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# Cohesive Zone Modeling of Particle-Binder Interfaces

**DR. TIMOTHY C. MILLER, SR. MATERIALS RESEARCH ENGINEER**

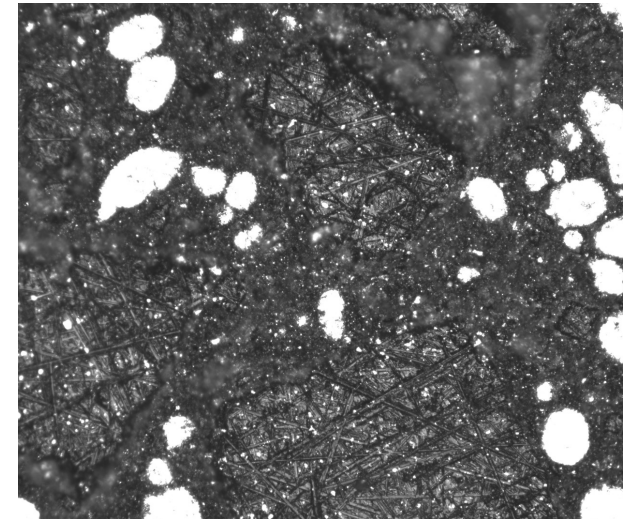
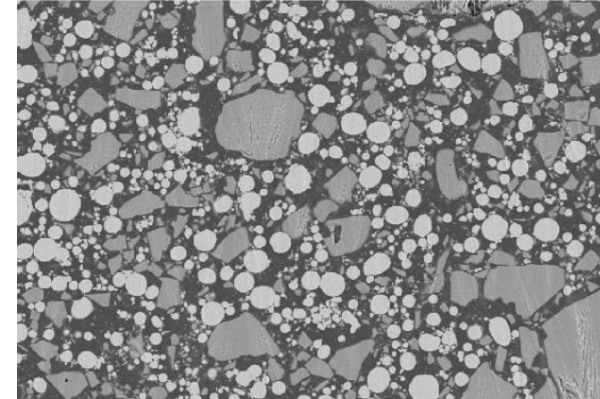
**AFRL/RQRP 6-16 DECEMBER 2021**

**JANNAF JOINT PROPULSION CONFERENCE**



# Background: Special Features of Highly-Filled Composites (HFCs)

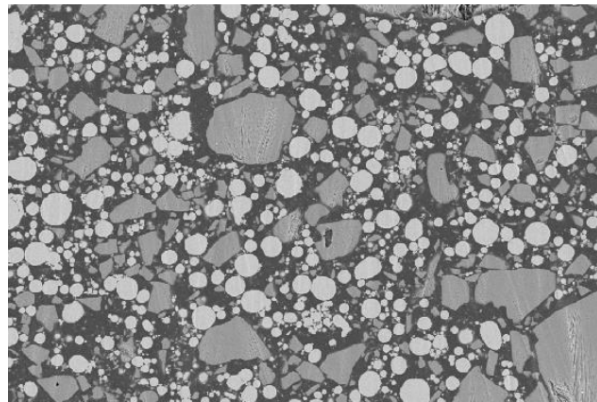
- **They are *highly-filled*.** Most micromechanics constitutive theories were formulated with a *dilute concentration assumption* – the embedded inclusions (fibers, voids, or particles) were far from each other and had no effect on each other. This is not the case with HFCs.
- **They have *interfaces*.** – since composites are combinations of two or more materials, separated by *interfaces* – these interfaces are usually the weak point in the microstructure, and a deep understanding of these interfaces is needed but not yet fully developed.
- **Time and temperature dependence.** The matrix material is frequently viscoelastic, so *time and temperature effects* become very important.



# Background: Damage in Highly-Filled Composites

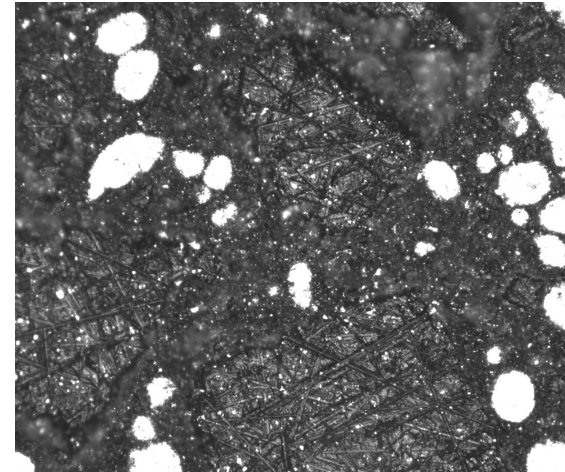
*Dewetting* within solid propellants is one of the major phenomena associated with degradation of the propellant.

*Delaminations* at the propellant-liner-insulation interfaces is another source of solid rocket motor failure.

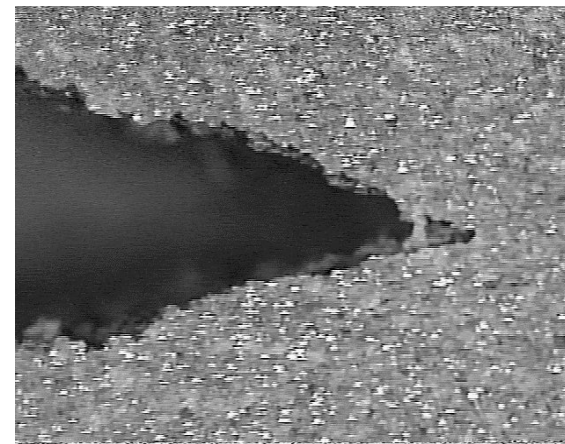


350  $\mu\text{m}$

**SEM microscope image** of solid propellant.



Olympus **optical microscope image** of solid propellant. The bright spots are aluminum while the larger “particles” are ammonium perchlorate.

