



SAND2011-2301P



# Adhesive Failure and the Aging of Adhesive Joints

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# Research Focus

## Materials

Polymers: coatings, underfills, encapsulants, foams, PWBs

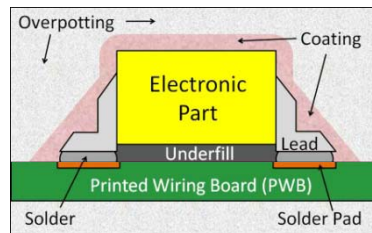
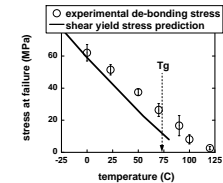
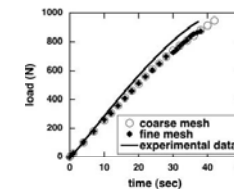


Solders: tin-lead, lead-free

## Performance Metrics

Solder Thermomechanical Fatigue  
Component Stresses

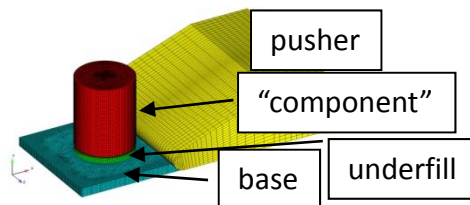
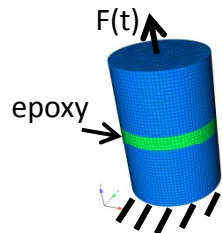
Polymer: Cohesive Failure/Adhesive Failure



**Surface Mount Technology Packaging:**  
Develop a fundamental understanding of how materials and design choices for packaging affect electrical components in thermal and dynamic loadings

## Predictive Tools

-Constitutive equation  
-Model Validation



## Processing Schemes

UV cure





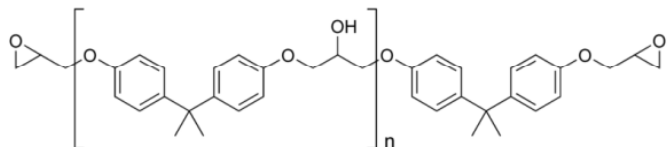
# Adhesive Joints



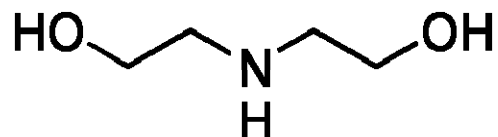
## Adhesive: 828/DEA

EPON® Resin 828

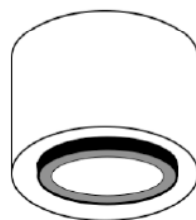
Diglycidylether of Bisphenol-A



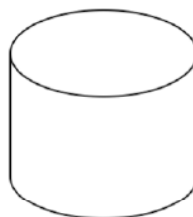
Diethanolamine



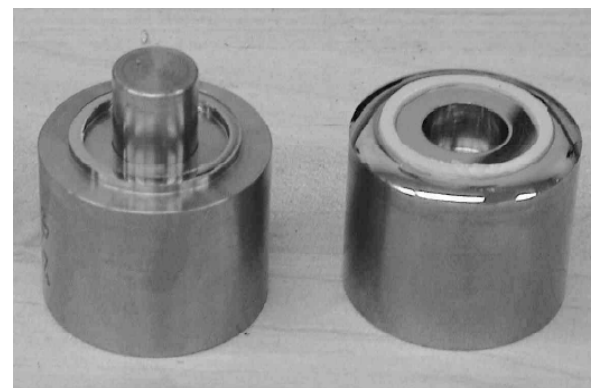
## Test Geometry: Napkin-Ring



Annulus with I.D. 0.65" and O.D. 0.75" so thickness of 50 mils. Height is also 50 mils



Bottom and top stainless steel plugs with 1 inch diameter.



de Bruyne and Houwink (eds) *Adhesion and Adhesives*, Elsevier, London p.92 (1951)

[http://www.sandia.gov/polymer-properties/828\\_DEA.html](http://www.sandia.gov/polymer-properties/828_DEA.html)



# Discussion Topics



- 1. Adhesive joint geometry and stress states**
- 2. Measuring and predicting the critical stress for debonding and how this changes with age in a humid environment**



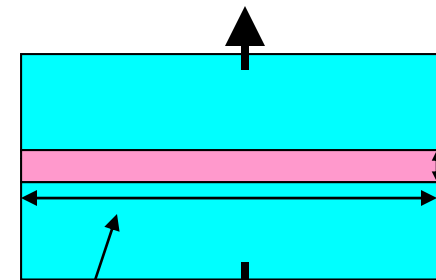
# Review of Test Geometry: Why the Napkin-Ring?



## Stress State of Adhesive During Cure and Cool-down

### ASTM Standards:

#### Butt Tension



Diameter  $\approx 1"$

#### Lap Shear



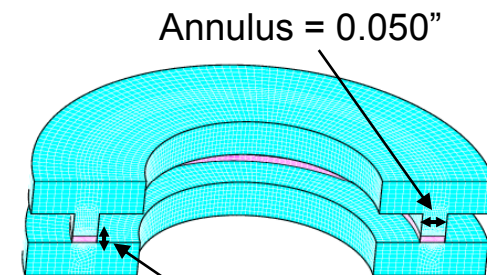
Length  $\approx 0.5"$

Thickness = 0.020"

Large diameter(length)-to-thickness ratio:  $\sim 25\text{-}50$

- large aspect ratio in epoxy bondline leaves little free surface to relieve shrinkage strains
- constrained volume generates significant residual stresses during cure and cooling before any load is applied

### Napkin-Ring



Annulus = 0.050"

Thickness = 0.020"

Relatively small annulus-to-thickness ratio:  $\sim 2$

- more free surface and less volume constraint
- minimal cure and thermal stress build-up in adhesive before test loading

**If residual stresses are high, then epoxy is closer to failure load**



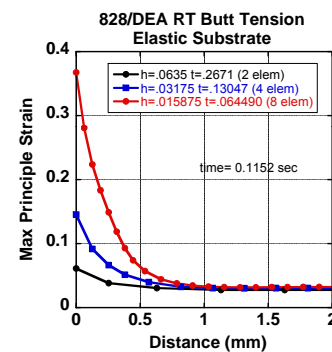
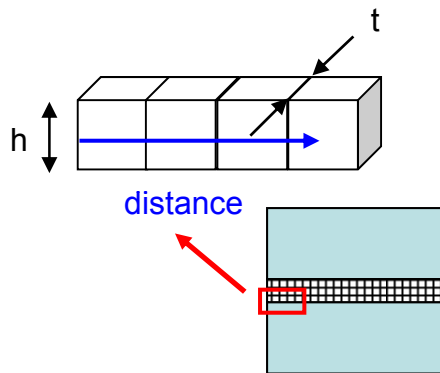
# Review of Test Geometry: Why the Napkin-Ring?



