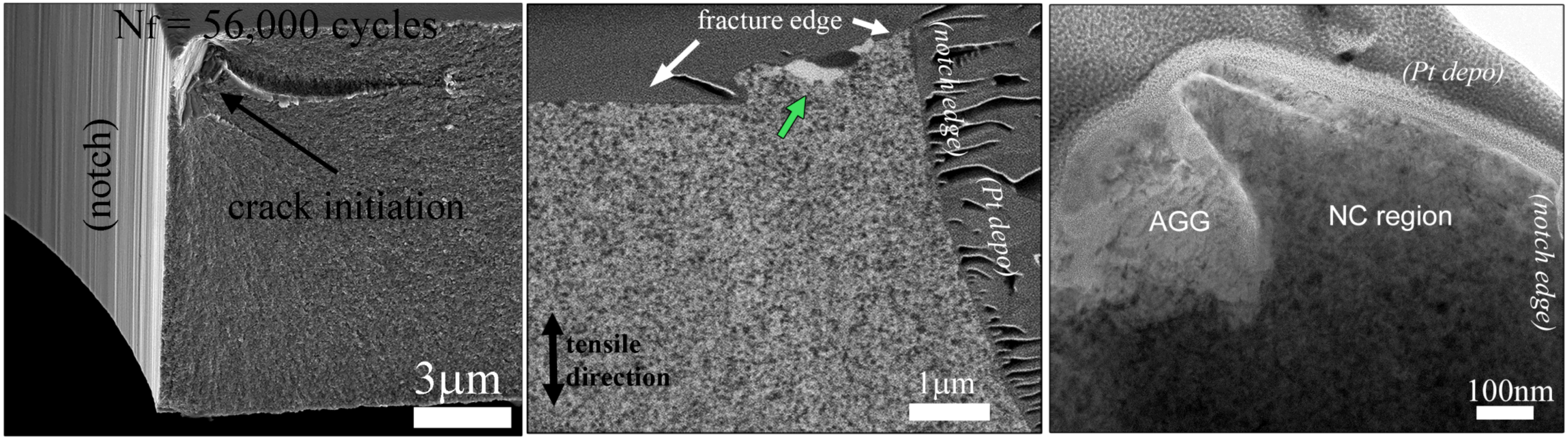




- The <u>10 µm notched</u> Lot1 and Lot2 specimens showed nearly identical fatigue performance
- The larger stress concentrations from the "primary" defect (the notch) overcame the effects of the "secondary" defects (the surface flaws)
- Fatigue is a weakest-link phenomenon – the highest stress concentration wins! (or loses…)

- We begin to see the potential effects of surface flaws and can start to characterize the *actual* stress concentration effects on the fatigue performance
- The inherent notch sensitivity (or "flaw" sensitivity) leaves these materials vulnerable if a pristine condition is not reached
- Even though monotonic tests showed no difference between the materials, fatigue tells a different story…

AGG was observed directly underneath crack initiation sites

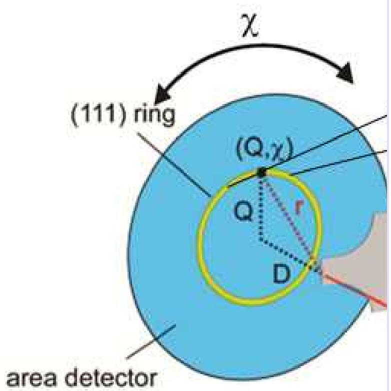


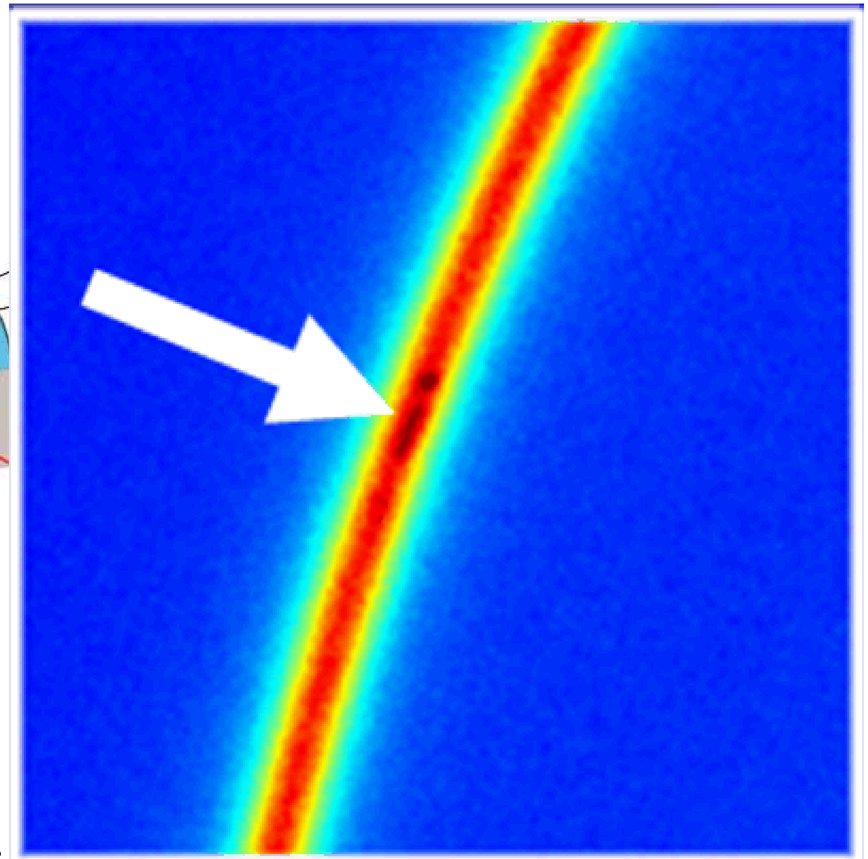
- Crack initiation regions consisted of faceted "blocky" features, similar to previous reports of fatigued AGG regions
- FIB cross-sectioning revealed 1-2 μm sized grains underneath the notches!

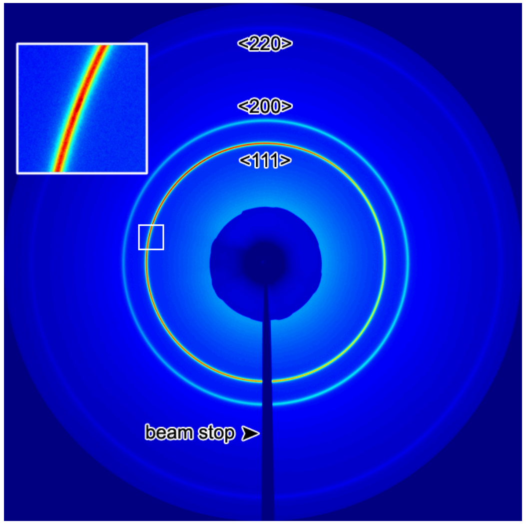
Well-developed regions of AGG were found underneath the fatigued notched regions! This validates our hypothesis that the AGG is a deformation process (not pre-existing)

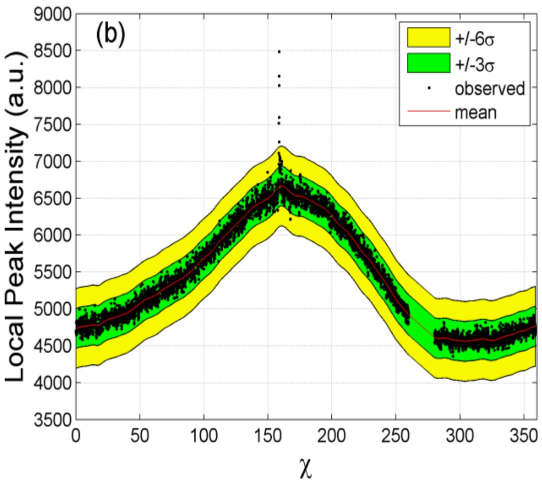
Are these processes cyclically induced or only a result of the final fracture?

Transmission synchrotron X-ray diffraction can provide means to detect AGG in a NC matrix