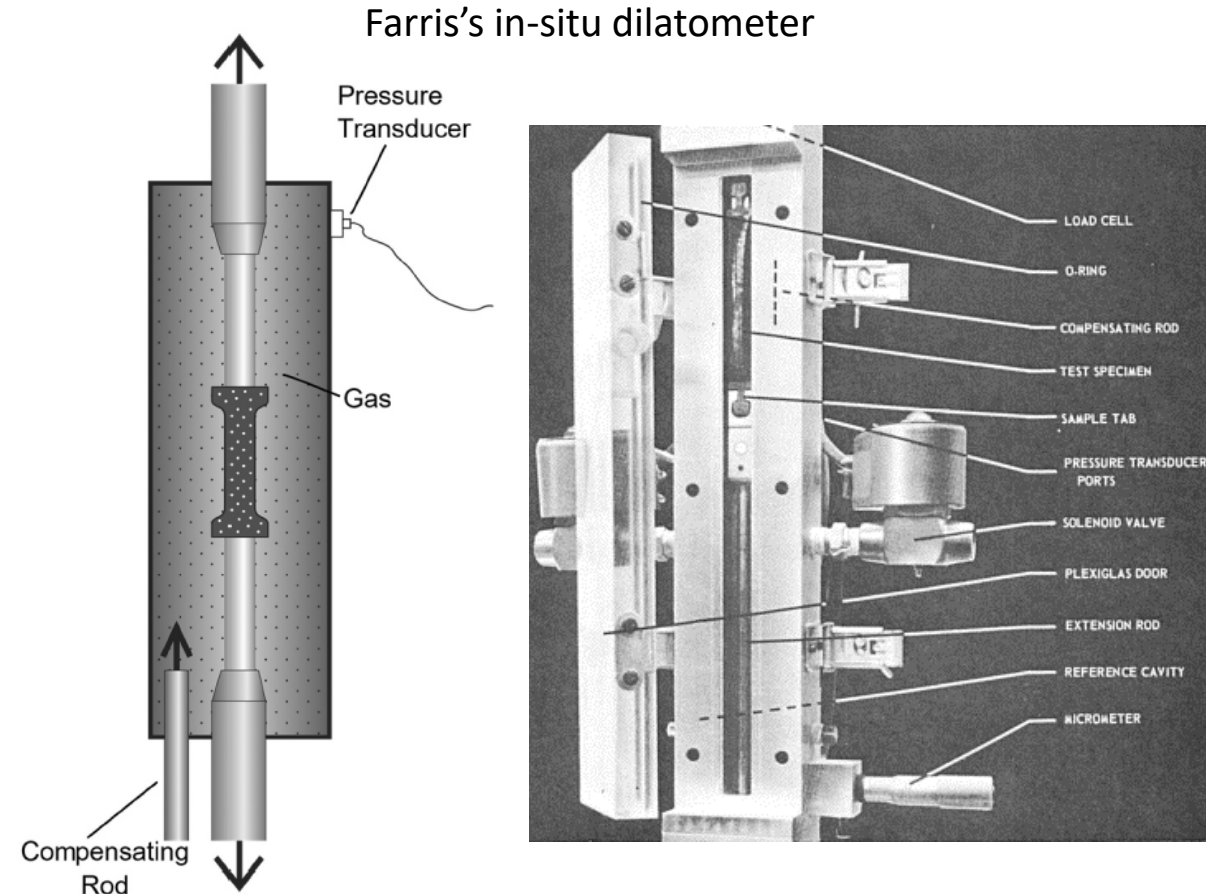


A macrocrack growing from the coalescence of voids that developed in the propellant as a result of dewetting.

Knowing how and why the particles separate allows us to develop accurate material models for failure.

# Background: Damage Measurement in Bulk Material

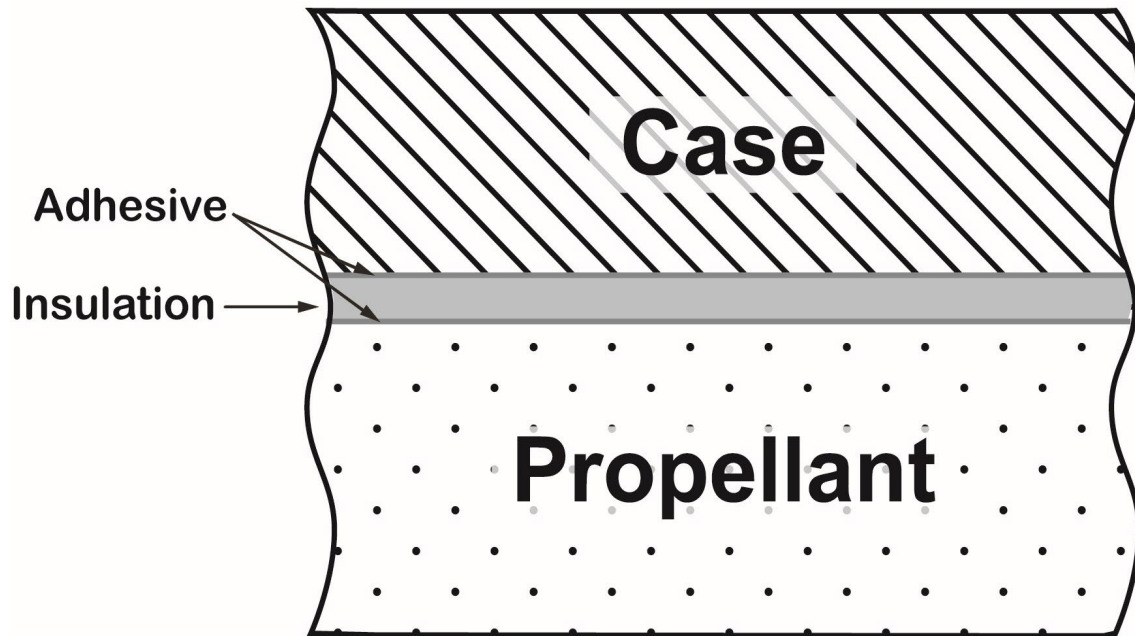
- **Dewetting** is the term used with propellants when the particles in the composite separate from the surrounding matrix material.
- Farris came up with a way to measure volume changes in a propellant specimen in the late 1960's – this gave information on when dewetting occurred.
- Problems with this approach:
  - It tells us nothing about what is happening at the interfaces, only that they are separating.
  - Farris's instrument only worked in uniaxial tension, whereas failure theories need to be conceived of and tested in a three-dimensional stress space.
- It would be really good to quantify the dewetting process **at the microstructural level**.



We can currently measure damage in highly-filled composites, but this is really only done at a bulk material level, rather than at a microstructural level.

# Background: PLI Interface Separations

## PLI (Propellant-Liner-Insulation) interface separations



These interfaces, found in solid rocket motors, are particularly hard to manage:

- They are important (grain unzipping is possible)
- They are hidden inside a structure
- Using ultrasonics to inspect them does not work (impedance mismatch)
- X-rays do not work well (density is not a characteristic of a debond)

Also, fracture mechanics is difficult:

- Sharp crack tips are not a reality
- Multiple thin layers means there is probably not a “singularity dominated zone” at the crack tip.
- Interfacial fracture mechanics is always tricky.