## Stress State of Adhesive During Load

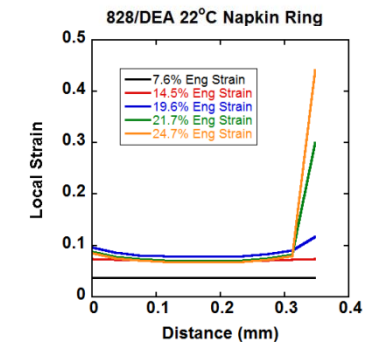
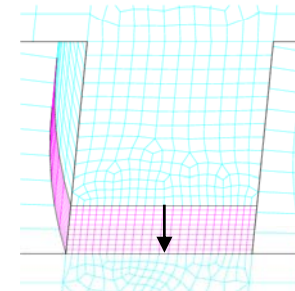
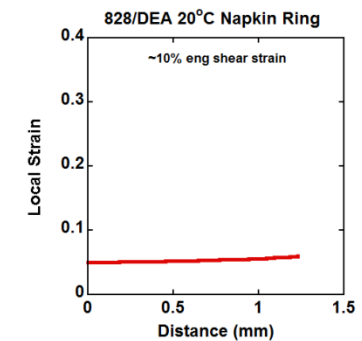
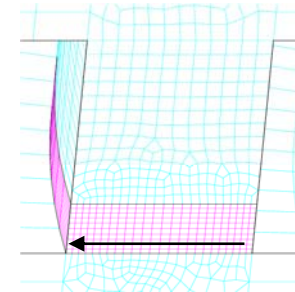
### ASTM Standards:

#### Butt Tension



- Sample has high stress/strain concentration at triple interface
- Model results are sensitive to mesh refinement at that point

### Napkin-Ring



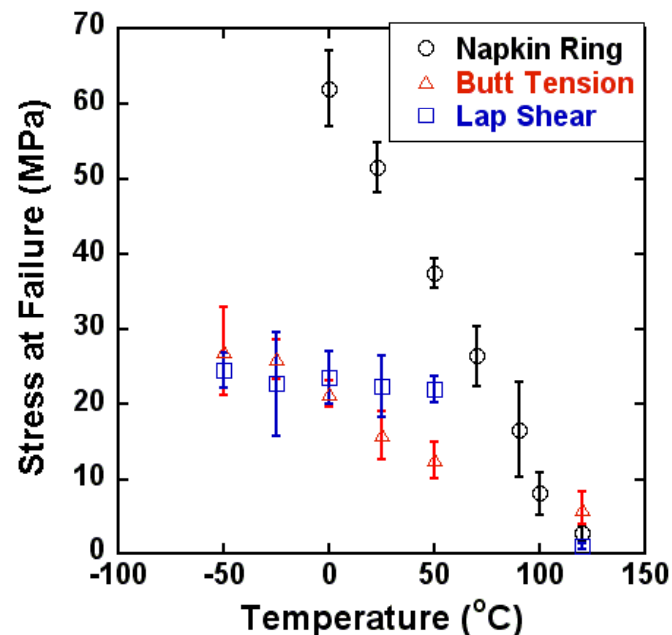
**Relatively uniform stress/strain  
distribution across entire bond**



# Review of Test Geometry: Why the Napkin-Ring?



Assume:  $\text{Stress} = (\text{Applied Load}) / \text{Area}$



- Cure/Thermal residual stresses in BT/LS are much higher than napkin ring, so epoxy begins test closer to failure point
- On loading BT/LS strains are concentrated at the triple interface rather than being distributed more uniformly like napkin ring
- BT/LS samples are more susceptible to edge defects