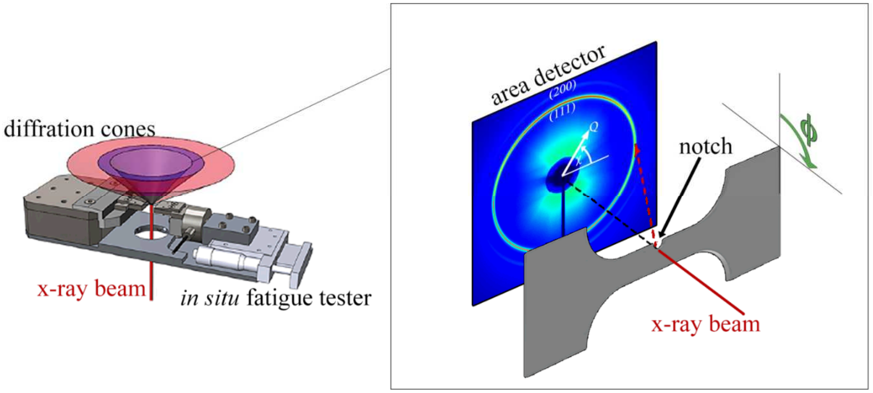


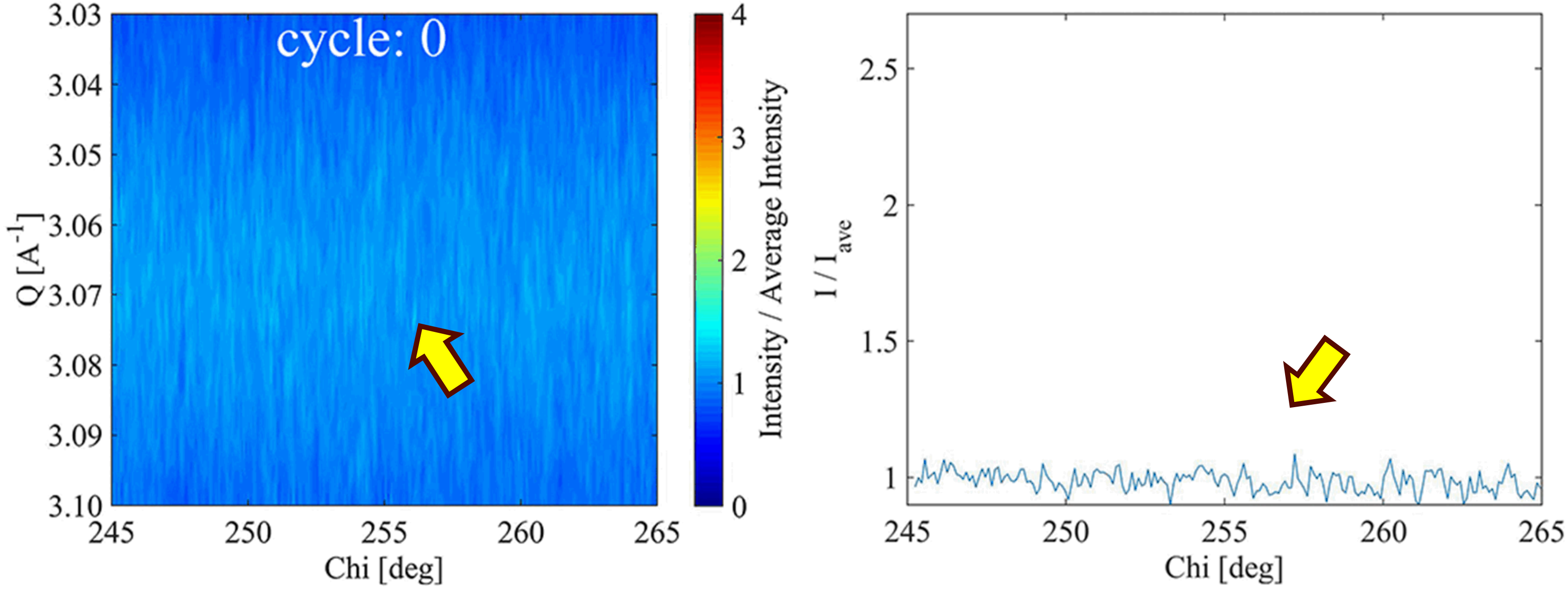




- Using this technique, we can identify clusters of abnormally large grains by statistically anomalous intensity spikes around the diffraction rings

In-situ scans showed the same anomalous peak present after 26,600 cycles, *30,000+ cycles* <u>*before final fracture*</u>!





Conclusions

- Micro-notches provided effective stress concentrations that dramatically reduced the fatigue resistance (notch sensitivities an order of magnitude larger than CG metals)
- The notches also led to localized deformation and fracture initiation
- *Stress concentrations are relevant for NC metals!* Most likely dependent on notch-to-grain size ratios…
- Abnormal grain growth (AGG) is a predominant precursor for crack initiation, the onset of which is likely dependent on the stress concentrations
- Understanding the AGG/crack initiation process will be paramount to devising means of improving the notch sensitivity in NC metals

The ultrahigh fatigue resistance of NC metals make them attractive for advanced applications. But, for these materials to be reliable we must understand the underlying deformation and fracture mechanisms, in addition to their sensitivity to flaws and defects.

Using a combination of micro-notched specimens and non-destructive detection of AGG during fatigue, we can begin to understand the origins, causes, and effects of stress concentrations on the mechanical properties in NC metals.

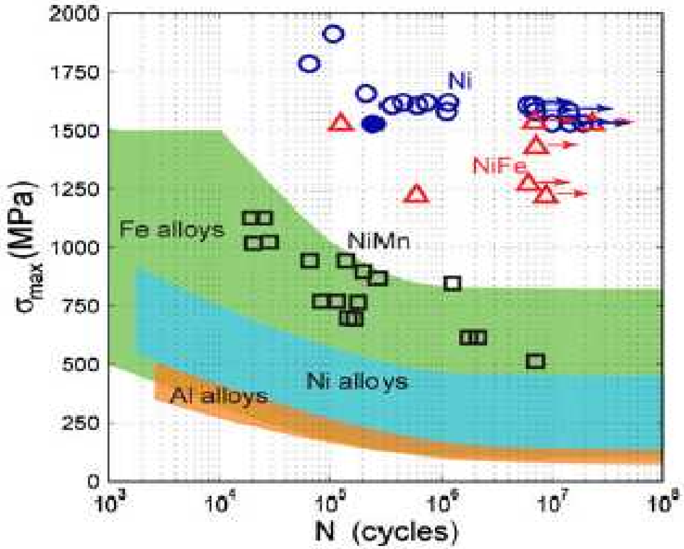
Acknowledgments

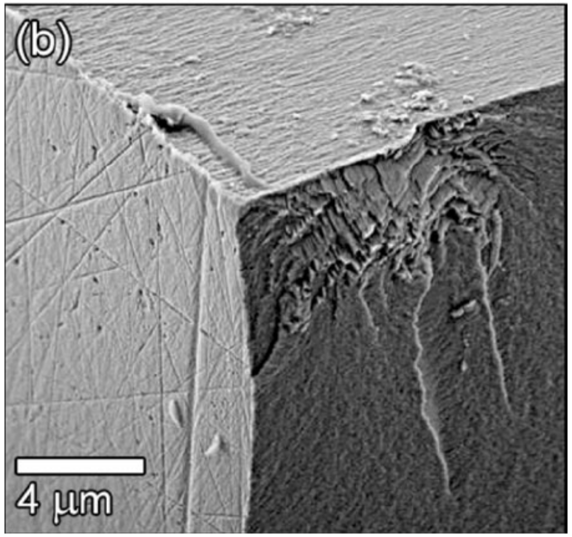
- This work was funded by the U.S. DOE, Office of Basic Energy Sciences, Division of Materials Science and Engineering
- A portion of the microscopy and FIB work was performed at the Center for Integrated Nanotechnologies (CINT)
- Synchrotron work was performed at beamline 11-3 at Stanford Synchrotron Radiation Lightsource (SSRL)
- Thanks to Apurva Mehta, Doug Van Campen, Bill Mook, Michael Rye, and Amy Allen

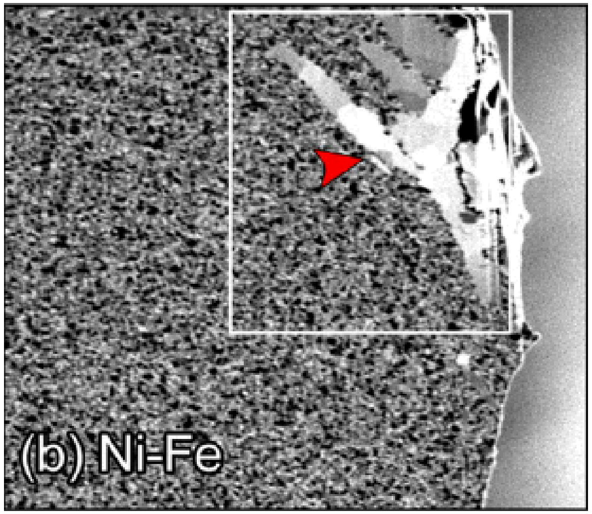
Extra Slides

The suppression of dislocation-based mechanisms can lead to dominant GB-related responses, e.g. AGG

Boyce, Met. Mat. Trans. A, (2011)

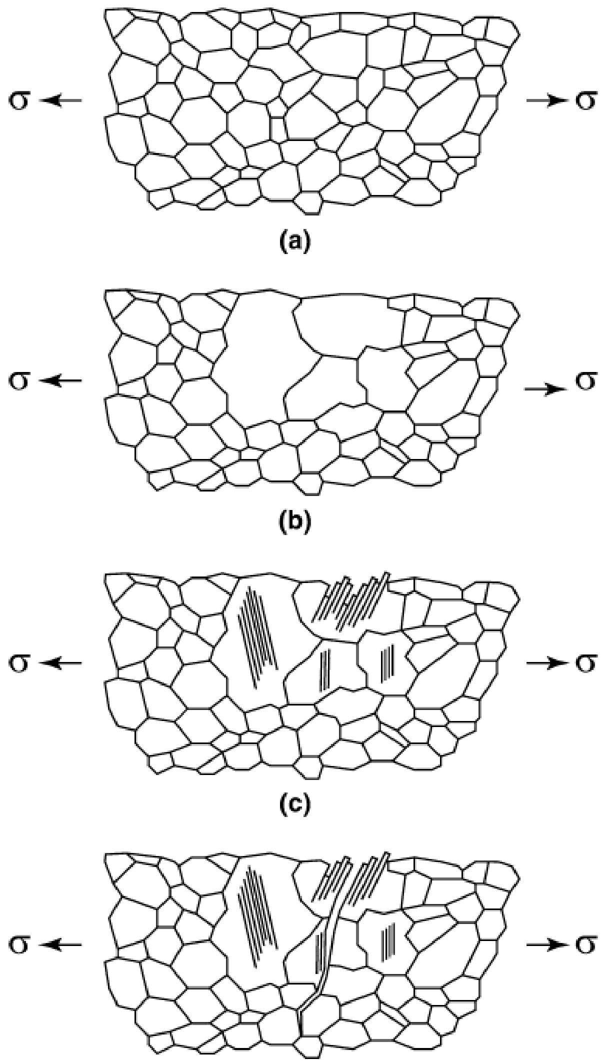






- High fatigue strengths are commonly observed in NC metals
- This is usually attributed to the inhibition of dislocation-based plasticity (e.g. persistent slip bands) and thus, a delay in the crack initiation and fracture
- With this suppression of "traditional" fatigue deformation and fracture, new GB related mechanisms arise