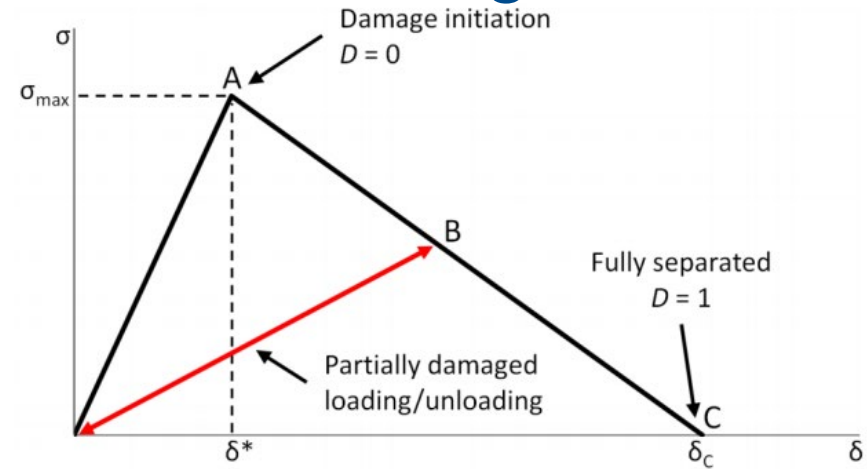
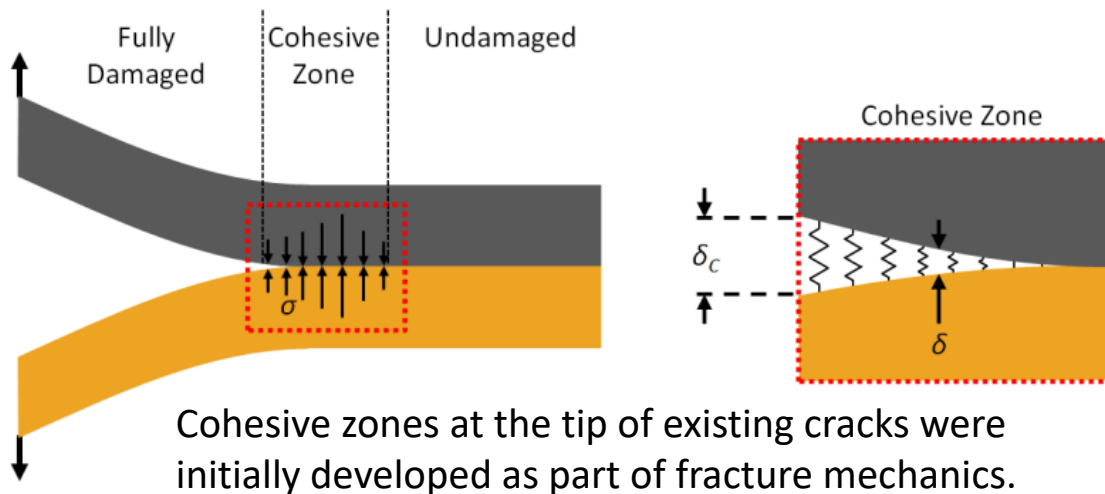
There's a huge payoff if we can model the failure of these interfaces better: more accurate failure predictions means better service life predictions. This applies to missiles and also to bombs.

# Background: Cohesive Zone Modeling

Interfaces separate due to applied stresses; the particular relationship between tractions and displacement separation can be modeled with a *traction separation law*.

Determining the parameters for the traction separation law requires experimental data near or on the interface.

Once characterized, the model can be used to predict interfacial separation for multiple debonding scenarios.



Bilinear traction-separation law for cohesive zone elements.

Cohesive Zone Modeling is an alternative approach that evolved from fracture mechanics, but is distinct:

- It uses a damage parameter rather than a crack size assumption
- It does not need to presume an initial crack size and geometry
- It works better with complex geometries and loads, especially interfacial separations.

Cohesive Zone Modeling (CZM) is more useful than fracture mechanics in complex situations such as those found along interfaces.

# Background: Applications

Solid rocket motors

Dewetting

Propellant-liner-insulation delaminations

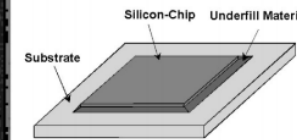
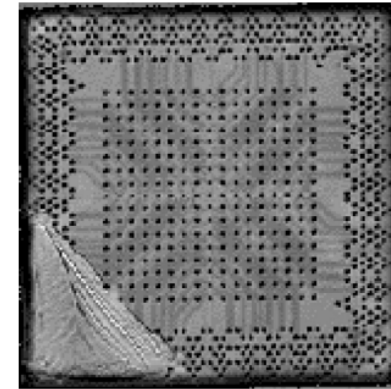
Geologic and civil engineering materials

Rock strata

Concrete

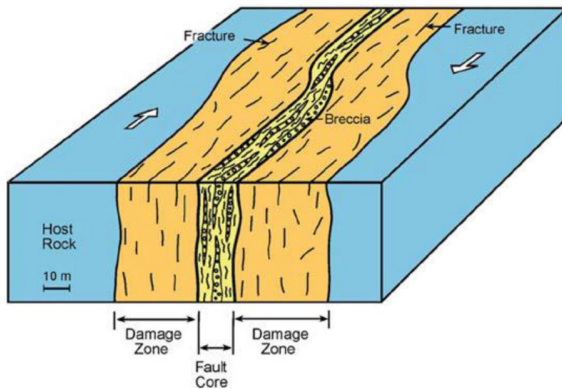
Asphalt

Layered materials (e.g., printed circuit boards)



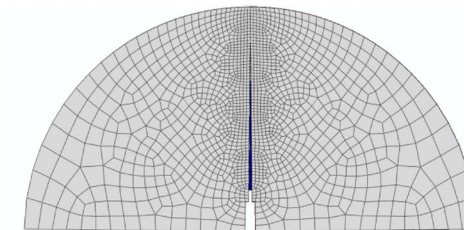
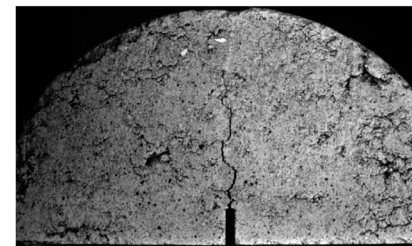
Electronic packaging (circuit board failure)

Nied, 2003, doi:10.1109/TDMR.2003.820623



Rock strata (earthquake fault lines)

IEAGHG, "Criteria of Fault Geomechanical Stability during a Pressure Build-Up, 2015/04, April, 2015



Asphalt pavement (potholes)

Liu et al., 2019, <https://doi.org/10.3390/ma12081278>



# Background: GOTCHA Chart

**Overall Goal:** A more fundamental understanding of the behavior of failure phenomena in highly-filled composites.

**Objective 1:** Investigate the microstructural failure phenomena along particle/matrix interfaces in a highly filled composite  
What affects the onset of particle dewetting? (bonding agent, particle size, volume fraction, temperature, loading rate)

**Objective 2:** Investigate the special circumstances of a propellant-insulation delamination  
What makes a kissing debond propagate? (bond strength, material properties, mode-mixity, temperature, loading rate)

## Technical Challenge

Digital Image Correlation analyzes the displacements of speckle patterns. To work properly, the speckles must be much smaller than the objects that you are investigating.