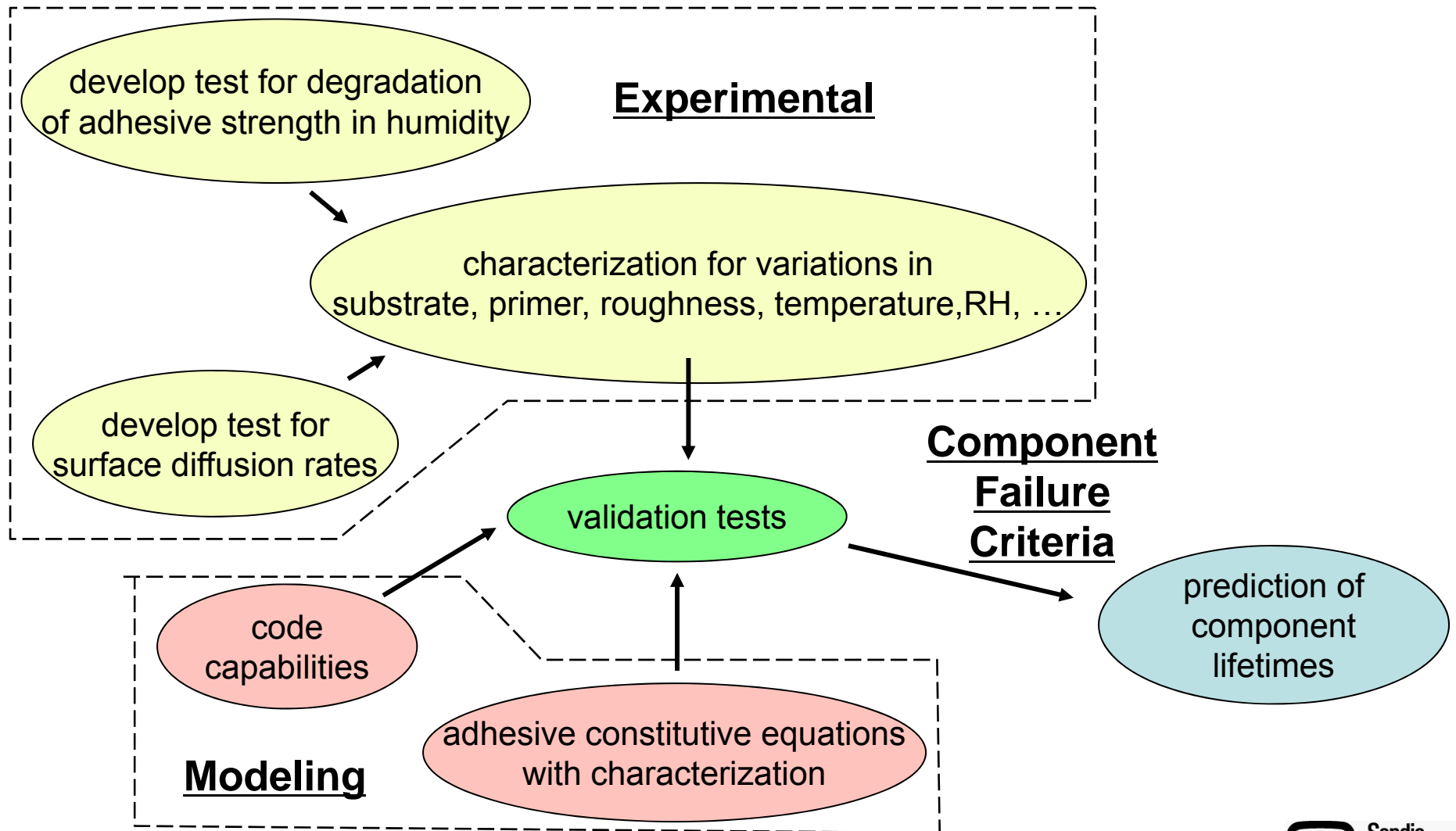
# Discussion Topics



- 1. Adhesive joint geometry and stress states**
- 2. Measuring and predicting the critical stress for debonding and how this changes with age in a humid environment**



# Adhesive Joint Aging in Humid Environments

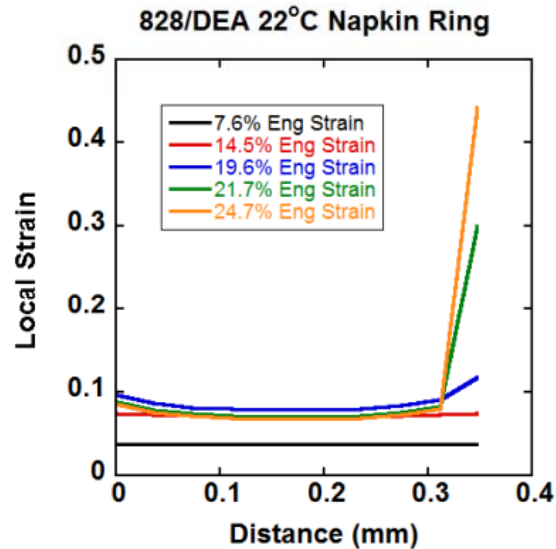
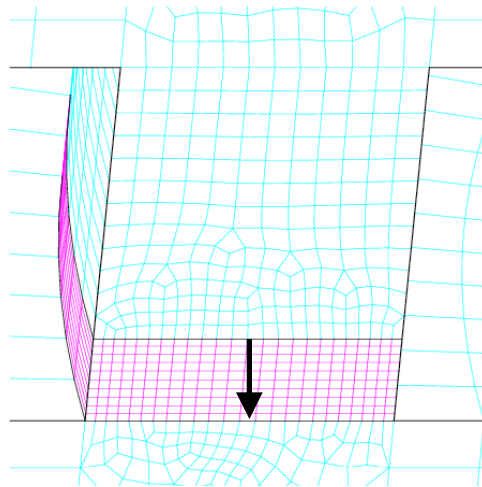




# Dry Adhesive Failure on Napkin-Rings



## Predicting the Critical Stress/Strain for Debonding



### Failure Mechanism:

- Highest stress 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

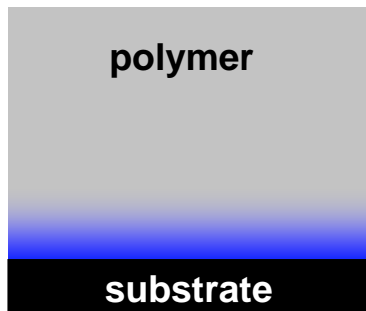
**Failure is cohesive, although it visually appears adhesive**



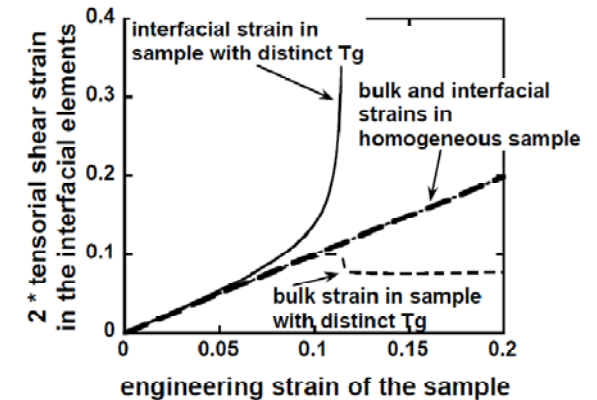
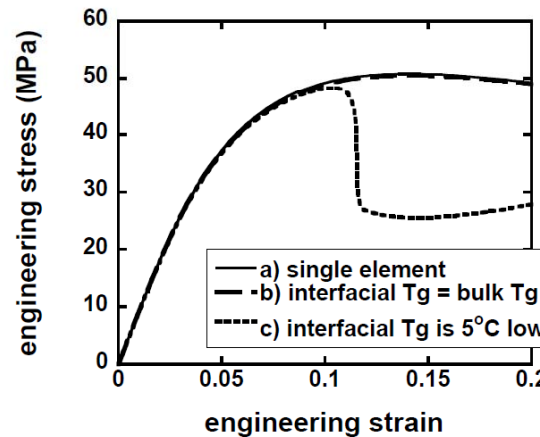
# Dry Adhesive Failure on Napkin-Rings



