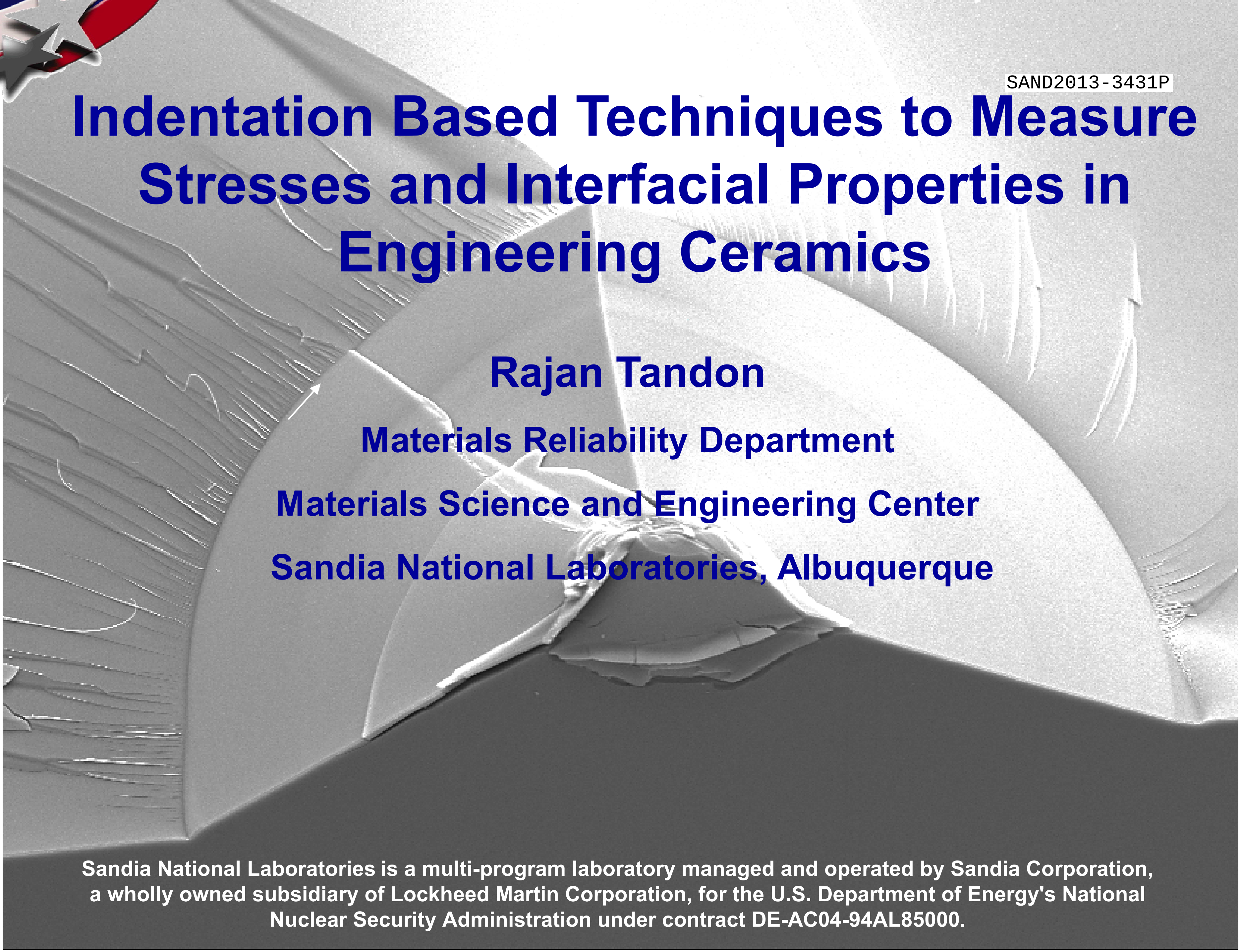
SAND2013-3431P

Indentation Based Techniques to Measure Stresses and Interfacial Properties in Engineering Ceramics



Rajan Tandon

Materials Reliability Department

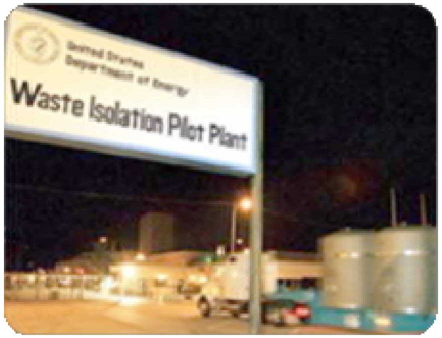
Materials Science and Engineering Center

Sandia National Laboratories, Albuquerque

Sandia's Sites

Albuquerque, New Mexico

Carlsbad, New Mexico



Tonopah, Nevada

Livermore, California

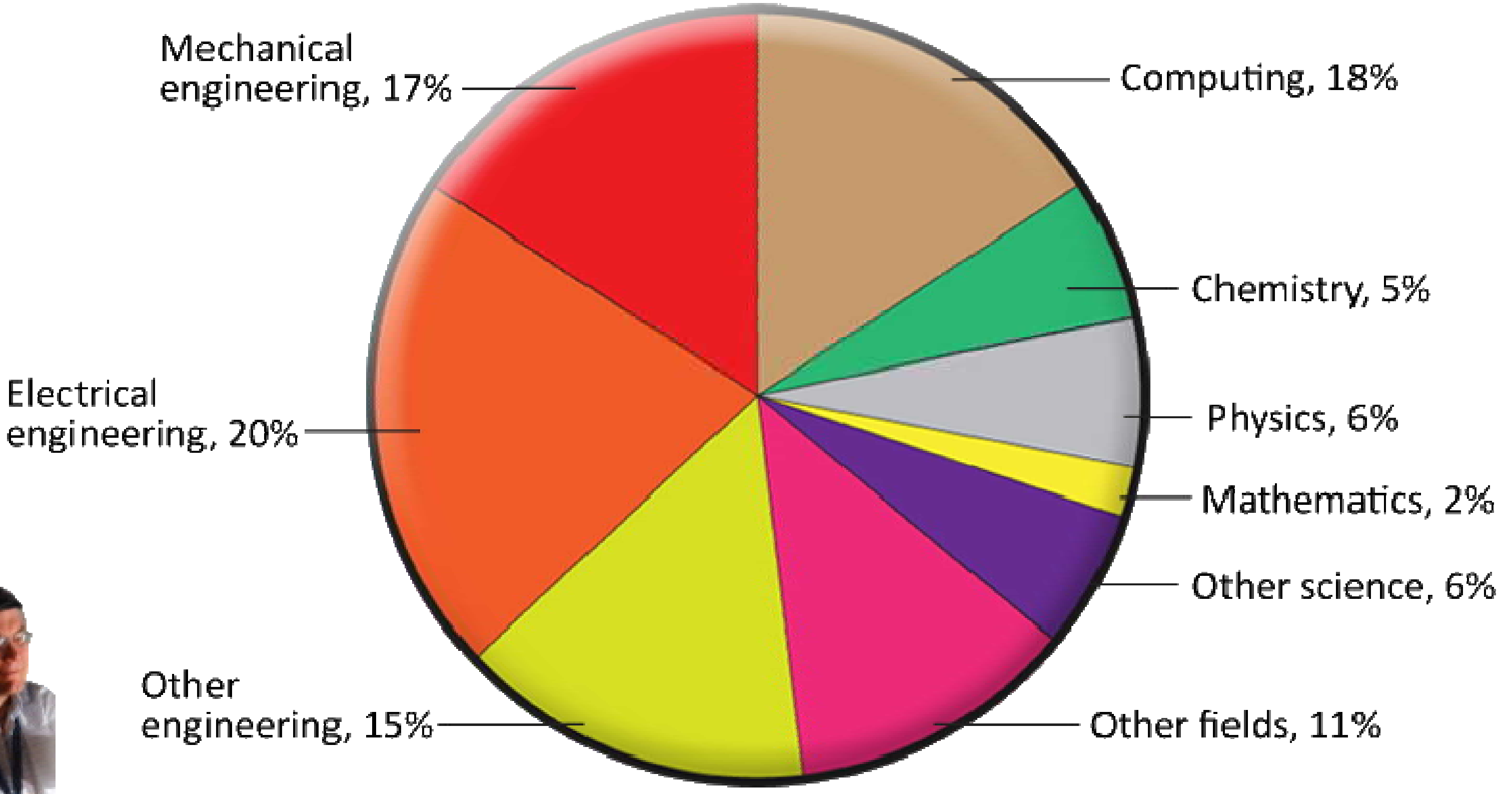
Amarillo, Texas

Kauai, Hawaii

Our Workforce

- **Onsite workforce: 11,711**
- **Regular employees: 9,238**
- **Gross payroll: ~$981M**

Data for FY12 through end of September



Sandia National Laboratories

Sandia's Missions

Sandia's National Security Missions:

Nuclear Weapons: Ensure a safe, secure, & reliable nuclear deterrent.

- Sandia designs and integrates ~6000 parts for several weapons
- Ceramics used in active & passive electronic, seals, optics
- Sandia manufactures and procures all types of ceramic materials

Alumina, LTCC, PZT, ZnO, Glasses, ferrites, quartz, KDP, other oxides...

Defense Systems and Assessments: Help maintain U.S. military weapon superiority/intelligence.

Borides and carbides for high T, SiC, B_4C for armor, fibers, optical mat.

Homeland Security: Help protect against terrorism through advanced technology

Cerium oxide fragmentation, Quartz swirl chamber for decontamination

Energy and Infrastructure Assurance: Ensure clean, abundant, & affordable energy (**SOFC, hi-voltage capacitors, solar)**

Nonproliferation: Reduce proliferation of weapons of mass destruction & threat of accidents.

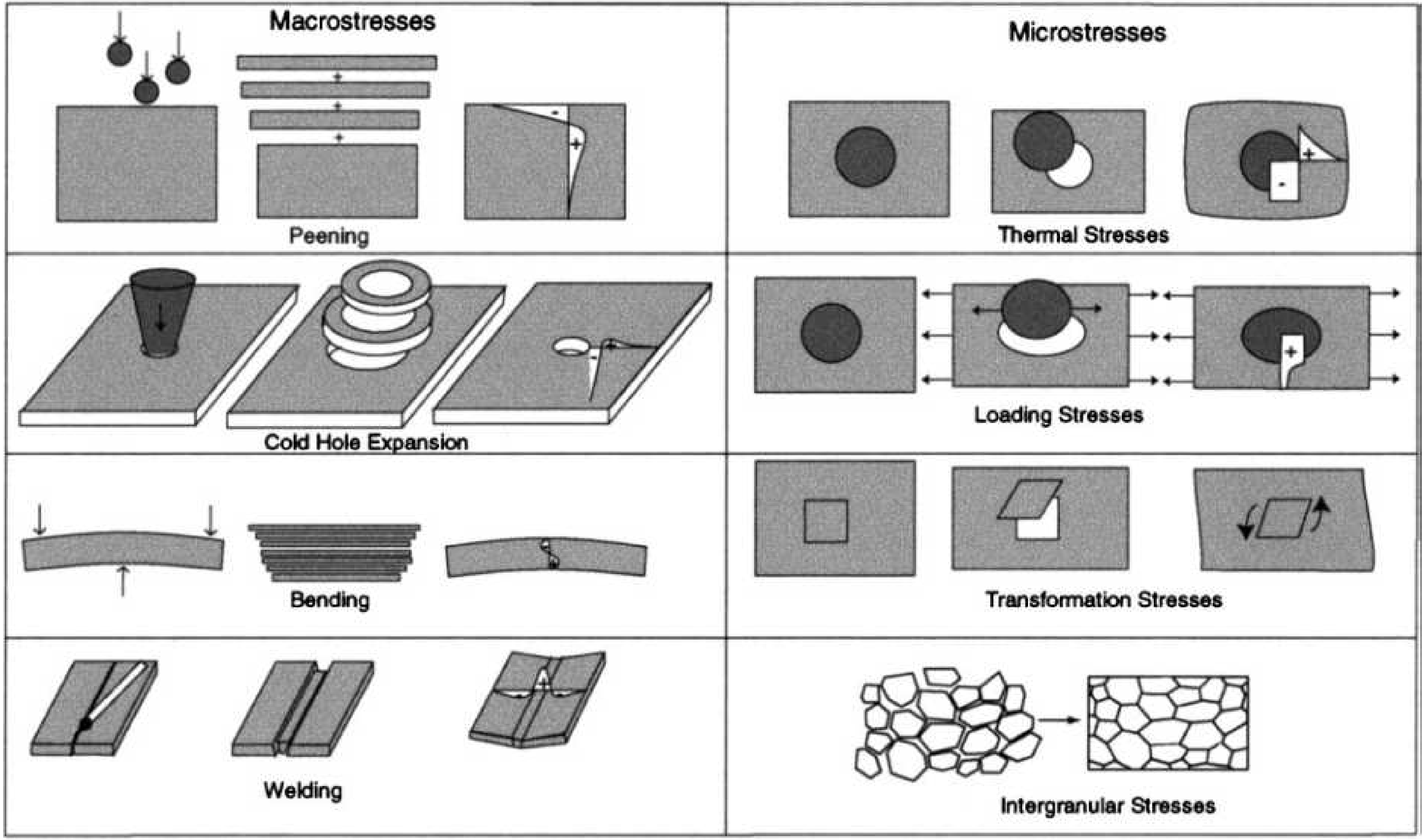
Science, Technology and Engineering underpins all the missions

Outline

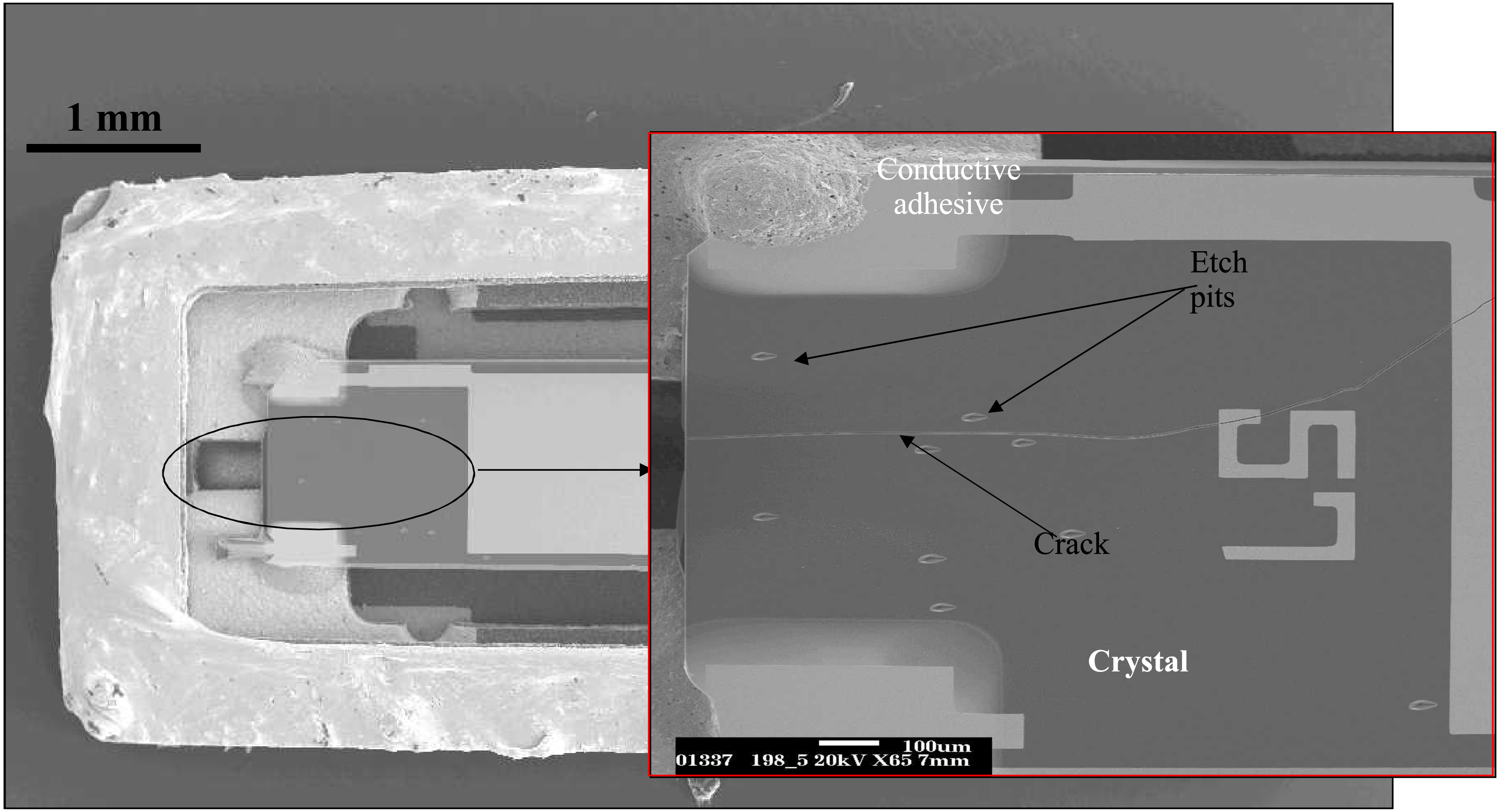
- **Localized (residual) stresses are important**
  - **Examples of material failures from residual stresses**
- **Why measure stresses?**
  - **Use of Vickers indentation for residual stress measurements**
- **Method to use Curved Cracks to Measure Stress**
  - **Fracture mechanics analysis, and use in LTCC-gold via**
- **Development of Cube-corner Indentation Method**
  - **Experiments on stressed glasses (calibration)**
  - **“Quarter- penny” cracks; Fracture Mechanics Analysis**
  - **Volume Resolution of technique, Practical Use & Limitations**
- **Nano-indentation based Method**
  - **Load displacement curves on stressed surface are shifted**
- **Hertzian Indentation**
  - **Local strength measurements**
  - **Interfacial toughness measurement**

Residual stresses arise from misfits...

... between different regions or different phases when they are bonded together (figure from Withers and Bhadeshia, Mat. Sci. and Tech., 2001)

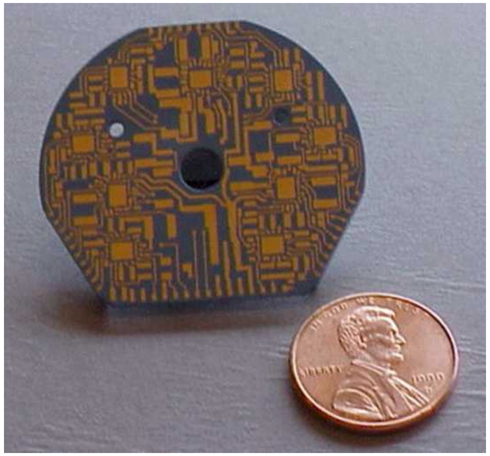


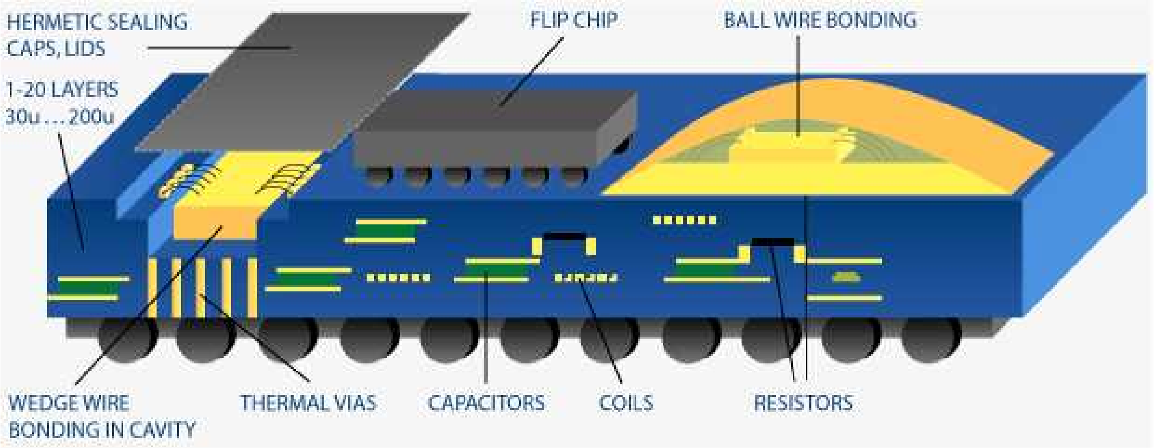
Quartz Resonator Cracking

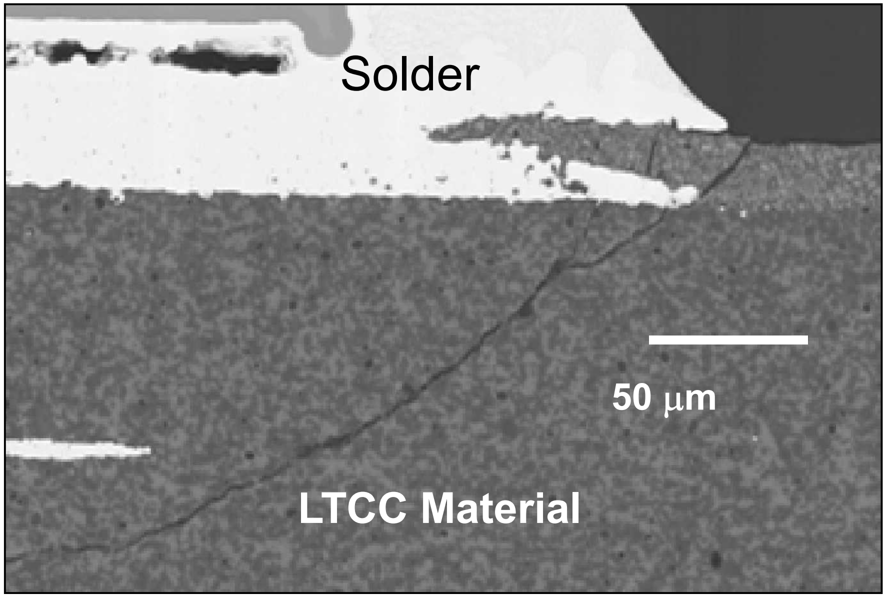


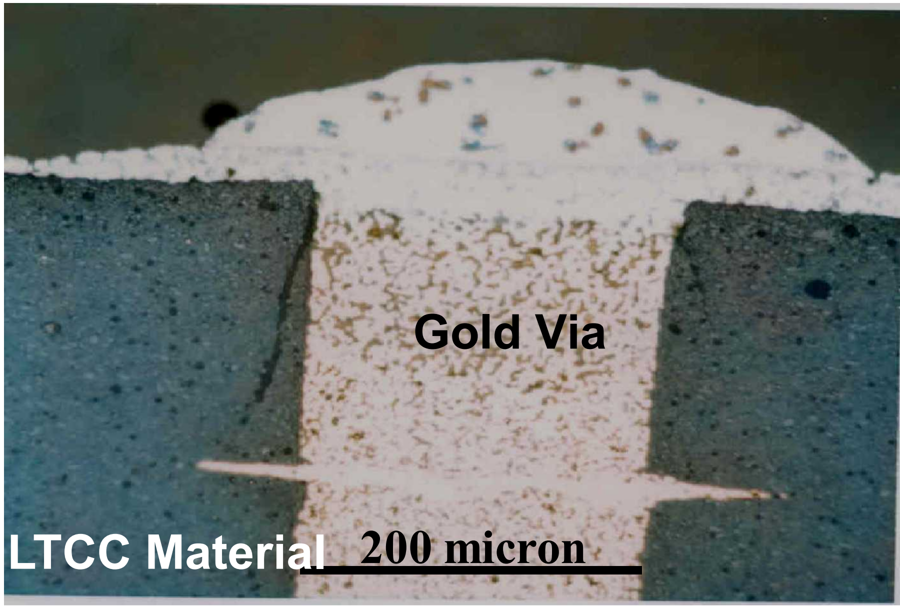
-Cracking fractures the electrical lines
- Polymer adhesive causes stress