The depth of field of the optical microscope prevents acquisition of good data.

Too many variables to investigate in the given time frame.

## Approach

Use non-conventional approaches to speckling, quantum dots or finely dispersed particulate matter, as a means of speckling.

Work with the digital image correlation vendor to determine proper optics and approaches for this work.

Work incrementally, varying the most important parameters first.

# Background: Problem, Approach, Payoff

## Problem

- During storage or transport, particles in highly-filled composites can separate from the surrounding material, leading to damage that is hard to detect, predict, and characterize.
- Strains near the particle/matrix interfaces are hard to determine experimentally.
- The variables that characterize particle/matrix separation are not well understood.

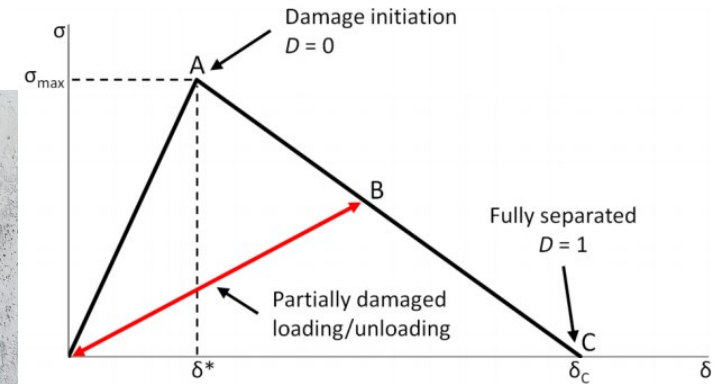
## Approach

- Apply dispersion of nanoparticles to surface of specimen.
- Using **two-dimensional digital image correlation** and optical microscopy, obtain deformations in microstructure while stretching the material.
- Determine stresses and strains near the interfaces.
- Analyze results to obtain parameters for cohesive zone modeling.

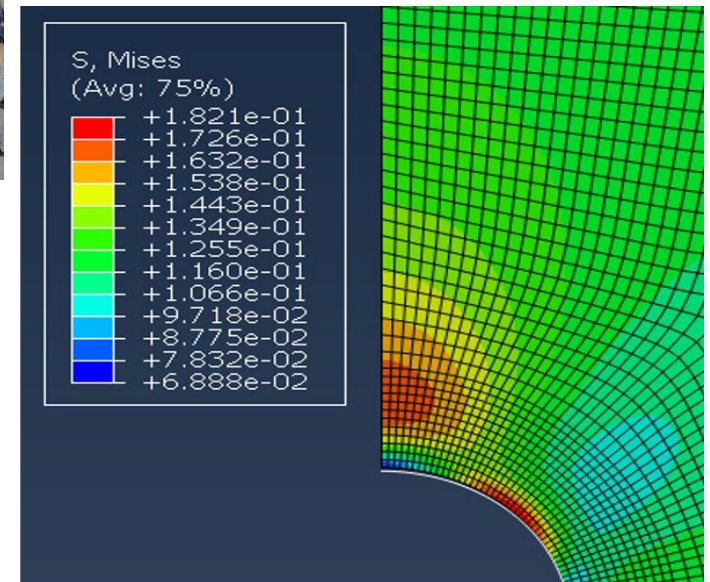
Validate with auxiliary testing.

## Payoff

- In-depth understanding of how particles separate from matrix. This gives propellant developers insight into strength and service life of newly developed materials.
- The ability to predict the tendency for adherends to separate under realistic loading conditions – this gives engineers the ability to determine failure modes and to predict service life.



Bilinear traction-separation law for cohesive zone elements.





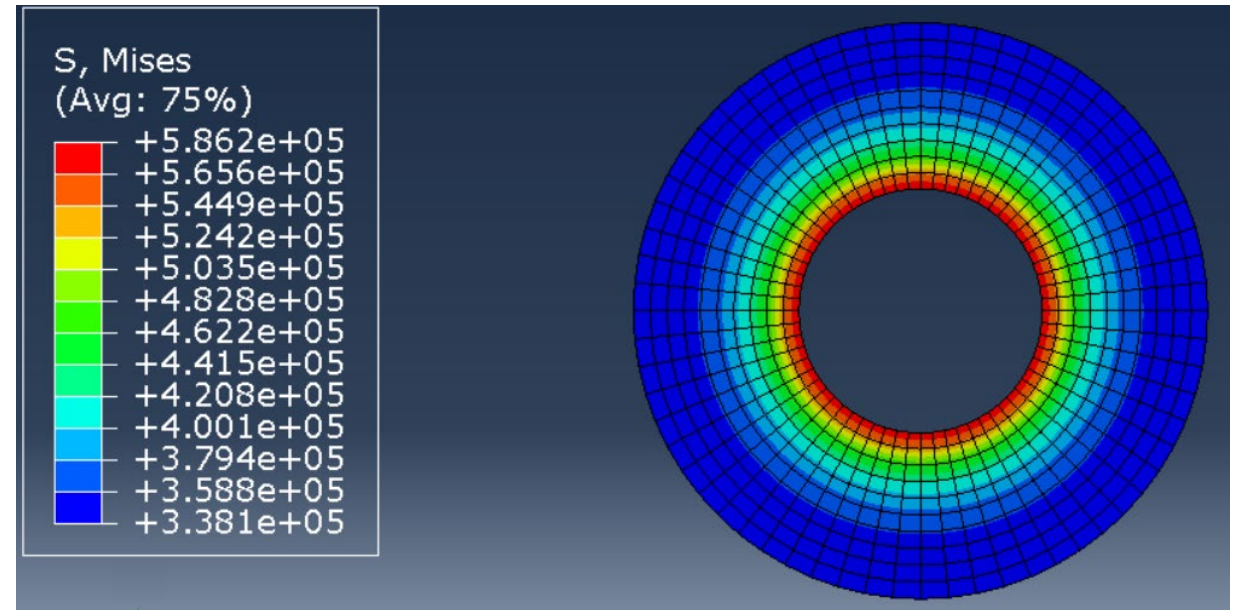
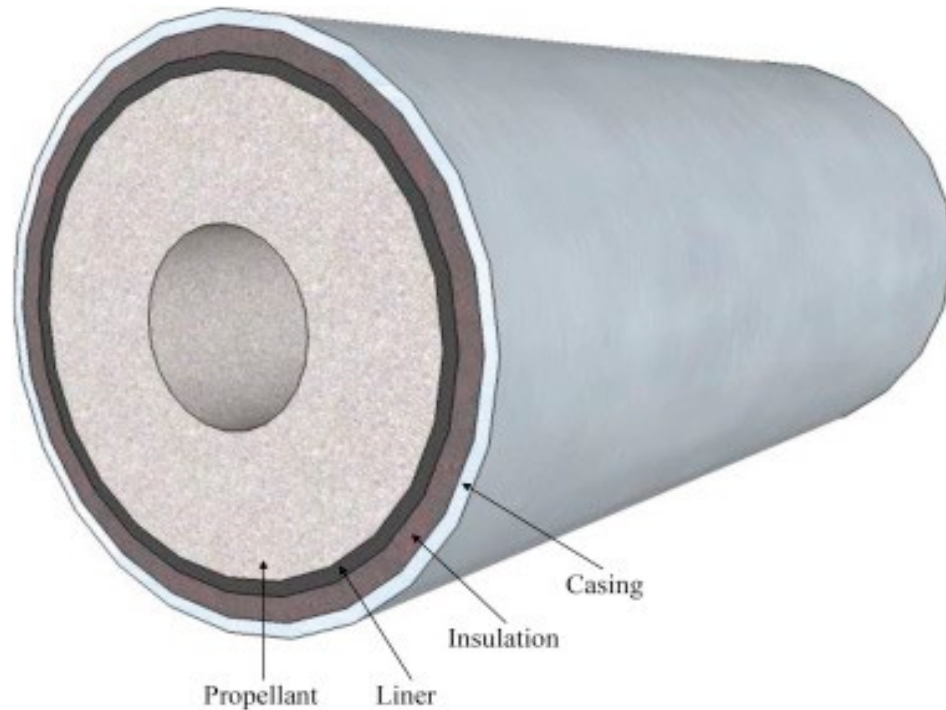
## Background: Technical Approach (Step-by-Step)

