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Interfacial Toughness: Variation with Surface Roughness and Test Temperature

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Tested two epoxy materials

EPON® Resin 828 cured with DEA (diethanolamine)

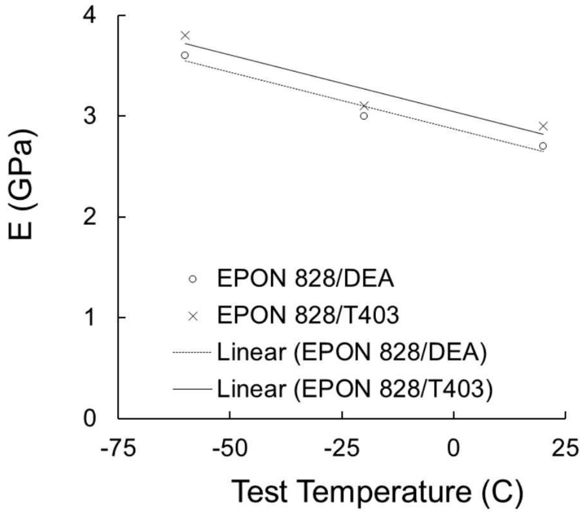
- 100:12 pbw mix ratio.
- Cure cycle: 24hr at 45°C, ramp to 71°C in $6\,^{1}/_{3}$ hr, hold at 71°C for 5 hr, cool down to RT.

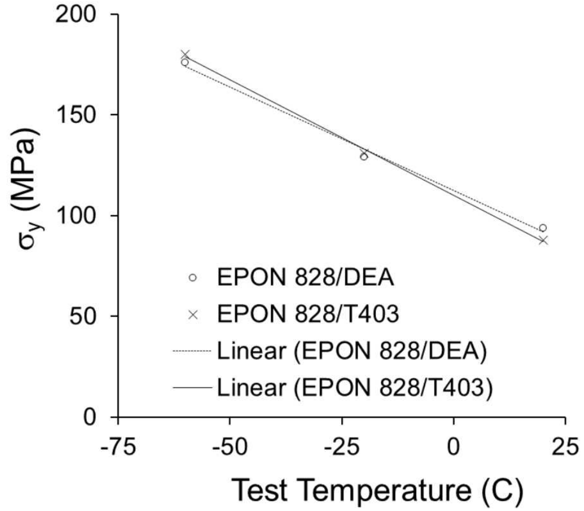
EPON® Resin 828 cured with Jeffamine® T-403 (polyetheramine)

- 100:43 pbw mix ratio.
- Cure cycle: 24hr at 23°C, followed by 3hr at 50°C, followed by 15hr at 80°C, cool down to RT.

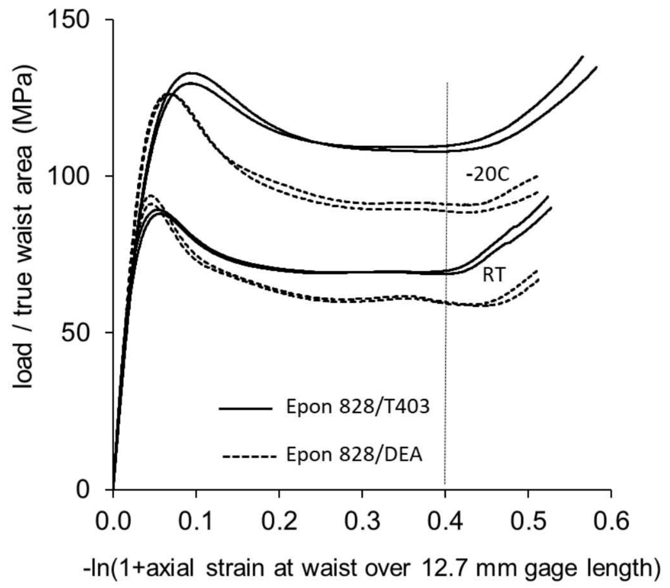
Compression plug stress-strain data
(nominal strain rate = 0.001/s)

	EPON 828/DEA T_g=70 °C		EPON 828/T403 T_g=85 °C	
T (C)	E (GPa)	σ_y (MPa)	E (GPa)	σ_y (MPa)
RT	2.7	94	2.9	88
-20	3.0	129	3.1	131
-60	3.6	176	3.8	180