Cracks in Various Low-temp. cofired ceramic



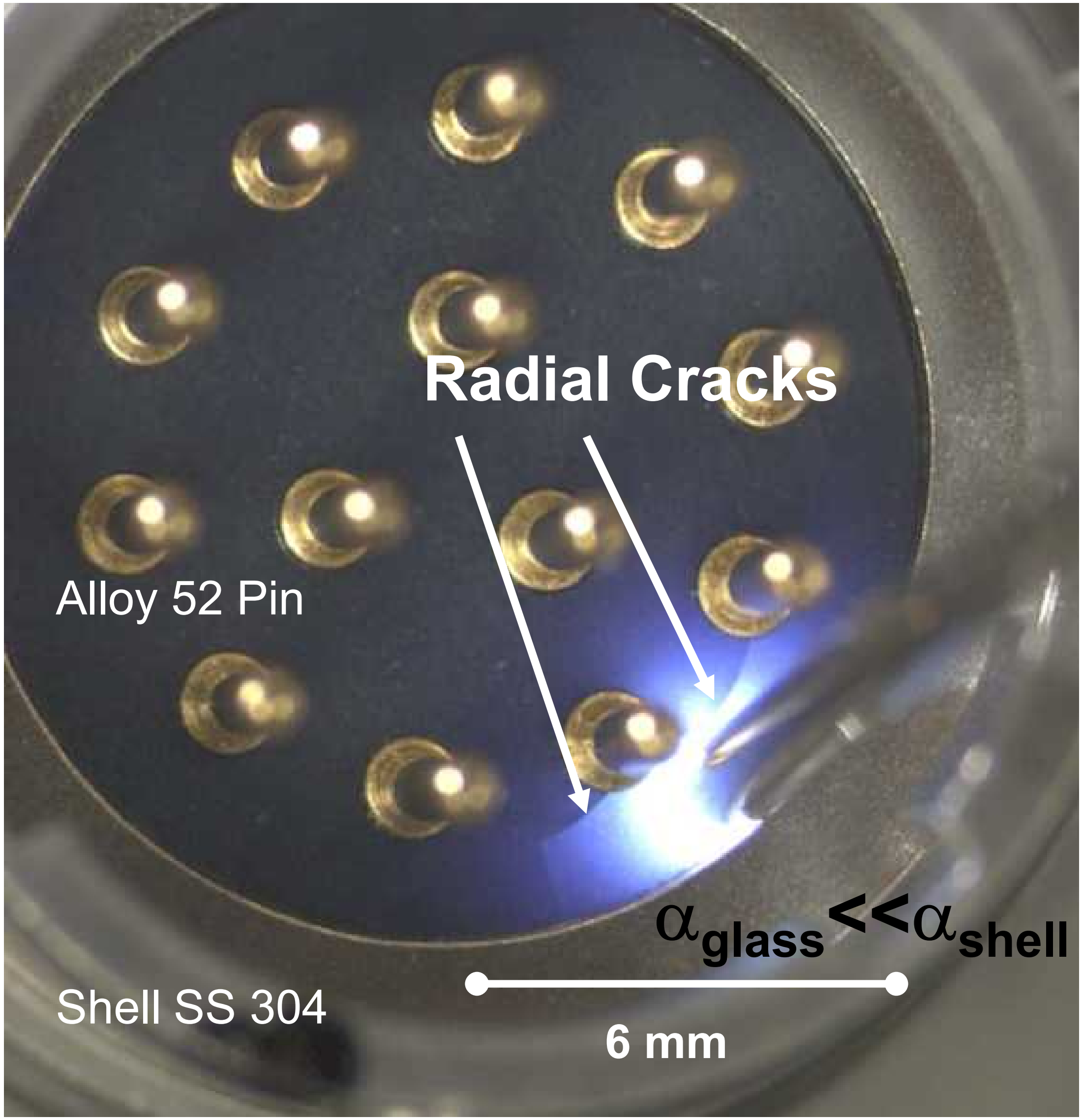






- Metal-ceramic thermal expansion mismatch causes stress

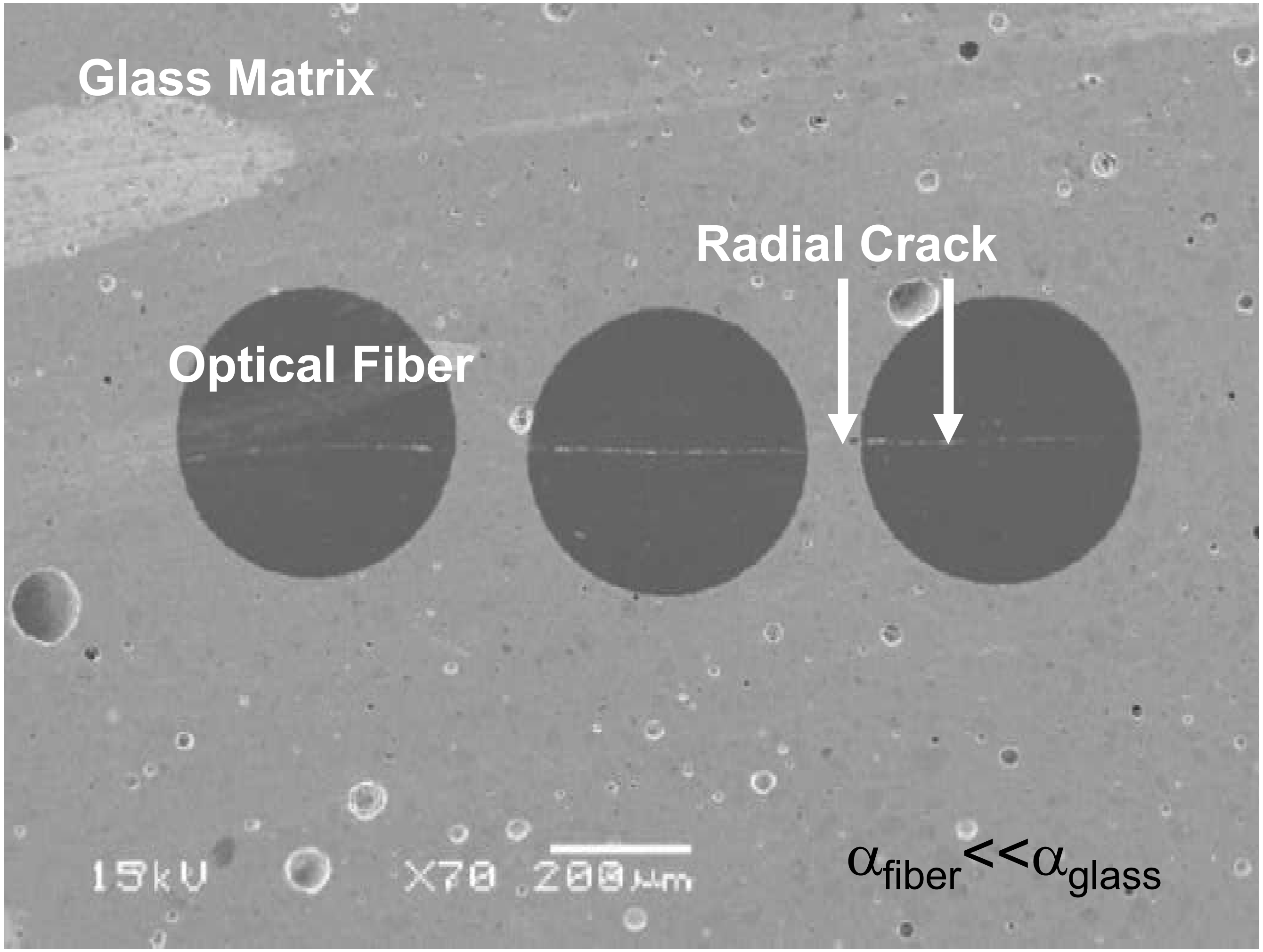
Cracks in Multi-Pin Glass-to-Metal Connector



- Leakage and electrical breakdown issues

Radial Cracks Within and Outside Fiber

- Loss of power-transmission functionality



$\alpha_{fiber} << \alpha_{glass}$

All glass seal

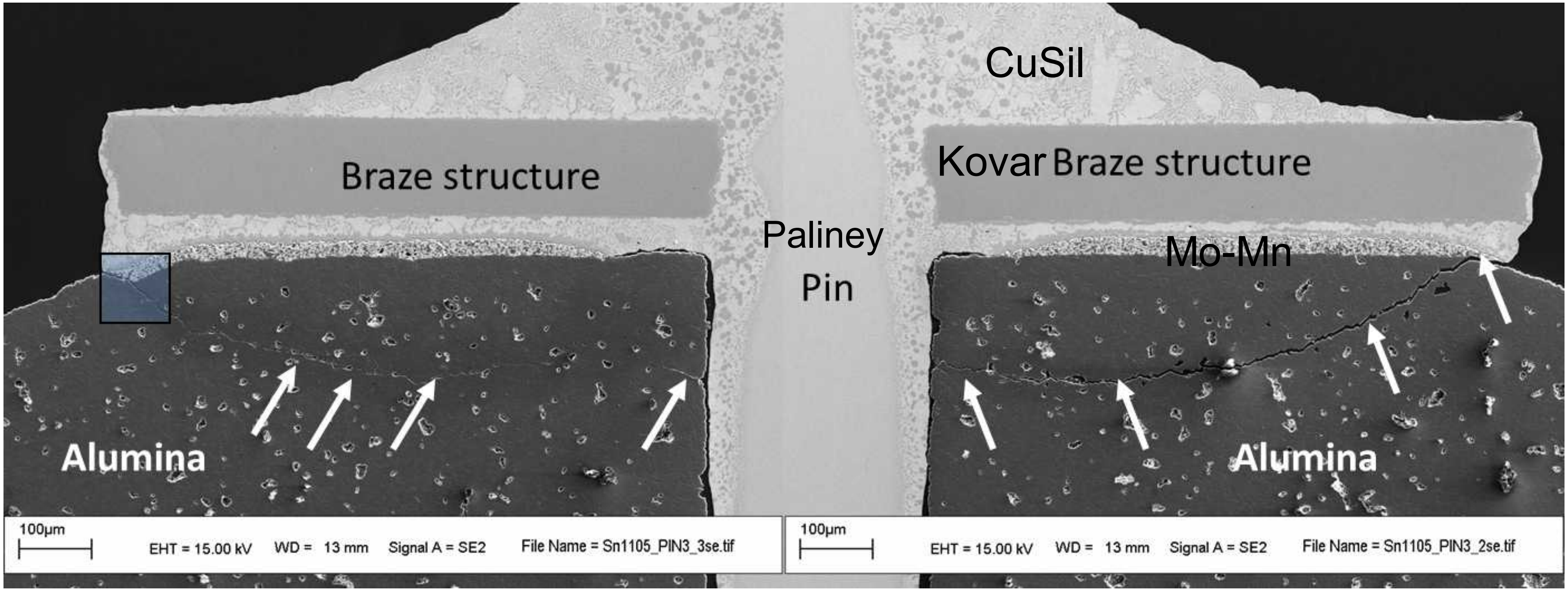
Outline

- Localized stresses are important
    - Examples of material failures from residual stresses
- **Why measure stresses?**
    - **Use of Vickers indentation for residual stress measurements**
- Method to use Curved Cracks to Measure Stress
    - Fracture mechanics analysis, and use in LTCC-gold via
- Development of Cube-corner Indentation Method
    - Experiments on stressed glasses (calibration)
    - “Quarter- penny” cracks; Fracture Mechanics Analysis
    - Volume Resolution of technique, Practical Use & Limitations
- Nano-indentation based Method
    - Load displacement curves on stressed surface are shifted
- Hertzian Indentation
    - Local strength measurements
    - Interfacial toughness measurement

Sandia
National
Laboratories

Why do we have to measure stresses?

Brazed alumina



Many of the temp. dependent properties of materials involved are not known. Constitutive inputs for FEA have questionable validity

Chemical reactions between materials change these values

Crack Geometries around Vickers

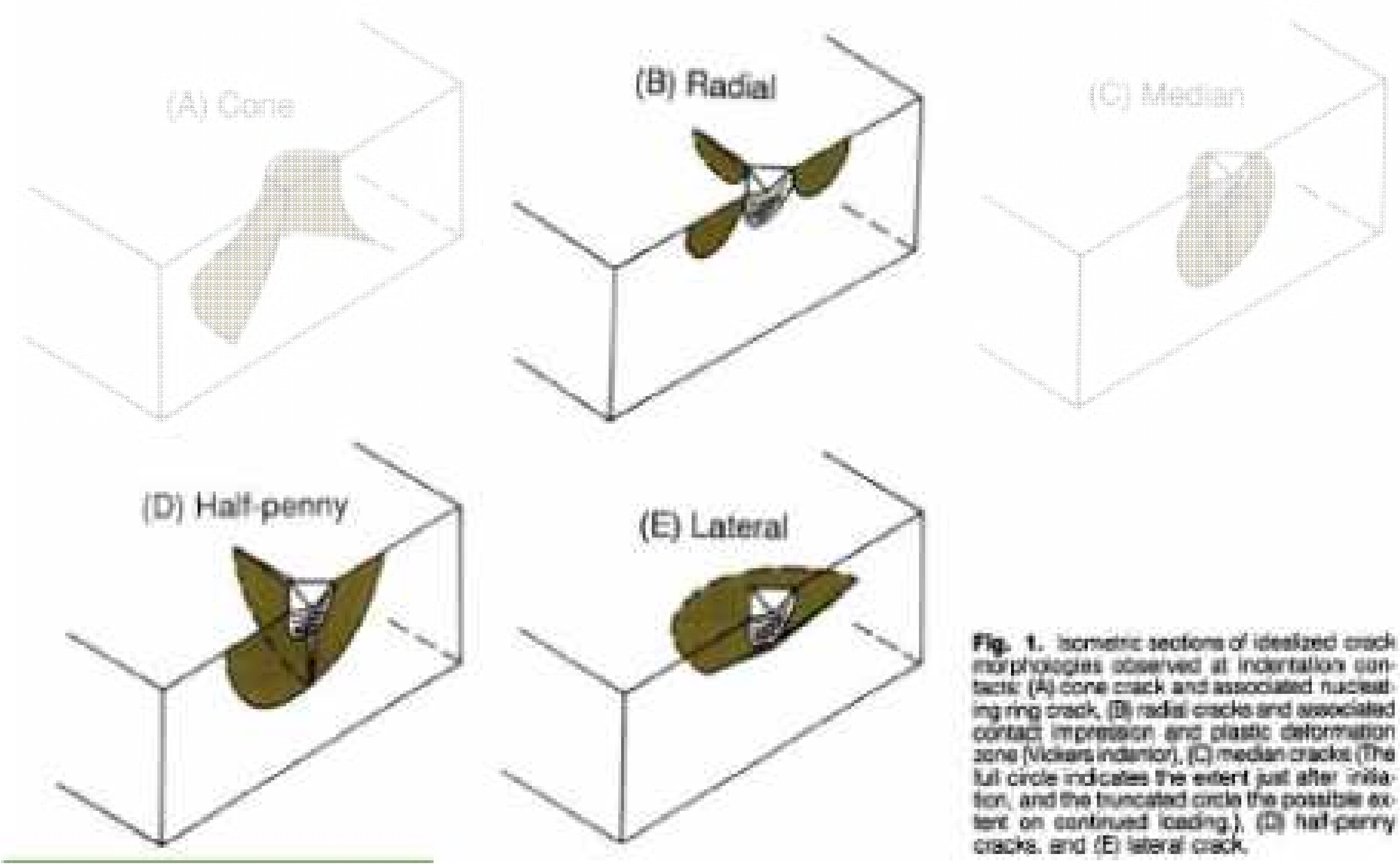
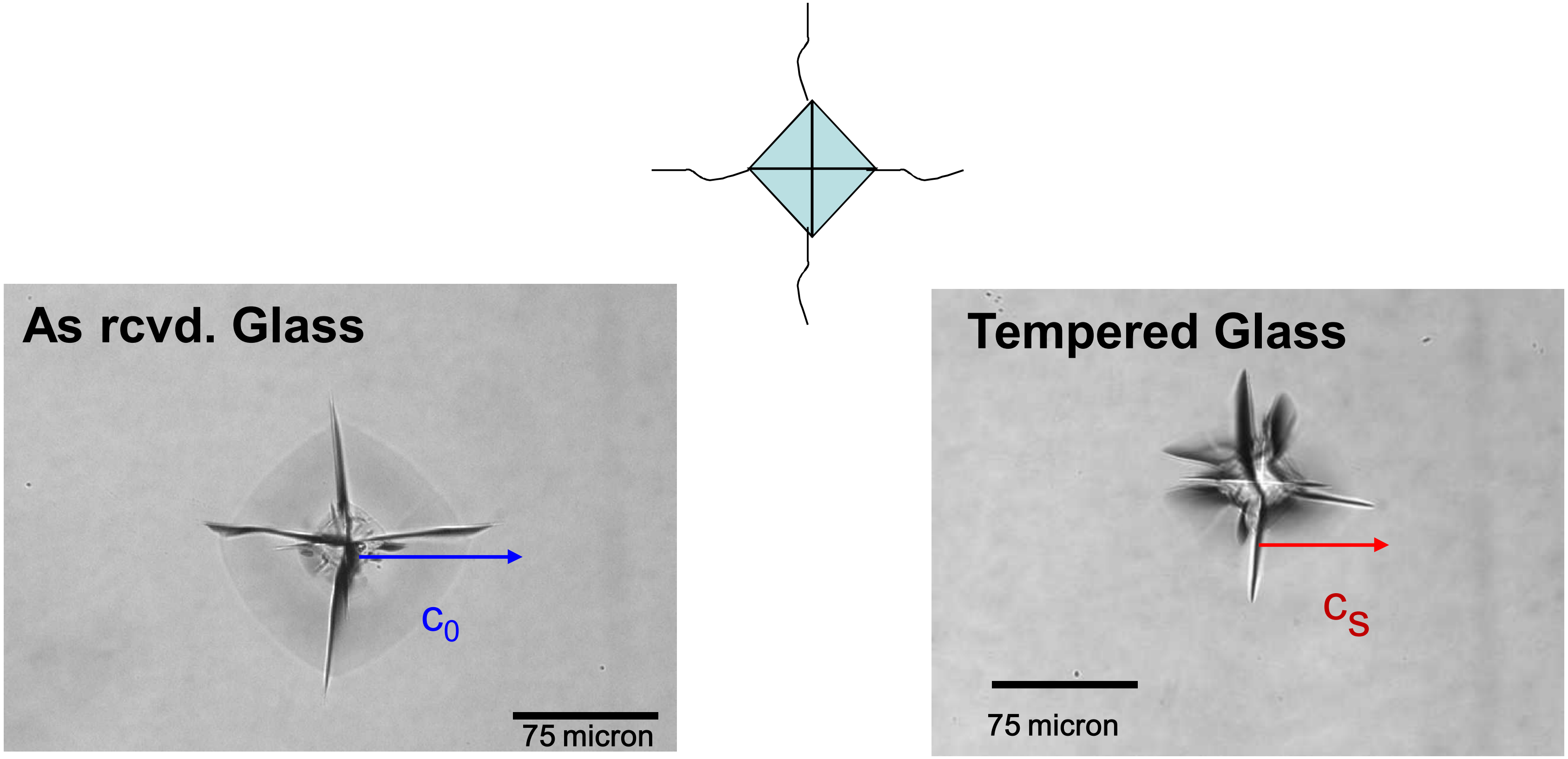


Fig. 1. Isometric sections of idealized crack morphologies observed at indentation contacts: (A) cone crack and associated nucleating ring crack, (B) radial cracks and associated contact impression and plastic deformation zone (Vickers indentor), (C) median cracks (The full circle indicates the extent just after initiation, and the truncated circle the possible extent on continued loading.), (D) half-penny cracks, and (E) lateral crack.

Cook and Pharr, J. ACerS., 1990

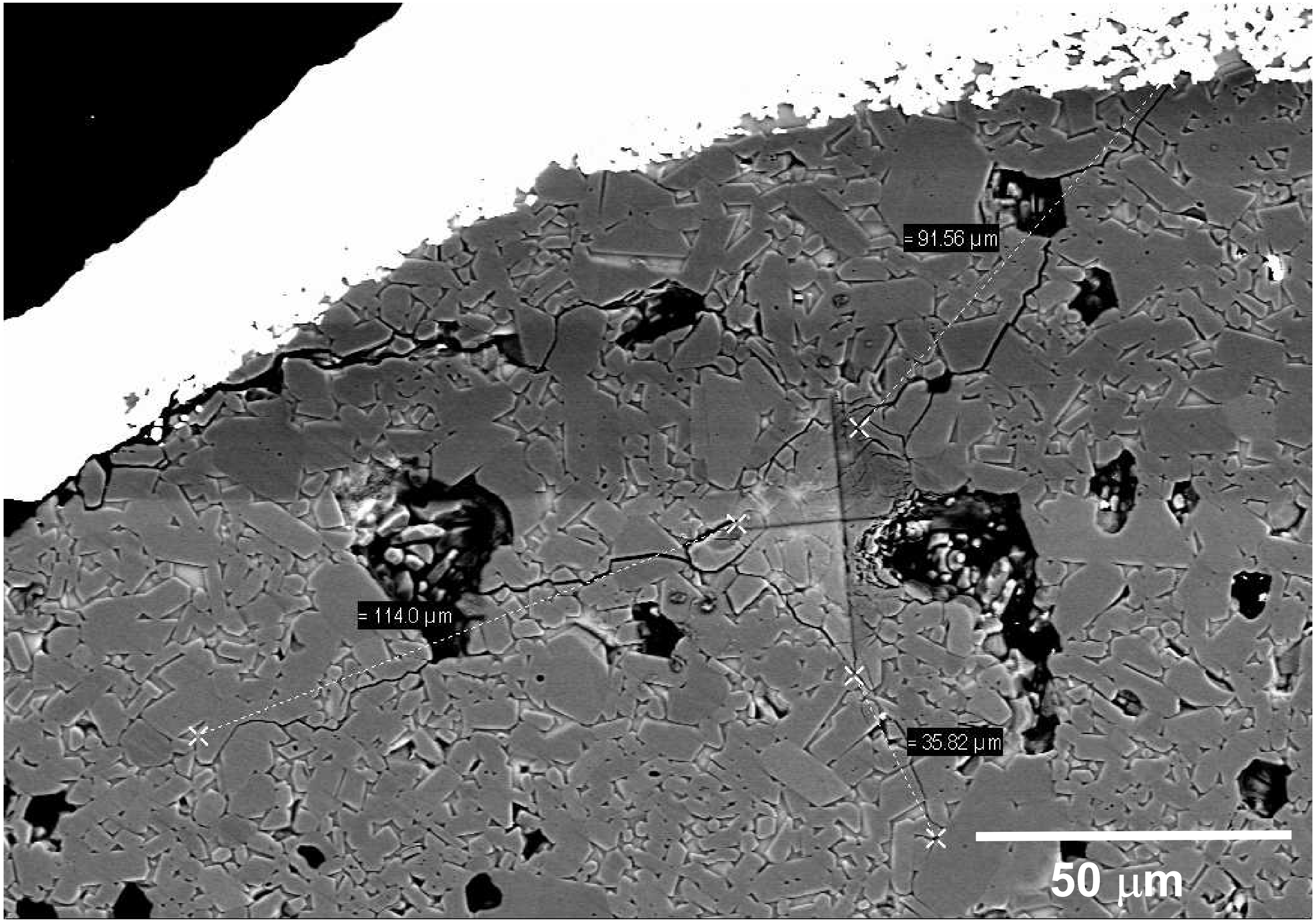
Hardness Impression Crack Measurement



For a compressively stressed surface, $c_s < c_0$

Δc can be used to calculate residual stress with some assumptions

Compare to ~50 μm in unstressed alumina

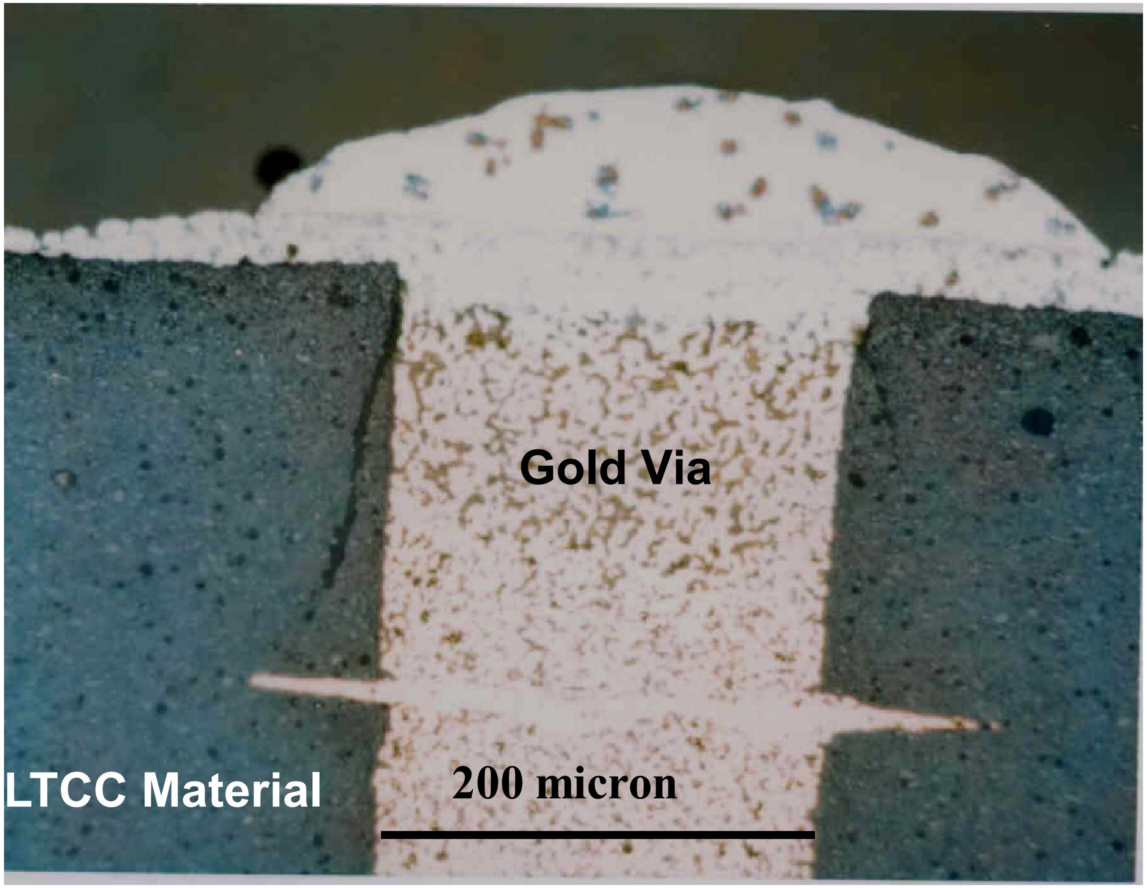


Drawbacks: Large size of indents; large load for crack initiation
Cannot get close enough to features of interest

