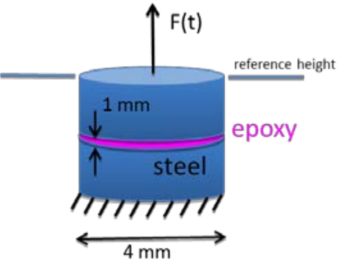
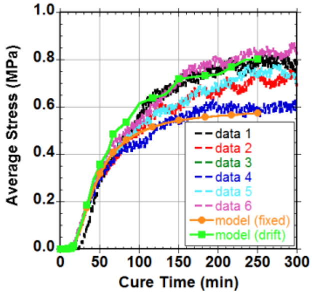
SAND2014-1972P

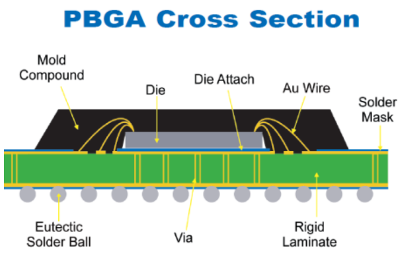
Exceptional service in the national interest

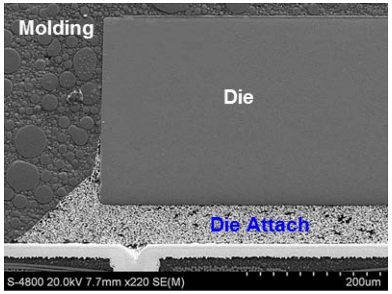
Research/Development





Applications





Problem Solving

Closely Coupled Experimental and Modeling Efforts on Polymer Structural Response: Research to Engineering Support

Jamie M. Kropka, Sandia National Laboratories, Albuquerque, NM

Materials Department Graduate Seminar, New Mexico Tech, March 14, 2014

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Modelers

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Sandia National Laboratories

Overview of the Talk

- What are polymers?
- How are polymers used at Sandia National Laboratories (SNL)
- Complexity of polymer structural response
- Approaches to model polymers, our vision and current tools
- In-progress adhesion work illustrating the collaborative relationship between experimentalists and modelers

Polymers: What are they?

Long chain-like molecules (macromolecules)

Synthesized from smaller molecules (monomers) during polymerization reaction

May be Linear (long threads), branched or crosslinked

May be amorphous, crystalline or some of each

“Thermoplastics”: not crosslinked (e.g., polycarbonate)

“Thermosets”: crosslinked systems (e.g., epoxies)

How are Polymers Used at SNL?

- Encapsulants for:
  - structural integrity
    - impact
    - vibration
  - high voltage isolation
- Adhesives or Underfills for:
  - bonding materials
  - mounting surface components
- Printed Circuit Boards:
  - orthotropic composites

thermosets

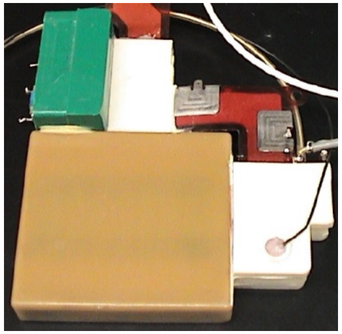
- Foams for:
  - energy dissipation
  - light constraints

- Plastic Parts for:
  - injection molded pieces

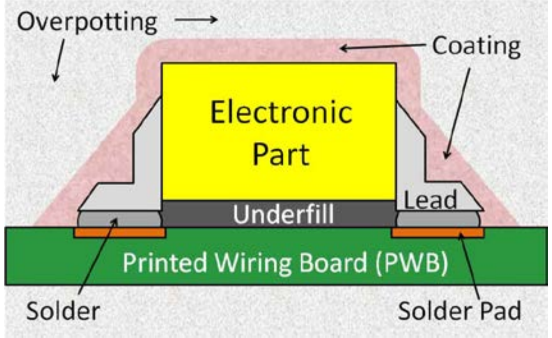
thermoplastics

- Gaskets and O-rings for:
  - sealing cavities
- Cushions, Pads, Coatings for:
  - stress relief
  - damping

elastomers

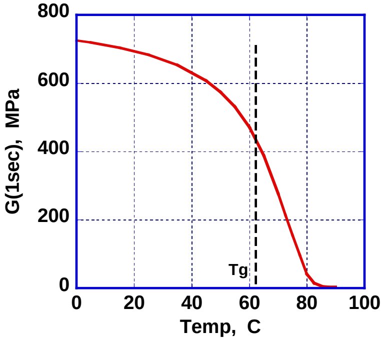


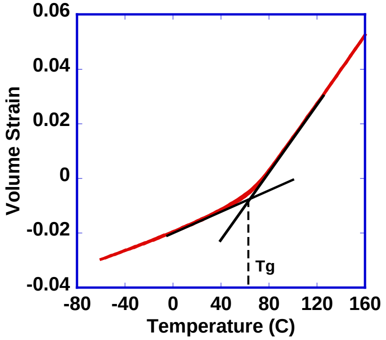
- Optimal use of polymers is not always obvious
- Poor choice of polymers can cause premature failures
- Modeling is important
- Must understand materials to represent them in models

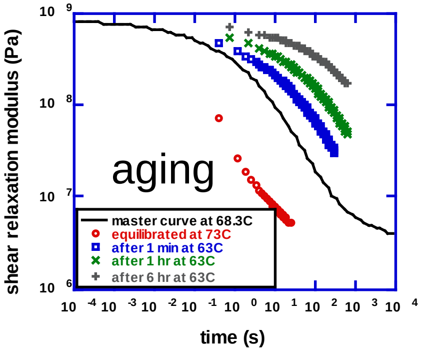


Polymers Are Complex Materials

They respond differently than metals and ceramics







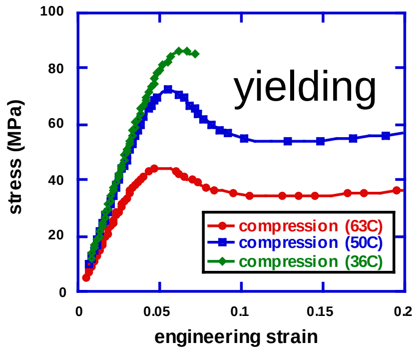


exhibit a glass transition:

- shear modulus can change by 2-3 orders of magnitude
- CTE can change by factor of 3

time dependent and nonlinear:

- relaxation rates vary with temperature and load

Behavior depends on thermal and strain histories

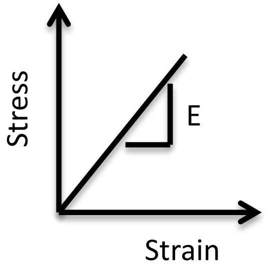
Performance predictions must be able to capture the full range of behavior for general thermo-mechanical loadings from manufacturing to failure.

- must be extensively validated
- computationally tractable

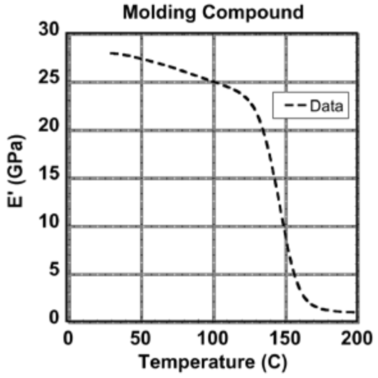
Evolution of Constitutive Representation of Polymers

Linear Elasticity

E, ν, α



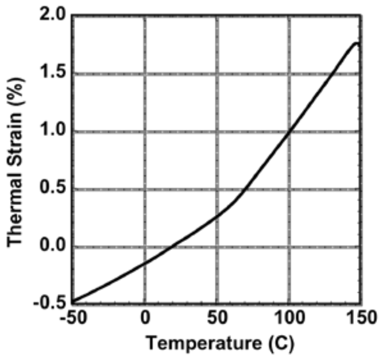
+ temperature dependencies



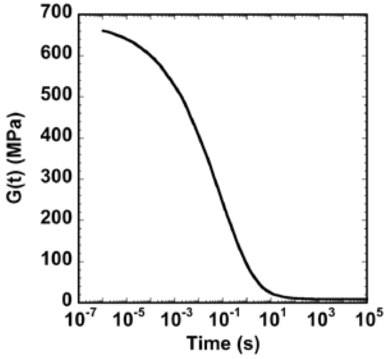
does not capture the physics of the material

Linear Viscoelasticity

Glass Transition



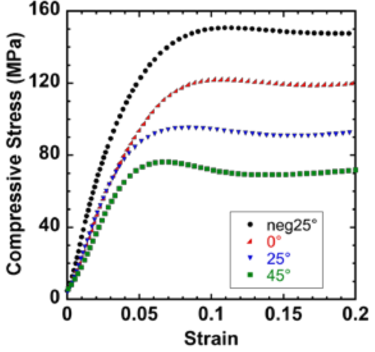
Stress Relaxation



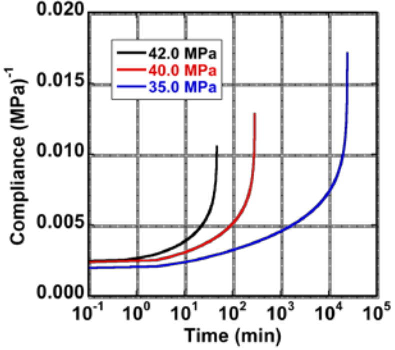
captures history dependence BUT misses "yield" or nonlinearity