Proposed mechanisms for crack initiation in NC metals



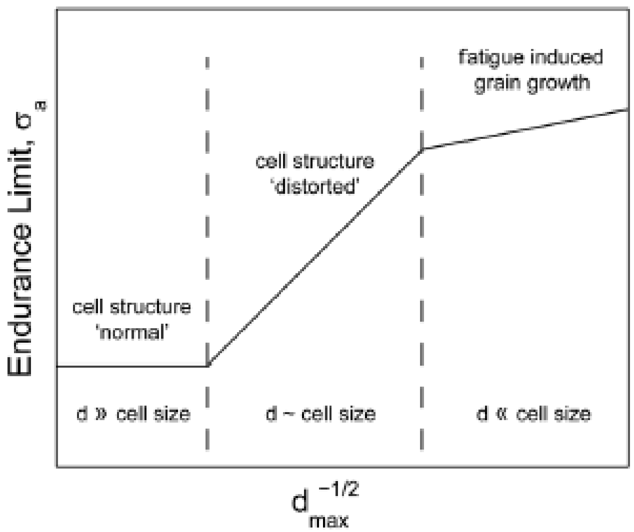
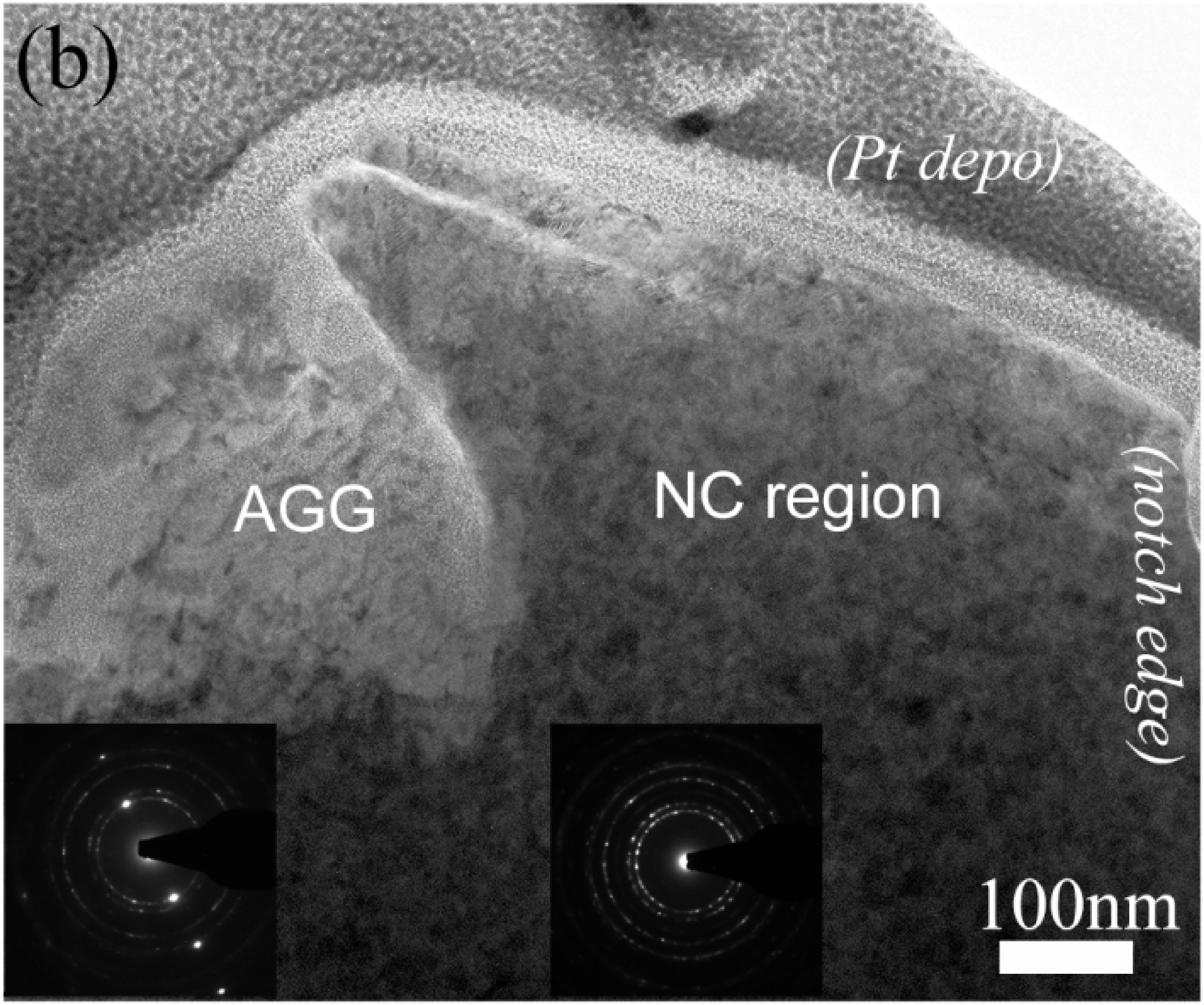
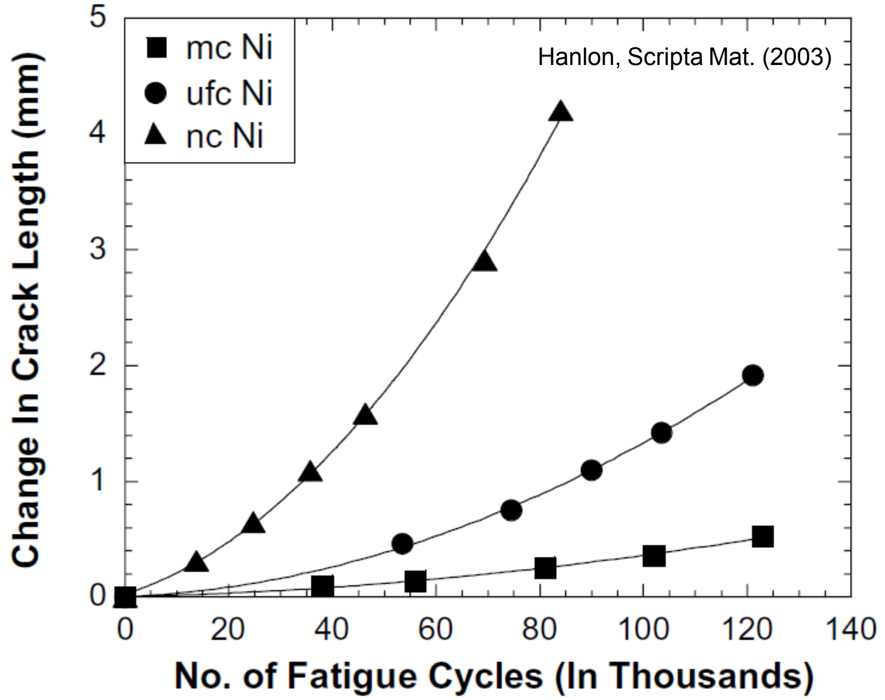


Figure 14. Schematic showing different scaling regimes for fatigue behavior with respect to the grain size.