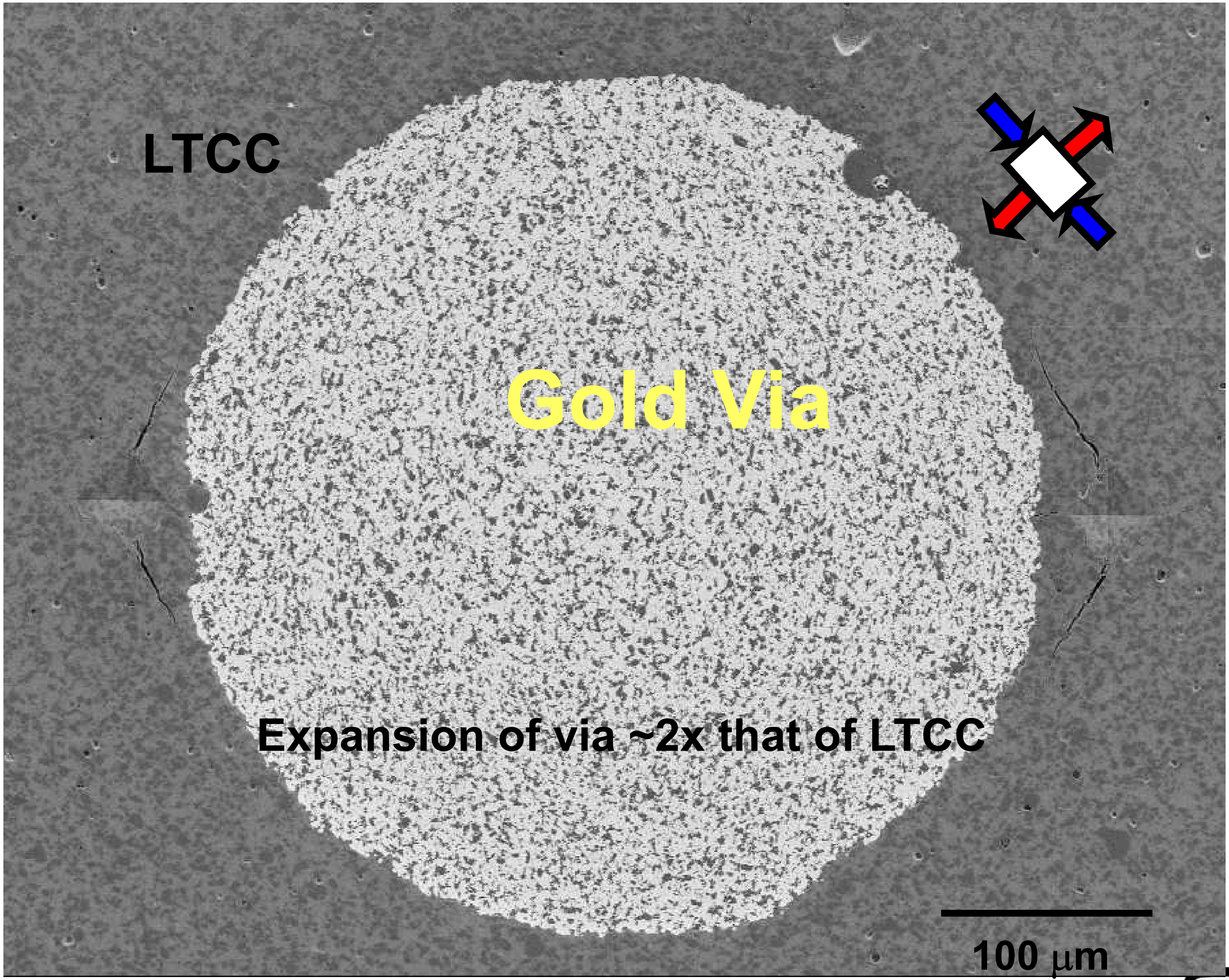
Outline

- Localized stresses are important
  - Examples of material failures from residual stresses
- Why measure stresses?
  - Use of Vickers indentation for residual stress measurements
- **Method to use Curved Cracks to Measure Stress**
  - **Fracture mechanics analysis, and use in LTCC-gold via**
- Development of Cube-corner Indentation Method
  - Experiments on stressed glasses (calibration)
  - "Quarter- penny" cracks; Fracture Mechanics Analysis
  - Volume Resolution of technique, Practical Use & Limitations
- Nano-indentation based Method
  - Load displacement curves on stressed surface are shifted
- Hertzian Indentation
  - Local strength measurements
  - Interfacial toughness measurement

Cracks in LTCC Materials near Metal Vias



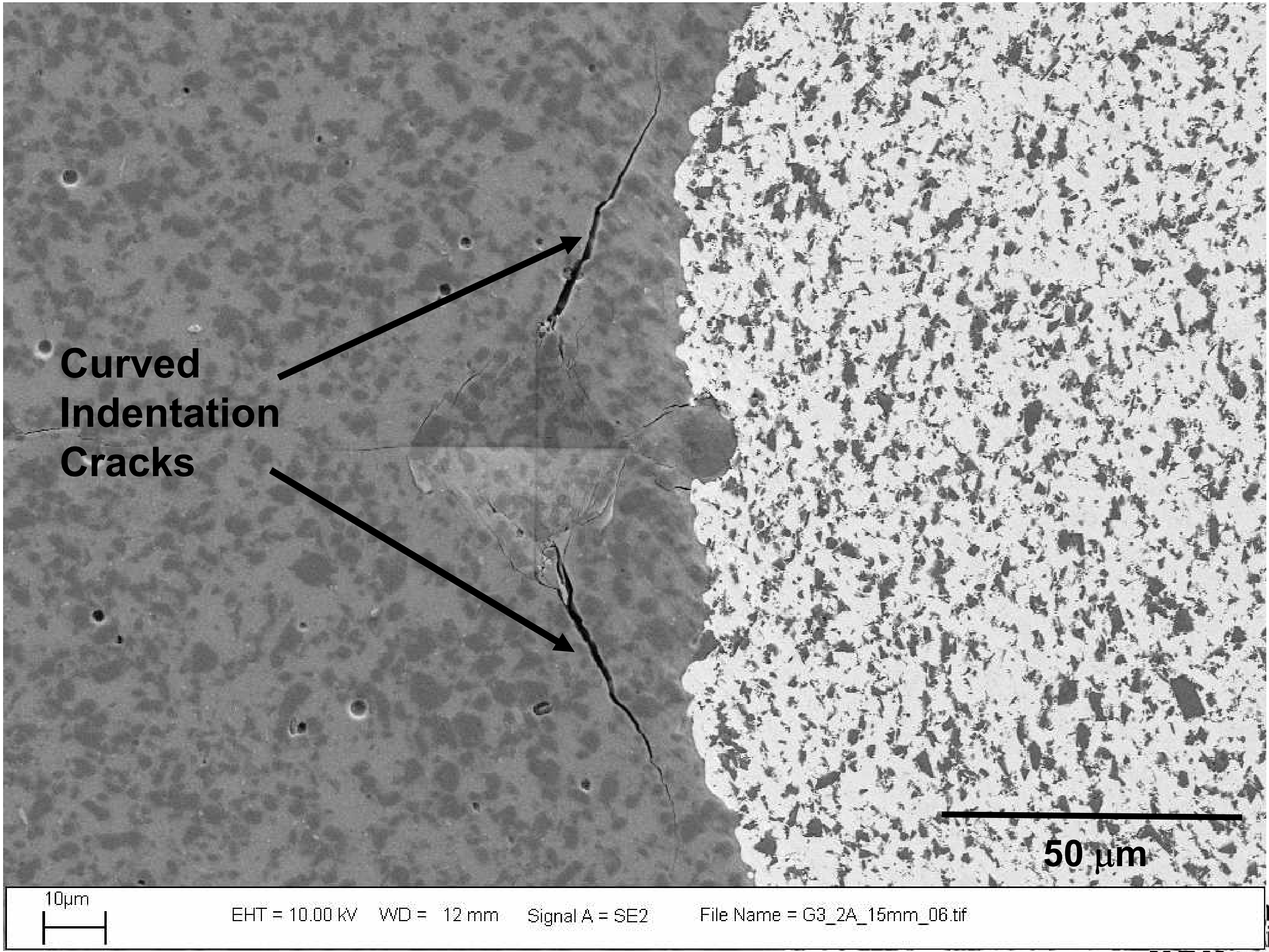
Curved Cracks Near Vias



Expansion of via ~2x that of LTCC

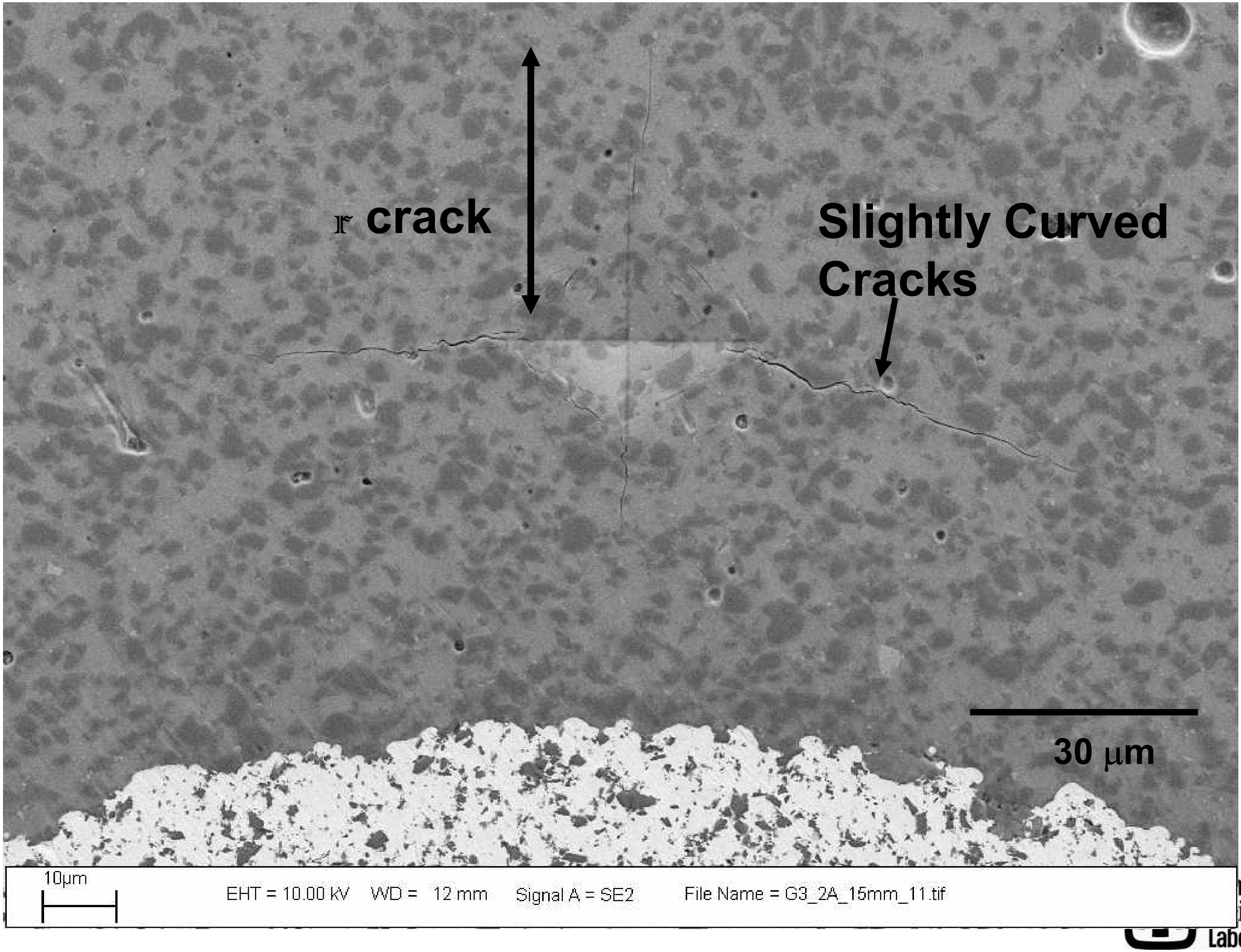
Stress states generated during cooling from processing

Curved Cracks Near Vias



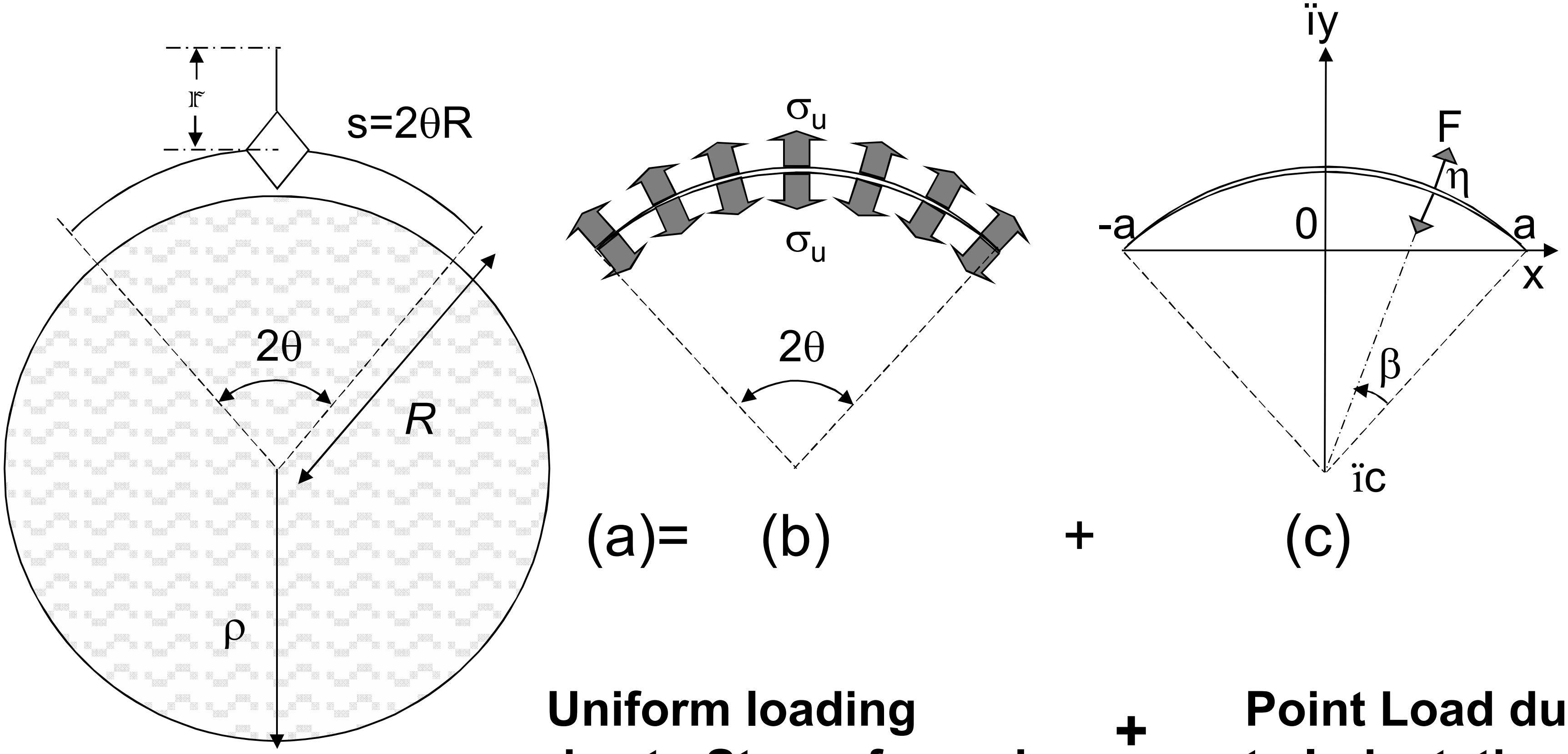
Curved Indentation Cracks

Curvature Decreases further from Via



Fracture Mechanics Idealization

Loading acting on arc crack



Uniform loading due to Stress from via

+

Point Load due to Indentation Impression

Stress Intensity Factors

$$K_{I-u} = \frac{\sigma_u \sqrt{\pi R \sin\theta}}{1+\sin^2\frac{\theta}{2}} \cos\frac{\theta}{2}$$

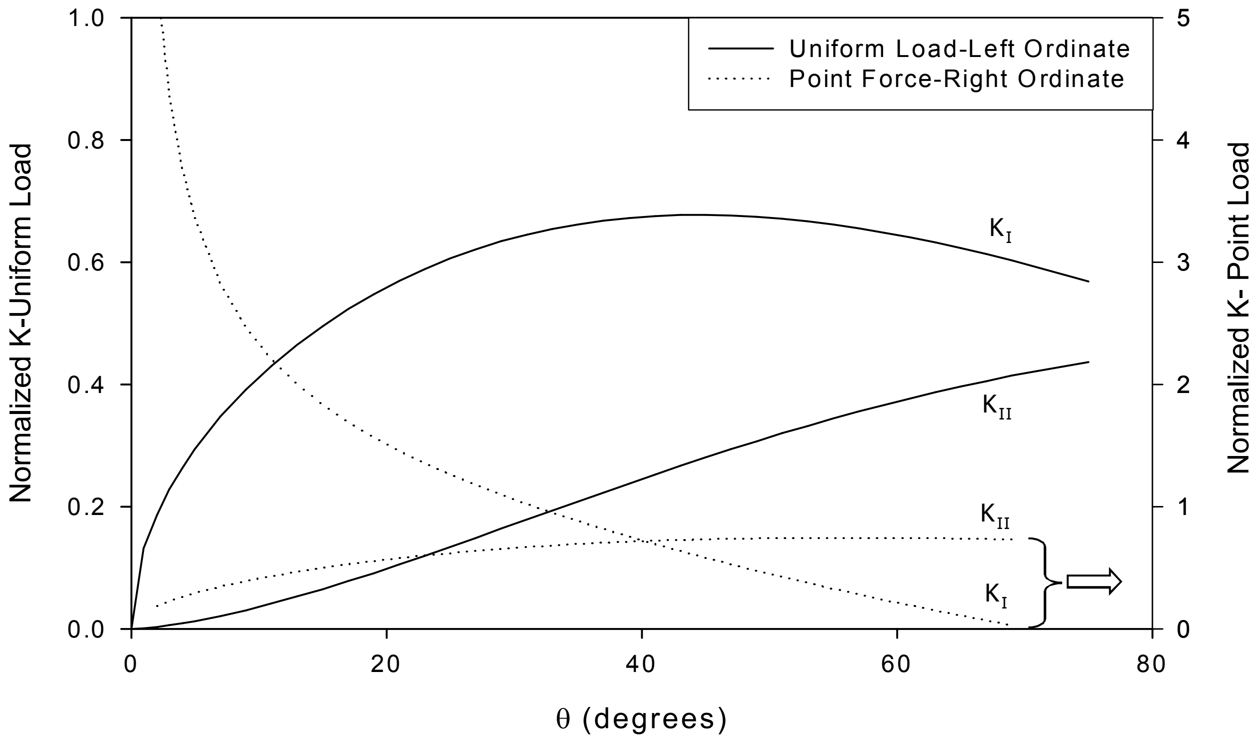
$$K_{II-u} = \frac{\sigma_u \sqrt{\pi R \sin\theta}}{1+\sin^2\frac{\theta}{2}} \sin\frac{\theta}{2}$$