A “Weak” Interfacial Layer will further Promote Failure at the Interface



Entropic,  $\Delta T_g \sim 5^\circ\text{C}$



Same failure mechanism, exacerbated by reduced polymer  $T_g$  at interface

## Failure Mechanism:

- Highest stress 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

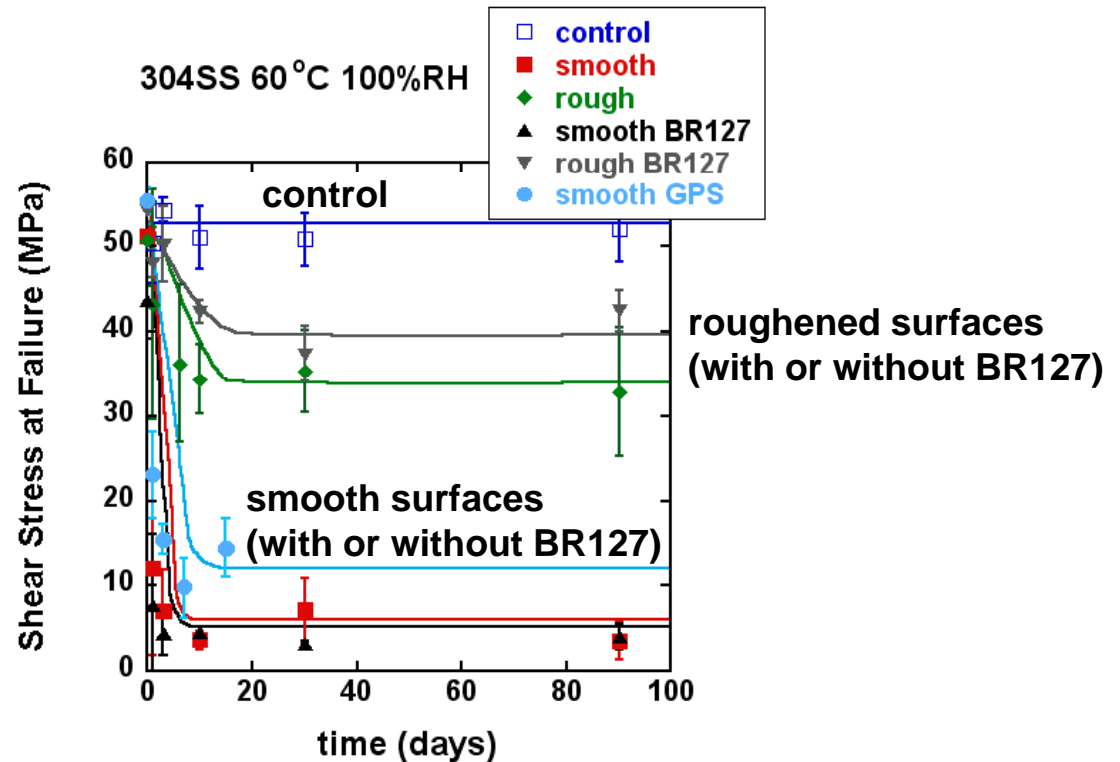
Failure is cohesive, although it visually appears adhesive



# Wet Adhesive Failure on Napkin-Rings



## 828/DEA bonding 304SS of varied surface preparations



Effects of humidity can be measured:

- Equilibrium effect of water on bond strength reached in days
- Surface roughness critical in determining magnitude of water effect on bond strength

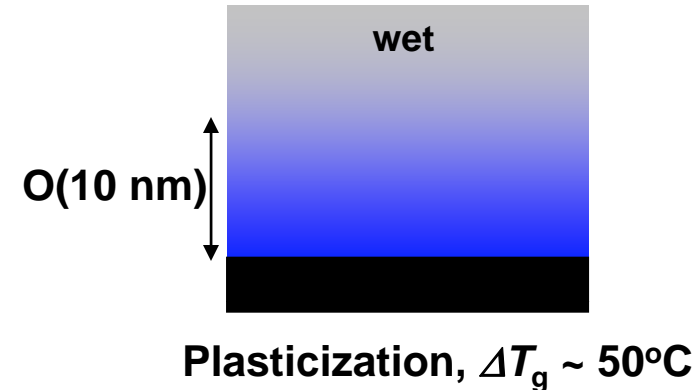
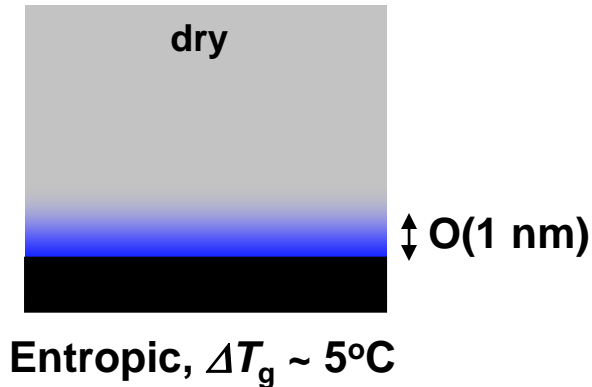
Adolf, D. B., *Predicting Stresses in Thermosets*,  
Sandia National Laboratories, Albuquerque, NM, 2010.