AGG in NC Metals

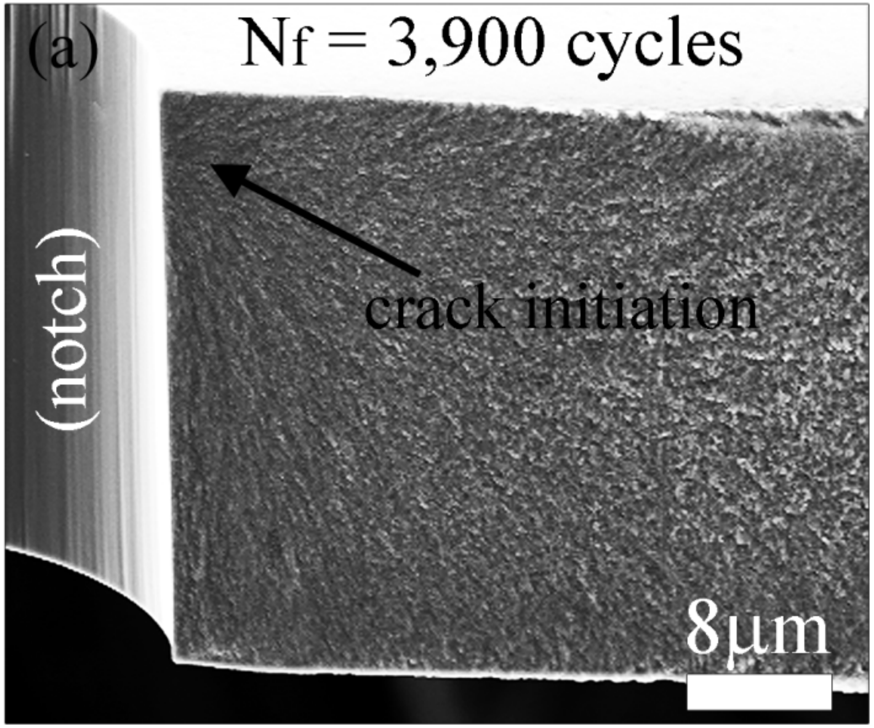

(b)
(Pt depo)
AGG
NC region
(notch edge)
100nm

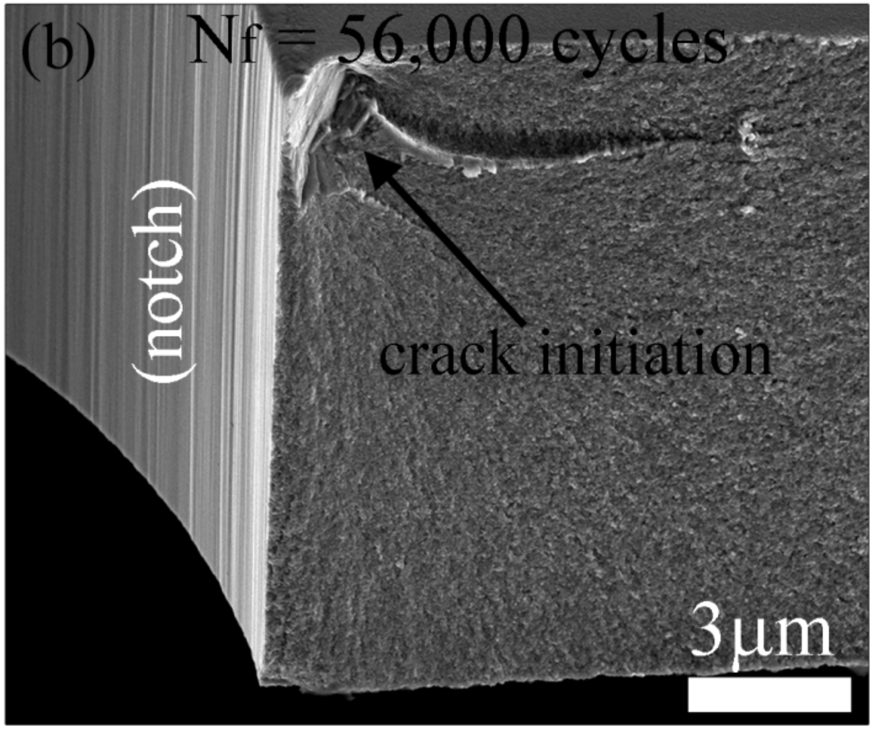
Fracture toughness of NC metals has been shown to be much lower than for ufg and CG counterparts



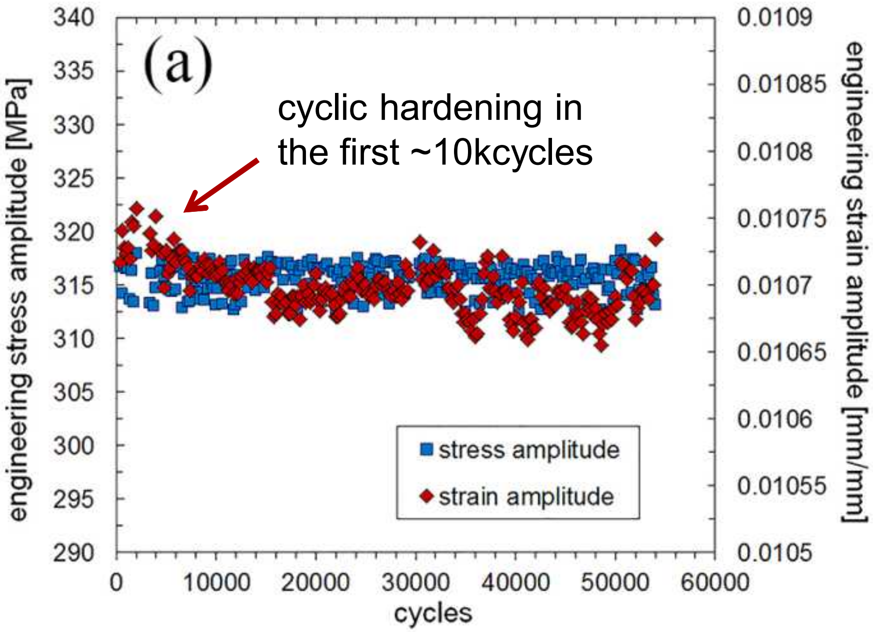
In samples that fractured > ~10,000, the initiation zones consisted of "blocky" protrusions (not present in samples with Nf<10,000 cycles)

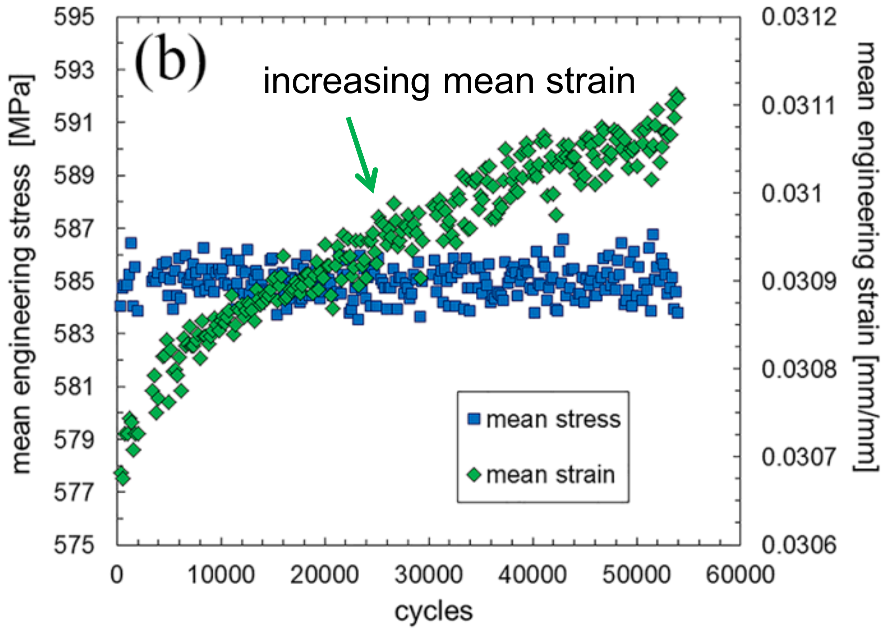
Possible change of mechanisms? Possible time-dependent (or cyclic dependent) behavior?





Some cyclic hardening was observed, in addition to monotonic "ratcheting" throughout the fatigue life



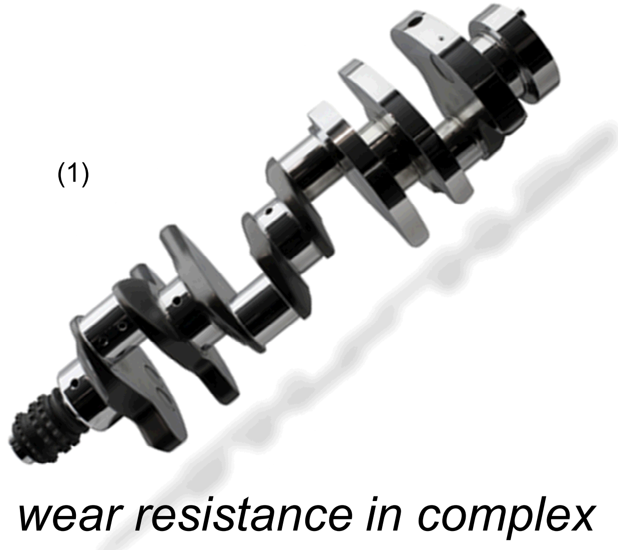


- Cyclic hardening is expected based on work-hardening effects observed during monotonic tension (likely some dislocation activity)
- Monotonic accumulation of strain (separate from the cyclic hardening effects) indicates possible boundary-related activity (e.g. Coble creep mechanisms)

Sample Preparation

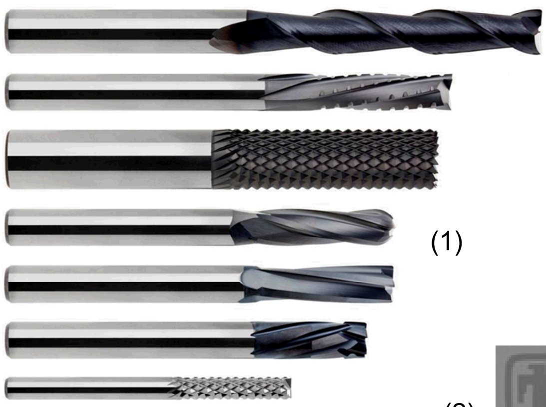
Current & potential applications for NC metals

(1)



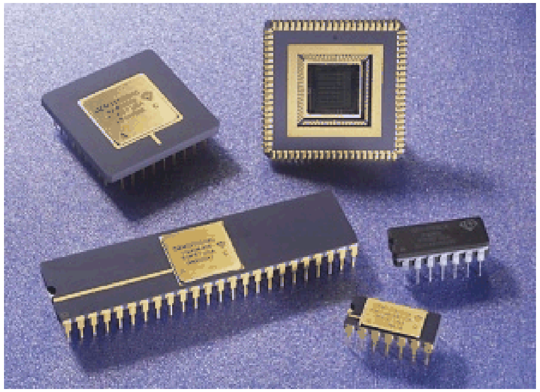
wear resistance in complex geometries

Ultra-hard thin films



(1)

(3)



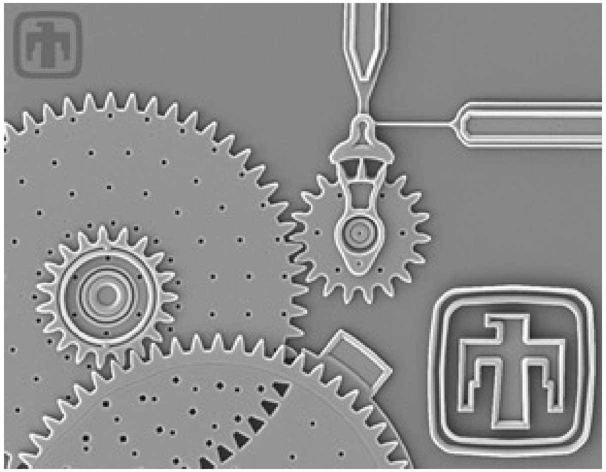
Microcircuits

(1)