Uniform Loading

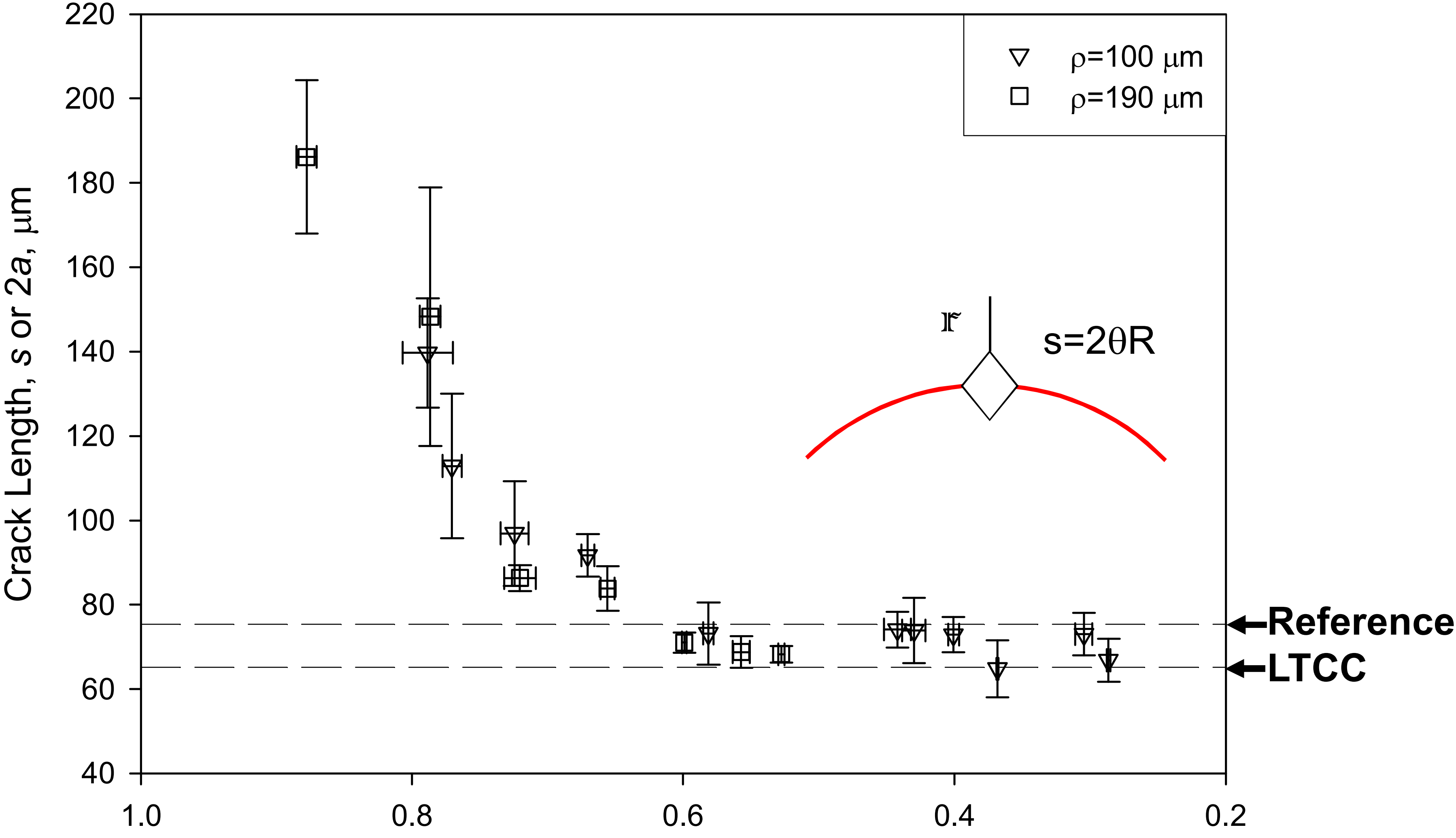
$$K_{I-p}\Big|_{\pm a} = \frac{F}{\sqrt{\pi R \sin\theta}} \left[\frac{\cos\theta}{2} - \frac{1}{2} + \frac{2\cos^4\frac{\theta}{2}}{\left(3-\cos\theta\right)} \right]$$

$$K_{II-p}\Big|_{\pm a} = \frac{F}{\sqrt{\pi R \sin\theta}} \left[\frac{\sin\theta}{2} + \frac{2\cos^3\frac{\theta}{2}\sin\frac{\theta}{2}}{\left(3-\cos\theta\right)} \right]$$

Point Loading

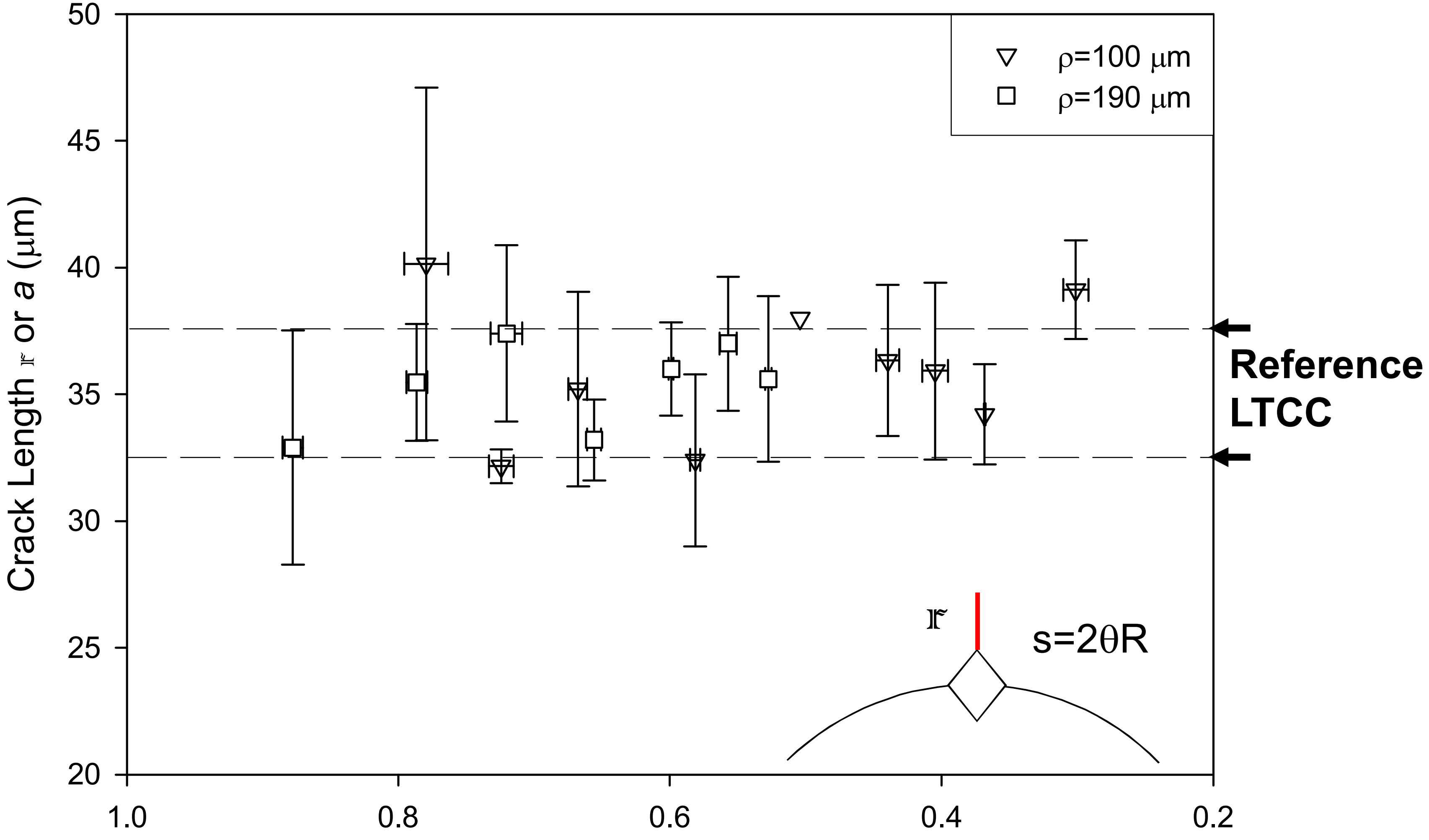


Crack Length Measurements



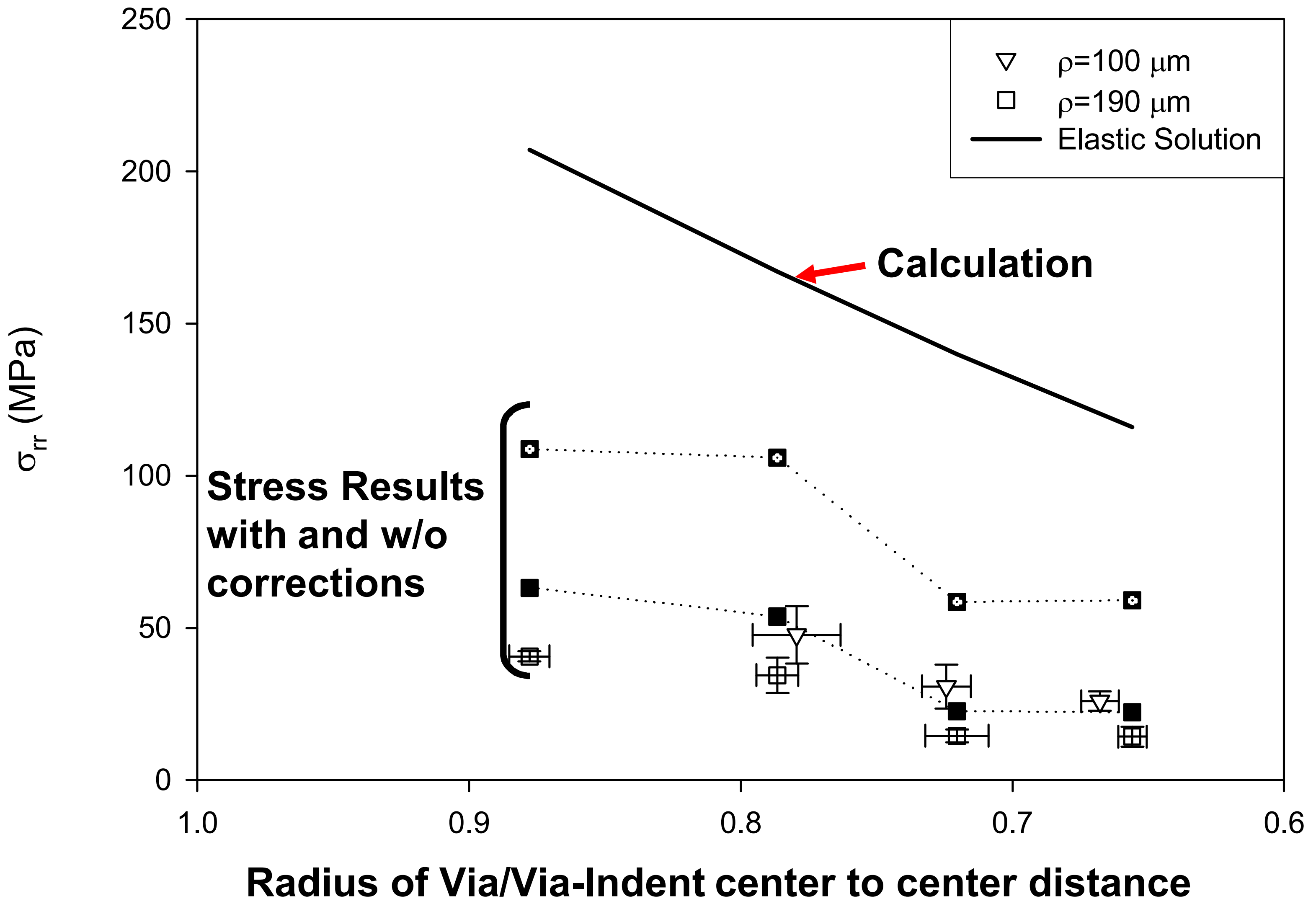
Map of tensile stress near via

Crack Length Measurements



No compressive hoop stress !

Stress Calculations

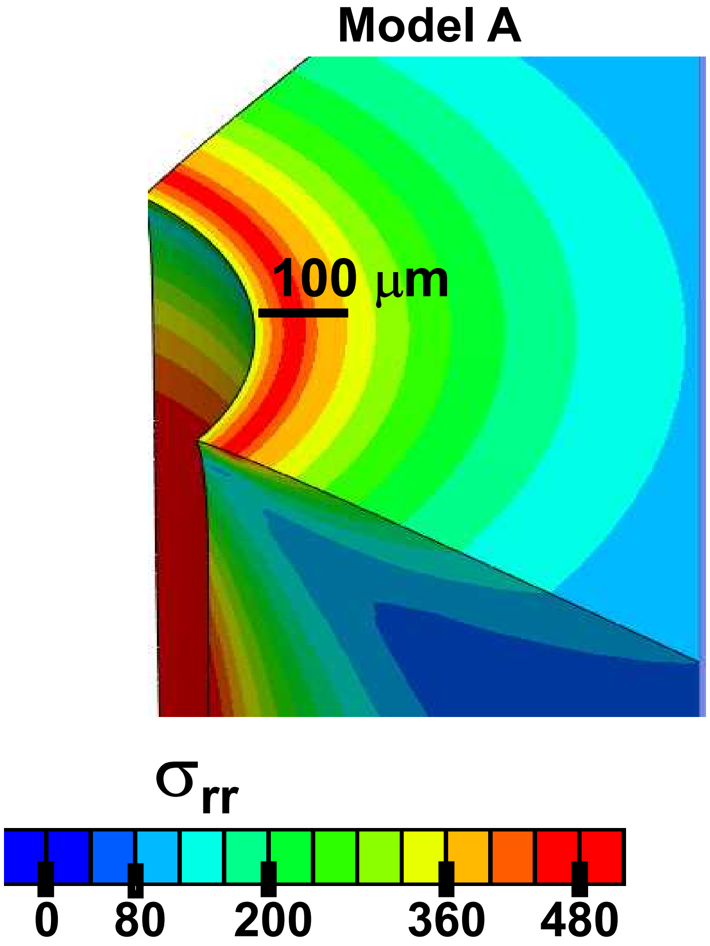


Stresses significantly lower than calculated

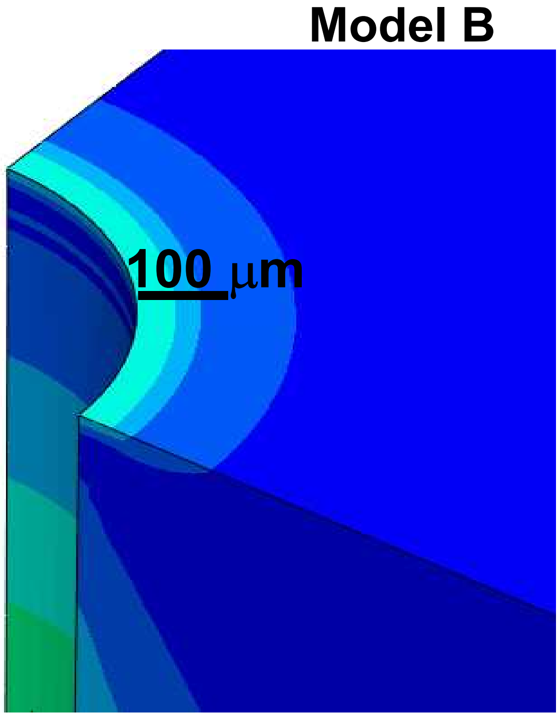
Conclusions

- Cause of discrepancy: Plastic deformation in gold via

Elastic Via



Elastic-Plastic Via



Peak Stress:

Model A ~550 MPa

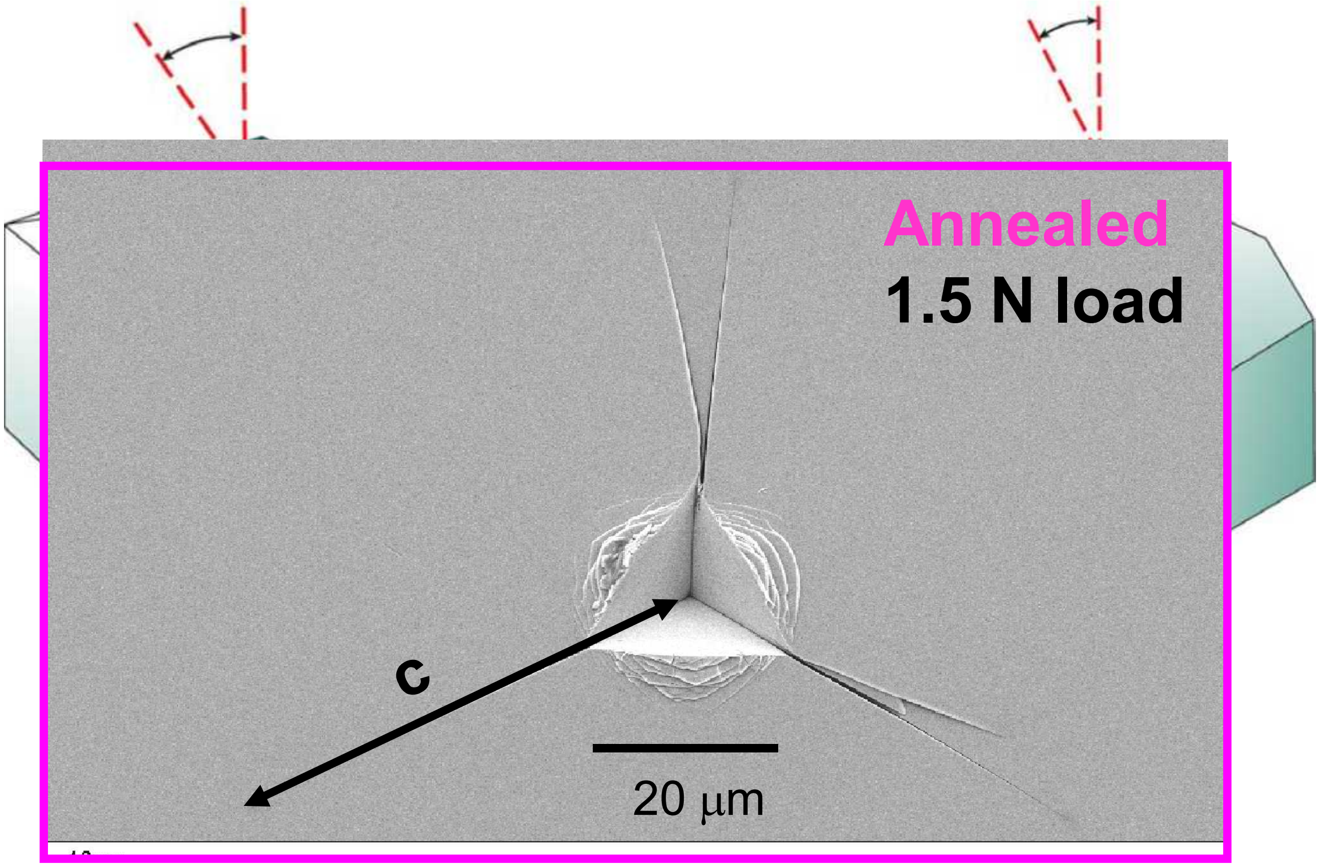
Model B ~160-120 MPa

Outline

- Localized stresses are important
  - Examples of material failures from residual stresses
- Why measure stresses?
  - Use of Vickers indentation for residual stress measurements
- Method to use Curved Cracks to Measure Stress
  - Fracture mechanics analysis, and use in LTCC-gold via
- **Development of Cube-corner Indentation Method**
  - **Experiments on stressed glasses (calibration)**
  - **"Quarter- penny" cracks; Fracture Mechanics Analysis**
  - **Volume Resolution of technique, Practical Use & Limitations**
- Nano-indentation based Method
  - Load displacement curves on stressed surface are shifted
- Hertzian Indentation
  - Local strength measurements
  - Interfacial toughness measurement

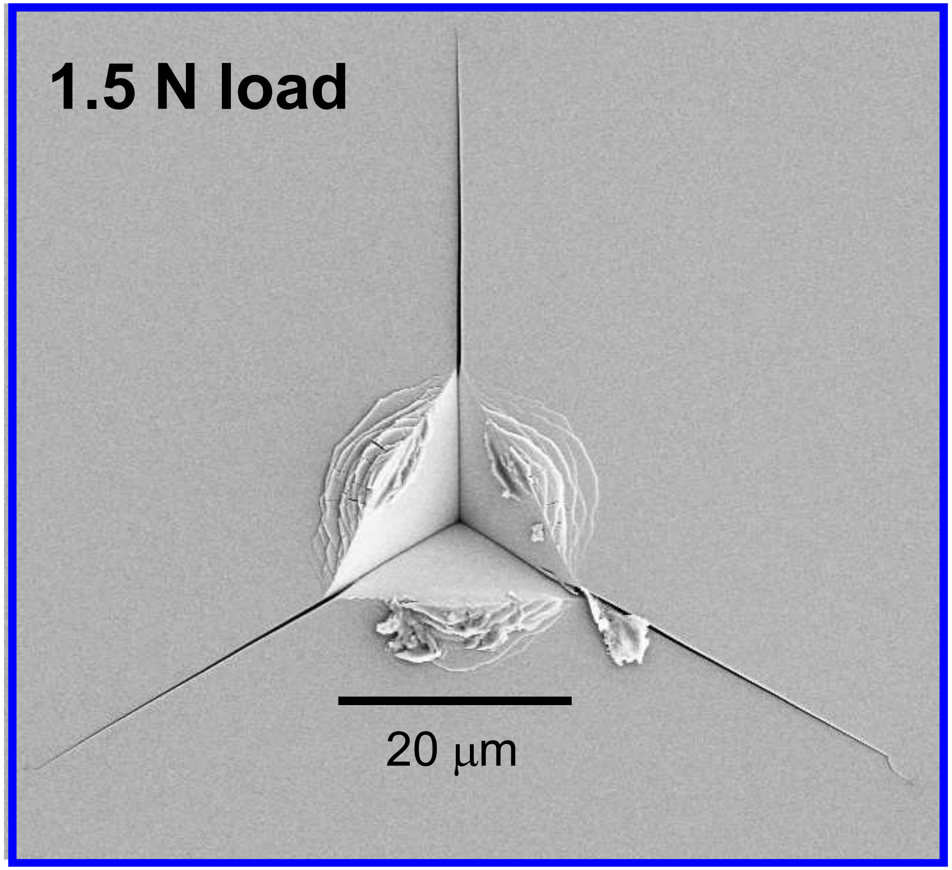
Cube Corner Indentation

Cracks develop at low loads & small impression size
Allow stress measurement over small volumes

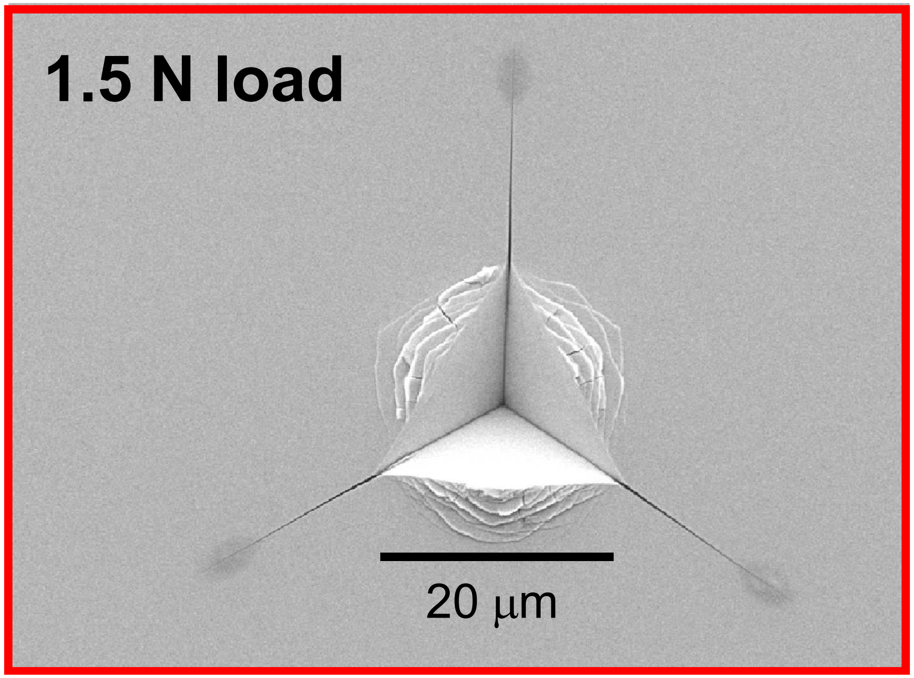


Indentation on Two Stressed Glass Surfaces

Heat Strengthened (partial temper)