Nonlinear Viscoelasticity

Yielding

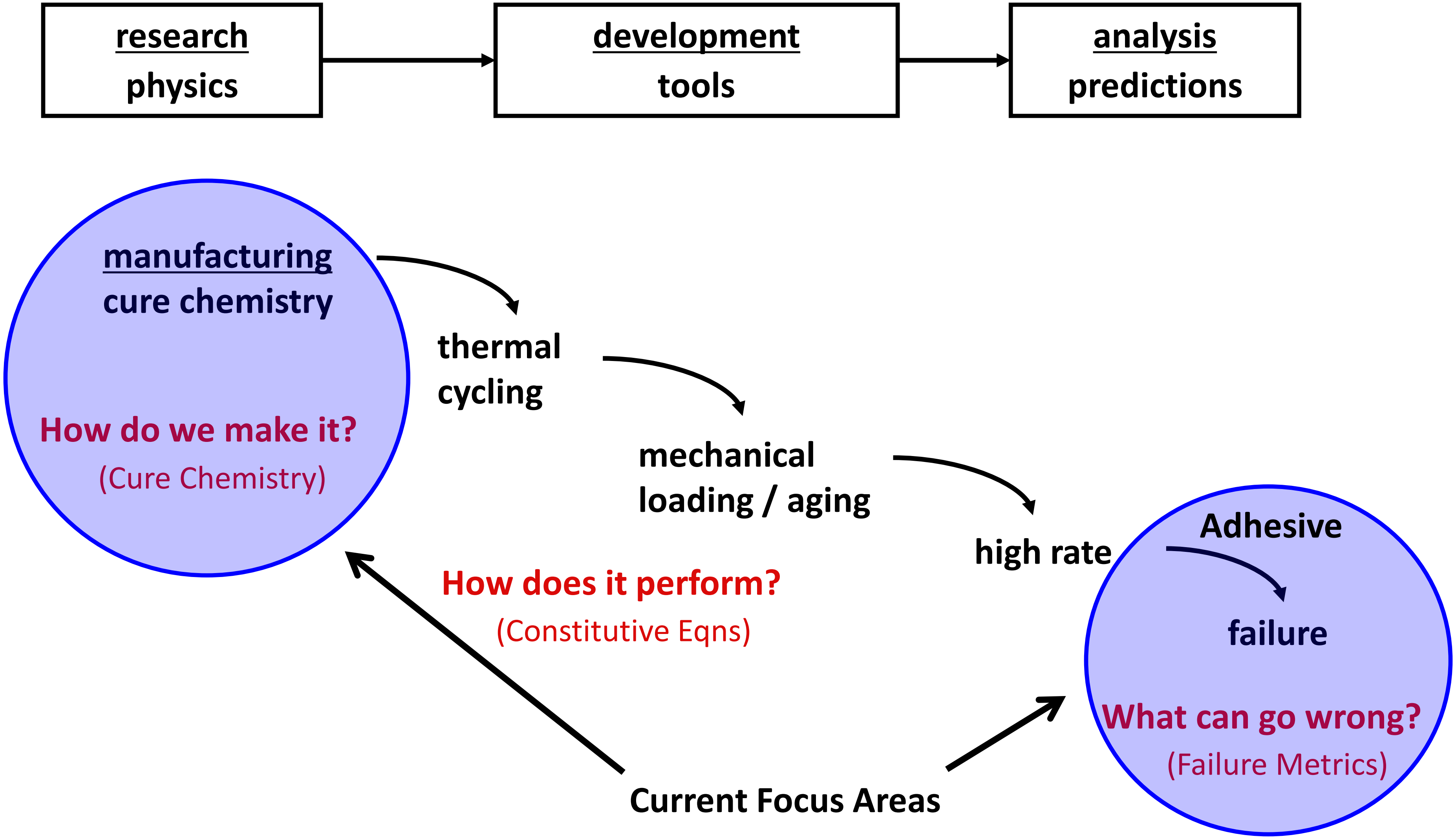


Creep



Our Vision: Validated Model-Based Life-Cycle Engineering

Polymer Nonlinear Viscoelastic (NLVE) Model



research
physics

development
tools

analysis
predictions

manufacturing
cure chemistry

How do we make it?
(Cure Chemistry)

thermal
cycling

mechanical
loading / aging

high rate

How does it perform?
(Constitutive Eqns)

Adhesive

failure

What can go wrong?
(Failure Metrics)

Current Focus Areas

Brief History of SNL Polymer Nonlinear Viscoelastic Model

Potential Energy Clock (PEC) Model[1]

Thermodynamically consistent nonlinear viscoelastic model capable of predicting glassy polymer behavior such as volume/enthalpy/stress relaxation, physical aging, creep and "yielding"—zero parameter non-linear theory

Challenging to calibrate and computationally intensive

↓

Simplified Potential Energy Clock (SPEC) Model[2]

Phenomenological but retained comparable predictive capability as PEC

Easier to parameterize, less computationally intensive, and more suitable engineering tool for design analyses

↓

PEC or SPEC Cure Model

Additional terms to account for effects of cure shrinkage and changing reference state

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[1] Caruthers, J. M.; Adolf, D. B.; Chambers, R. S.; Shrikhande, P. *Polymer* **2004,** 45, (13), 4577-4597.

[2] Adolf, D. B.; Chambers, R. S.; Neidigk, M. A. *Polymer* **2009,** 50, (17), 4257-4269.

Hierarchy of Polymer Material Characterization for Modeling Nonlinear Viscoelasticity (NLVE)

Other Options not Possible

Bare Bones Approach

Measure:

1. calorimetric Tg
2. filler volume fraction

Model Parameterization:

Estimate NLVE response based on universal properties and rule of mixtures approach

Limitations/Potential Errors:

- Must be rigid fillers (e.g., alumina, silica, mica…)
- Breadth of relaxation spectra
- Nonlinear material clock

Material Evaluations

Quick and Dirty Approach

Measure:

1. filler volume fraction
2. thermal strain versus temperature
3. elastic shear modulus versus temperature

Model Parameterization:

Estimate NLVE response based on universal properties and rule of mixtures approach. Compare predictions to data. Ability to tweak relaxation spectra and prefactors to better match predictions to data.

Limitations/Potential Errors:

Lack definition of clock for nonlinear relaxations

Critical Encapsulants/Adhesives

The Whole Shebang

Measure:

1. filler volume fraction
2. thermal strain versus temperature
3. elastic shear modulus versus temperature
4. compressive stress-stain through yield at multiple temperatures
5. shear mastercurve
6. creep at multiple temperatures and stress levels
7. bulk modulus versus temperature

Model Parameterization:

Populate material specific SPEC NLVE model

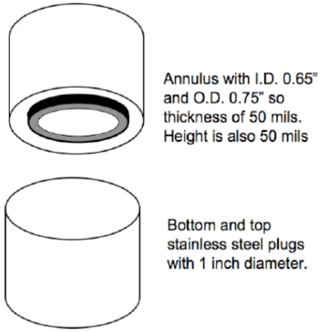
Advantage:

Model can now predict yielding AND (physical) aging with more confidence

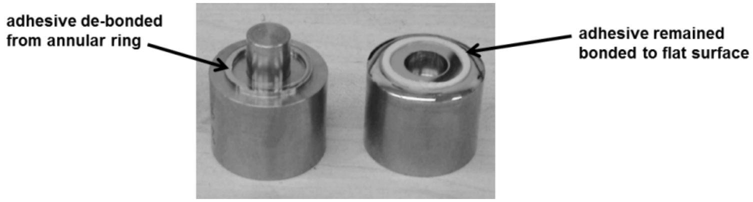
Role of Residual Stress on the Strength of Adhesive Joints

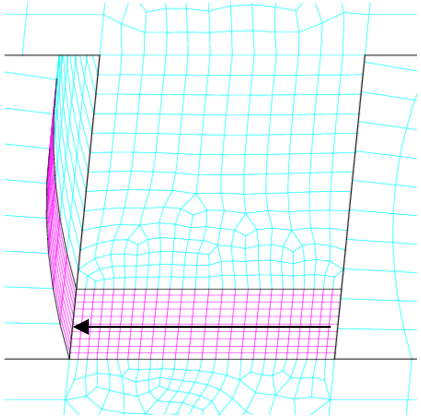
Joint geometry and stress distribution under load

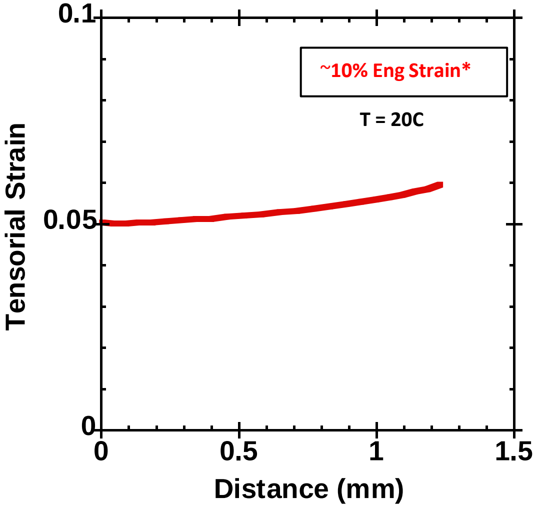
Napkin-Ring (NR)

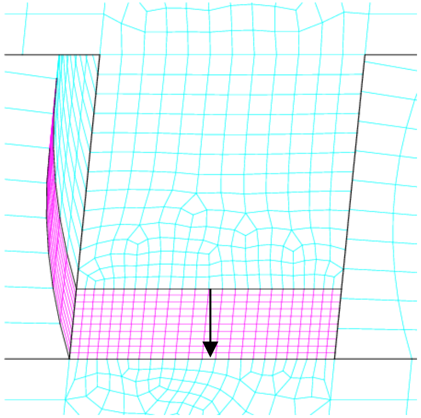


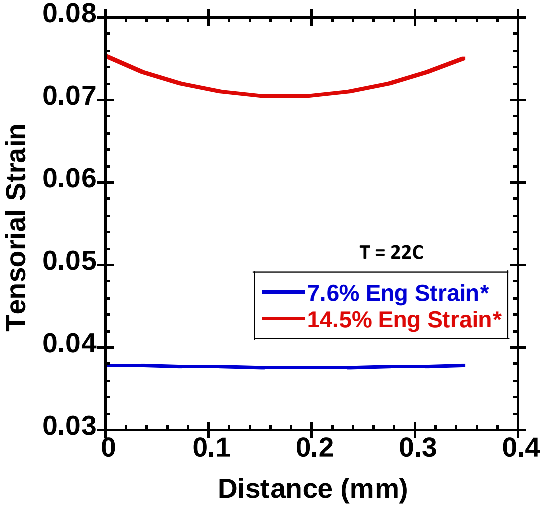
After Failure











The absence of severe strain gradients enables computational assessment of the joint using the Simplified Potential Energy Clock (SPEC)[1] nonlinear viscoelastic (NLVE) polymer model in finite element analyses (FEA)