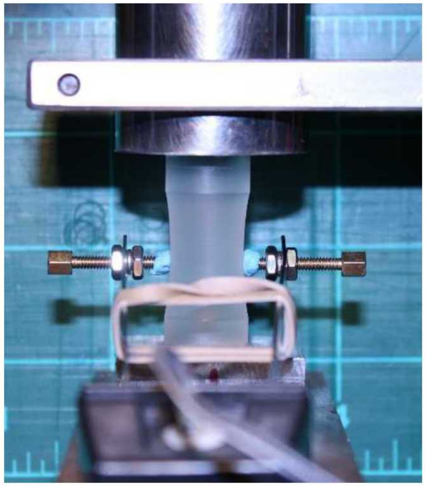


Large strain compressive response



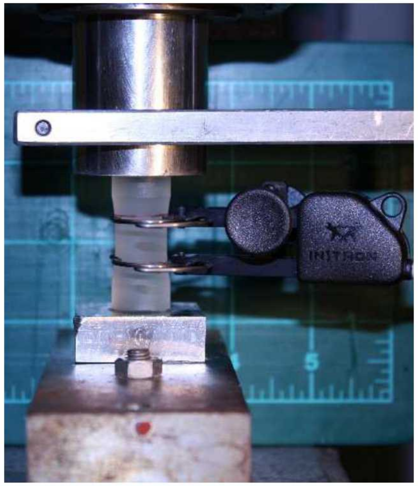
- Initial yield, strain-softening, a lower stress plateau, and finally hardening at large strain (tests terminated when strain ~0.5).
  - strain hardening begins at ~ $\varepsilon = \varepsilon_h > 0.4$, and is not strongly dependent on σ_y.
- softening generates localized deformation, so results depend on specimen geometry and loading.
  - nevertheless, overall shape should reflect yield strength, the post yield stress plateau, and the strain at which final strain hardening occurs.