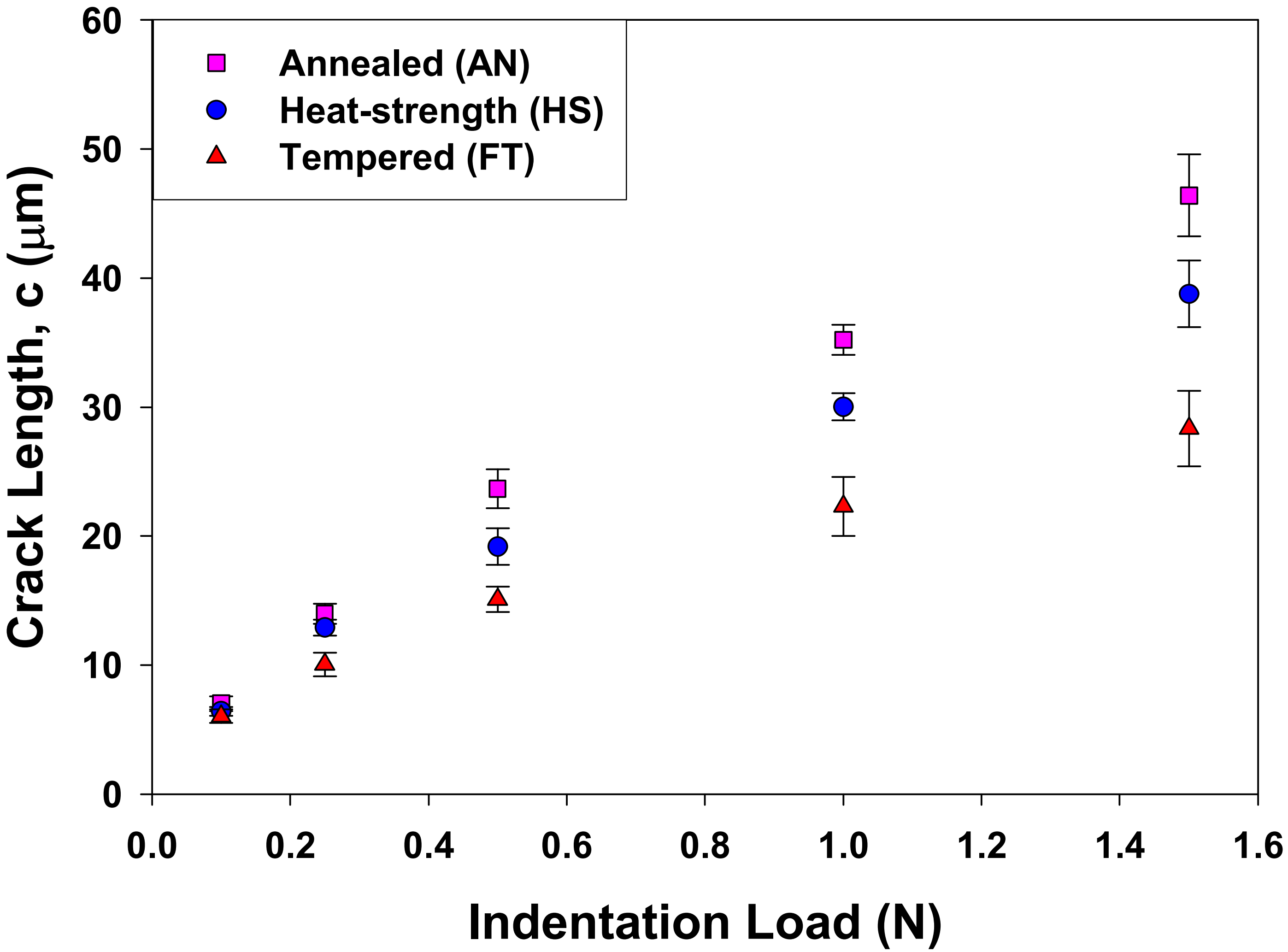


Fully Tempered



Crack sizes are significantly smaller

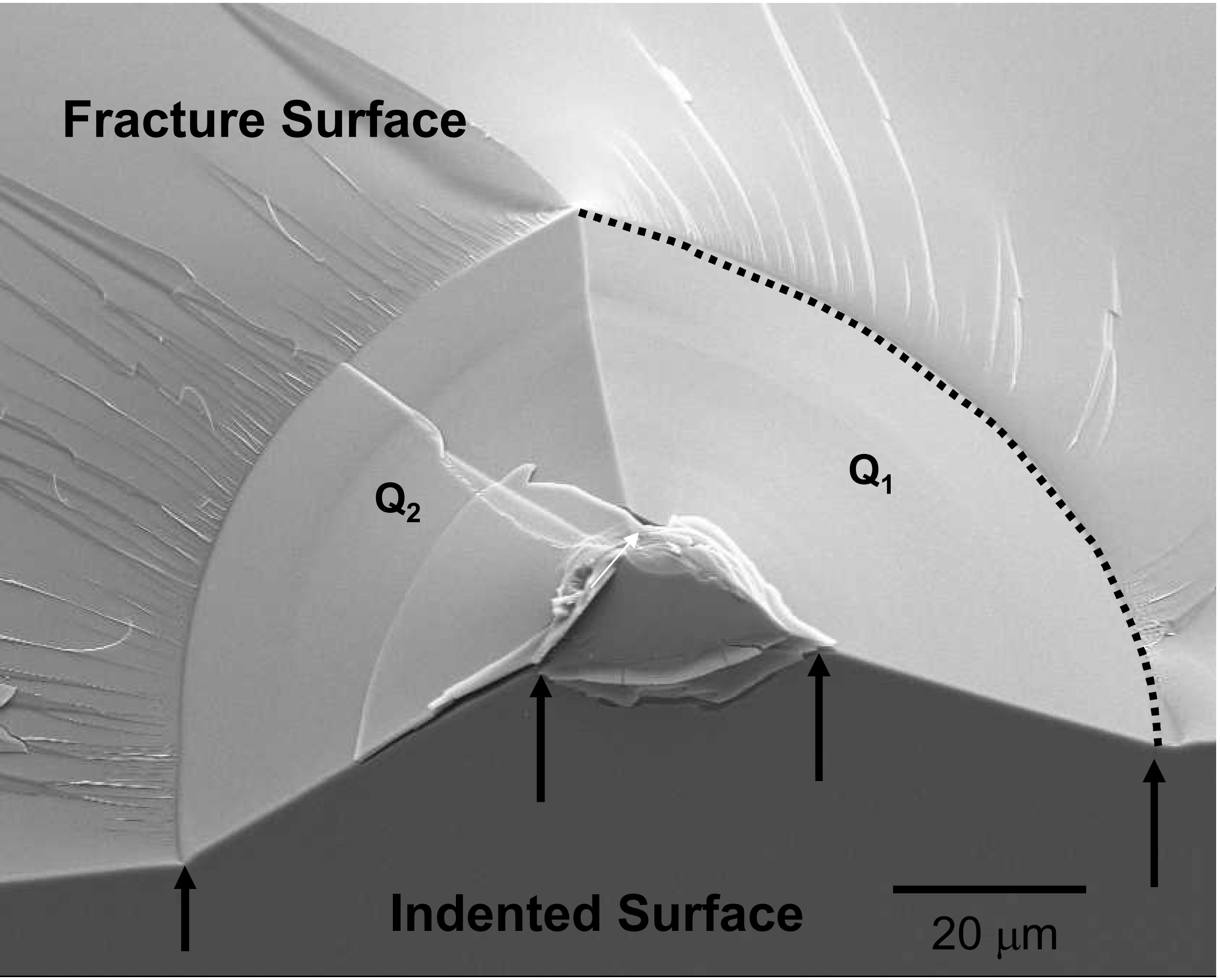
Crack Lengths Lower on Stressed Glasses

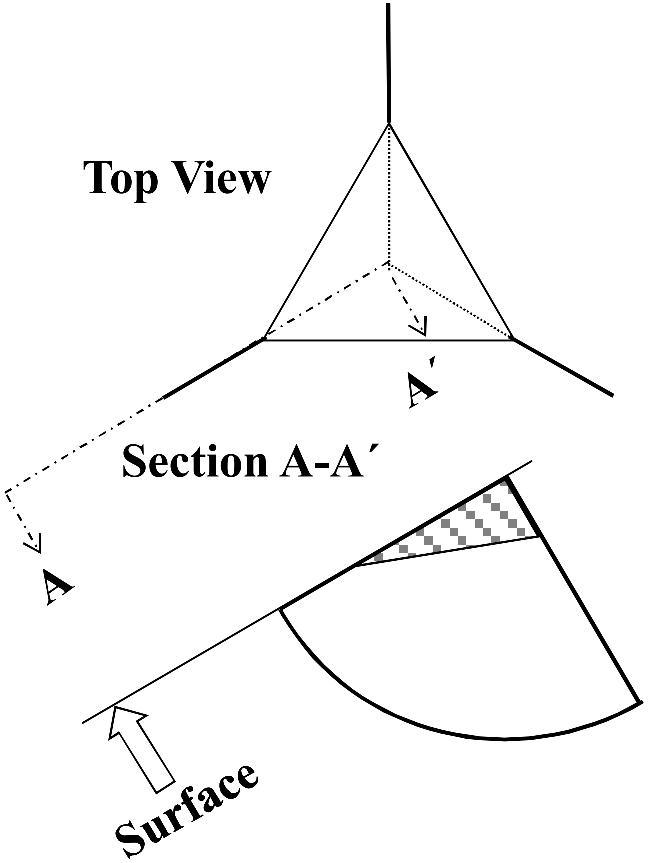


Crack length difference increases with increasing load/with increasing crack length

Quarter Penny Crack: High Load

Annealed: 2 N Load





Stress Intensity Superposition

$$K_{Net} = \chi \frac{P}{c^{3/2}} - 2f(\alpha)|\sigma_S|\left(\frac{c}{\pi}\right)^{1/2}$$

Elastic-plastic Mismatch Term:

–χ is ~ 2 times that for Vickers
-Half-penny crack assumption (?)

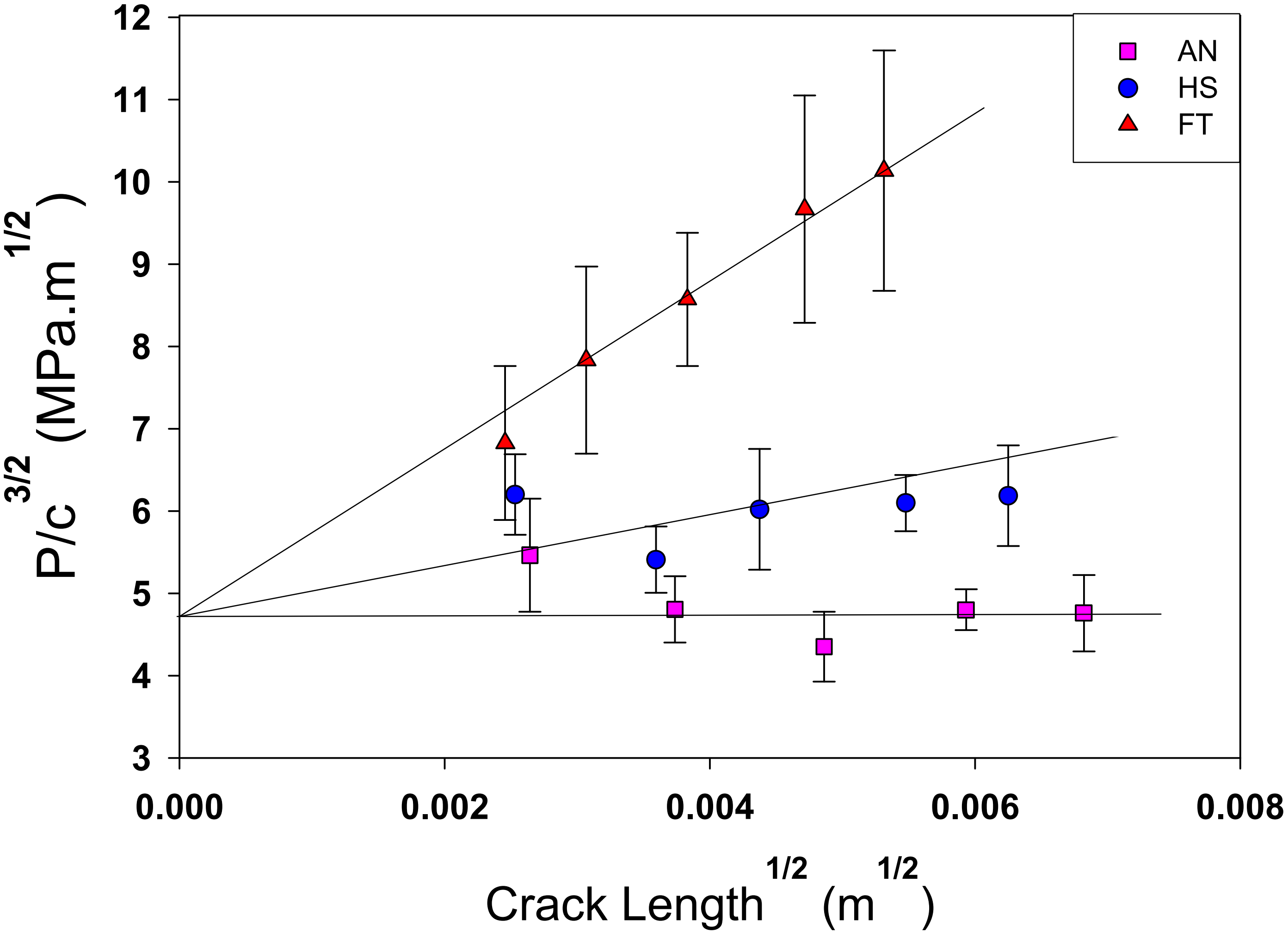
Residual Stress, σ_s, term:

-f(α) for quarter-penny using a weight function approach~1.13
- Kese and Rowcliffe, f(α)~0.53

$$\frac{P}{c^{3/2}} = \frac{P}{c_0^{3/2}} + \frac{2f(\alpha)|\sigma_S|\left(\frac{c}{\pi}\right)^{1/2}}{\chi}$$