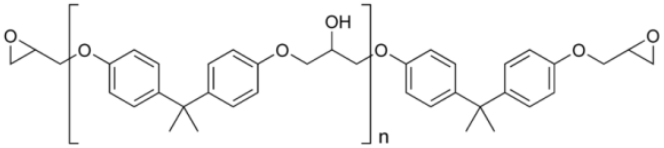
Eng Strain ~ 2(tensorial strain)

[1] D.B. Adolf, et al., *Polymer*, 2009, 50, pp 4257-4269

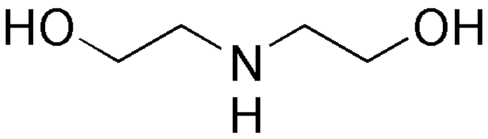
Adhesives used in napkin-ring bonded joints

828/DEA[1]

EPON® Resin 828
Diglycidylether of Bisphenol-A



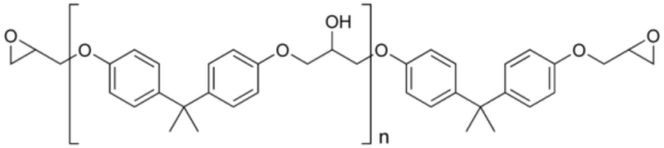
Diethanolamine



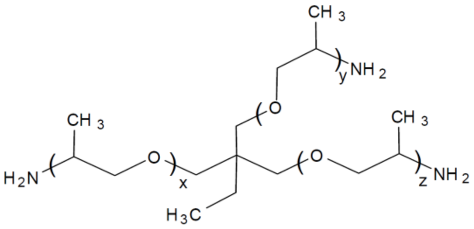
T_g ~ 70C

828/T403[2]

EPON® Resin 828
Diglycidylether of Bisphenol-A



Jeffamine® T-403 Polyetheramine



(x+y+z) = 5-6

T_g ~ 80C

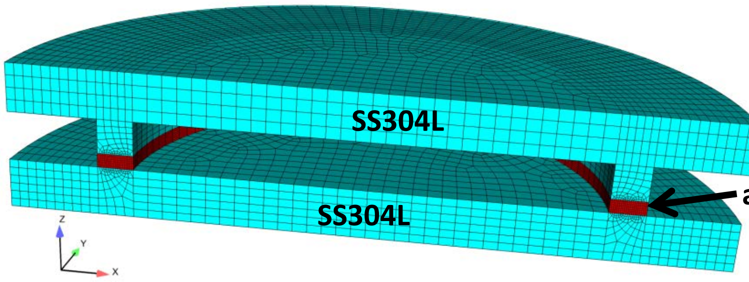
---

[1] Mix ratio, cure and typical properties can be found at: http://www.sandia.gov/polymer-properties/828_DEA.html

[2] Mix ratio (100:43 pbw 828:T403), cure, and more can be found in SAND2013-8681

Macroscopic stress-strain response of joint

½ Symmetry NR

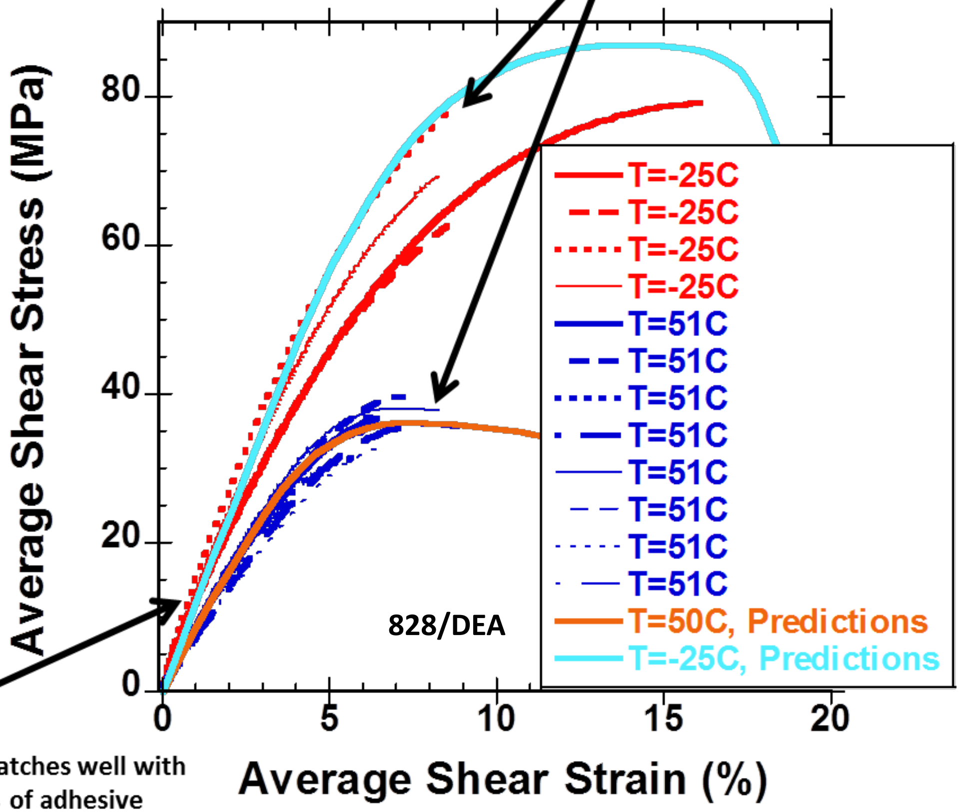


adhesive
0.020” thick
OD = 0.75”
ID=0.65”

Model the test details*:

- Stress free at T=75C
- Cool to T=23C at 0.5C/min
- Rotational displacement at 5 degrees/min

joint failure

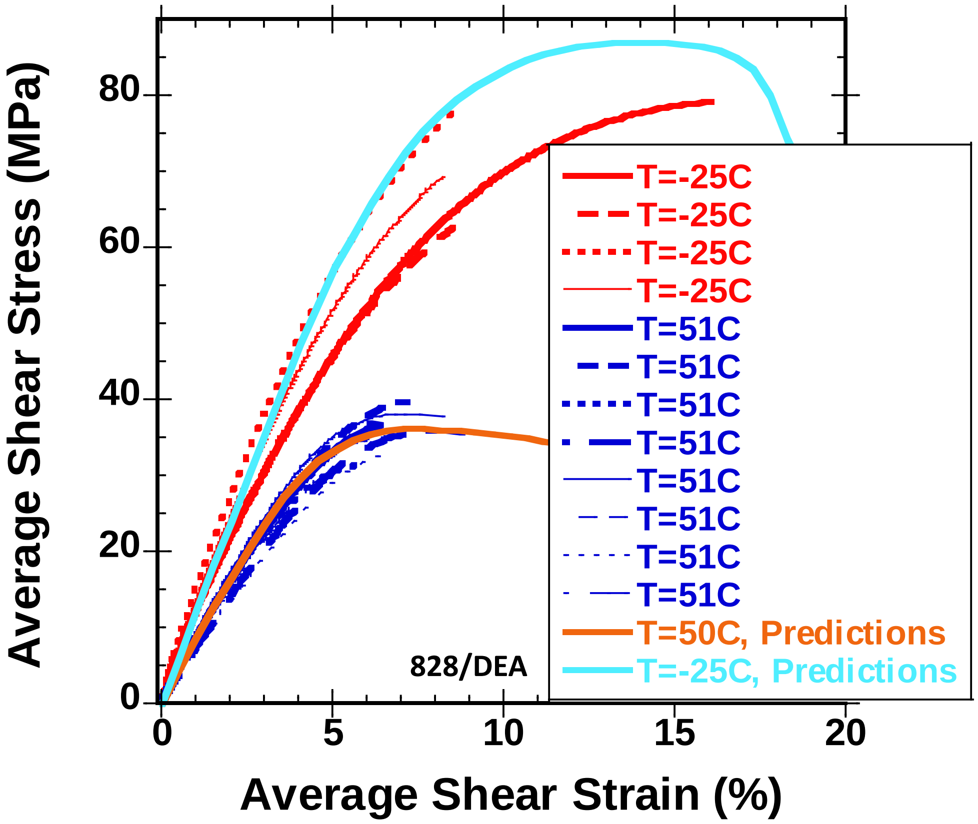


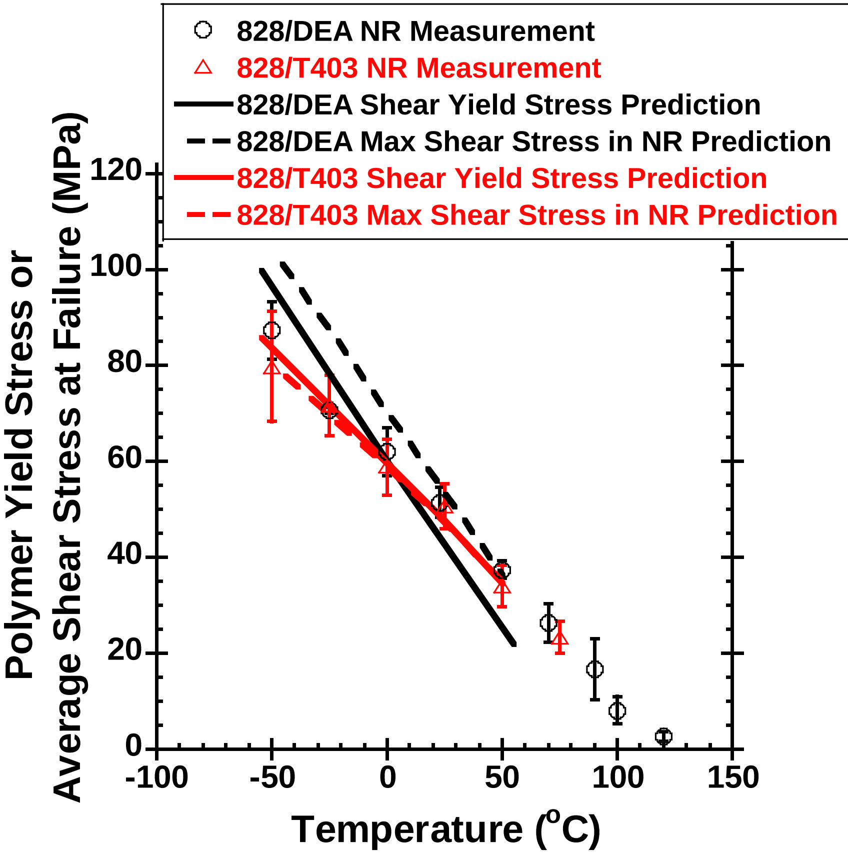
initial slope matches well with shear modulus of adhesive

Predictions track measurements well to joint failure

*Note that not every test detail is captured in the model. For instance, adhesive geometry is assumed square with sharp corners in the model and the adhesive spreads along the flat surface of the bottom of the joint in the experiment.