waisted specimen
min radius 7.2 mm

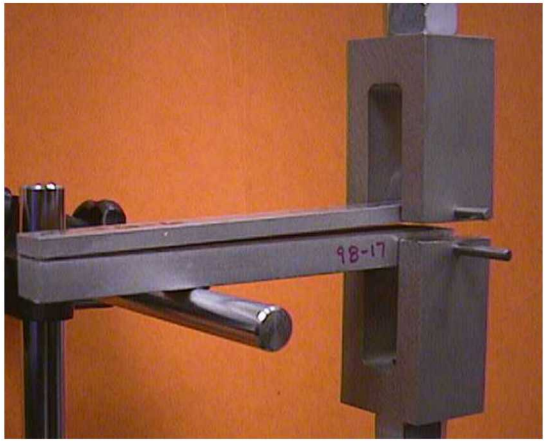


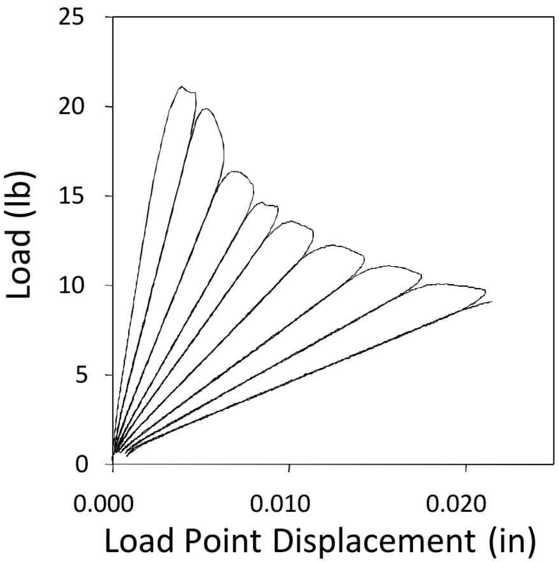
measured change in diameter

measured axial displacement over a 12.7 mm gage length



Asymmetric Double Cantilevered Beam Sandwich specimen

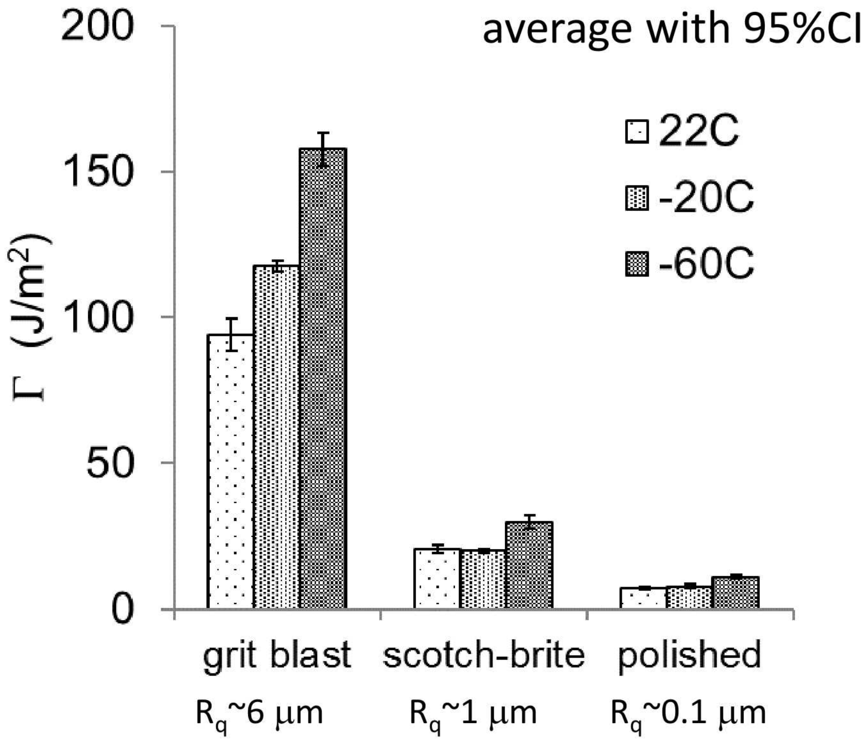




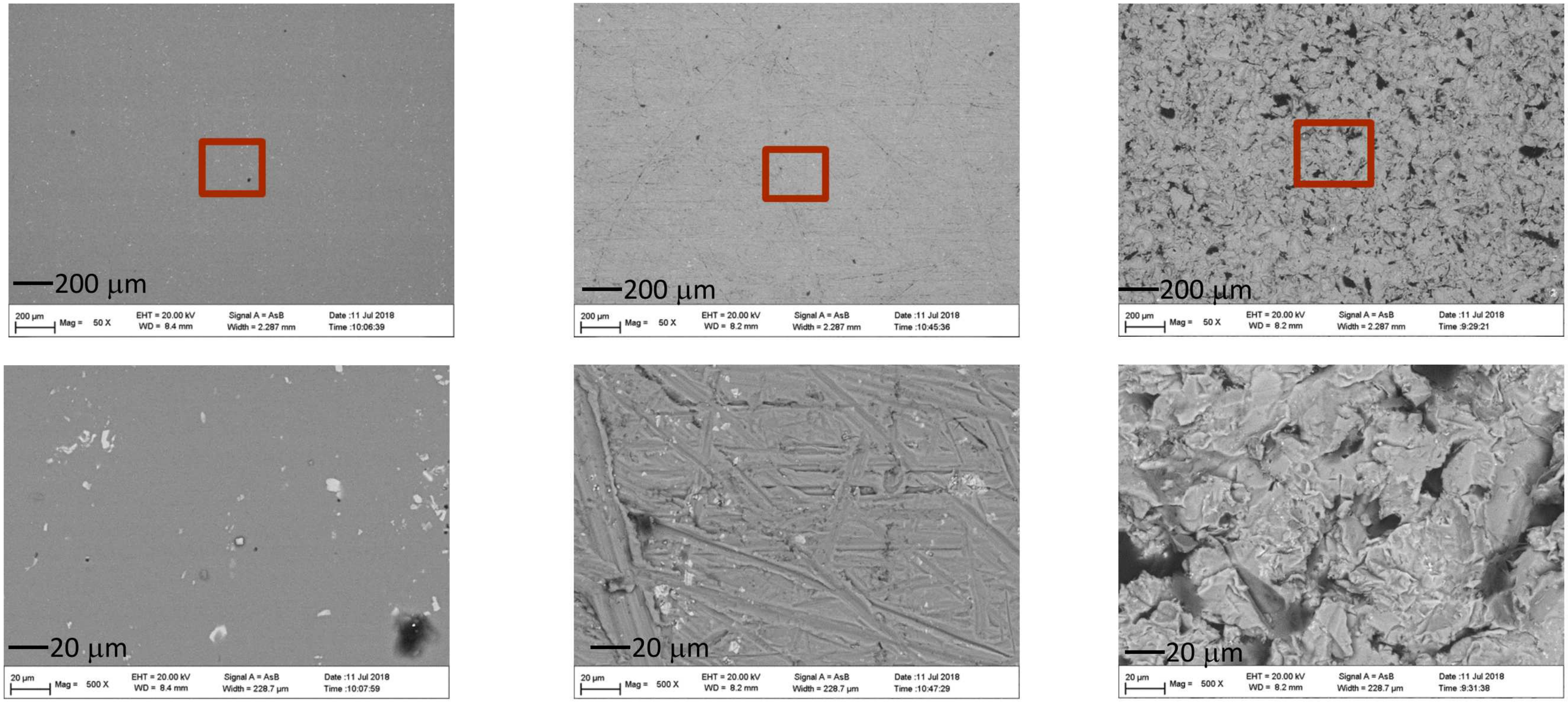
- Can make multiple Γ measurements per specimen (crack propagates stably).
- Use unloading compliance to determine crack length.
- Produces a predominantly Mode I crack-tip loading with a slight tendency to push the crack towards the interface ($\Psi_{l=10\mu m} = -15^o$ for a 0.5 mm bond).