Residual stress obtained from slope

Stress Results Obtained



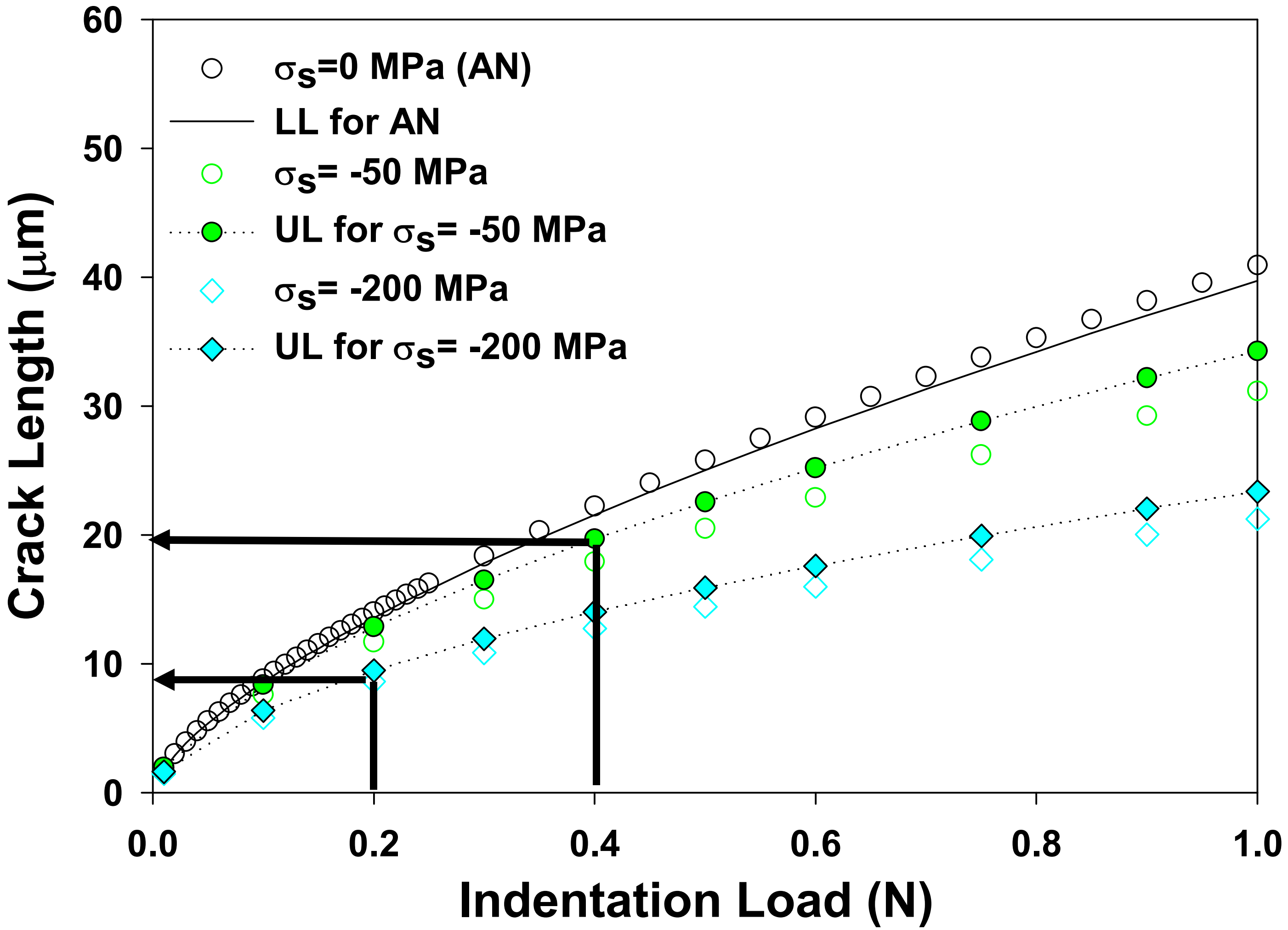
Heat Strengthened: -30 MPa

Tempered: -110 MPa

Higher stresses can be resolved over a smaller volume

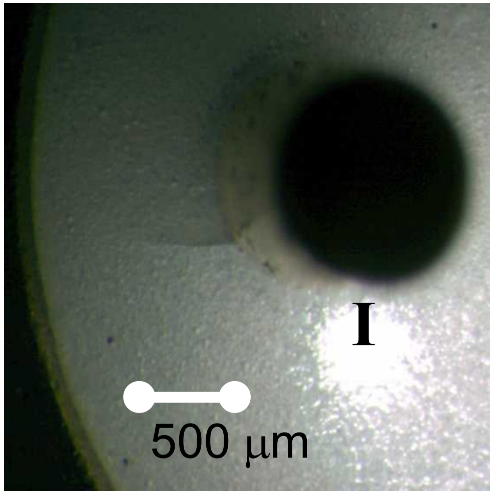
-30 MPa stress resolvable over 25 μm^3

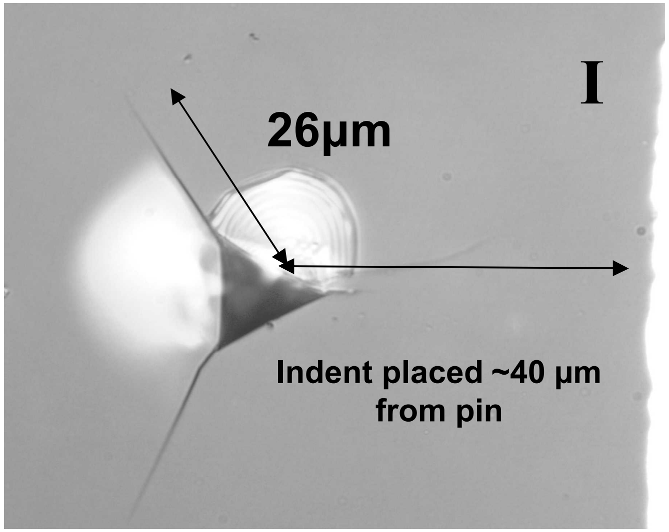
-110 MPa stress resolvable over 20 μm^3

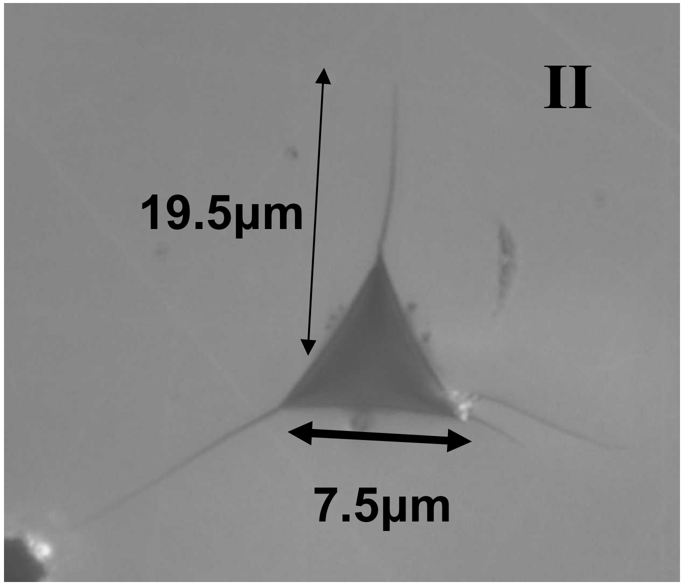


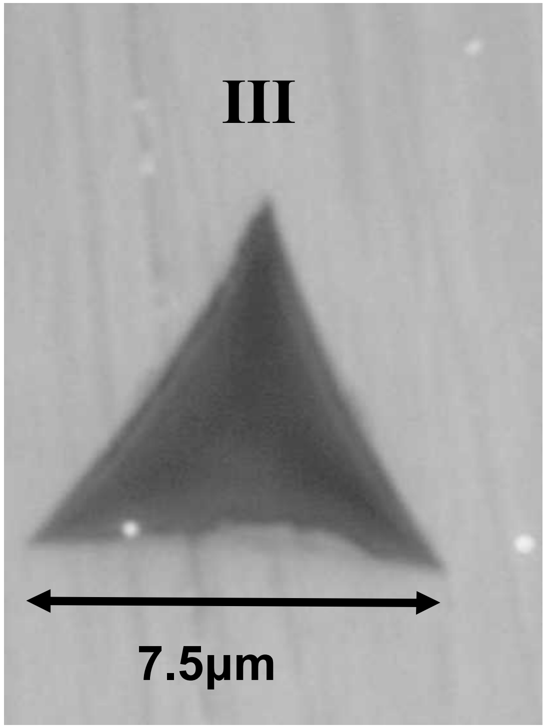
Sandia National Laboratories

Successful Use of Cube Corner in Glass-to Metal Seal Design and Production



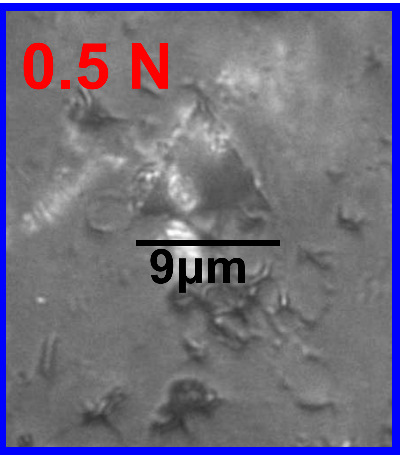


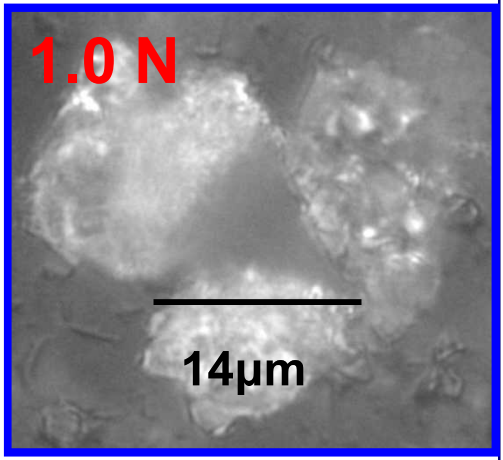


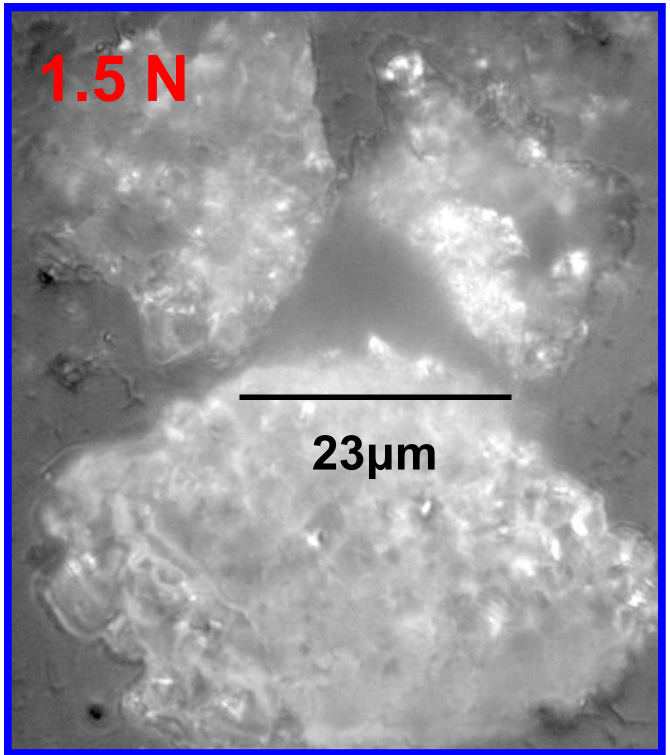


Limitations of use of Cube Corner

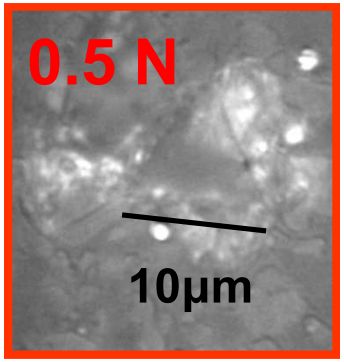
94% alumina

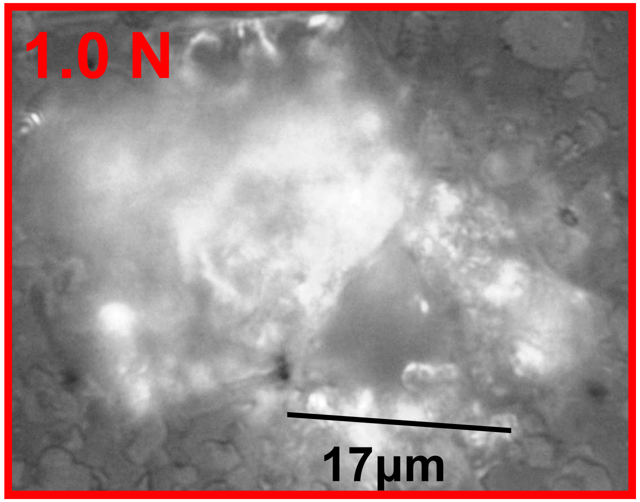


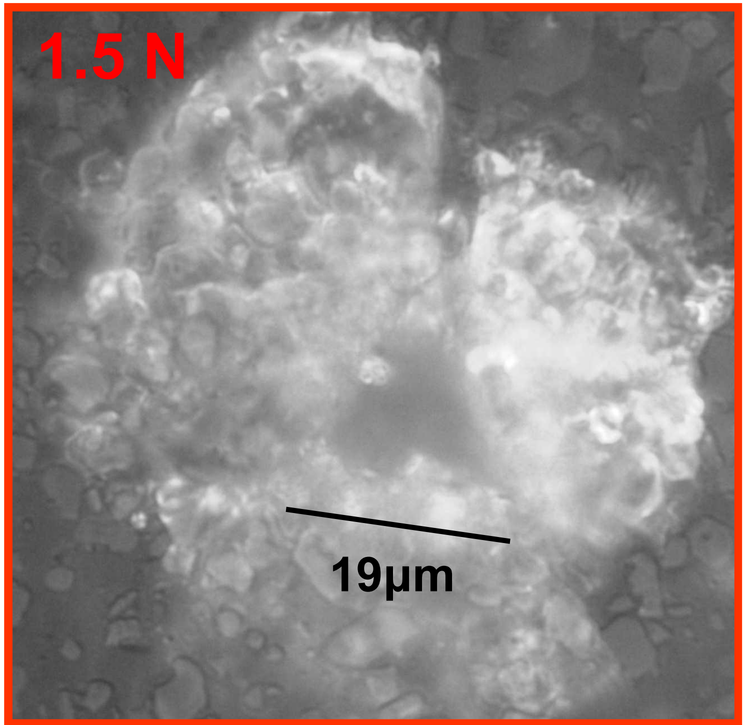




LTCC







Conclusions

-Cube corner indentation appears to be a good technique for measuring stresses in glasses

-As compressive stress magnitude increases, volume in which stresses can be sampled decreases

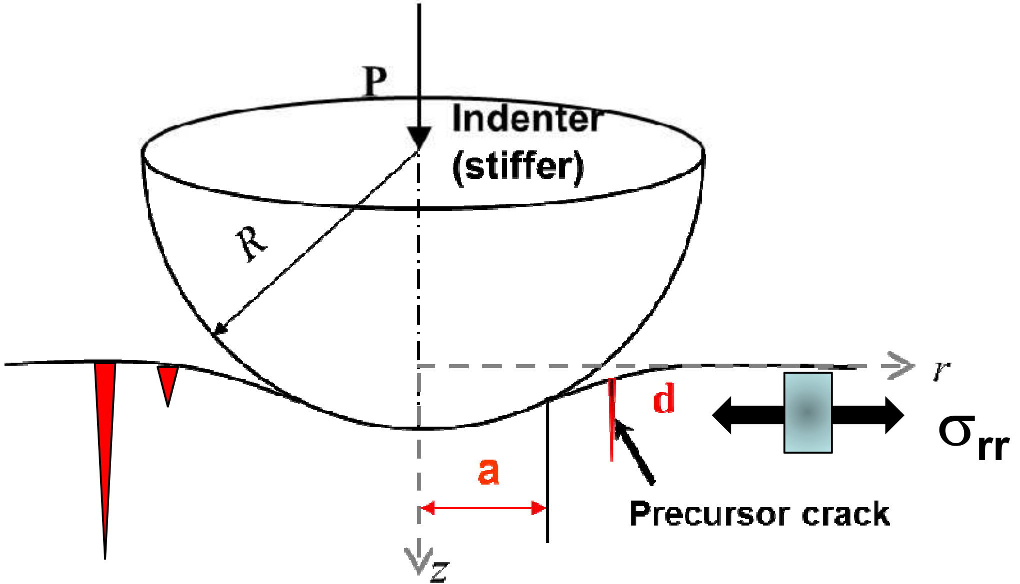
- Compressive stress measurements in ~10μm^3 appears possible

-Polycrystalline measurements may not be possible

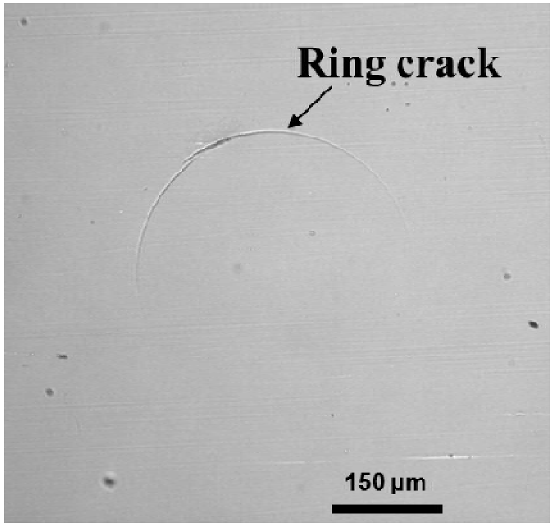
Outline

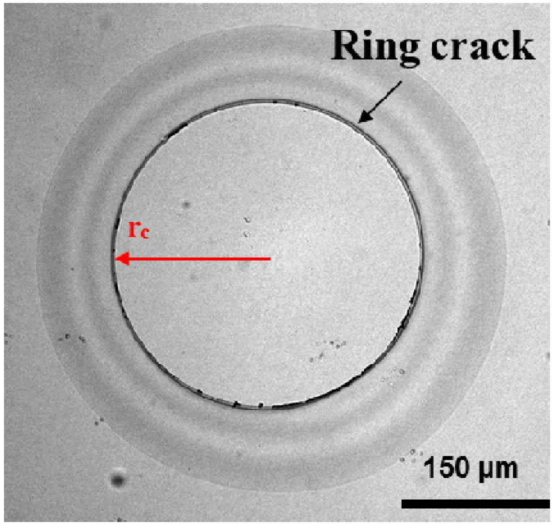
- Localized stresses are important
  - Examples of material failures from residual stresses
- Why measure stresses?
  - Use of Vickers indentation for residual stress measurements
- Method to use Curved Cracks to Measure Stress
  - Fracture mechanics analysis, and use in LTCC-gold via
- Development of Cube-corner Indentation Method
  - Experiments on stressed glasses (calibration)
  - “Quarter- penny” cracks; Fracture Mechanics Analysis
  - Volume Resolution of technique, Practical Use & Limitations
- Nano-indentation based Method
  - Load displacement curves on stressed surface are shifted
- **Hertzian Indentation**
  - **Local strength measurements**
  - **Interfacial toughness measurement**

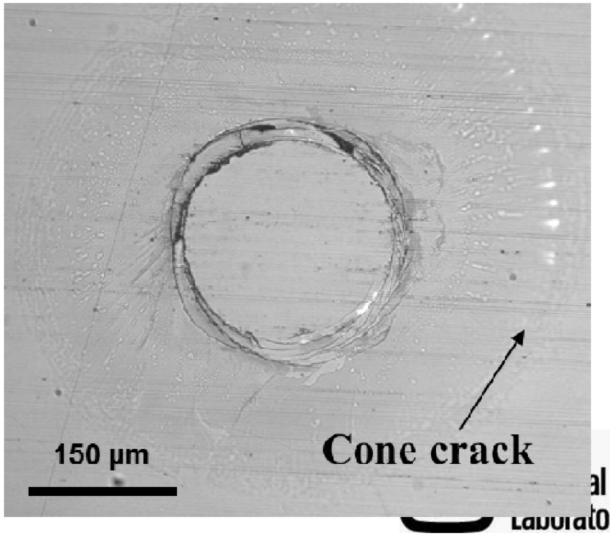
Hertzian Indentation



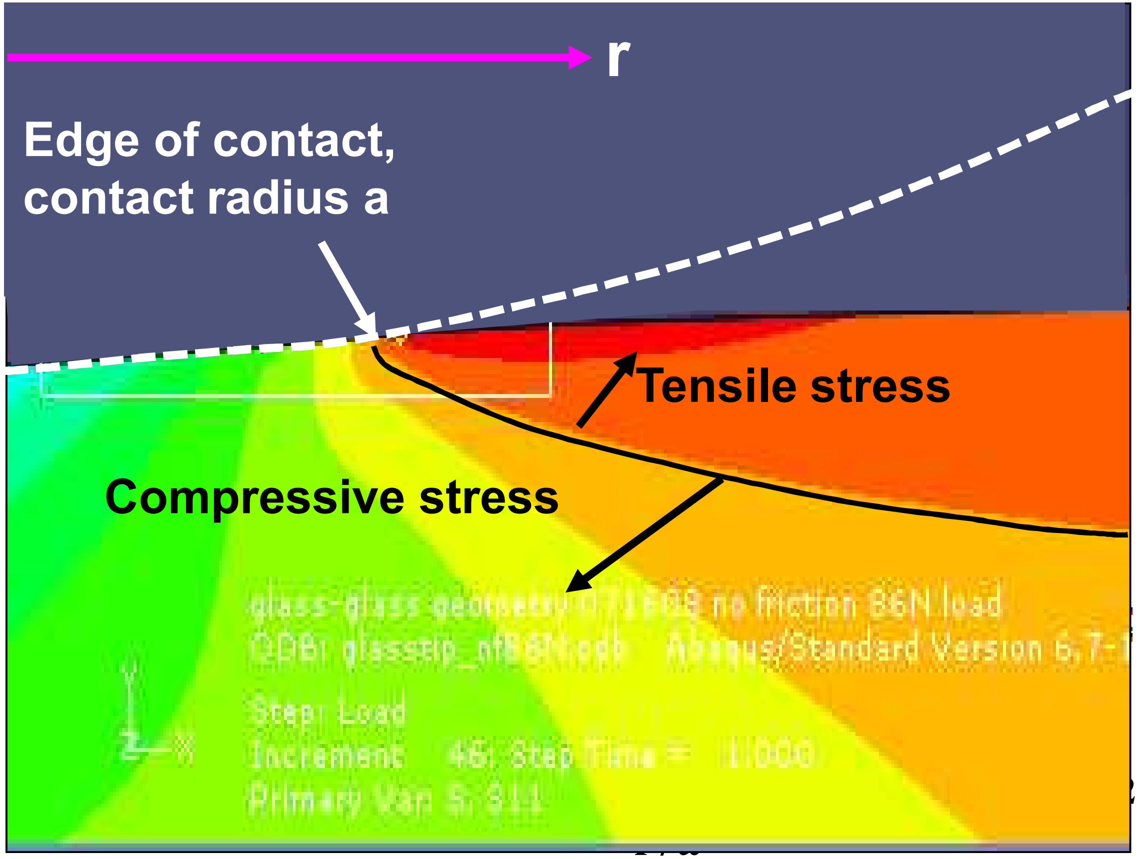
- Microscale-strength test
- Adhesion Measurements
- Localized strength
- Flaw size distribution







Analytical, FE Results for Radial Stress with and w/o friction



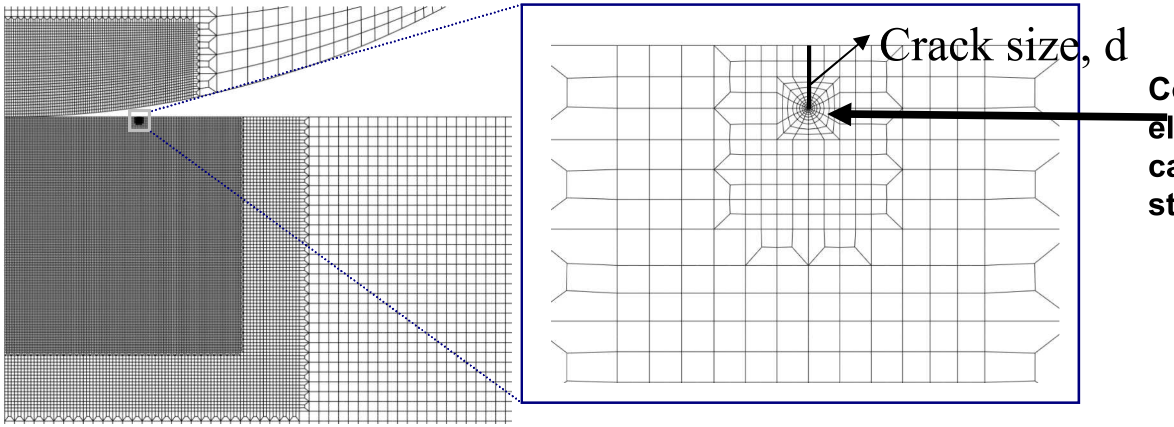
Friction is important for elastically dissimilar contacts
Friction shifts the maximum in the stress farther from contact edge
Friction also leads to reduction in normalized stress

Paliwal B , R Tandon et al., Assessing the Hertzian Indentation Approach for Measuring Fracture Toughness
J. Amer. Ceram. Soc., Volume 94, Issue 5, May 2011

Analytical and FE Approach to K

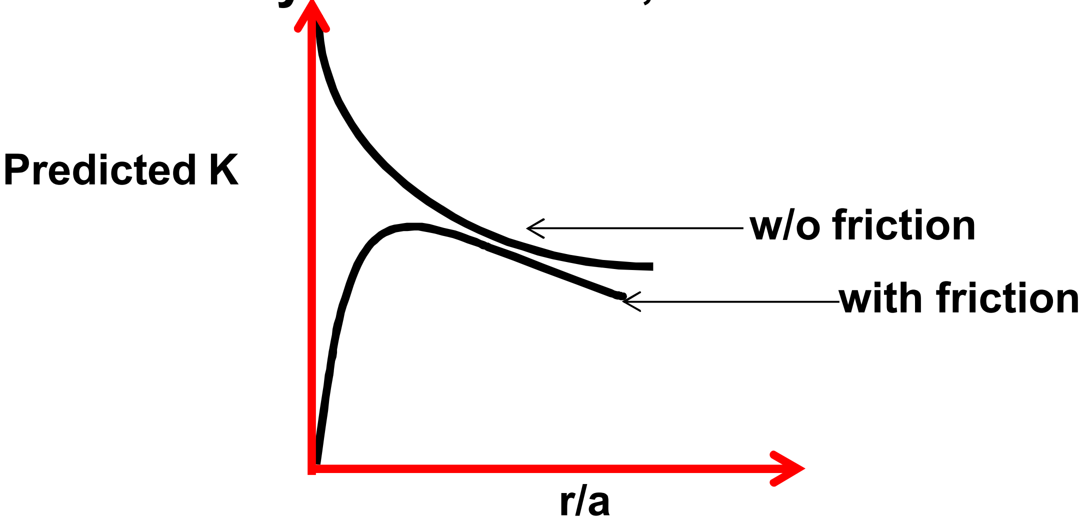
Knowing the radial stress distribution in the presence of friction, mode I stress intensity factor can be written as

$$\frac{K_I}{p_0\sqrt{\pi a}} = \frac{2}{\pi}\sqrt{\frac{d}{a}}\int_0^{d/a}\frac{\sigma_{rr}(r_c,z)/p_0}{\sqrt{(d/a)^2-(z/a)^2}}\,d(z/a)$$



Collapsed quad elements to capture stress singularity

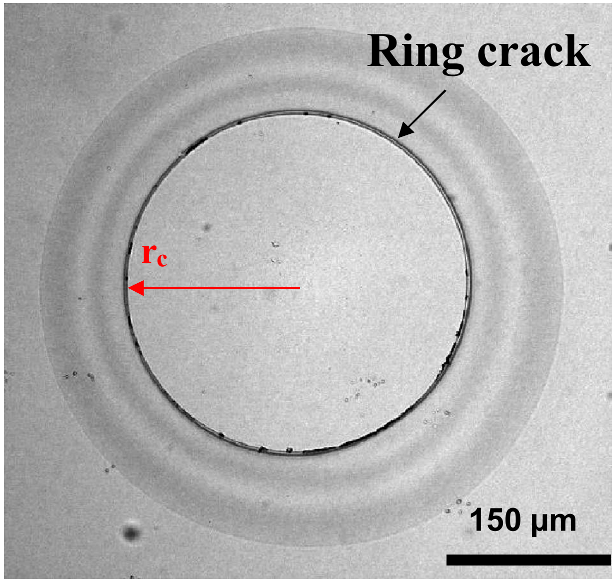
For extremely small cracks, d/a<0.03



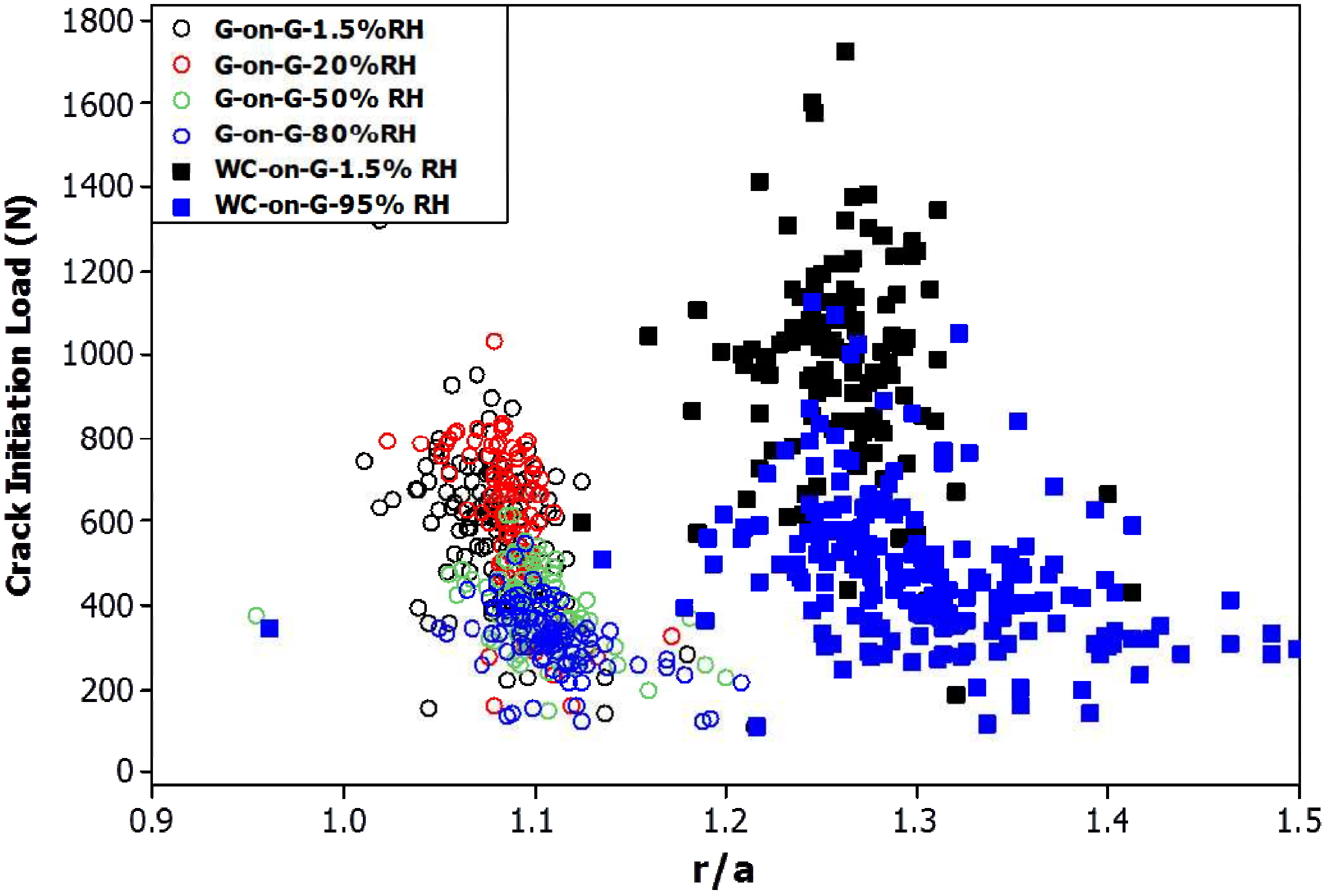
Experimental Approach

- >500 tests WC-glass (dissimilar contact), R=0.8 mm- 5.8 mm
- >500 tests Glass-glass (similar contact), R=0.8 mm- 5.8 mm
- Various controlled RH's, loading rate=0.2 mm/min, 10-15 sec per test
- Acoustic emission was used to measure the load at cracking
- Friction coefficient of glass/ WC was measured (μ=0.15 in sliding test)