1. Use digital image correlation with a **very small speckle pattern** and an optical microscope. Apply the very small speckle patterns to **both** PLI specimens and filled composites.
2. Get the strains (using DIC and an optical microscope) under various loadings and for various parameters (e.g., bonding agent).
3. Use the strains to determine good values for parameters of a “traction separation law”.
4. Use the traction separation law in finite models of experimental specimens.
5. Use the real experimental specimens to verify the validity of this approach.
6. Find the limits of applicability of this approach through further experimentation.



# Activities

# Activities: Development of Solid Rocket Motor Finite Element Model

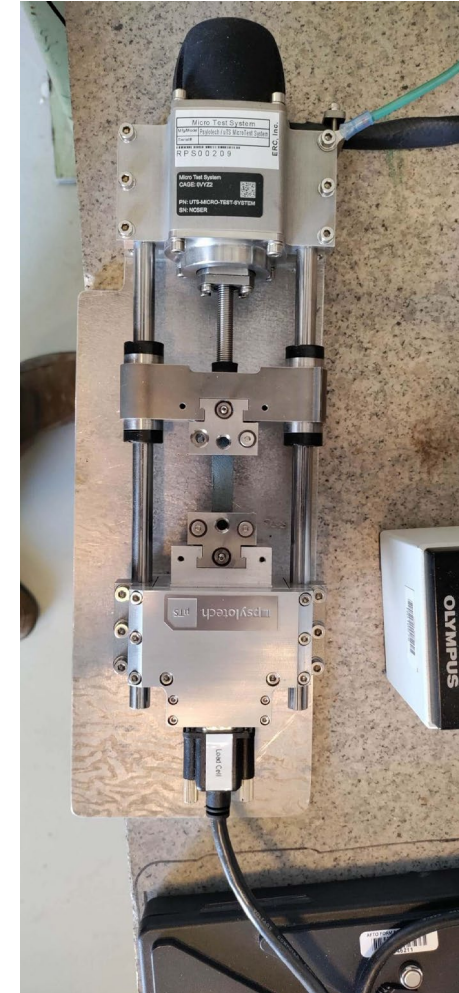


A solid rocket motor finite element model was developed for later use. The constitutive models incorporating cohesive zone modeling can be “dropped” into the model when they are developed.

# Activities: Initial Equipment Adjustments

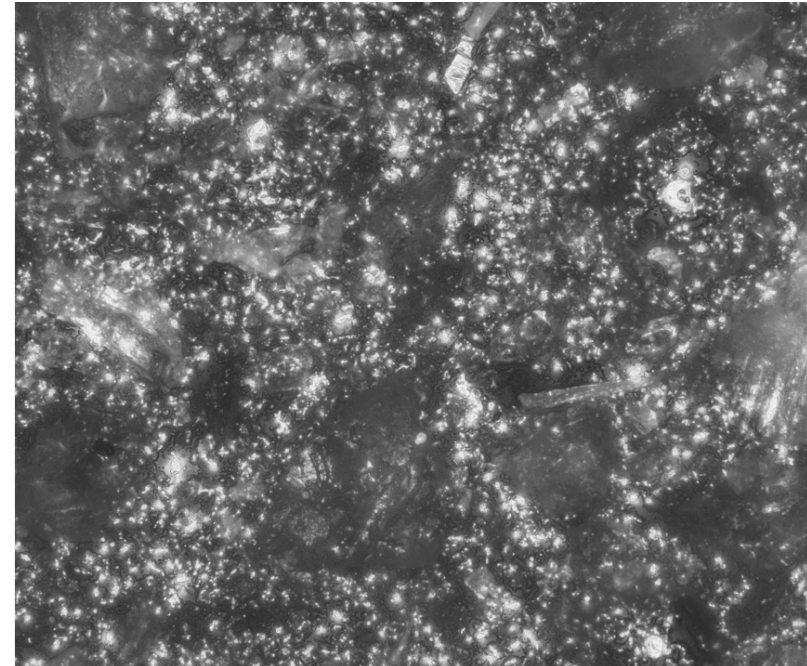
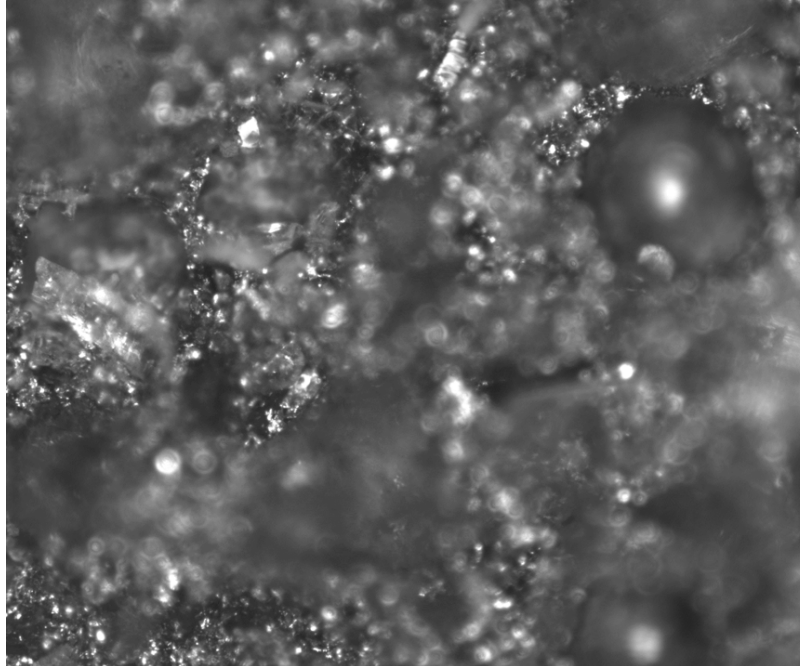


The original microscope setup employed a 10X microscope objective, which was not optimal – by modifying the tensile tester stage so that longer, thicker specimens could be used, the standoff distance for the stage was increased. The result was that 20X and 50X microscope objectives could be used.