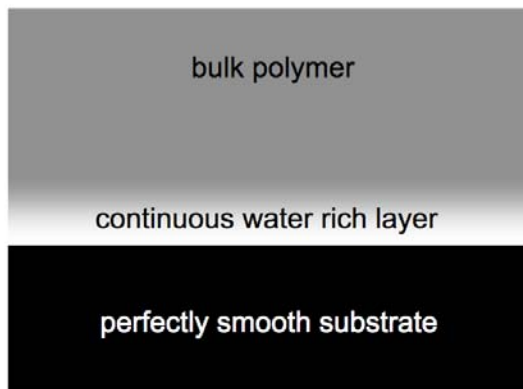
# Wet Adhesive Failure Mechanism



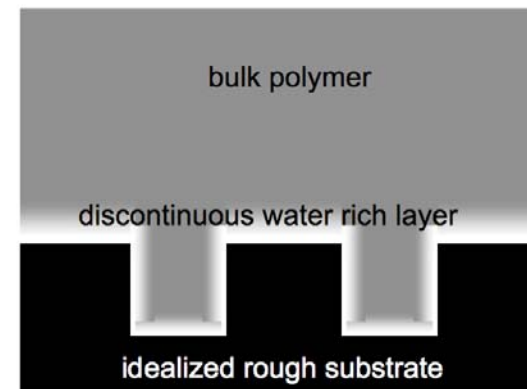
## Reduced $T_g$ : wet vs. dry



## Moisture Effects on Bond Strength: smooth vs. rough surface



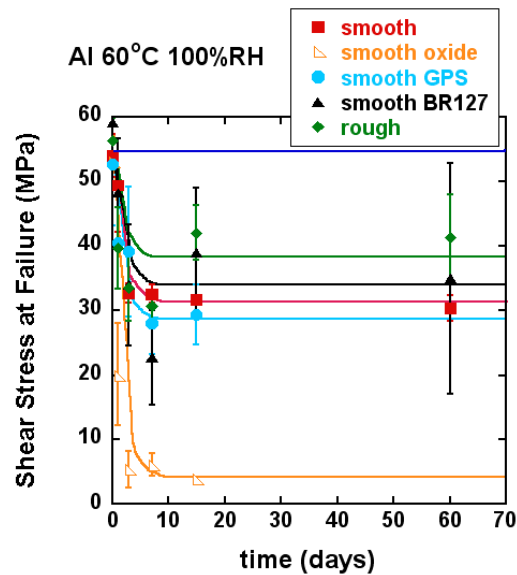
Clean break across reduced  $T_g$  interface



Bulk polymer must be traversed



# Wet Adhesive Failure: Adherend Composition



decrease on rough aluminum

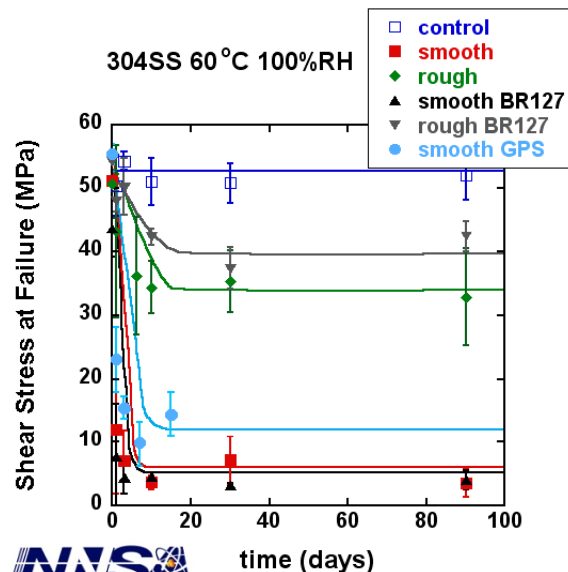
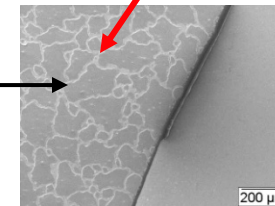
decrease on smooth aluminum  
(with or without GPS or BR127)

decrease on  
smooth oxide



unbonded area

bonded area



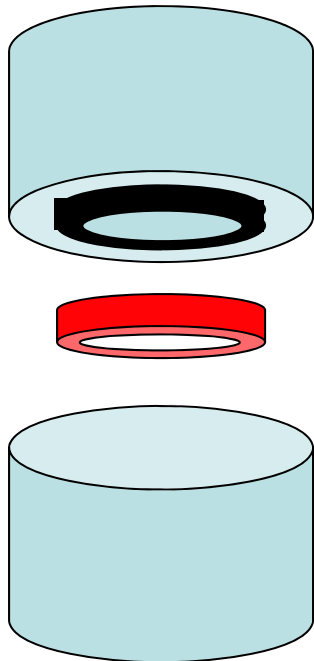
decrease on rough steel  
(with or without BR127)

decrease on smooth steel  
(with or without GPS)

- decrease in strength on smooth surfaces is less for Al than steel
- GPS does not minimize loss of adhesive strength
- oxide layer on Al significantly decreases wet strength



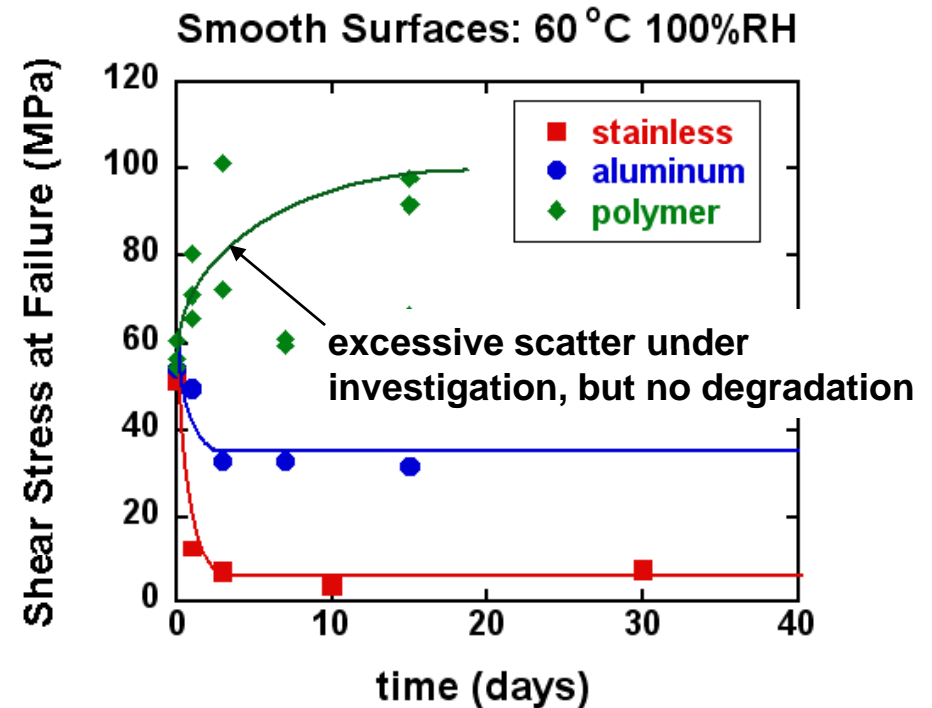
# Wet Adhesive Failure: Adherend Composition