-Cracking locations were measured

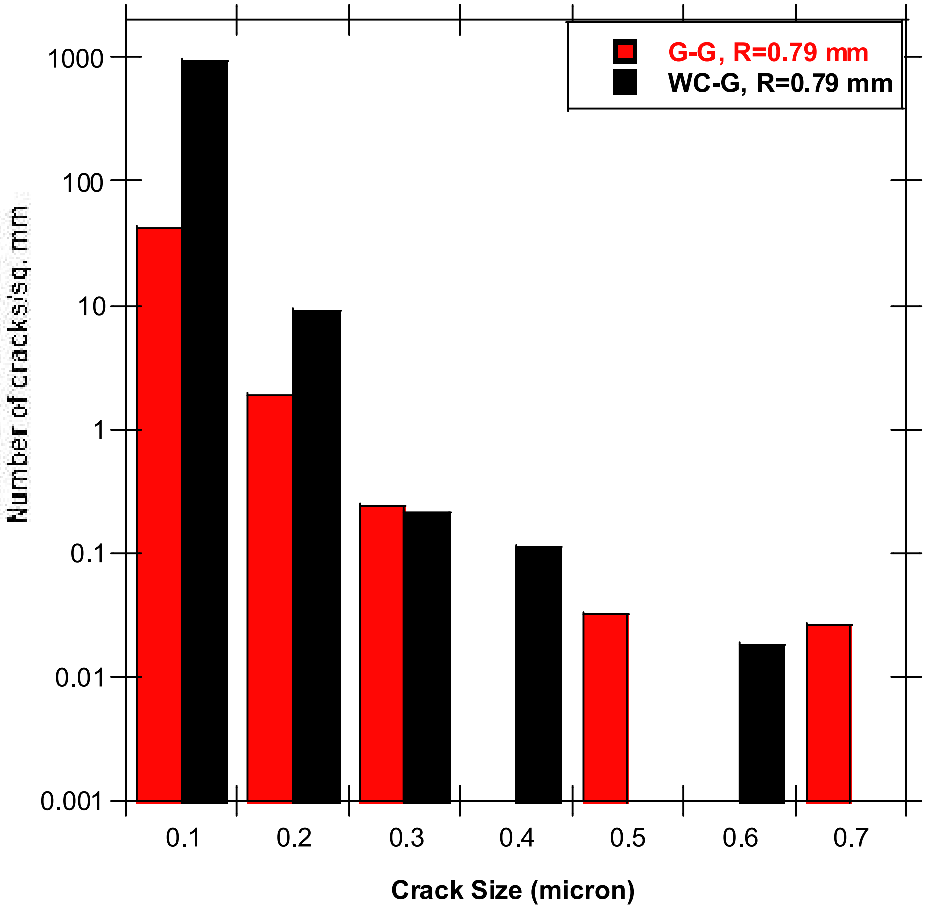


Effects of Humidity and Friction



- Cracking locations are shifted well away from r/a=1 for WC-on-G
- Consistent with the value of peak stress and K being shifted away from contact edge for dissimilar contact
- Effect of humidity is to shift these further due to SCG

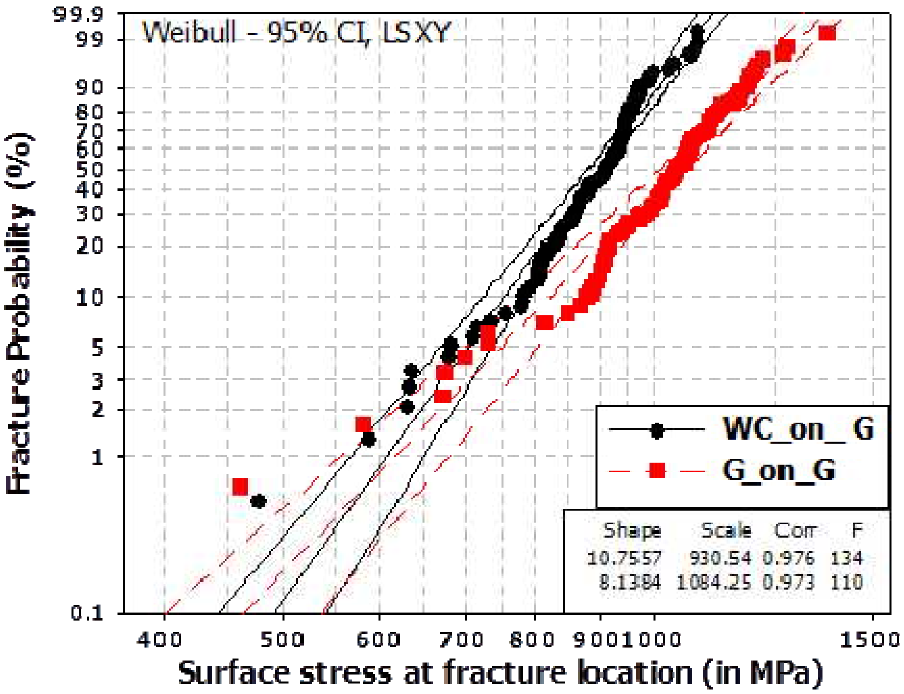
Materials Parameters Obtained



Crack Density on Glass Surface

Relevant to blast or penetration simulation

Micro-strength Test on a Surface



Shape	Scale	Corr	F
10.7557	930.54	0.976	134
8.1384	1084.25	0.973	110

Micro-strength >> Macro-strength

Relevant to fracture initiation in electronic packaging

Interfacial Delamination

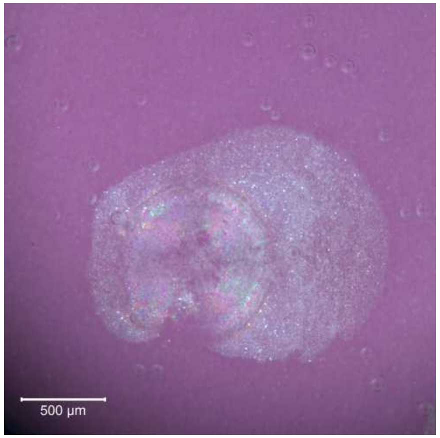
Hertzian loading can be used to create well-defined delaminations on bi-material interfaces

Delamination size can be used to estimate toughness of interface

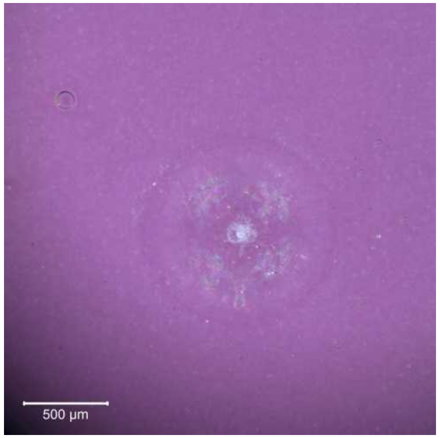
Delamination - Alumina coated with epoxy

C3 50kg

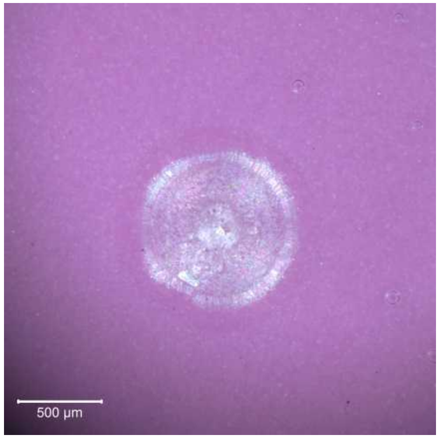
No delam



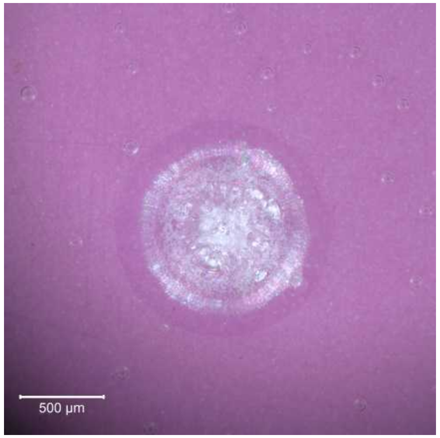
-55C



-30C

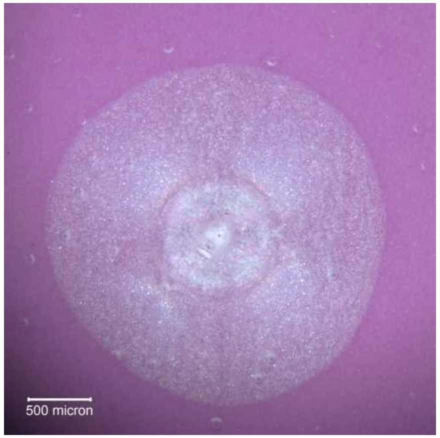


0C



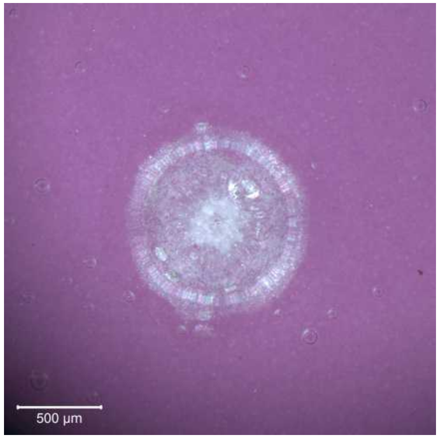
21C

C3 80kg



-55C

-30C



0C

21C

Summary

Measurements from curving cracks may be used to estimate stresses near interfaces with proper accounting for crack geometry and elastic mismatches

Measurements of stresses at ~10-20 micron resolution using cube corner indentations in glasses are possible

Spherical indentations are extremely useful for measuring

- localized strength
- crack density
- interfacial toughness

Development and Practical Applications of Indentation Based-Techniques to Measure Stresses and Interfacial Properties in Brittle Materials

Rajan Tandon
Materials Reliability
Materials Science and Engineering Center,
Sandia National Laboratories, Albuquerque, NM 87185

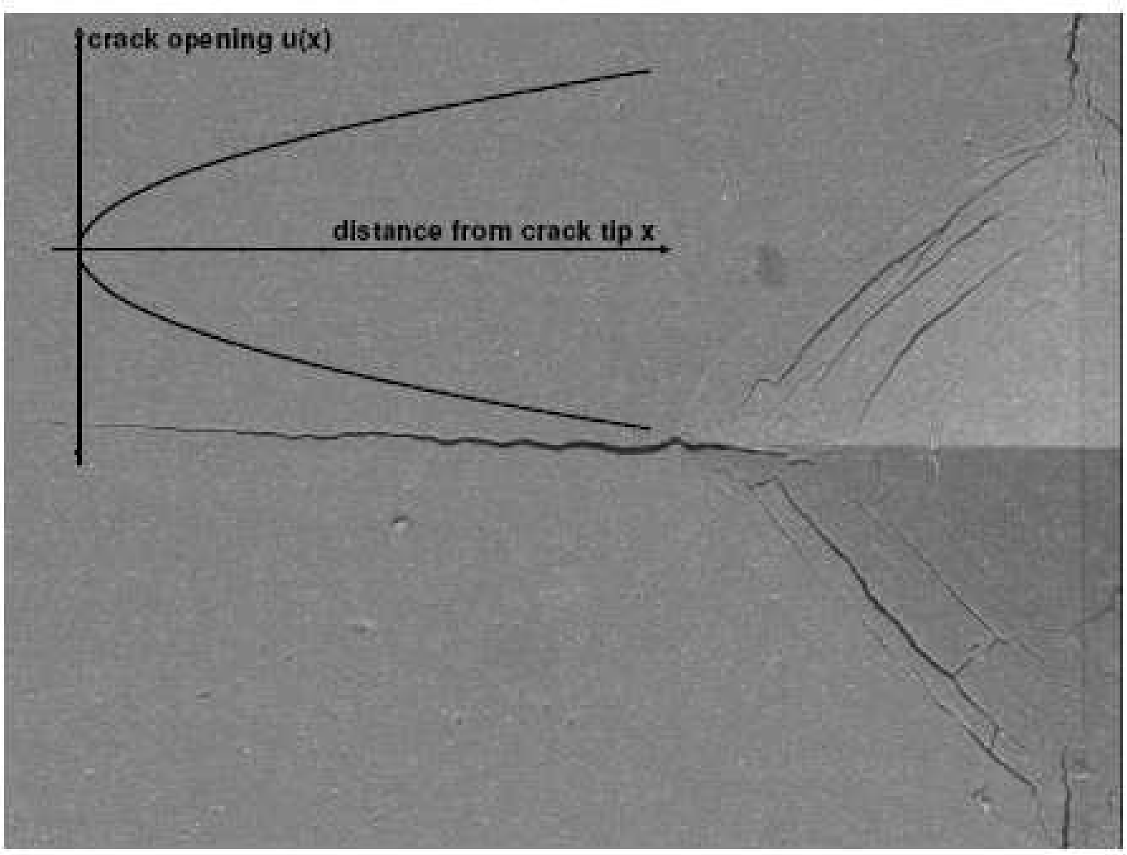
Engineering ceramics and glasses are widely used in electronic packages, hermetic seals and optical components. For component functionality, the ceramic or glass is often associated with a metal or polymer, and in many cases, a strong bond is desired across this interface. Elastic and thermal expansion mismatches between the different materials create a state of residual stress in the ceramic/glass, which in extreme cases, manifest as failures during manufacturing or testing. Many of these failures occur close to, or at the interface. Thus, an understanding of the residual stress state and interfacial bonding properties is essential for the reliability and lifetime prediction of ceramics/glasses.

Despite the availability of enormous computing resources, predictions of these characteristics are not straightforward. For a model to accurately predict stresses in the ceramic/glass, knowledge of the physics and chemistry of the problem is a pre-requisite. For instance, chemical reactions at interfaces often create compounds whose properties are largely unknown. Viscous deformations in polymers and glass/glass-filled ceramics and the plastic behavior of metals strongly impact the residual stresses near interfaces. Therefore, precise information about the thermal environments during manufacturing and testing, and temperature-dependent thermal and viscous/elastic/plastic mechanical properties are needed for accurate prediction. For many engineering materials, these properties and proper constitutive behavior are unknown over the range of temperatures of interest. Additionally, the interfacial bonding characteristics are influenced by numerous factors, some intrinsic (such as surface roughness, chemical affinity etc.), and some extrinsic (such as cleanliness), which makes failure predictions very difficult.

Our work, therefore, has focused on deploying techniques for measuring residual stresses and interfacial properties in components. Indentation approaches are generally easy to implement, and provide materials property information with meso/microscale resolution. For estimating bulk behavior, we have made extensive use of indentation crack lengths using a sharp diamond indenter. These cracks in bulk materials are straight, and the difference in crack lengths between the stressed surface (in the component) to an unstressed surface can be related to the stress state using fracture mechanics. Near interfaces where extremely localized information is needed, this simple approach runs into at least two limitations: (1) near curved interfaces, indentation cracks are curved, and (2) stresses are sampled over the entire crack length, which in many cases is of the order of several hundred microns. In the first part of the talk, a fracture mechanics approach that allows estimation of stresses from measurements of the curved crack lengths near curved interfaces is described. The analysis results are applied to measure stresses as a function of radial distance from a metal-ceramic interface in an electronic packaging material. In the second part of the talk, two additional approaches to measure stresses, over progressively smaller length scales, are described. One approach is to use a cube-corner indenter, which due to its acuity, generates cracks at extremely low loads. A demonstration of the technique to sample stress in an ~10 micron region is demonstrated, and the successful implementation of the technique in component design is presented. A nano-indentation approach that may allow sampling stress across ~5 micron regions is then discussed. In the third part of the talk, a spherical indentation approach to measure interfacial toughness is described, and preliminary experimental results are presented.

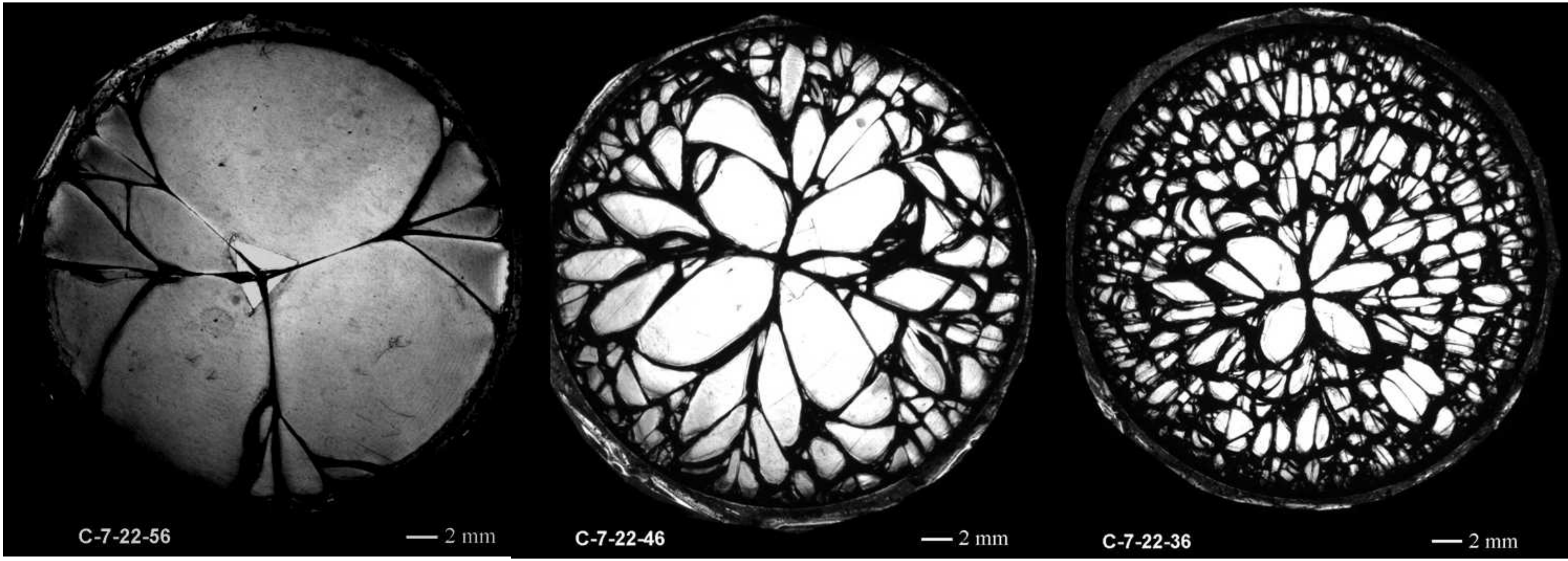
Motivation for COD work

Crack Opening Displacement (nm)



Differences in COD might provide estimates of stress

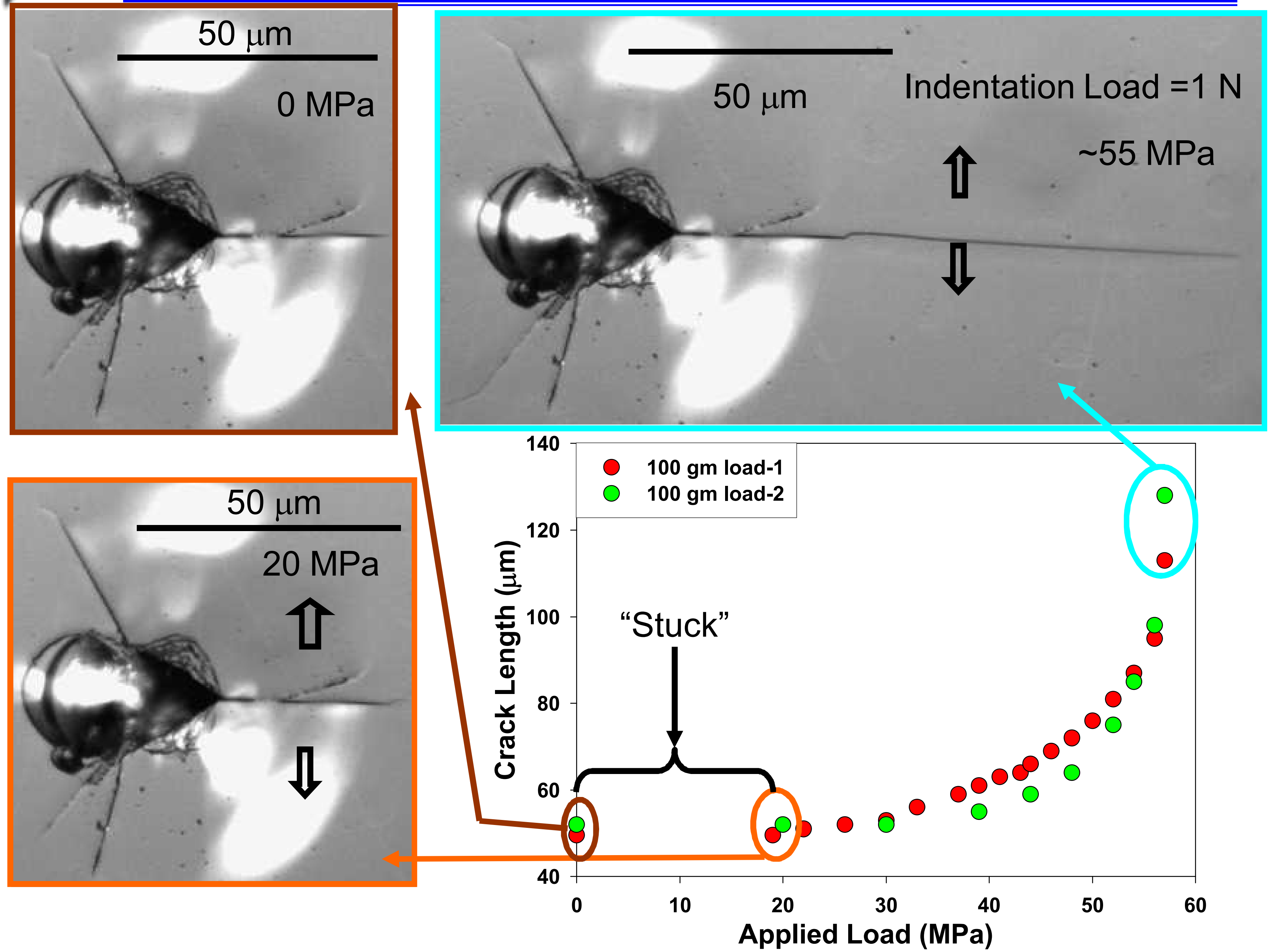
Motivation for Ion-Ex. work



Field-assisted ion-exchange might enhance the design space for such applications

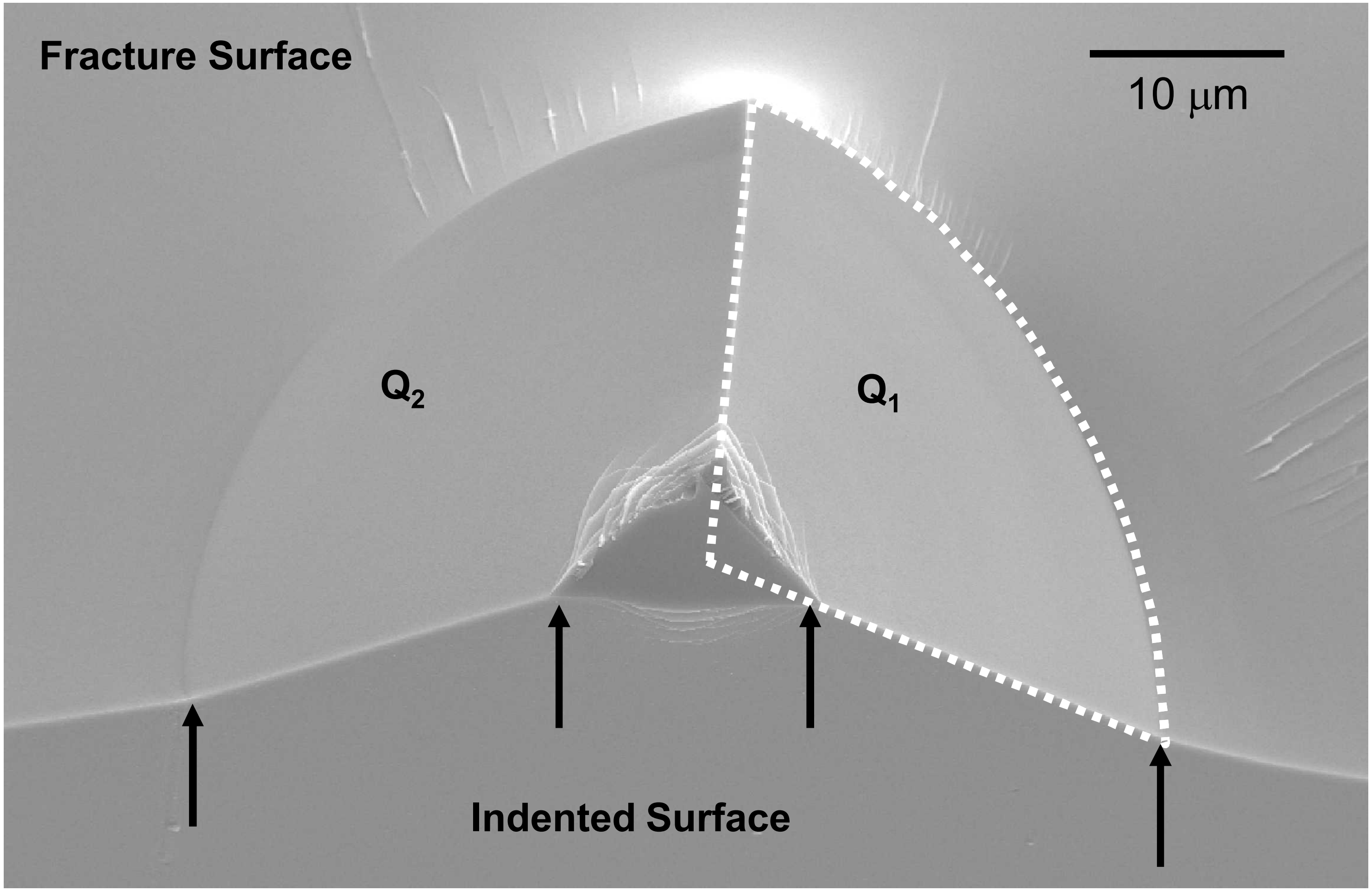
- Looking for "<u>things that go bang</u>" with temperature rise