Temperature dependence of joint failure stress





Experimental failure stress of the joint correlates well with both the predicted shear “yield stress” of the polymer and the maximum predicted shear stress in a torsional loaded NR joint—failure in NR joint is associated with shear yielding in polymer

Do these plots change as the residual stress state in the adhesive changes?

Altering residual stress state in napkin-ring joint

1. Alter width-to-thickness ratio of bond-line

No significant changes in joint strength resolved

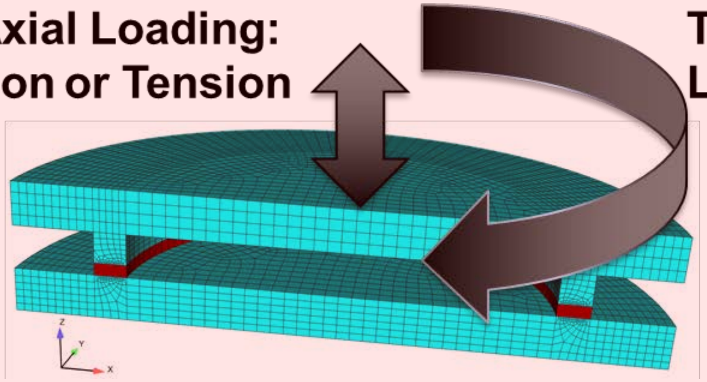
-residual stress predictions were well below the initiation of “yield”

2. Axially fix adherends during processing

No significant changes in experimental joint strength resolved

-even predictions suggested < 20% change in maximum shear stress sustainable in NR joint over experimentally practical limits

3. Combined axial and torsion loading of the joint



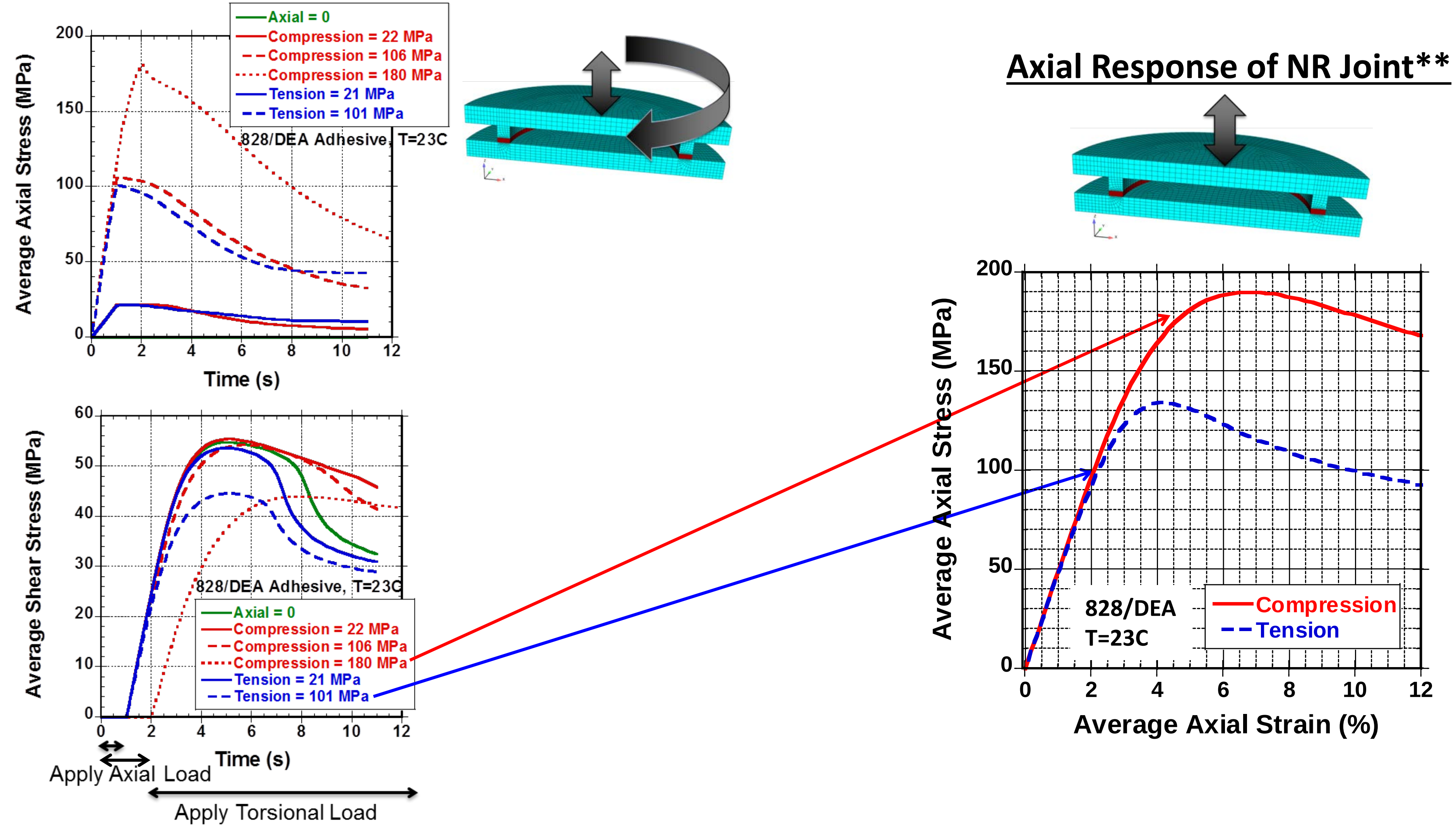
Axial Loading: Compression or Tension

Torsional Loading

Napkin-Ring Cross Section

Axial and Torsion Loading of NR: Predictions

Combined Axial (strain control) and Torsion Load*



Axial Response of NR Joint**

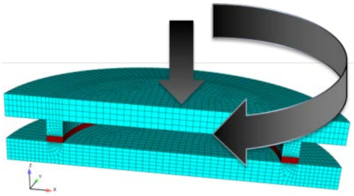
Axial load must approach “yield” of the adhesive in order to reduce the maximum predicted shear stress under torsion

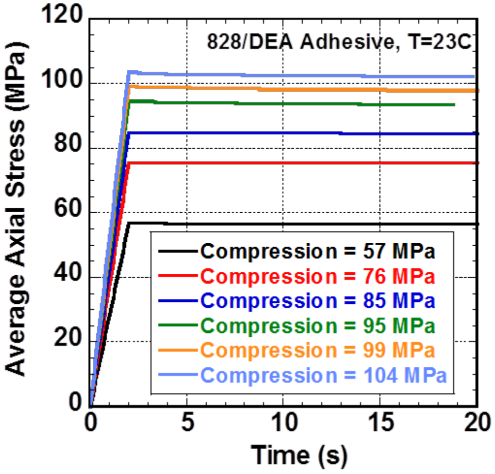
*strain controlled axial deformation in 1-2 s followed by strain controlled rotational displacement at 5 degrees/min

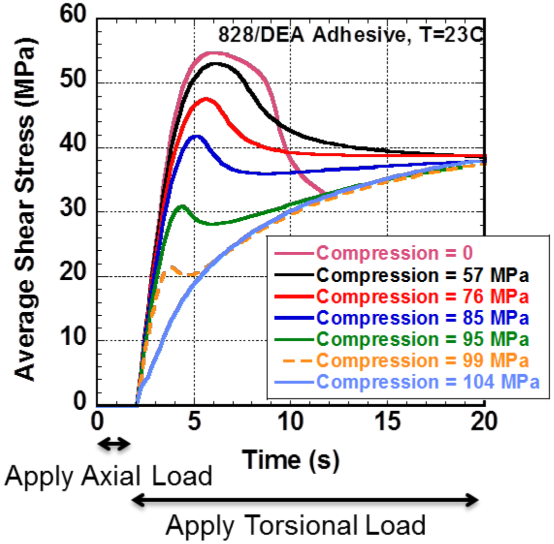
**strain controlled axial deformation at 0.025 (in/in)s^{-1}