upper plug with annulus  
alumina-filled epoxy ( $T_g \sim 160^\circ\text{C}$ )

bonded adhesive  
unfilled epoxy ( $T_g \sim 70^\circ\text{C}$ )

lower plug  
alumina-filled epoxy ( $T_g \sim 160^\circ\text{C}$ )



polymer-polymer interfaces (e.g., adhesive to composite) show no degradation

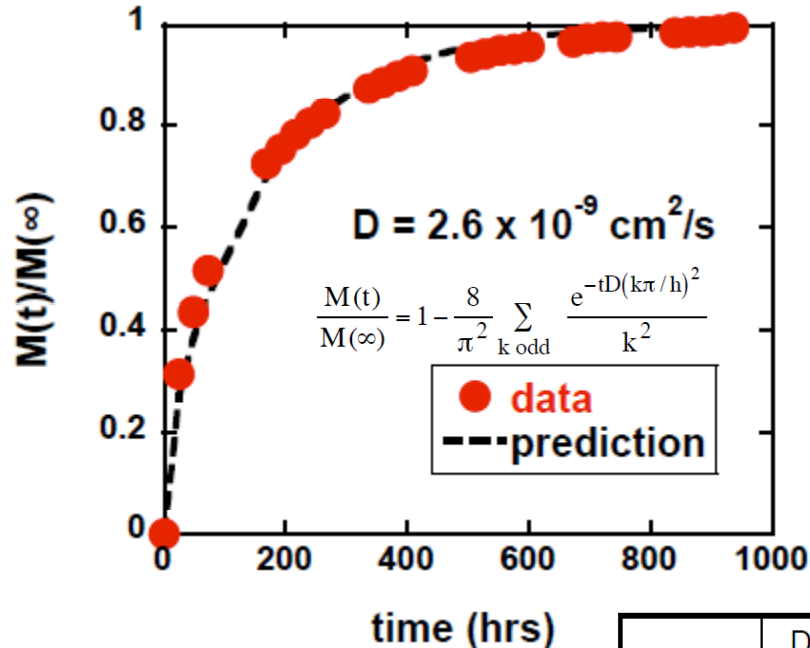
- no thermodynamic driving force for water to migrate to interface



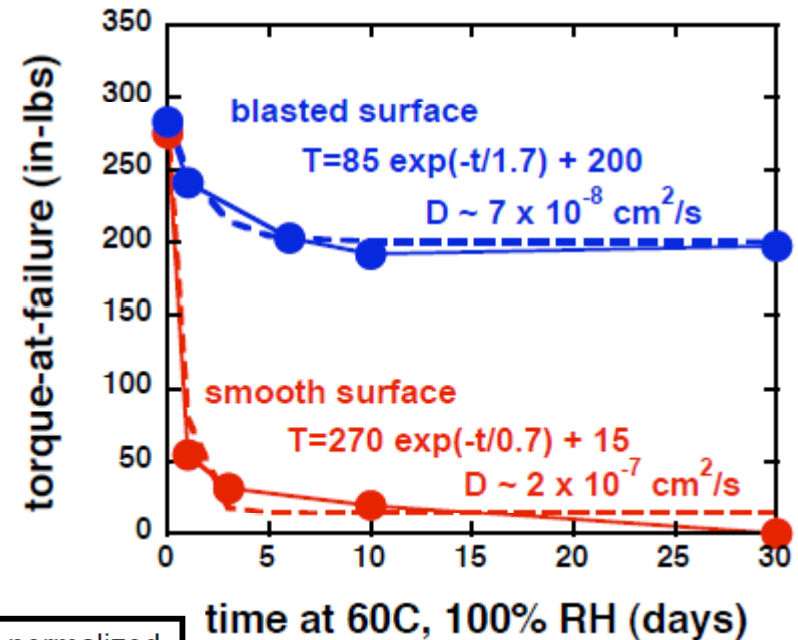
# Predicting Humid Joint Aging: Water Diffusion



## Mass Gain: 60°C 100% RH



## Napkin Ring Adhesion: 60°C 100% RH



	D (cm <sup>2</sup> /s)	normalized to bulk D
smooth	$1.6 \times 10^{-7}$	62
rough	$6.8 \times 10^{-8}$	26
bulk	$2.6 \times 10^{-9}$	1

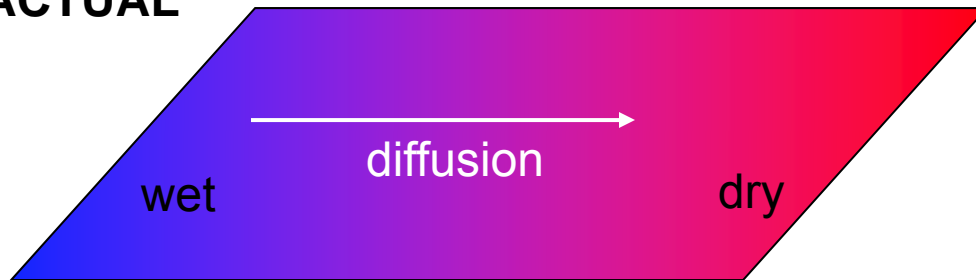
**Interfacial diffusion ~50 times faster than bulk diffusion**