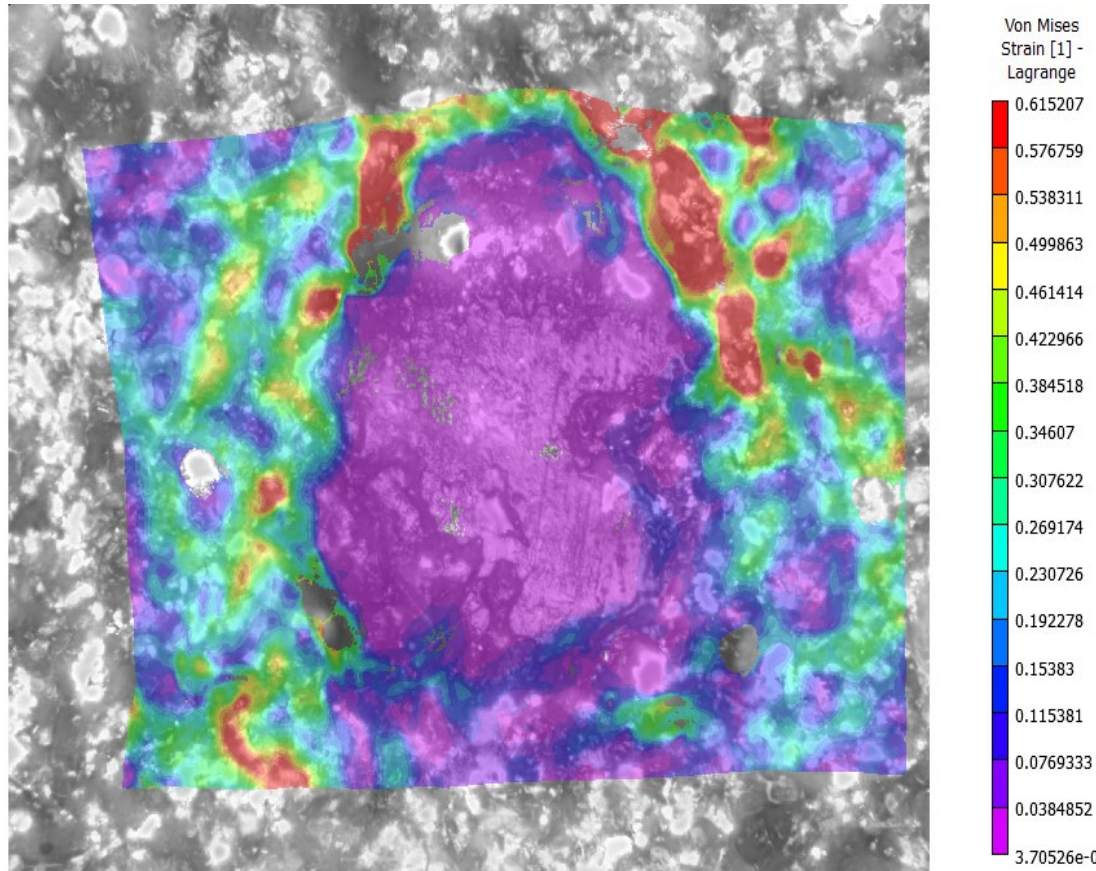


# Activities: Blurring Reduction (Compositing) Algorithm



Because of the narrow depth-of-field for optical microscopes (compared to the surface roughness of propellant), a blurring reduction algorithm was needed to obtain clear pictures for subsequent analysis.

# Activities: Compositing Algorithm



Strain field around large particle in inert HTPB-bound composite propellant (20x magnification)

At 20X, the depth of field was not adequate to capture all of the image clearly. Since smoothing the specimen more would cause potential problems, a *compositing algorithm* was developed:

- Specimen images were captured at fifteen different elevations;
- For each elevation, a weight was assigned to each pixel based on its blurriness (sharper = higher weight);
- For each pixel, the overall greyscale value was the sum of the fifteen weighted pixel values.

Note: At 50X, this approach did not work well.

# Activities: Dewetting and Strain Fields Captured Successfully

## Initial capture and analysis

of images and strain fields surrounding a large particle.

Issues:

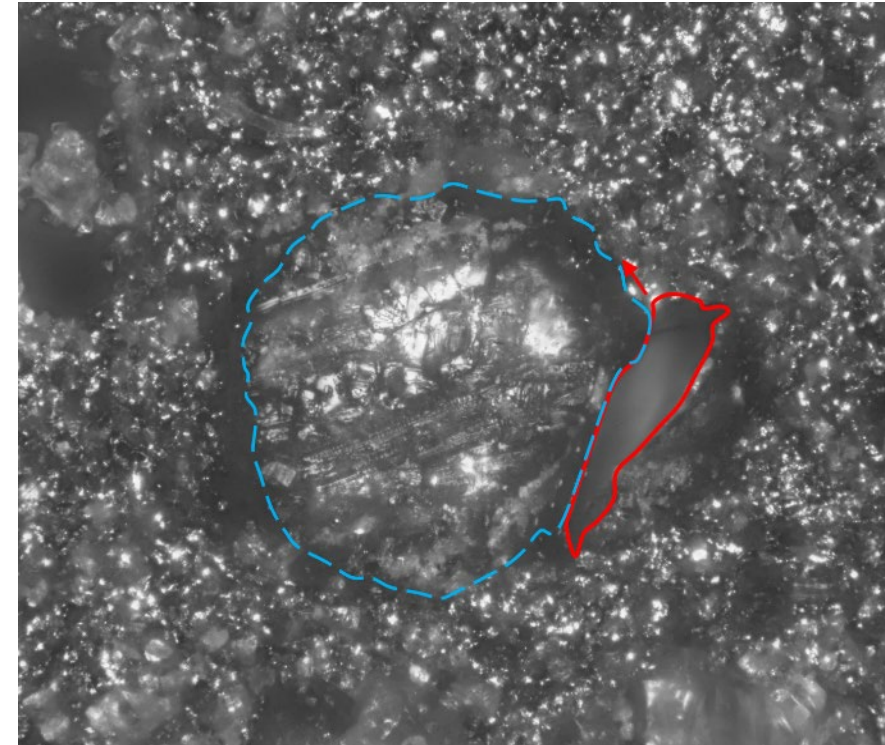
- Decorrelation right where you want it least;
- Real interfaces are not sharp, but models such as cohesive zone models presume two distinct materials separated by a sharp interface.