Axial and Torsion Loading of NR: Predictions

Combined Compression (stress control) and Torsion Load*





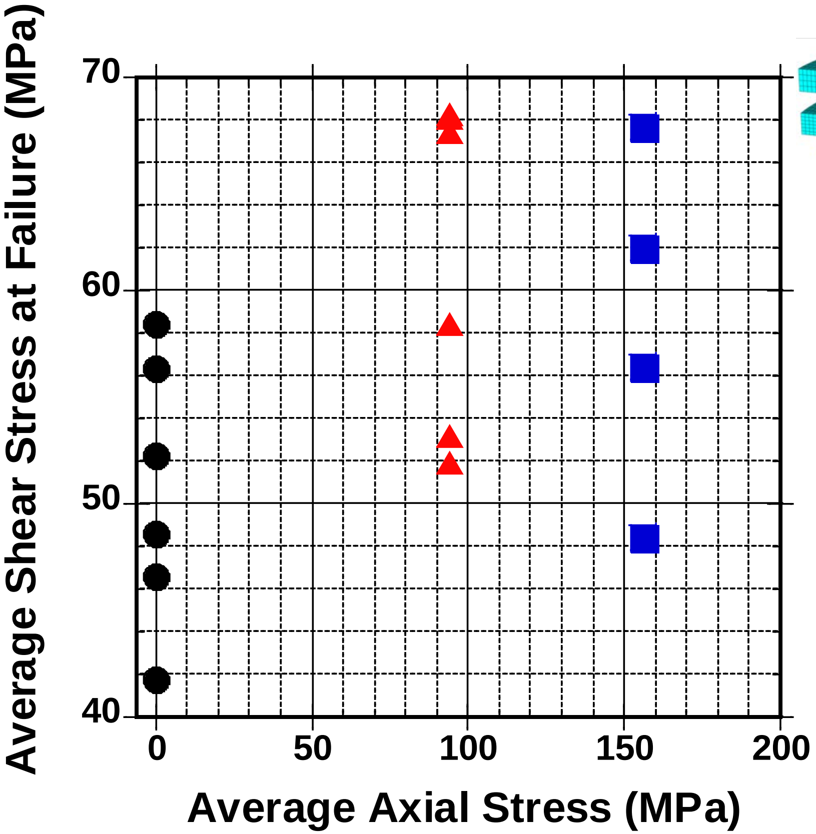


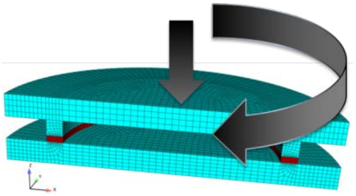
Maximum predicted shear stress under torsion is reduced at smaller compressive loads when axial load is force control

* stress controlled axial load ramped over 2 sec followed by strain controlled rotational displacement at 5 degrees/min

Axial and Torsion Loading of NR: Experiments

<u>Combined Compression (stress control) and Torsion Load*</u>





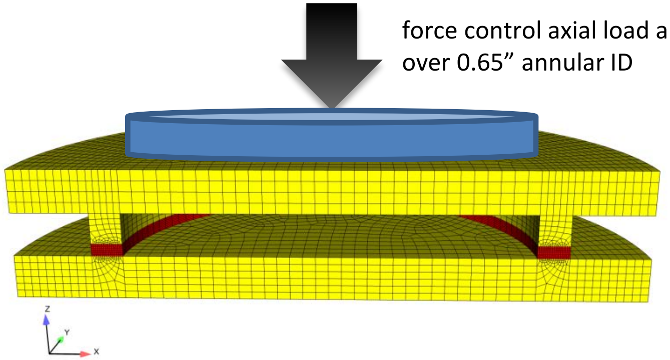
No drop in joint failure stress with compressive loads up to 156 MPa

Why the apparent disagreement between predictions and measurements?

*stress control axial load ramped over 10 seconds, followed by strain controlled rotational displacement

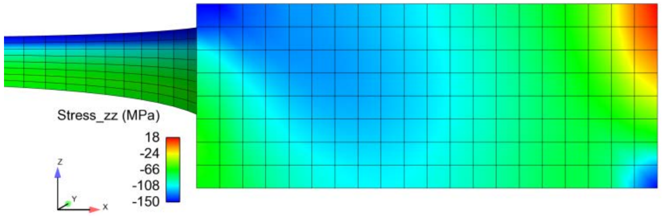
Model sensitivity to the boundary value problem specified

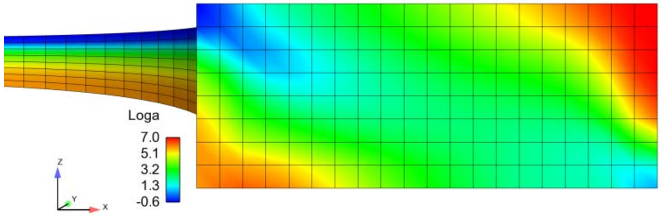
Thin Adherends*



force control axial load applied over 0.65” annular ID

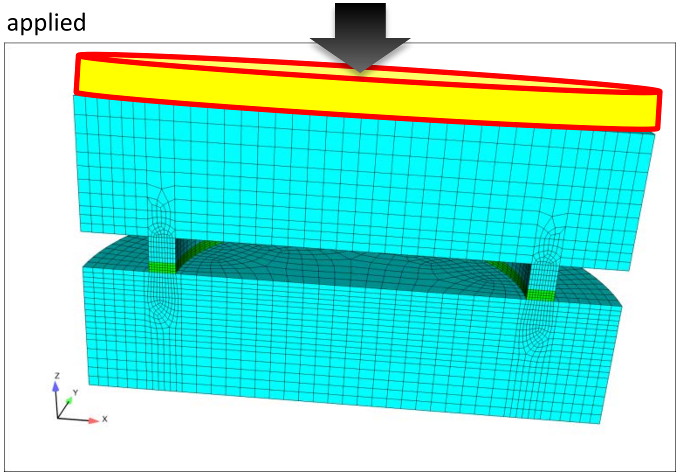
Average Axial Stress: 94 MPa



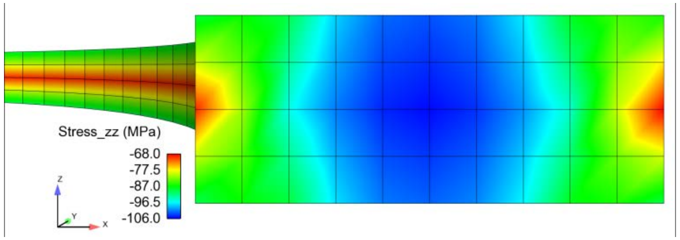


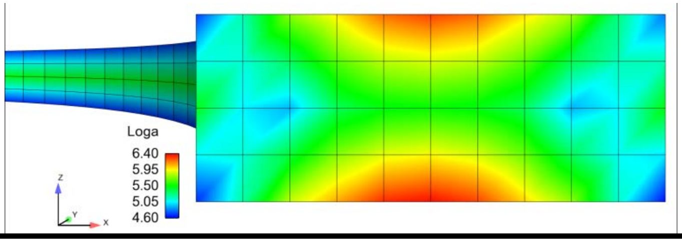
Thick Adherends**

force control axial load applied over entire 1” surface



Average Axial Stress: 94 MPa





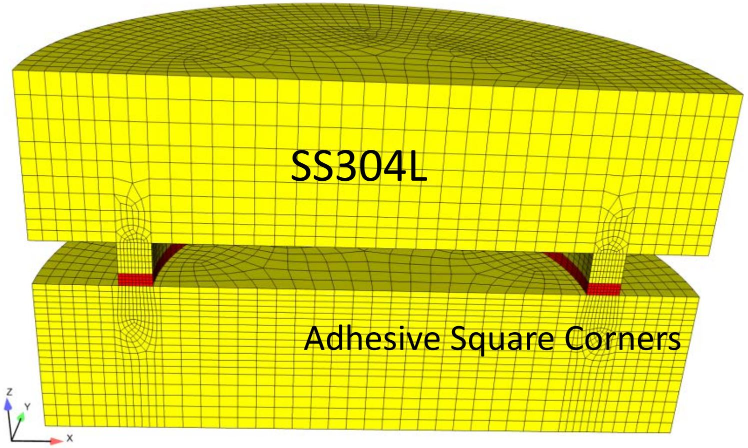
Thin adherend deforms under force control axial load applied to inner annular area of adherend and initiates “yielding” (small Loga) at inner corner of epoxy, unlike experiments

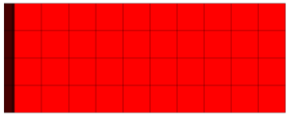
*828/DEA adhesive
**828/T403 adhesive

Details Matter!

Geometric Discrepancy Between Models and Experiments

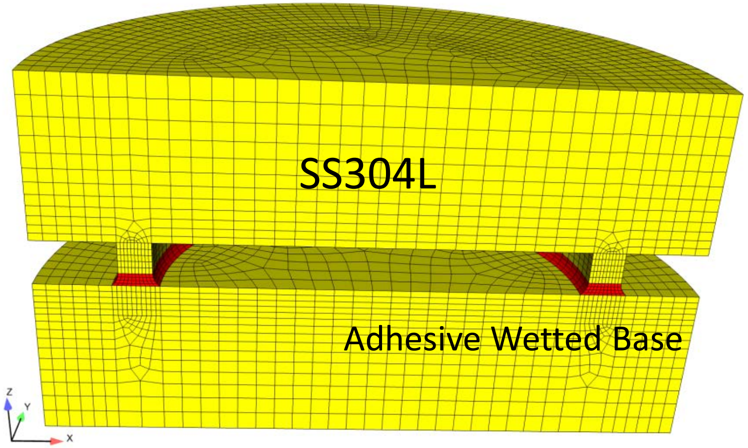
Idealized Geometry in Initial Calculations

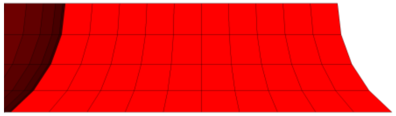




Adhesive Section:
Square Corners

More Realistic Geometry





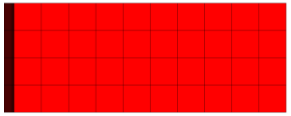
Adhesive Section:
Wetted Base

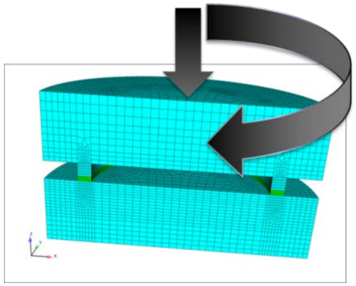
Model the test details:

- Stress free at T=90C
- Cool to T=20C at 0.5C/min

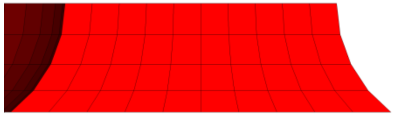
Geometric Sensitivity of Compression and Torsion of NR*