Solid/dry lubricant coatings

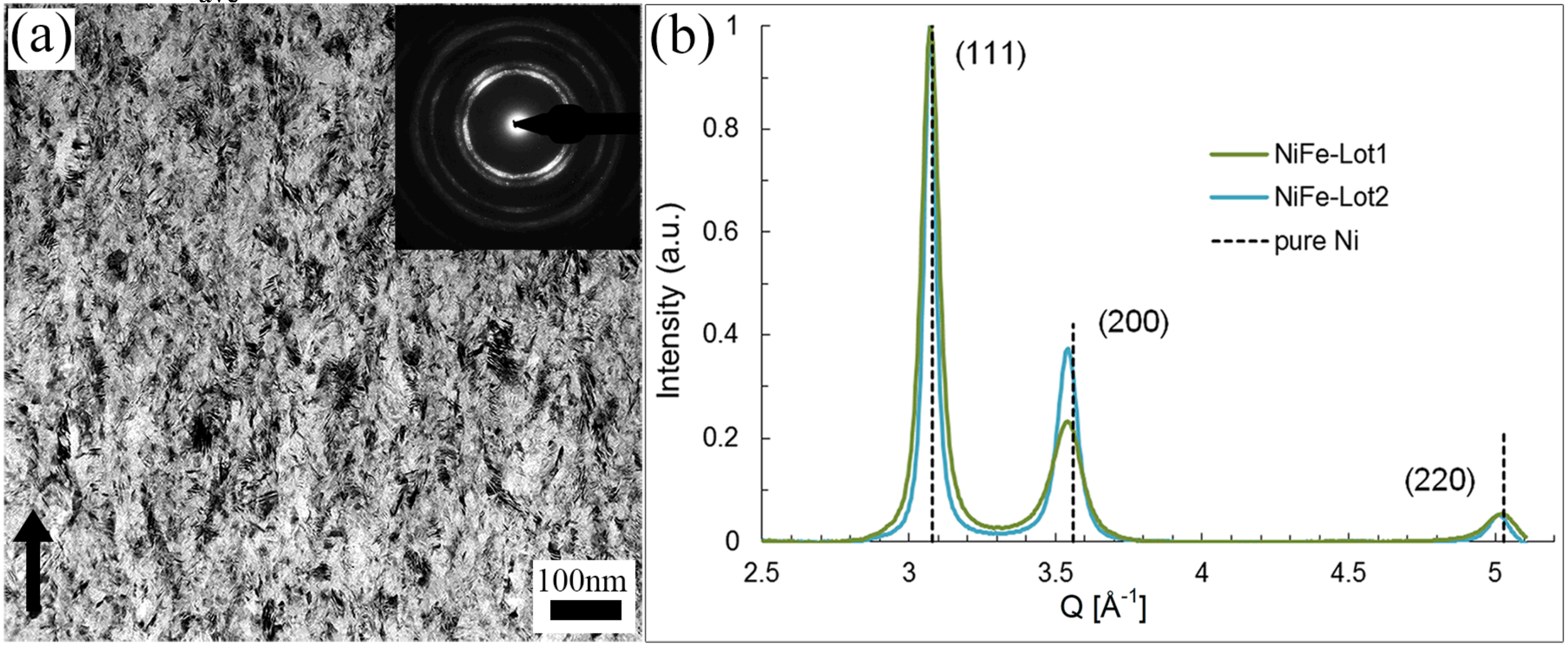
(1) www.enduracoatings.com
(2) www.lotvacuumamerica.com
(3) www.sri.com

(2)

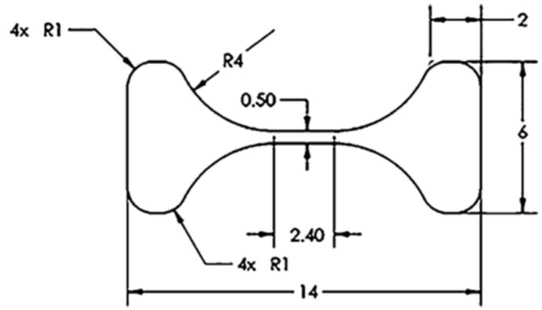


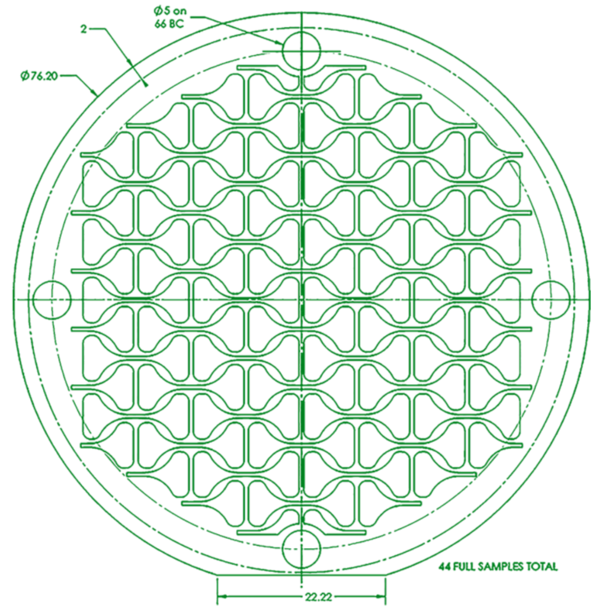
MEMS & Micromachines

d_{ave}=10-20 nm



(a)
100nm
(b)
(111)
(200)
(220)
NiFe-Lot1
NiFe-Lot2
pure Ni
Intensity (a.u.)
Q [Å^{-1}]
1
0.8
0.6
0.4
0.2
0
2.5
3
3.5
4
4.5
5



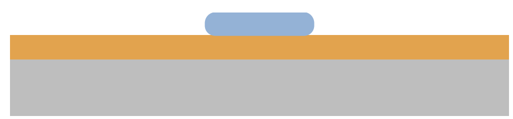


Nickel/iron chemistry make-up	
$NiSO_4$ Nickel Sulfate	112 g/L
$FeSO_4$ Iron sulfate	5 g/L
$Na_3C_6H_5O_7$ Sodium Citrate	75 g/L
KSO_4 Potassium Sulfate	1.5 g/L
$C_7H_5NO_3S$ Saccharin	1 g/L