Γ for EPON 828/DEA epoxy-to-6061 aluminum interface

- Tested an epoxy/aluminum interface.
  - Epon828/DEA epoxy (100:12 pbw mix ratio 71°C cure).
  - 6061-T6 aluminum surfaces sonicated and cleaned with isopropyl alcohol prior to bonding.
- Both temperature and surface roughness have a strong impact on Γ.
  - Γ increases as test temperature decreases.



EPON 828/DEA epoxy-to-6061 aluminum fracture surfaces



Note: backscatter SEM images. Used EDS to identify al/epoxy regions. Then image thresholding where black=> carbon

Polished

- R_q <0.1 μm
- <1% carbon on aluminum fracture surface (4 specimens, analyzed - 3 regions/specimen)

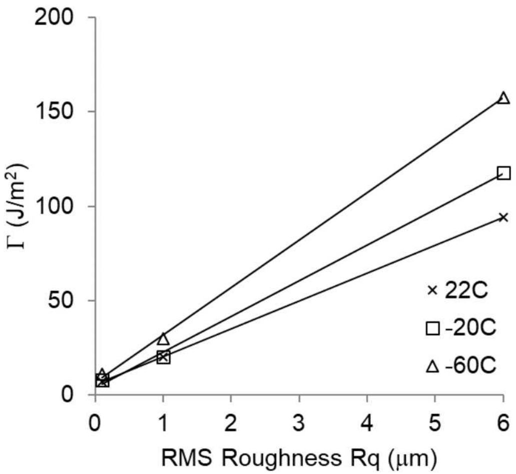
Hand roughened using a Scotch-Brite™ pad

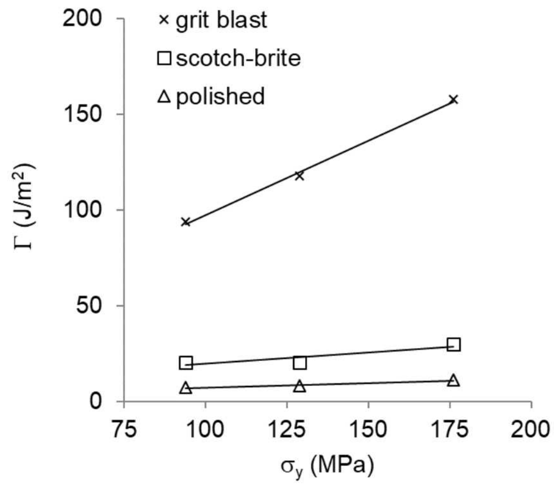
- R_q ~1.0 μm
- ~2-6% carbon on aluminum fracture surface (2 specimens analyzed - 3 regions/specimen)

Grit blasted

- R_q ~6.0 μm
- ~7-8% carbon on aluminum fracture surface (2 specimens analyzed - 3 regions/specimen)