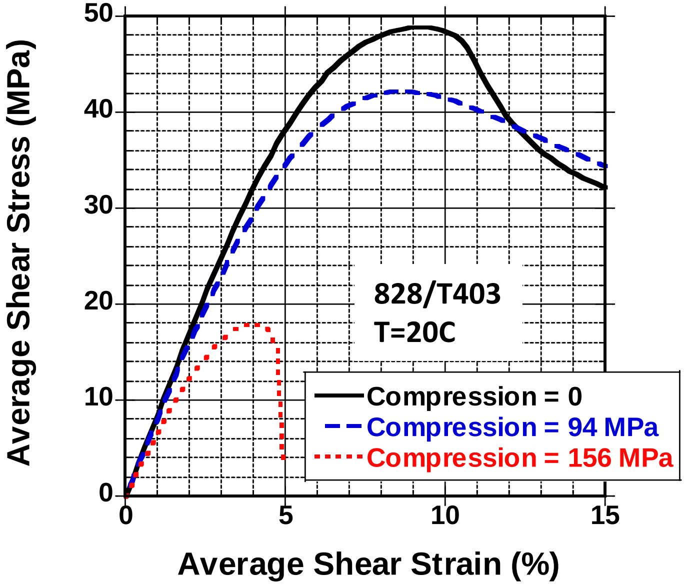
Adhesive Square Corners

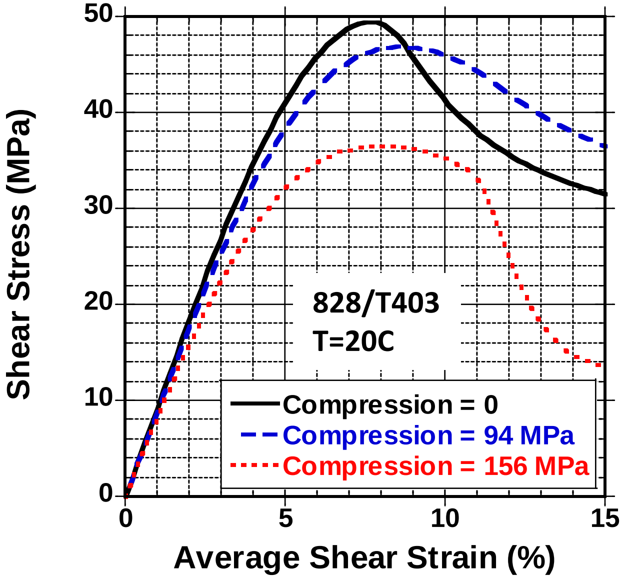




Adhesive Wetted Base





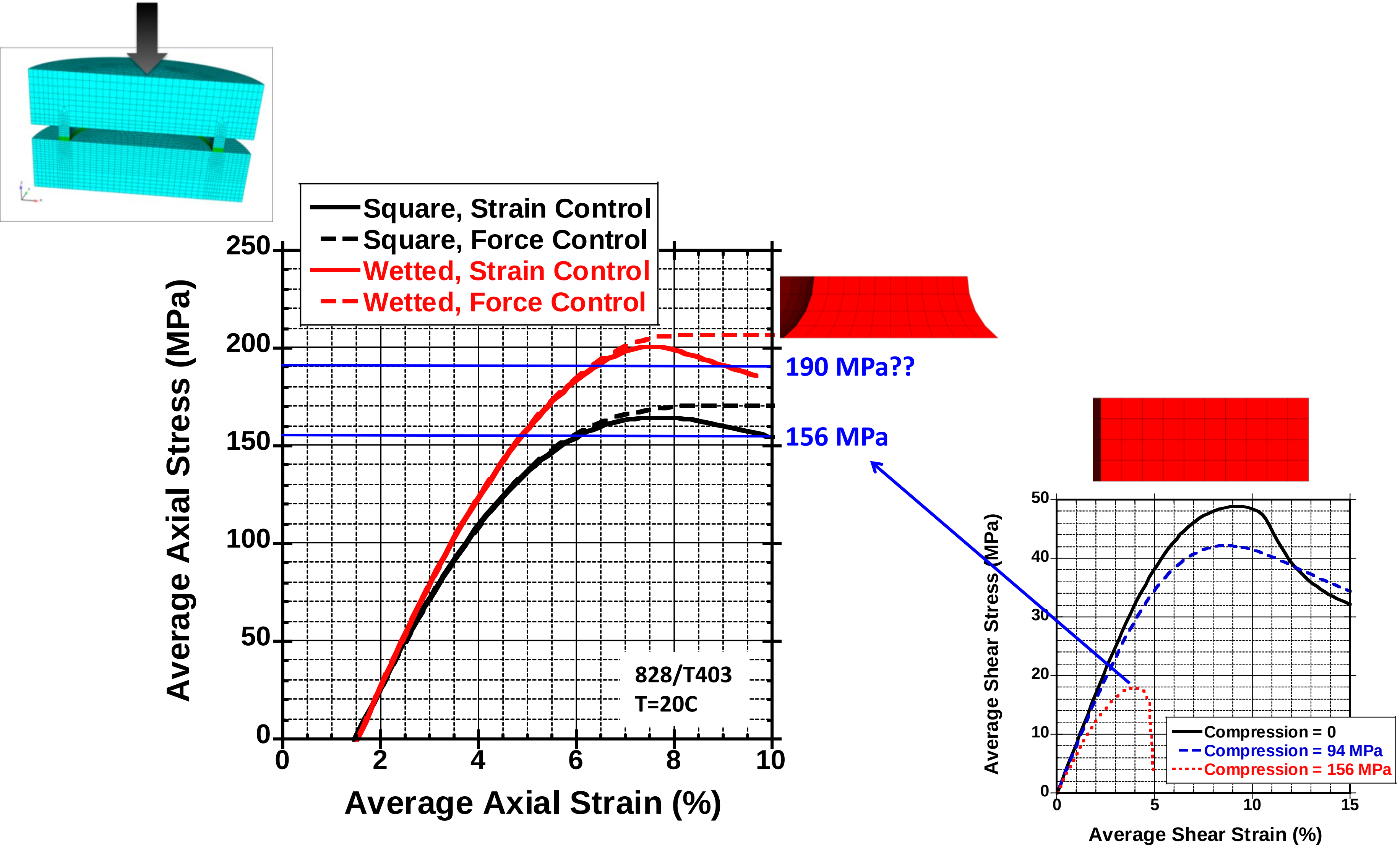


In wetted geometry the torsion response is less sensitive to the compressive load

Details Matter!

*stress controlled compressive load ramped over 10 seconds, followed by strain controlled rotational displacement at 1.31 deg/min

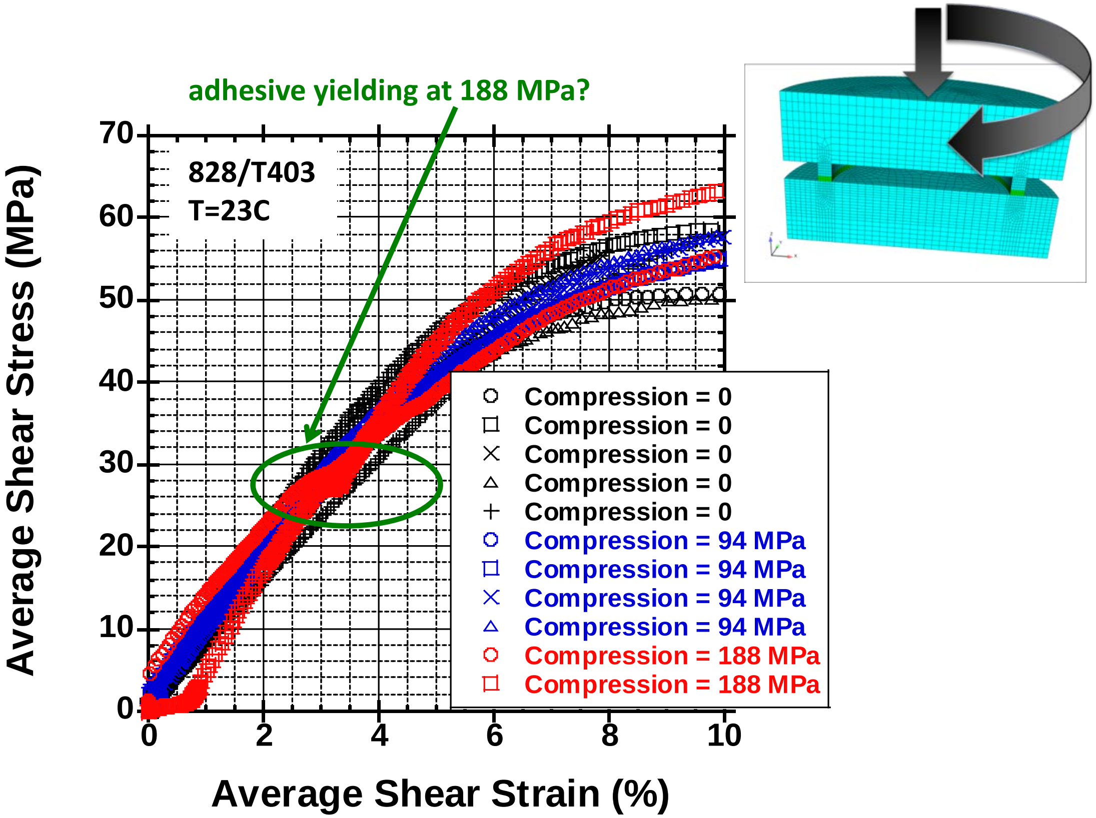
Geometric and Loading Sensitivity of Compression of NR



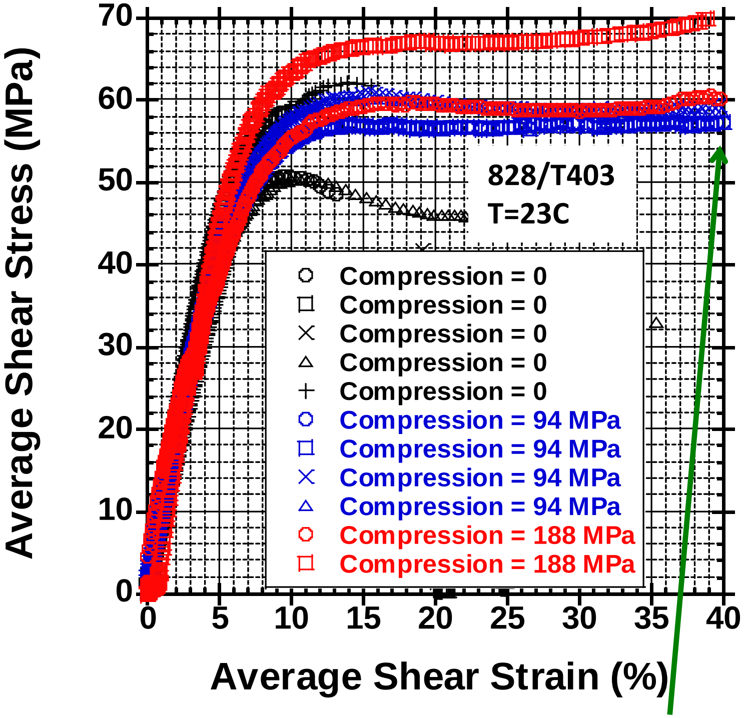
What if a higher compressive load is applied to the experimental wetted geometry before applying torsion?