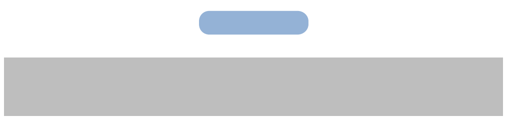
1. 100 nm deposition of Cu on Si wafer

2. Patterned front side of wafer

3. Electrodeposit material

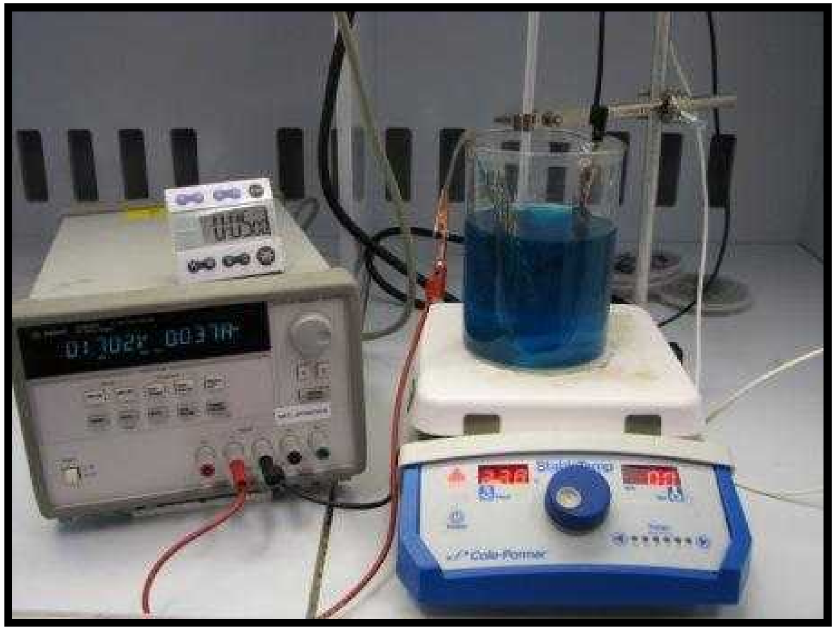


4. Remove photoresist

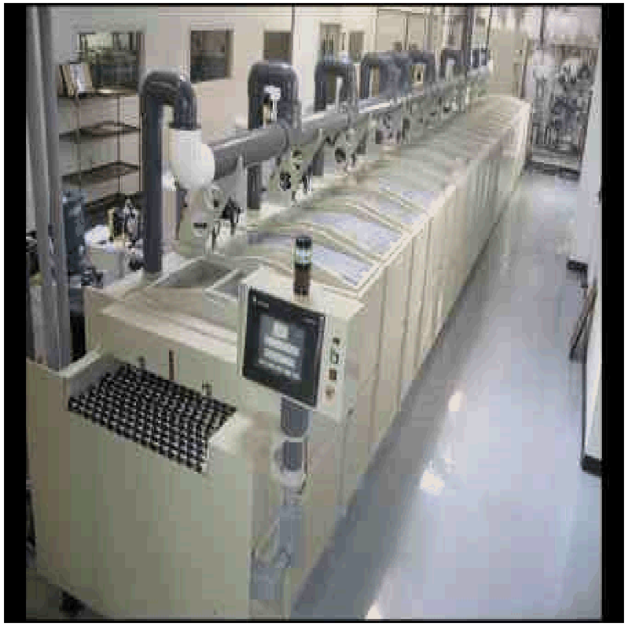


5. Lift-off Ni-Fe samples in Cu etch

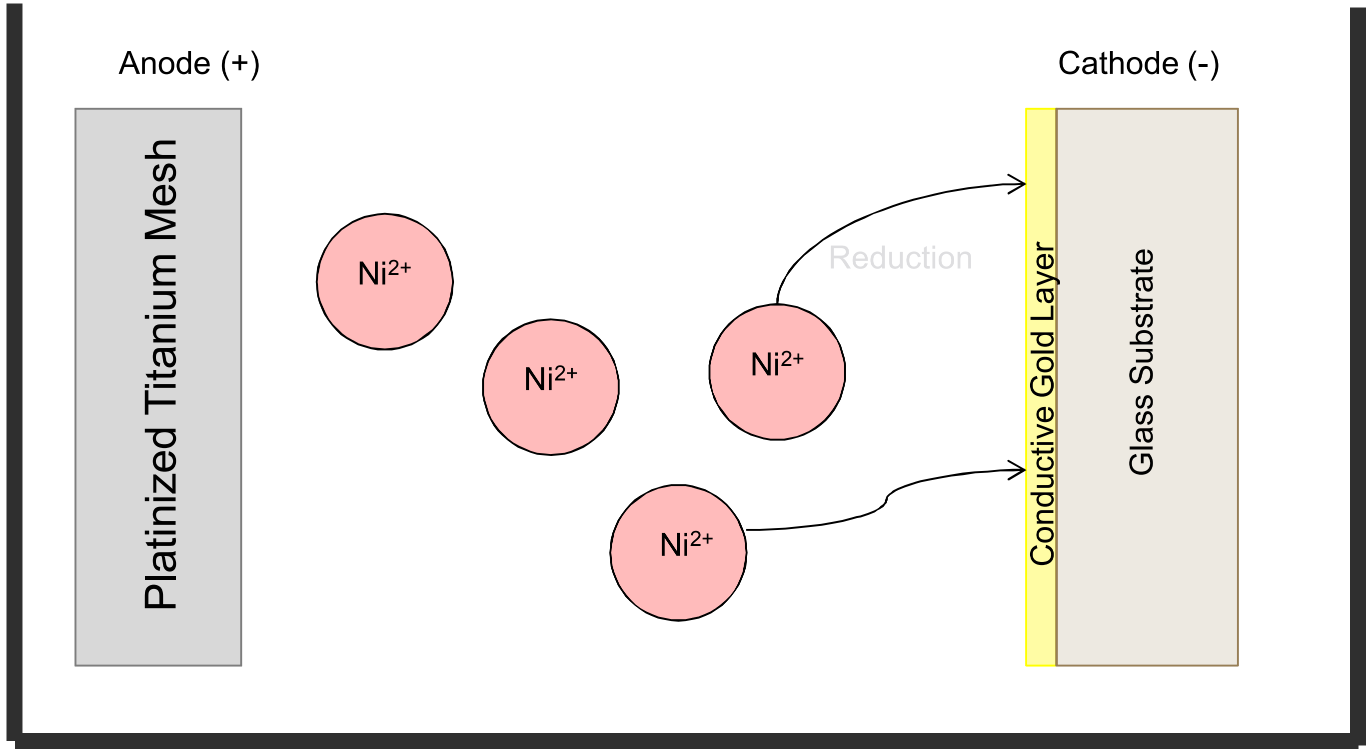
Bench Top to Industrial Plating



Bench Top Plating

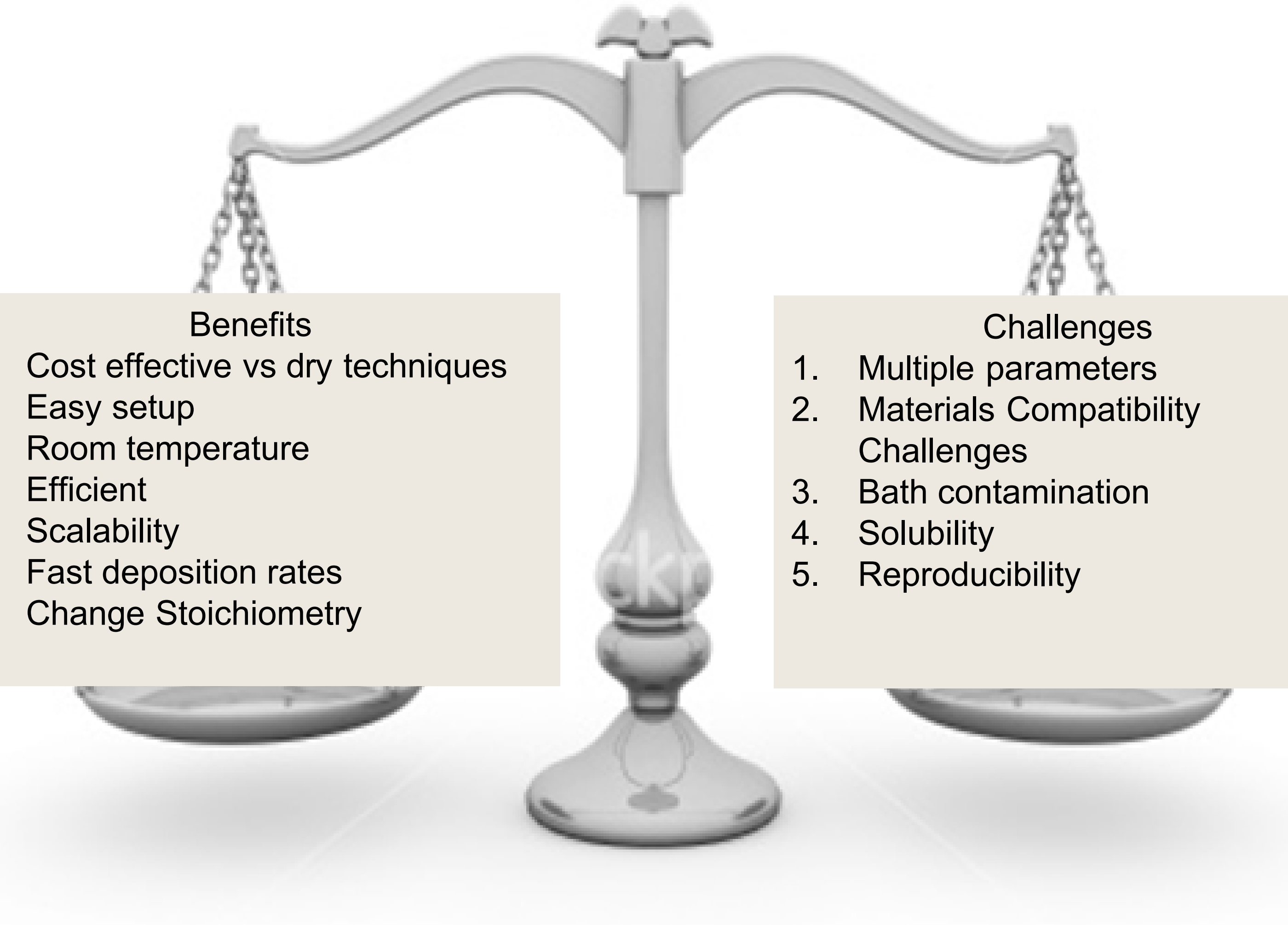


Industrial Plating Bench



Anode (+)
Cathode (-)
Platinized Titanium Mesh
Ni2+
Ni2+
Ni2+
Ni2+
Reduction
Conductive Gold Layer
Glass Substrate

ECD Benefits & Challenges



Benefits

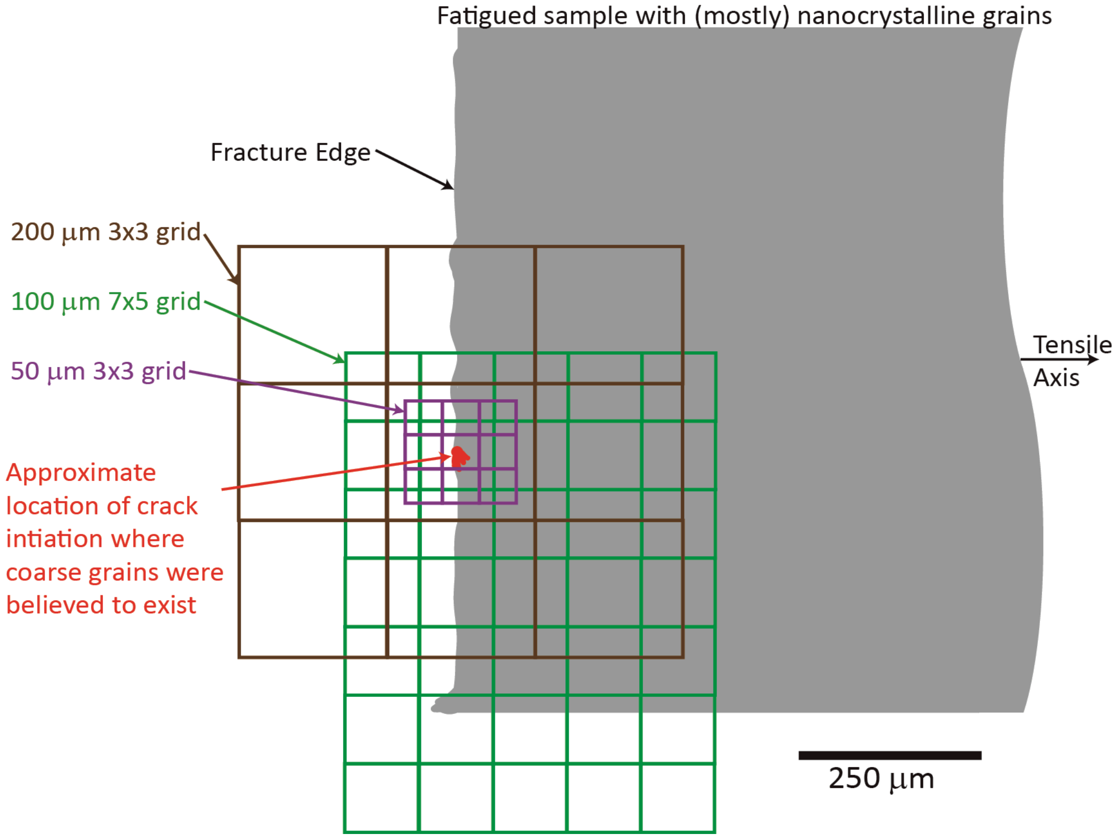
1. Cost effective vs dry techniques
2. Easy setup
3. Room temperature
4. Efficient
5. Scalability
6. Fast deposition rates
7. Change Stoichiometry