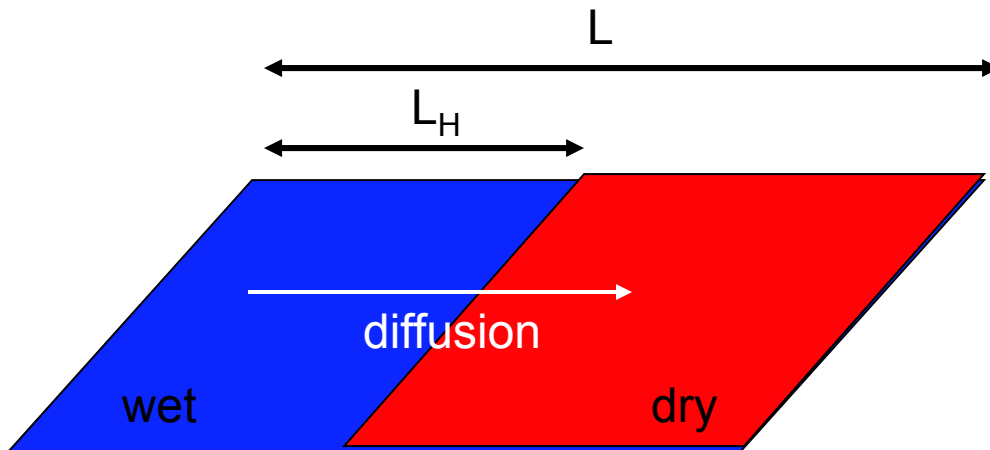
# Predicting Humid Joint Aging: Simplest Scheme



## ACTUAL



## IDEALIZED



“humid” strength  
= H

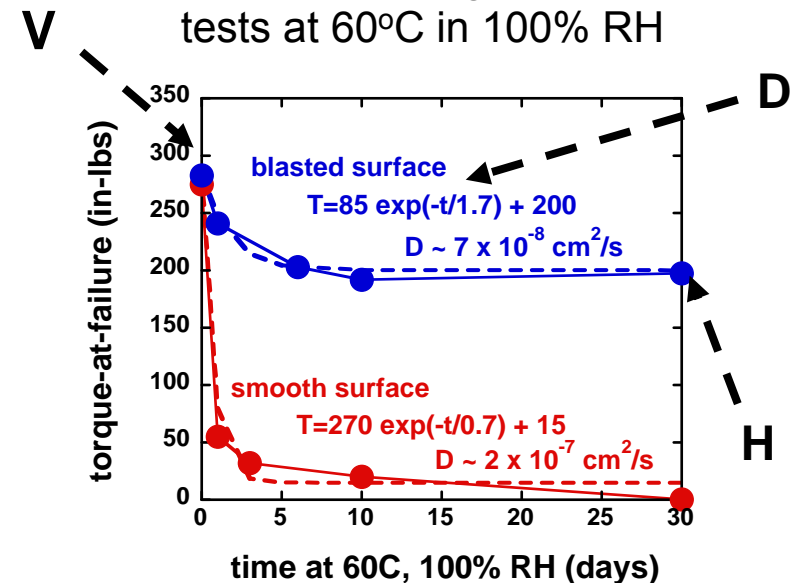
“virgin” strength  
= V

## EXAMPLE CALCULATION

$$\text{total strength} = H \left( \frac{L_H}{L} \right) + V \left( \frac{L - L_H}{L} \right)$$

$$\text{where } L_H = (Dt)^{1/2}$$

from napkin ring adhesion tests at 60°C in 100% RH

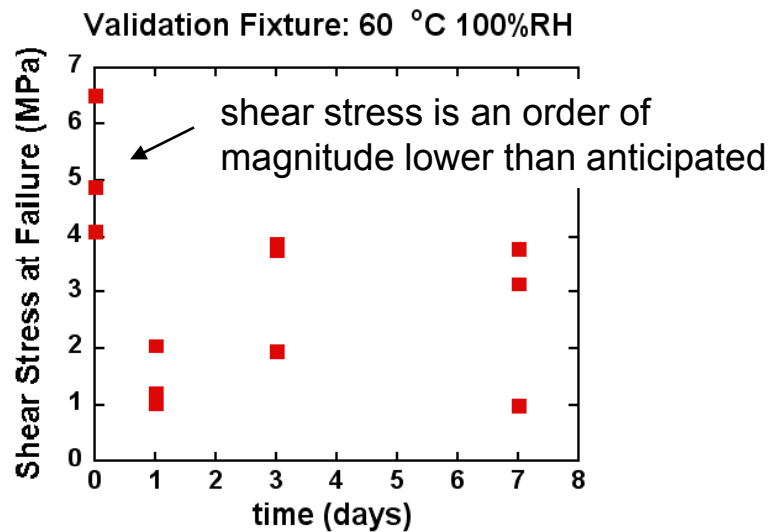
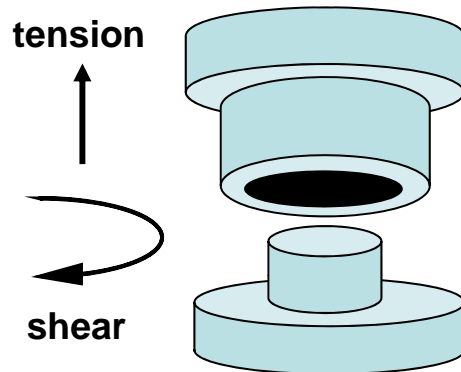




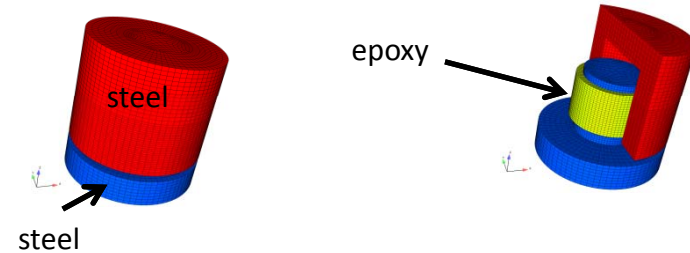
# Testing Predictions: Validation Geometry