Possible solutions:

- The particles are all unique, so look for one with sharper interfaces.

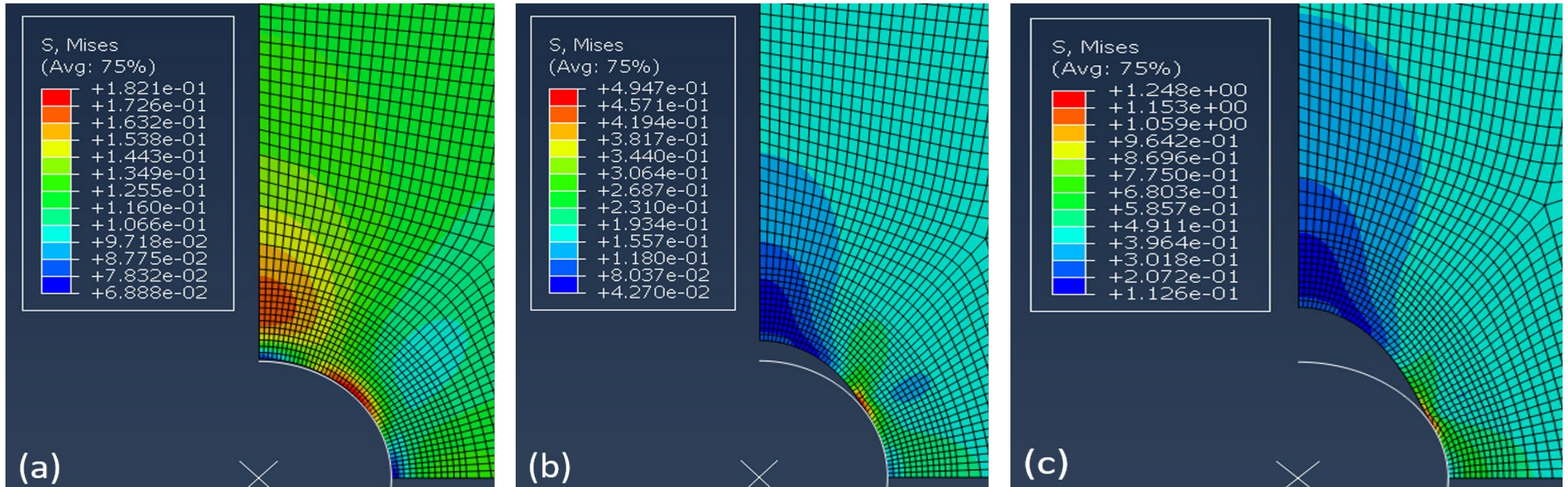
## Subsequent attempt

- Two image sequences of a large particle were captured; these use 10-20 images at progressive levels of specimen loading.



Crack along grain-polymer interface at 11mm specimen extension. Grain boundary (dashed blue line), crack edge (solid red line), and crack growth direction (red arrow) shown. Note also the dark region directly to the left of the grain, which experiences high strain but does not fully separate into a crack as on the right.

# Activities: Modeling the Interfacial Separation



Results of dewetting simulation for composite HTPB matrix surrounding a single rigid large particle (semicircle at bottom with X at center). Particle has radius 100  $\mu\text{m}$ ; stresses shown are in MPa. Applied far-field stress  $\sigma_\infty$  increases linearly over simulation time to a maximum of 0.423 MPa (about 60 psi).

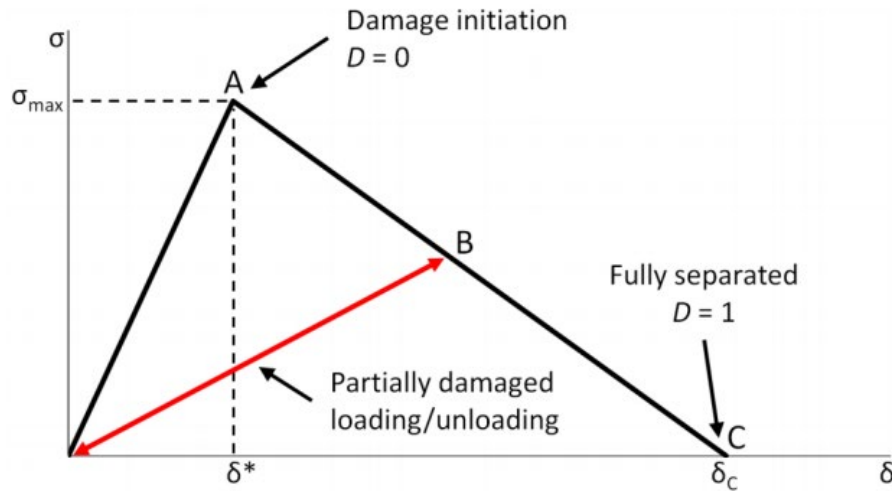


# Questions?



# Backup Slides

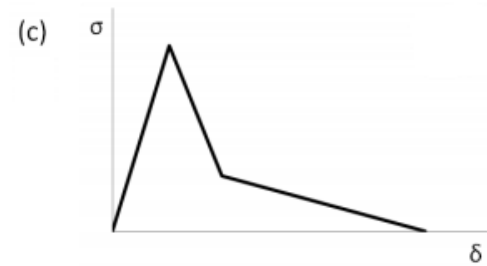
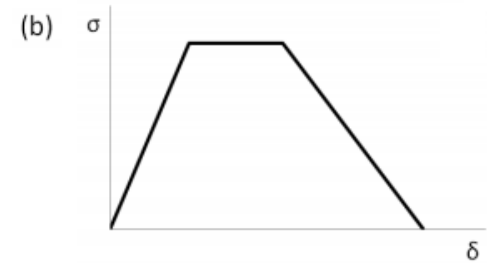
# Different Shapes for the Traction-Separation Law



**Bilinear traction-separation law for cohesive zone elements.(Krieger 2014)**

This “bilinear” traction separation law is composed of two lines – and is very popular, because it is characterized by only three parameters (one traction and two separations).