Limitations of use of Cube Corner

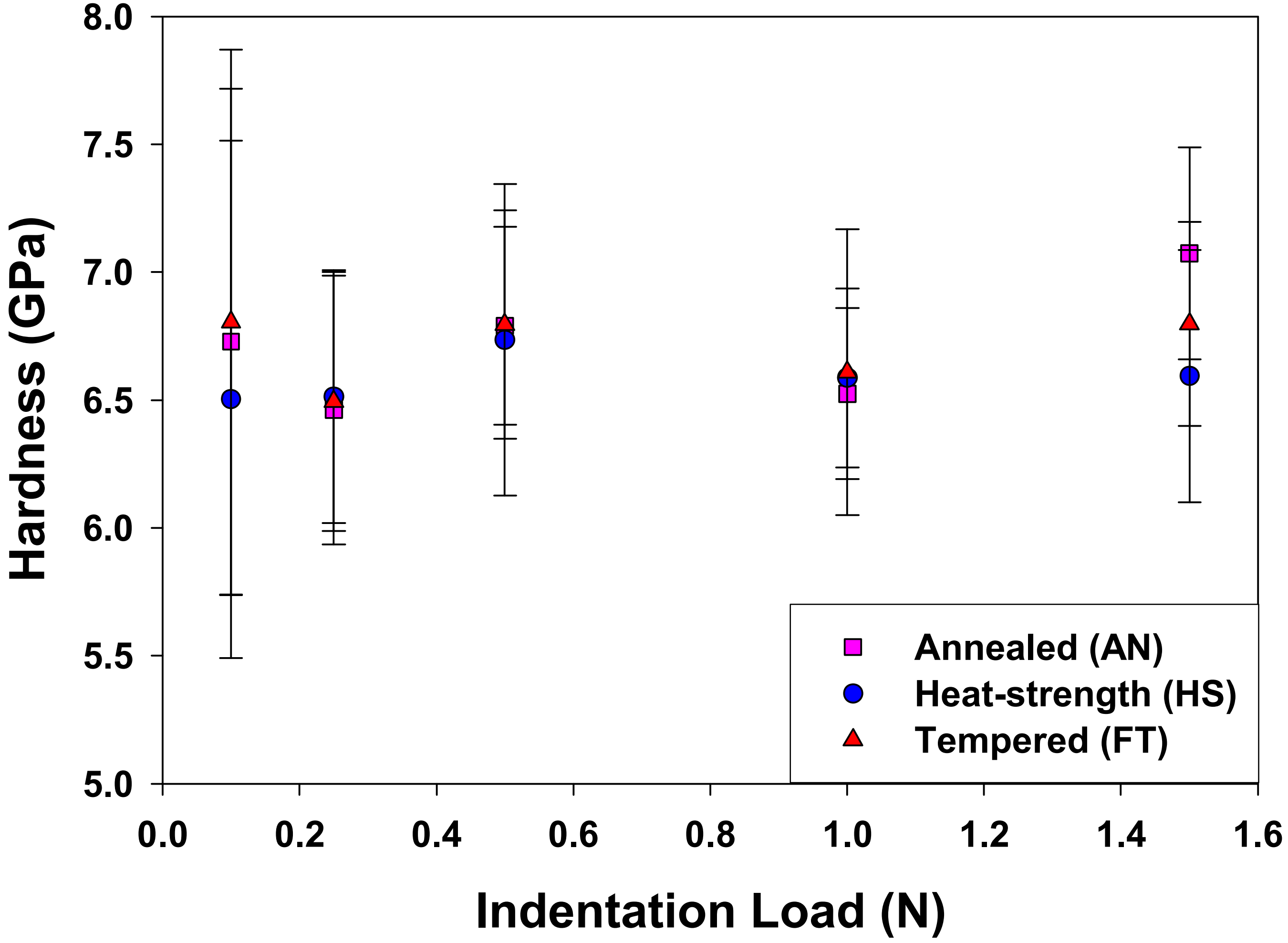


Quarter Penny Crack: Lower Load

Annealed: 0.5 N



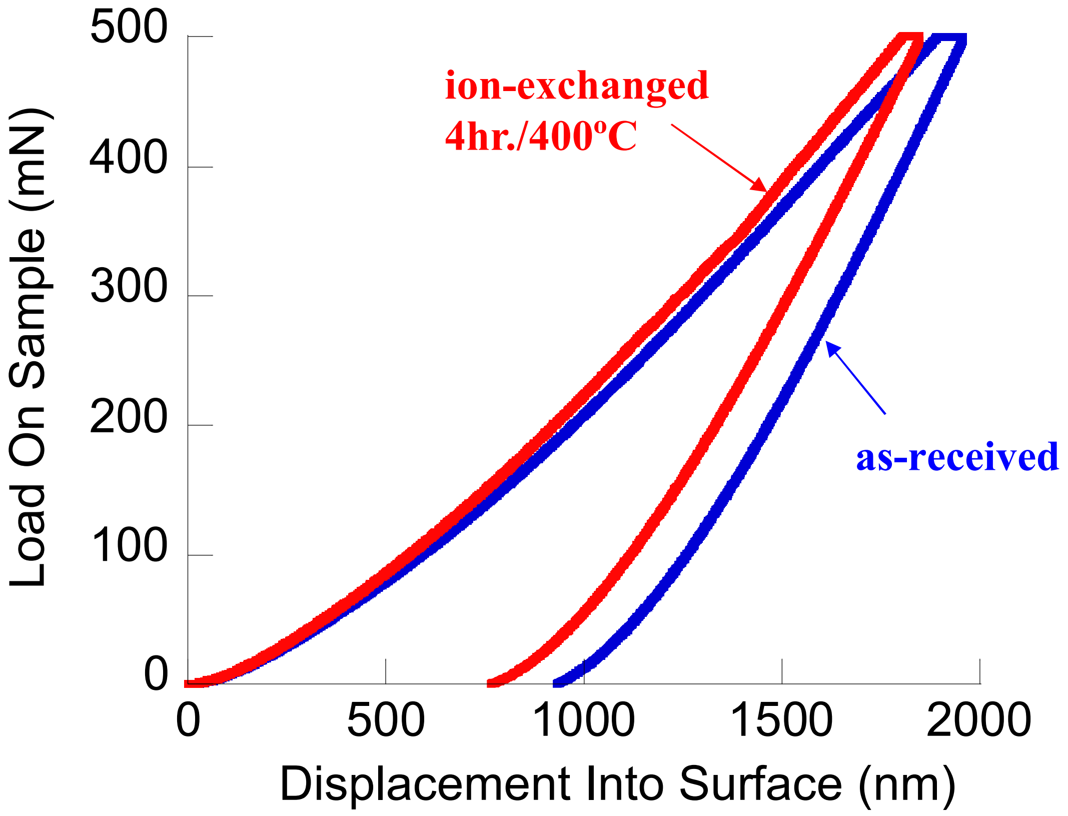
Hardness Values Unchanged



Outline

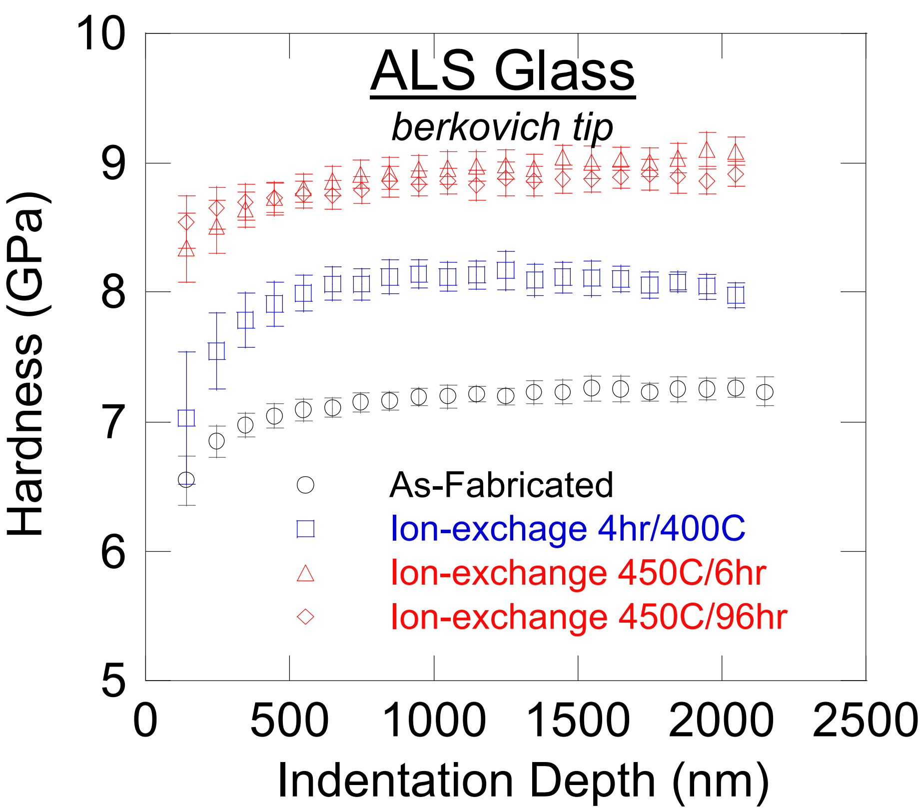
- Localized stresses are important
  - Examples of material failures from residual stresses
- Why measure stresses?
  - Use of Vickers indentation for residual stress measurements
- Method to use Curved Cracks to Measure Stress
  - Fracture mechanics analysis, and use in LTCC-gold via
- Development of Cube-corner Indentation Method
  - Experiments on stressed glasses (calibration)
  - “Quarter- penny” cracks; Fracture Mechanics Analysis
  - Volume Resolution of technique, Practical Use & Limitations
- **Nano-indentation based Method**
  - **Load displacement curves on stressed surface are shifted**
- Hertzian Indentation
  - Local strength measurements
  - Interfacial toughness measurement

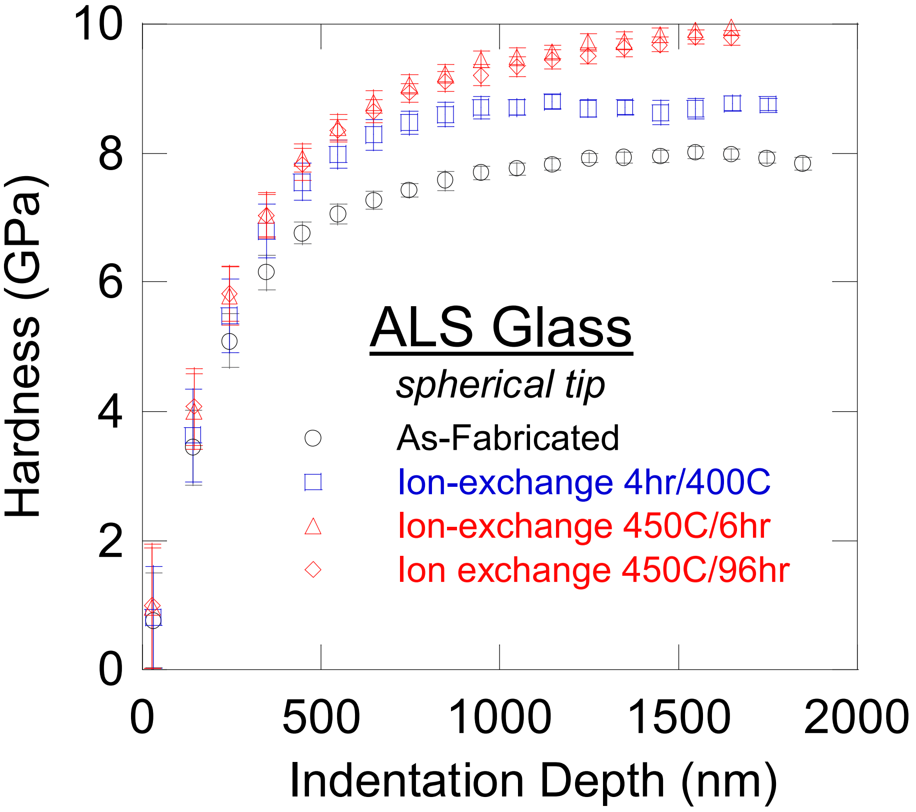
Nano-indentation may allow us to sample stress in much smaller volumes



- **A very noticeable and repeatable difference in measured indentation response between unstressed and stressed glass.**

CSM Results for sharp and blunt tips
Compressively stressed surfaces show higher hardness





Independently Estimated Stresses:

~-200 MPa: ALS Ion Exchanged 4hr/400C;

-300 MPa: ALS Ion Exchanged 6hr/450C ALS Ion-Exchanged 96hr/450C

Details of Finite Element Simulations

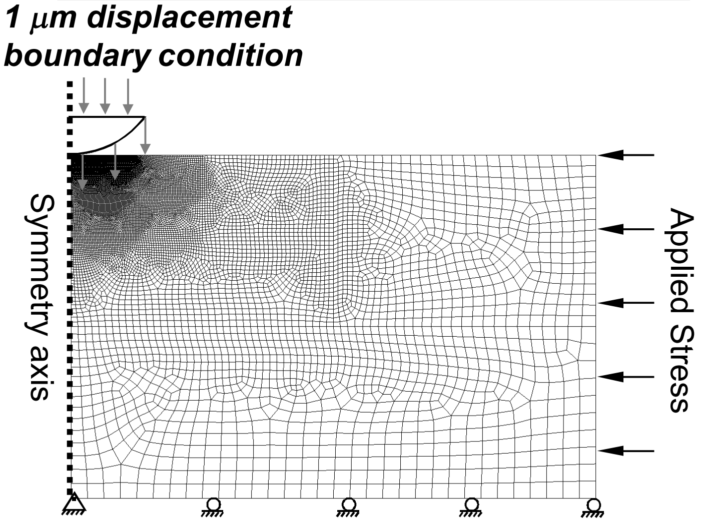
- **Displacement control: 1 µm depth**

	material properties	tip geometry	residual stress
GLASS	E=72 GPa σ_y=3 GPa	spherical- 10 µm radius	1) none 2) -500 MPa
	E=72 GPa σ_y=3 GPa	conical- 70.3º half-angle	3) none 4) -500 MPa
METAL	E=72 GPa σ_y=600 MPa	spherical- 10 µm radius	5) none 6) -500MPa
	E=72 GPa σ_y=600 MPa	conical- 70.3º half-angle	7) none 8) -500MPa

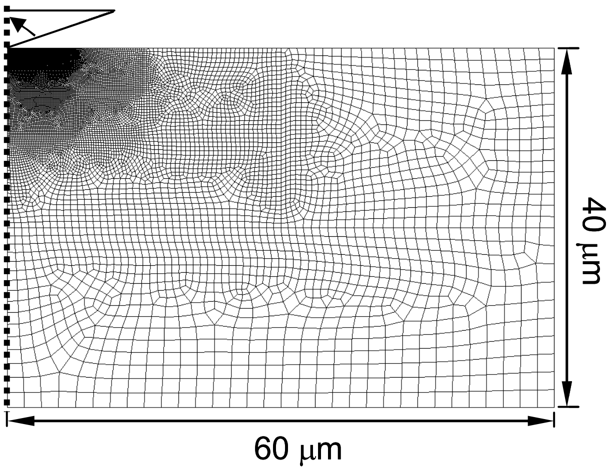
Simulation Details:

- Materials: elastic-perfectly plastic
- Frictionless contact
- *rigid tip*
- substrate 30x40 µm –fixed
- approx. 20,000 elements
- axisymmetric elements

Spherical Tip Radius=10 µm



Conical Tip Angle=70.3°

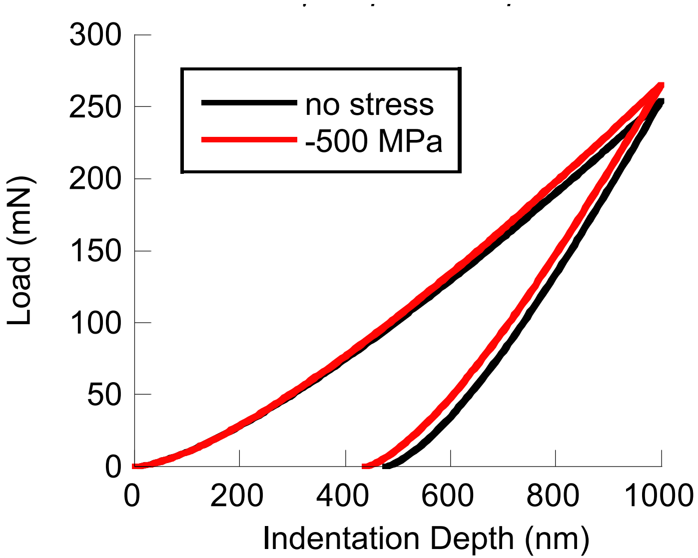


Simulation Result: Stressed substrates have different load-displacement response

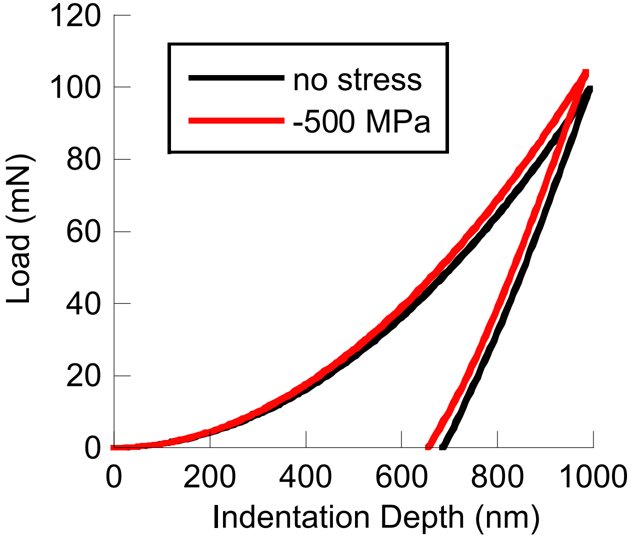
E= 72 GPa
σ_y= 3 GPa
ν=0.2

E/σ_y= 24, Low, Glass

Spherical tip

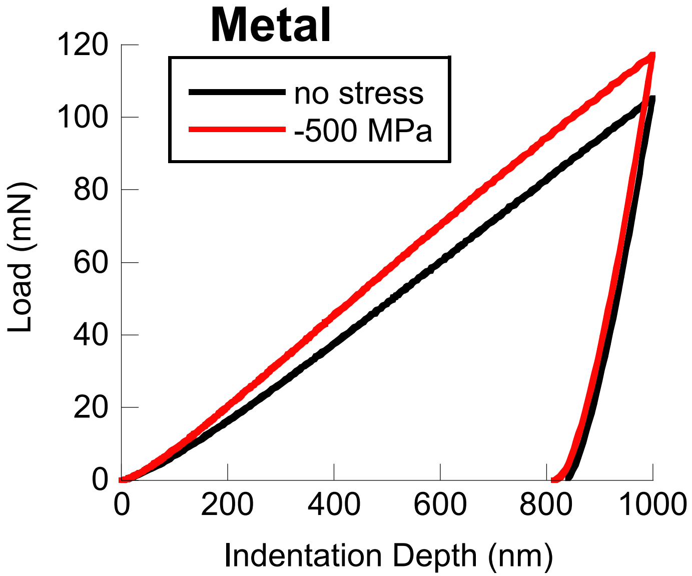


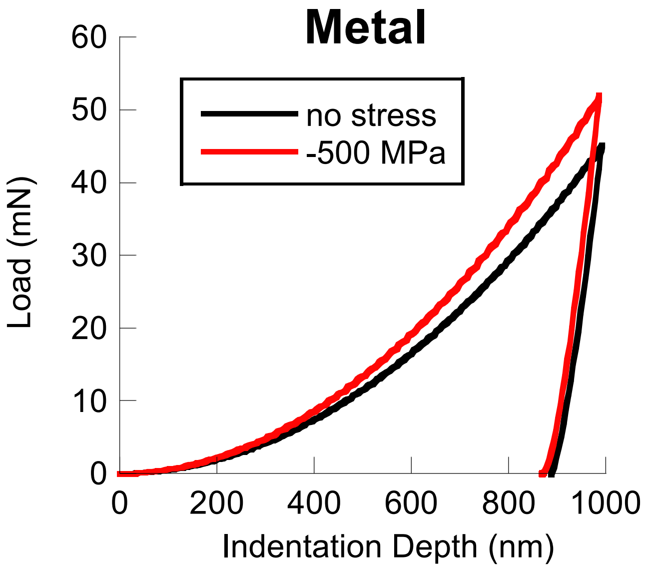
Conical tip



E= 72 GPa
σ_y= 600 MPa
ν=0.3

E/σ_y= 120, High, Metal





Conclusions

For brittle materials, low E/σ_y ratio
Experimental: Measureable hardness difference for spherical & conical
Simulations: This difference is an actual response to the stress (not due to pile-up)

For ductile material, high E/σ_y: Simulations only
Hardness differences in conical indentation completely due to pile-up
Spherical indentation in the elastic-plastic region show a difference

- Possibility of measuring surface stress states in small volumes exist
- Available formulations are not able to reproduce stress values either for the simulation or experiments