Axial and Torsion Loading of NR: Experiments

Combined Compression (stress control) and Torsion Load*



adhesive yielding at 188 MPa?



removing sample from instrument reveals that it is bonded after >40% strain at compressive loads!

More questions than answers:

- Does the "kink" at 188 MPa compression lead to damage?—next tests
- Why doesn't the joint fail under the high compressive loads???

*stress controlled compressive load ramped over 10 seconds, followed by strain controlled rotational displacement

Summary

- The absence of severe strain gradients in the napkin-ring joint enables quantitative analysis of the test using polymer constitutive models in finite element meshes. Failure of the joint matches well with adhesive yielding predicted by the Simplified Potential Energy Clock (SPEC)[1] nonlinear viscoelastic (NLVE) polymer model
- Applying an axial load to alter residual stresses before applying a torsional load:
  - Predictions suggest tensile loads on the joint must approach 100 MPa before the maximum predicted shear stress under torsion is significantly affected. Experiments show that joints fail under tensile loads of ~20 MPa.
  - Predictions suggest compressive loads can significantly affect the maximum predicted shear stress under torsion as the compressive load approaches "yield" of the adhesive. Experiments have generated more questions than answers. More to come…

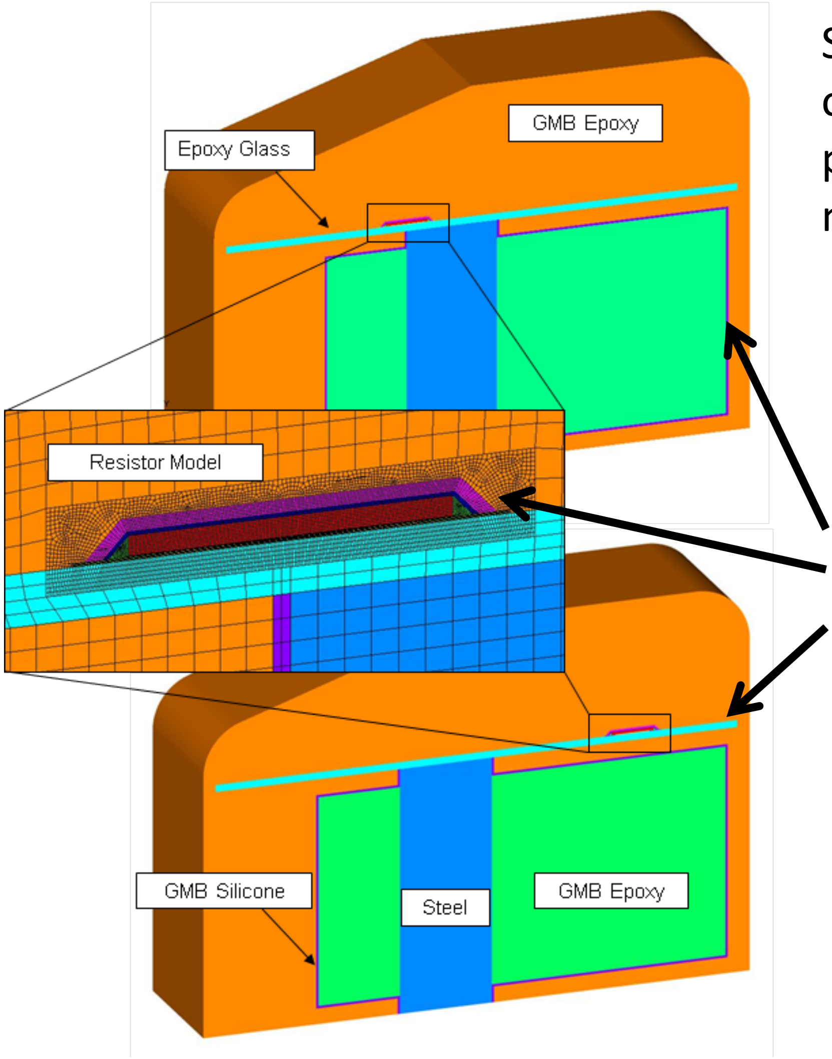
[1] D.B. Adolf, et al., *Polymer*, 2009, 50, pp 4257-4269

Final Remarks

1. SNL has a unique capability in characterizing and modeling the nonlinear viscoelastic behavior of polymers
   a. Methods exist to quickly evaluate new materials and develop constitutive representations (universal polymer model) that enable stress analyses of engineering designs
   b. New capabilities are being developed to represent contributions of cure and further refine failure metrics. These capabilities enable evaluation of problems that could not be addressed before and further refinement in the resolution of others.
2. The teaming that we have in the polymers area allows us to address problems that arise in engineering design more thoroughly than any of the individuals could alone, by leveraging a range of expertise
3. The combination of research, materials characterization, and problem solving efforts we engage in reinforce each other
   a. The development of new capabilities enables further resolution of engineering problems using science-based tools
   b. The inability to fully resolve problem solving efforts often demonstrates lack of capabilities and generates new research ideas
   c. Materials characterization efforts keep us aware of new trends in materials use and/or development and allows to evaluate whether current tools can capture the physics/chemistry that underlie the material responses

Extras

Failure at embedded interfaces



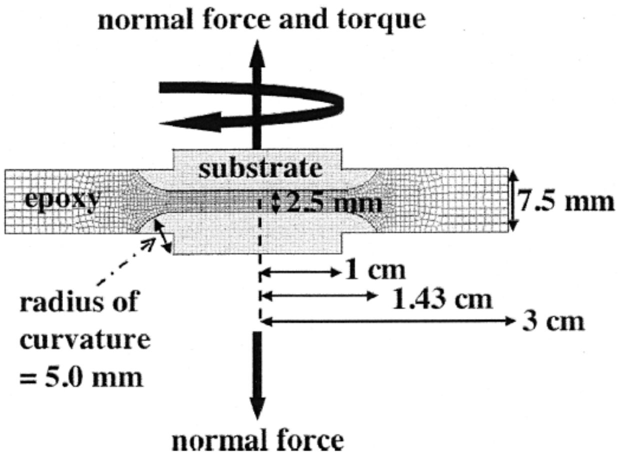
Surface mount electronics and other devices are often encapsulated in epoxy thermosets for protection against high voltage breakdown and/or mechanical shock and vibration

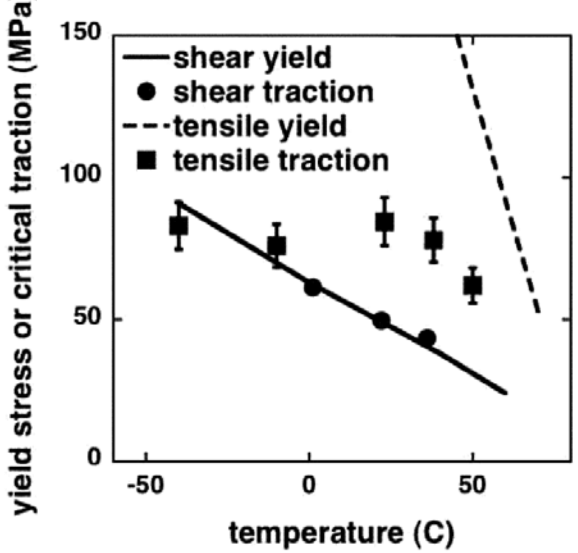
Encapsulation creates interfaces between dissimilar materials embedded within the external surface and maintaining interface integrity is key to protecting the electronic devices

Validated finite element models can predict stresses/strains at an interface, but what is the critical traction at which debonding initiates?