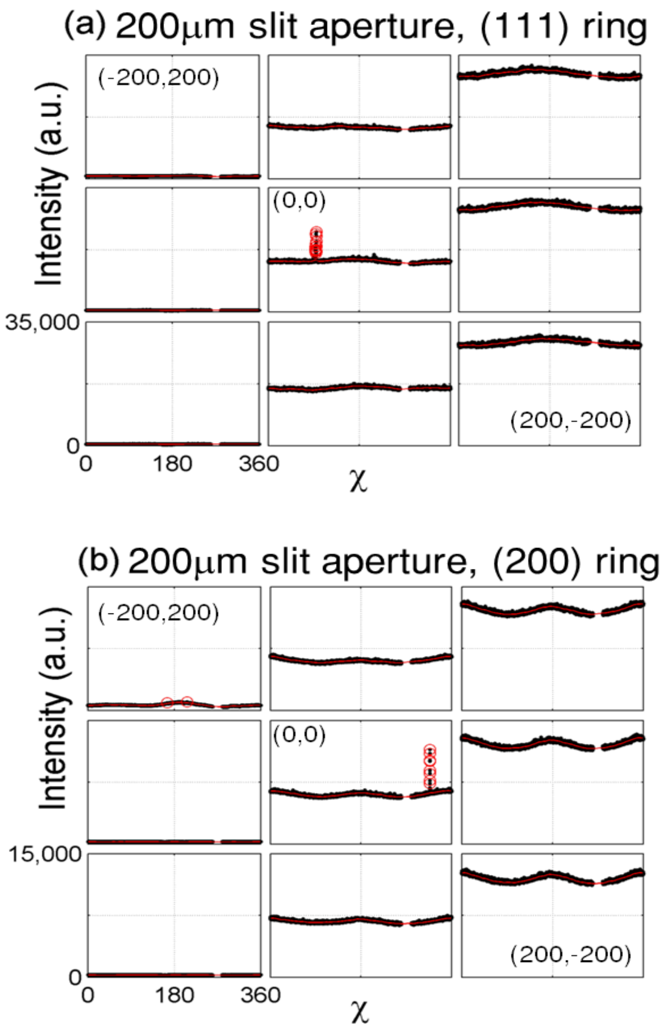
Challenges

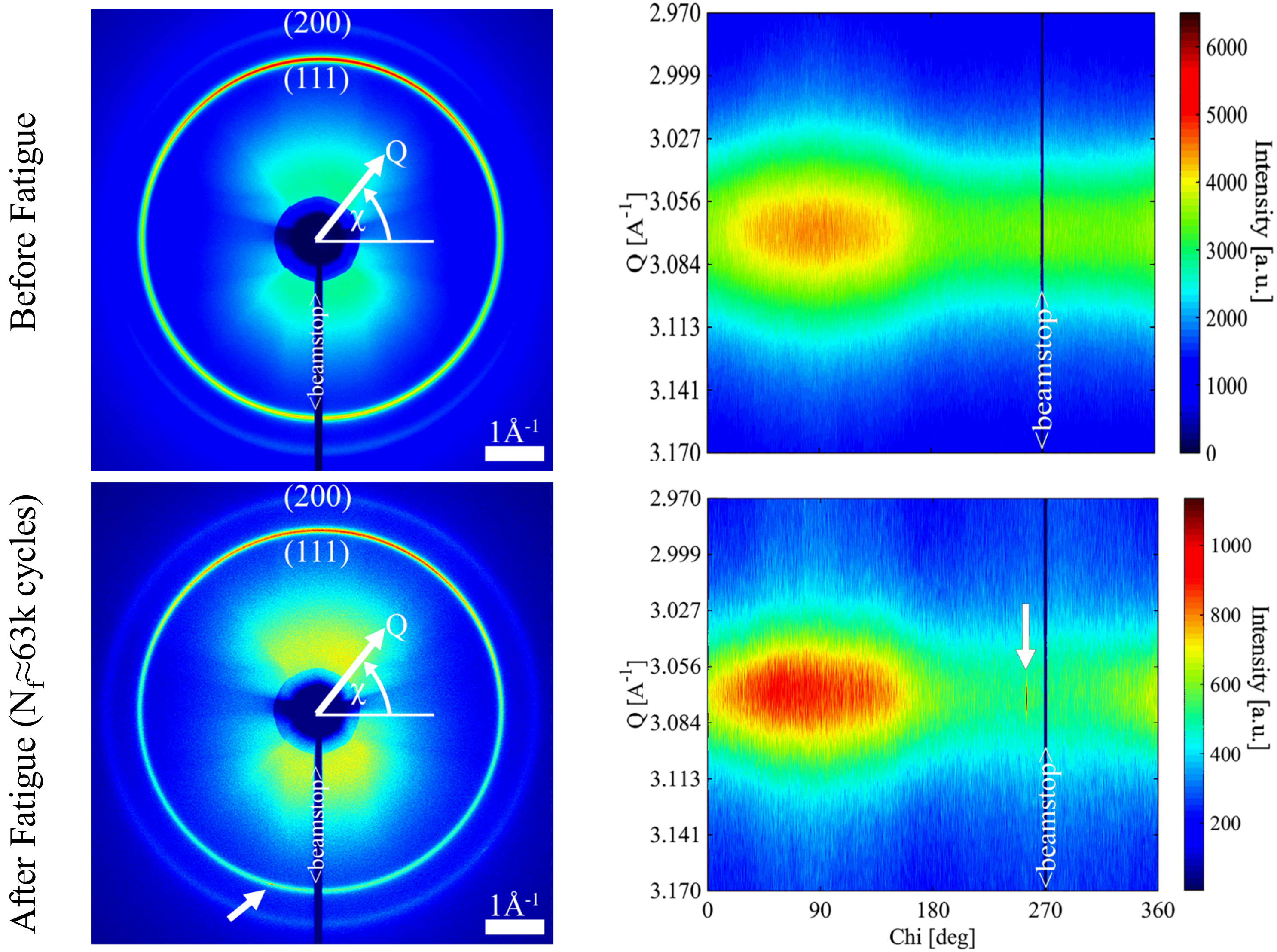
1. Multiple parameters
2. Materials Compatibility Challenges
3. Bath contamination
4. Solubility
5. Reproducibility

X-ray Diffraction

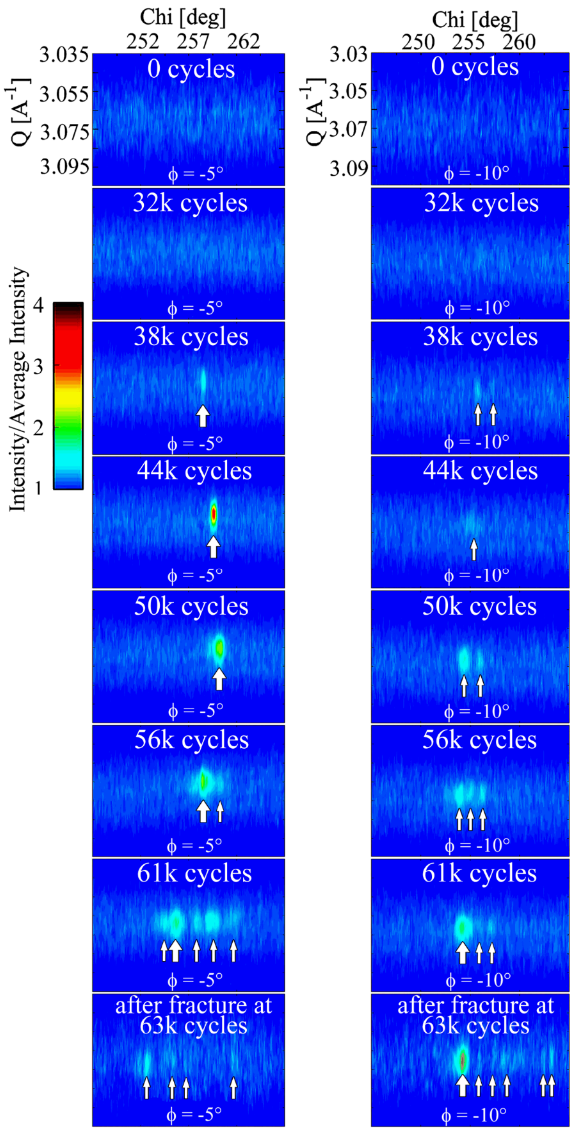
This x-ray technique identified the AGG region, despite only occupying < 0.0001% of the sampled volume!