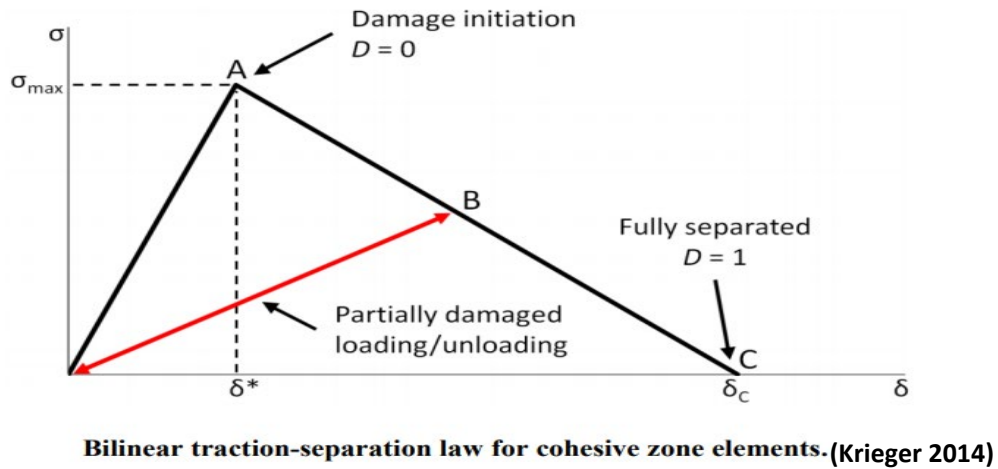
In reality, the “shape” of the separation law should match the actual conditions, so if the bilinear law is too simple, other shapes are also possible.



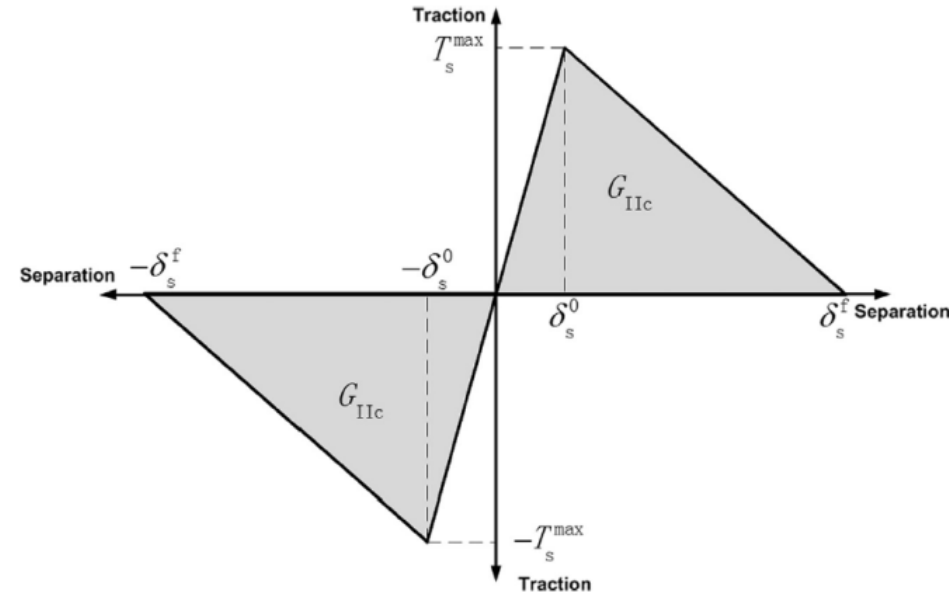
More complex shapes for the traction separation law include (a) exponential, (b) trapezoidal, and (c) trilinear

The simplest reasonable model will be used to model the interfacial separation phenomena.

# Different Traction-Separation Laws for Different Situations



This traction separation law is usually used for “Mode I” loading (pulling apart of two materials)



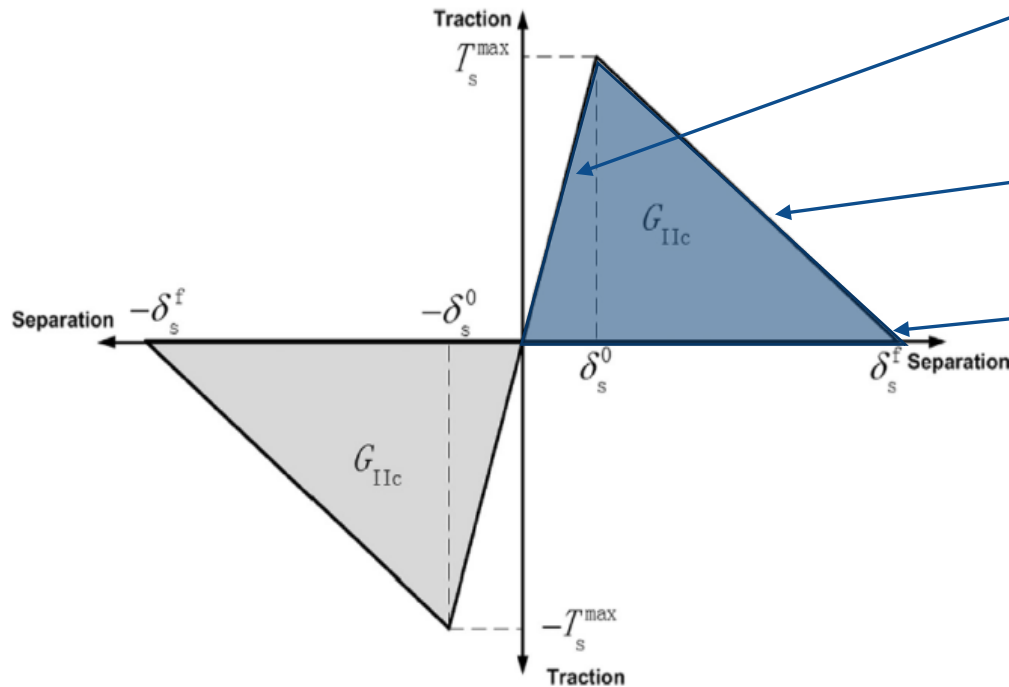
In shear, a similar law may apply – and in general, both modes *may* be combined using superposition for mixed mode loading.

For mixed-mode loading, things can get more complicated.

# If the particle matrix interfacial strength is so important, how do you model that?

## Cohesive zone modeling (CZM)

- The simplest traction separation law:
  - As you pull on an interface, it gets wider, but it also takes progressively more force to apply additional deformations.
  - After a certain point, the load reaches a peak, then begins to drop as damage takes place.
  - Once the load drops to zero, the two materials are separated.
- Separate sets of parameters will be needed for different modes of separation (tension vs. shear, for example)
- Getting the parameters from experimental data could prove difficult.
- More complex traction separation laws can be used if more fidelity is desired – a plateau can be implemented, for example.



Simple traction separation law. The area shown in blue is equal to the critical energy release rate from fracture mechanics