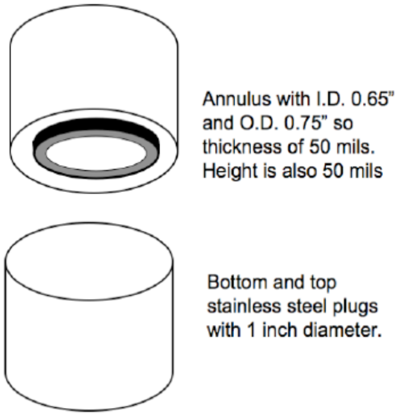
Test geometries to identify critical tractions for initiation of failure at embedded interfaces

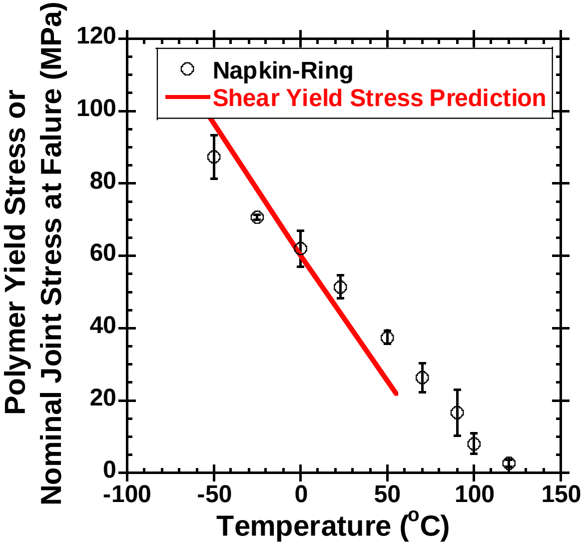
Saucer Geometry





Napkin-Ring Geometry



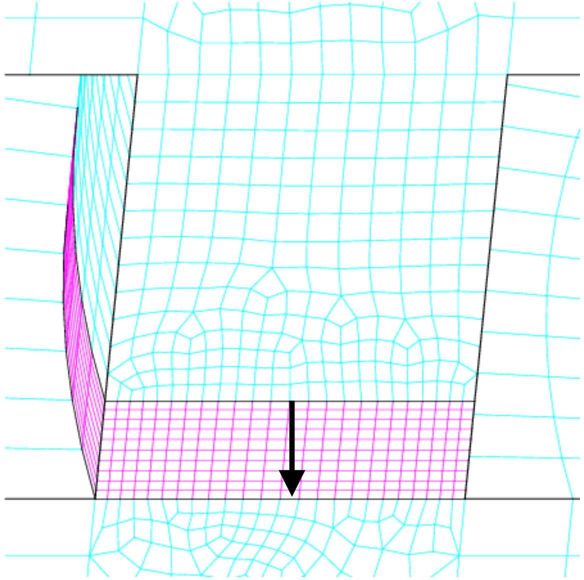


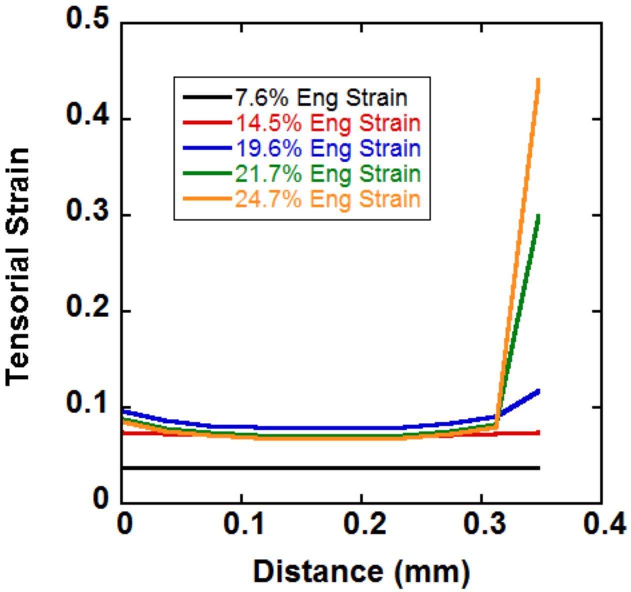
Shear traction at failure tracks the polymer shear yield in both geometries

D.B. Adolf, et al., *J. Adhesion*, 2006, 82, pp 63-92

D.B. Adolf, et al., *J. Adhesion*, 2007, 83, pp 85-104

Napkin-ring geometry: failure in “virgin” joint





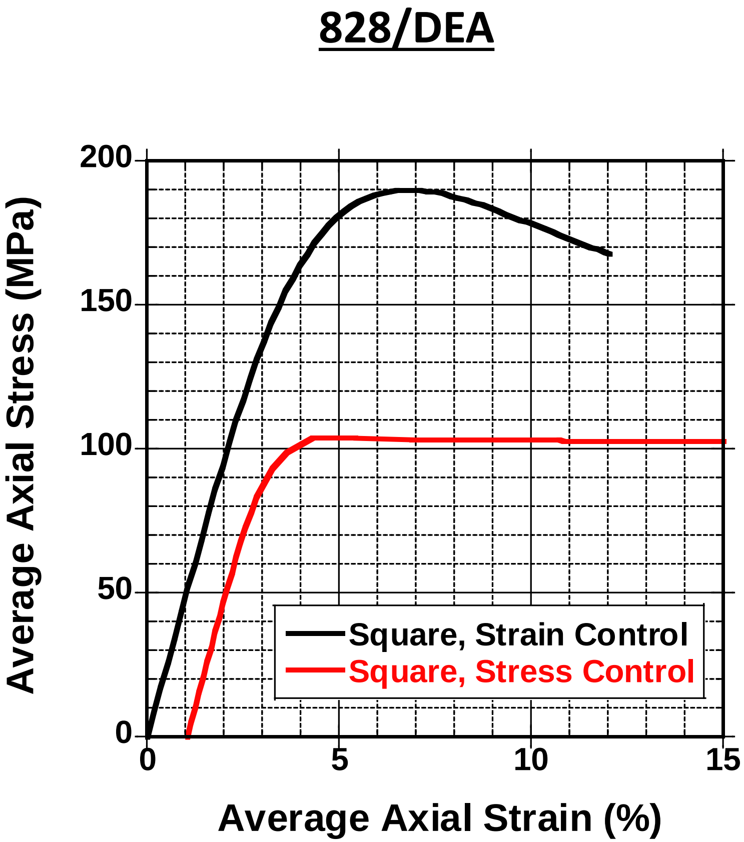
Eng Strain ~ 2(tensorial strain)

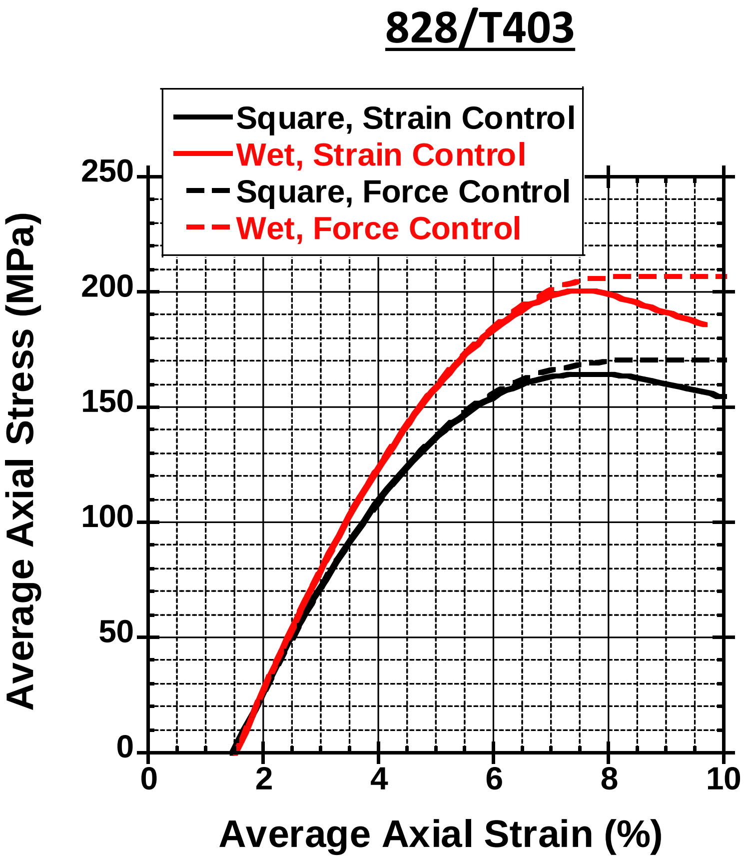
<u>Failure Mechanism:</u>

- Highest strain regions at bonding interfaces
- Highest adhesive confinement at lower interface
- Polymer in interfacial region shows highest strains and yields prior to bulk polymer
- Premature yield concentrates strain in the interfacial region
- High strain concentrations further increase relaxation rates until strain levels are no longer sustainable

<u>Note:</u> polymer cross-section is modeled with 90 degree corners, which is not the case in actual test samples. The details of the polymer geometry are anticipated to influence the location of failure (e.g., at interface with annulus vs. interface with flat surface). Analyses evaluating geometric sensitivities are currently underway.

Napkin-Ring Compression Predictions

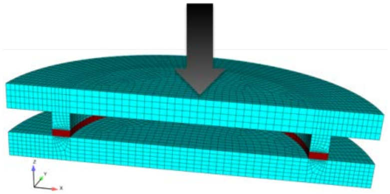


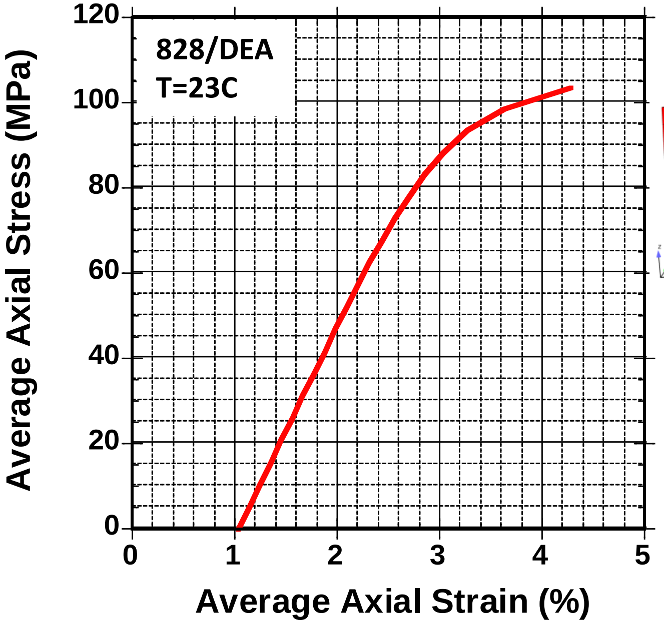


Strain Rate (strain control): 0.025 s^{-1}
Strain Rate (stress control): 0.011 s^{-1}

Strain Rate (all): ~0.002 s^{-1}

Compression Response of NR Joint*





Mesh Instability at ~107 MPa axial stress associated with adhesive yield