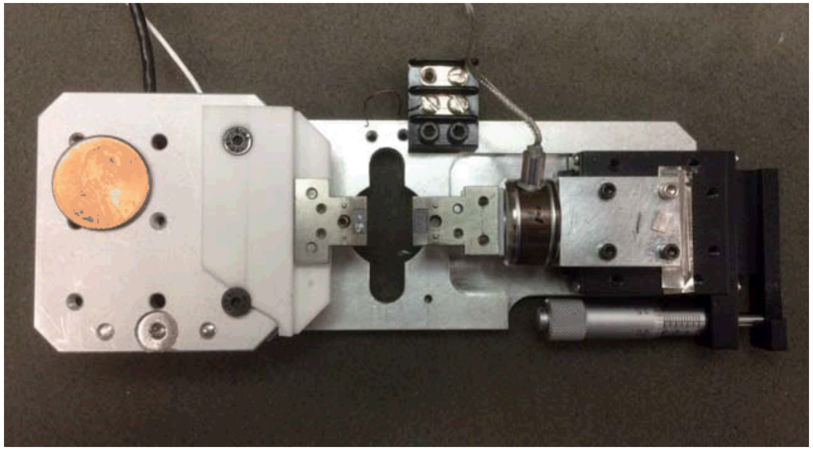




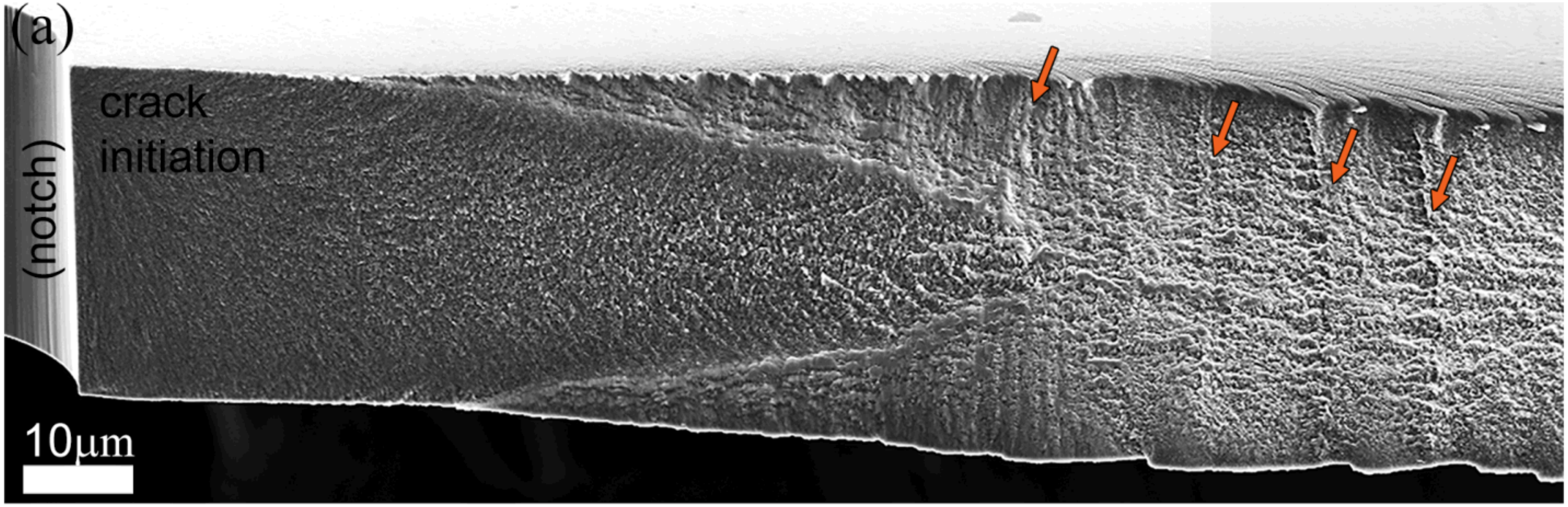
Before Fatigue
After Fatigue (N_f≈63k cycles)
(200)
(111)
Q
χ
<beamstop>
1Å^{-1}
Q [Å^{-1}]
2.970
2.999
3.027
3.056
3.084
3.113
3.141
3.170
Chi [deg]
0
90
180
270
360
Intensity [a.u.]
0
1000
2000
3000
4000
5000
6000
200
400
600
800
1000

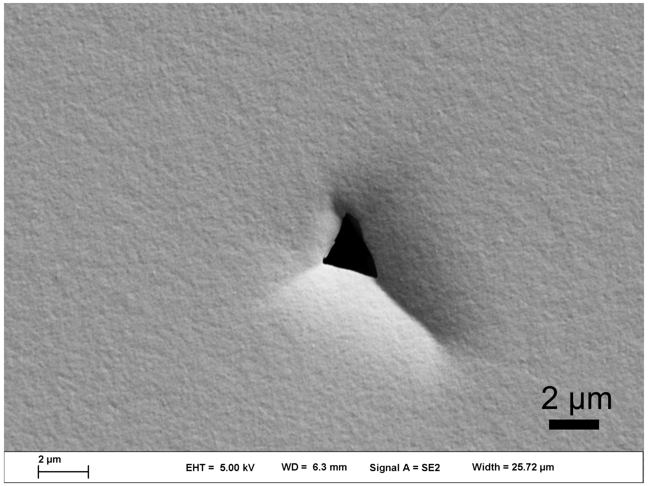

Chi [deg]
252 257 262
3.035
3.055
3.075
3.095
Q [A^-1]
0 cycles
32k cycles
38k cycles
44k cycles
50k cycles
56k cycles
61k cycles
after fracture at 63k cycles
ϕ = -5°
Intensity/Average Intensity
4
3
2
1
Chi [deg]
250 255 260
3.03
3.05
3.07
3.09
ϕ = -10°

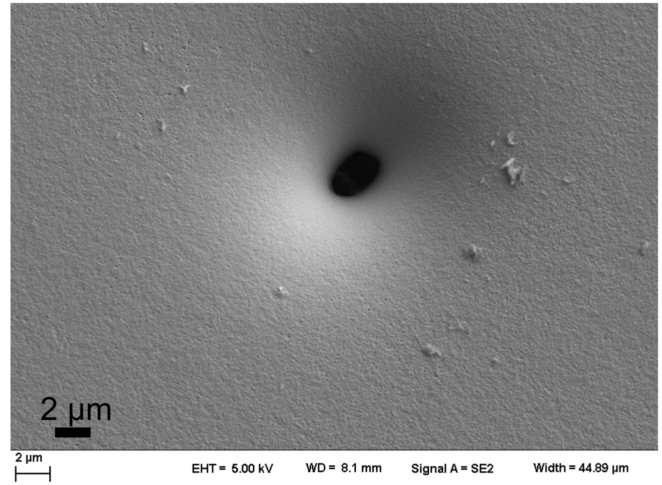
Force-controlled fatigue tests to failure were performed on the notched NC specimens

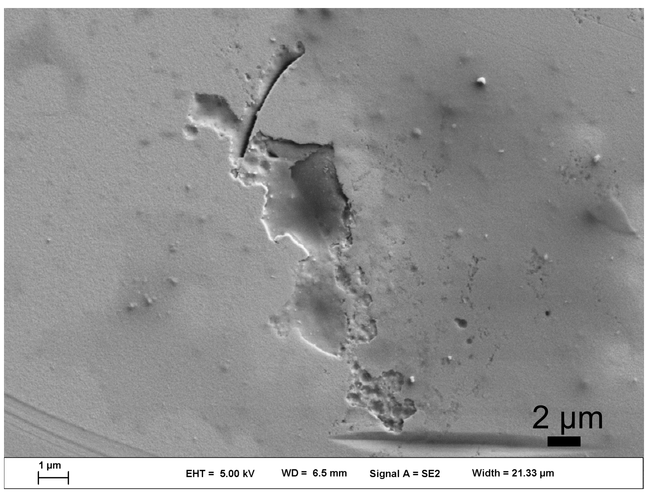


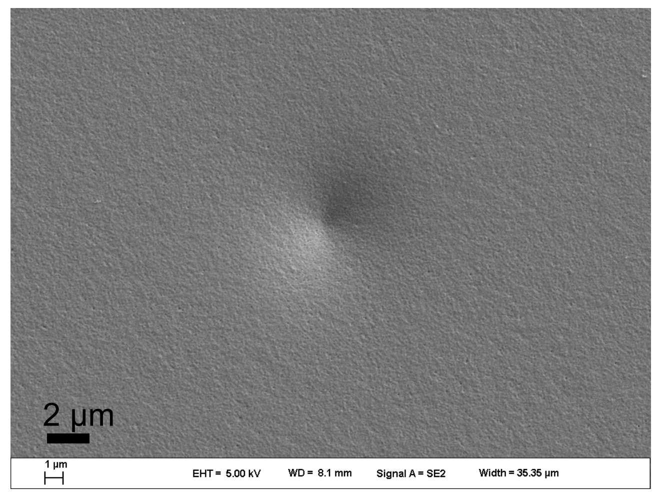
- Piezo-actuated custom tester
- R≈ 0.3 (f_{min}/f_{max}) [i.e. tension-tension fatigue, to avoid foil buckling]
- freq : 4-6 Hz (sine)




2 µm
2 µm
EHT = 5.00 kV
WD = 6.3 mm
Signal A = SE2
Width = 25.72 µm


2 µm
2 µm
EHT = 5.00 kV
WD = 8.1 mm
Signal A = SE2
Width = 44.89 µm


2 µm
1 µm
EHT = 5.00 kV
WD = 6.5 mm
Signal A = SE2
Width = 21.33 µm


2 µm
1 µm
EHT = 5.00 kV
WD = 8.1 mm
Signal A = SE2
Width = 35.35 µm