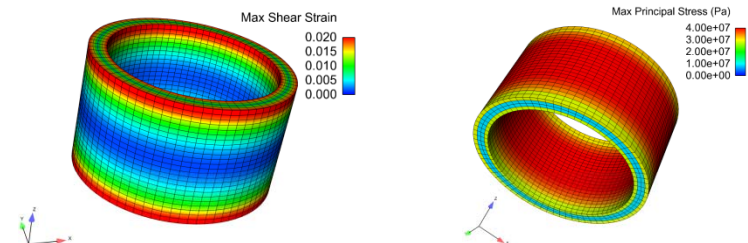
## Experimental



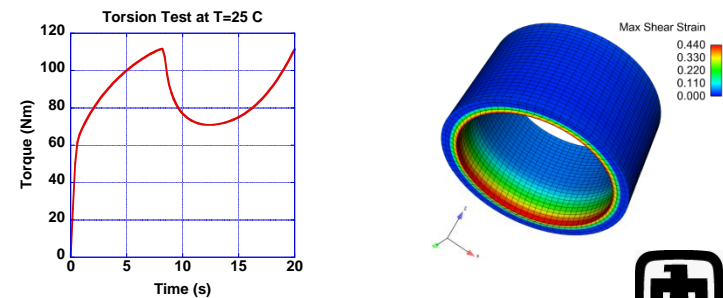
## Stress Analysis



## Thermal Stress/Strain

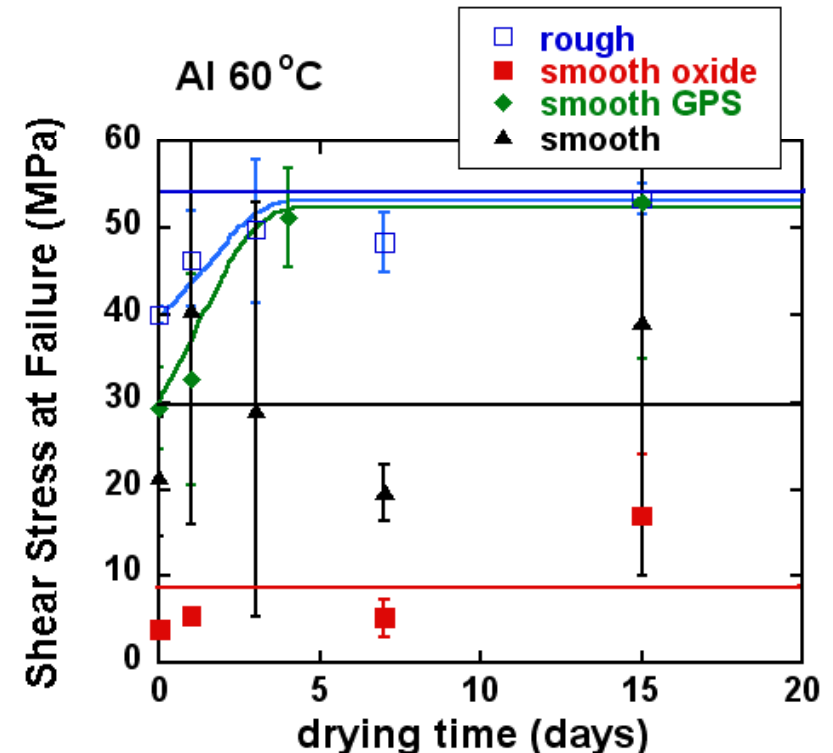
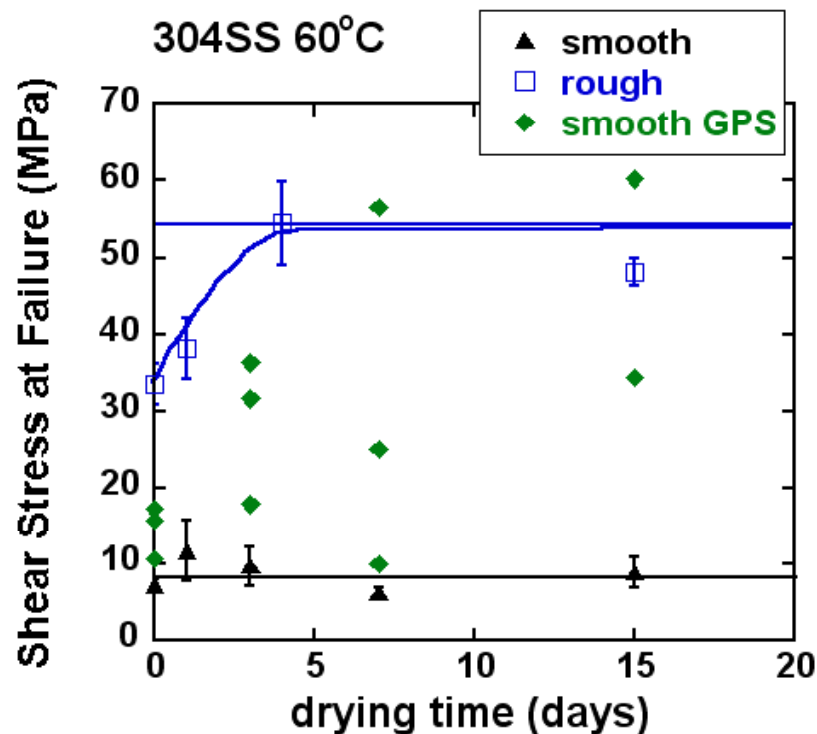


## Rotational Loading Strain





# Can Adhesive Joints be “Healed”



- Original strength is regained on rough surfaces
- No regaining of strength on smooth, unprimed surfaces
- GPS allows regaining of original strength on smooth Al but scattered results on steel



# Summary



- Failure isn't truly adhesive failure, typically the polymer fails cohesively and this can occur in a weak interfacial layer
- Loads to adhesive failure do depend on the joint geometry...choose carefully
- Napkin-Ring geometry is an excellent tool to characterize wet adhesive failure: fast (~2 weeks to equil.), directly yields stress-to-failure, simplicity allows mechanistic interpretation
- Bonding materials and surface preparation significantly affect the role of moisture on adhesive strength
- Degradation effects of moisture can be healed in some cases



# Back Up Slides



# Adhesion Task Four-Question Chart



## What are you trying to do in this task?

- Measure and predict the critical stresses for adhesive de-bonding
- Measure and predict the change in de-bonding stress when components age in dry and humid environments
- Relate the de-bonding stress to processing history

## What makes you think you can do it?

- Leverages previous SNL-funded research on measuring and predicting adhesive strength
- Adhesion working group involves DOE and DoD members to direct goals and share knowledge/experience

## What difference will it make?

- Component designs can be more robust if de-bonding stress margins are known
- Knowledge of aging mechanisms improve material selection for given environments
- Processes can be defined to improve adhesive strength

## What / When / To Whom Will You Deliver?

- Deliverables are metrics and procedures to measure and predict de-bonding
- Delivery will be staged to provide capability on successively more difficult systems
- Adhesion working group will identify a DoD contact to share capabilities