Γ for EPON 828/DEA epoxy-to-6061 aluminum interface depends on surface roughness and test temperature

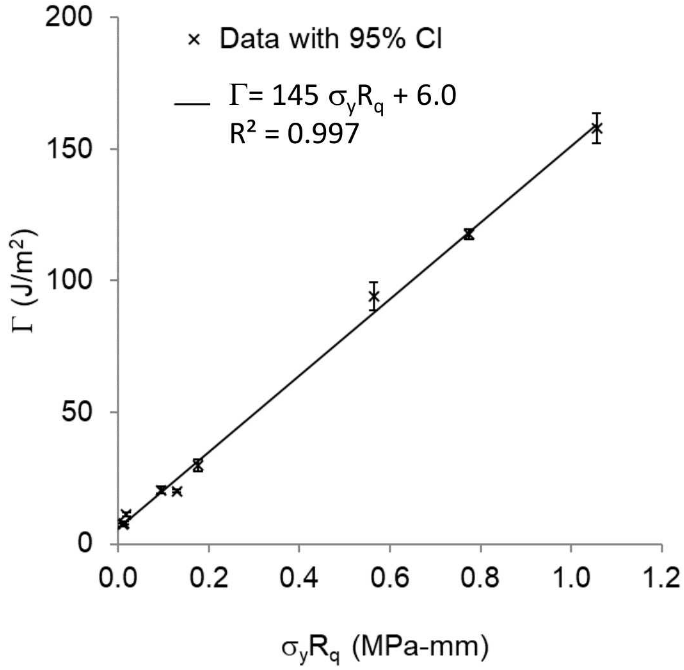




T (°C)	σ_y (MPa)	$\frac{\Gamma_{grit\ blast}}{\Gamma_{polished}}$	R_q (μm)	$\frac{\Gamma_{-60°C}}{\Gamma_{22°C}}$
22	94	12.9	0.1	1.52
-20	129	14.9	1.0	1.45
-60	176	14.2	6.0	1.68
average		14.0		1.55

- Converted test temperature to yield strength (linear relationship between σ_y and T)
- Γ varies ~ linearly with σ_y when R_q is fixed and Γ ~ linearly with R_q when σ_y is fixed.
- ~factor of 14 increase in Γ when R_q is increased from 0.1 μm to 6 μm (~independent of test temperature)
- ~55 % increase in Γ when test temperature is decreased from 22°C to -60°C (~independent of roughness level).
- Simplest relationship consistent with observation: $\Gamma = \Gamma_o + C\sigma_y R_q$ where Γ_o is the toughness of a smooth interface and C is a proportionality constant.

Dependence on surface roughness and test temperature EPON 828/DEA epoxy-to-6061 aluminum interface



T (°C)	σ_y (MPa)	R_q (μm)	$\sigma_y R_q$ (N/mm)	Γ (J/m²)	St Dev Γ (J/m²)	# data pts
23	94	0.1	0.01	7.3	0.6	7
23	94	1	0.09	20.5	1.4	7
23	94	6	0.56	94	4.3	5
-20	129	0.1	0.01	7.9	0.7	8
-20	129	1	0.13	20.1	1.0	10
-20	129	6	0.77	117.6	1.4	5
-60	176	0.1	0.02	11.1	1.3	14
-60	176	1	0.18	29.8	3.1	10
-60	176	6	1.06	157.7	10.3	15

Plot mean with 95% confidence interval

Γ for EPON 828/T403 epoxy-to-6061 aluminum interface

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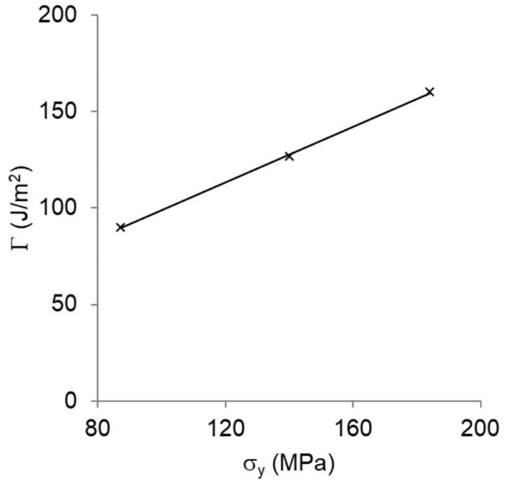
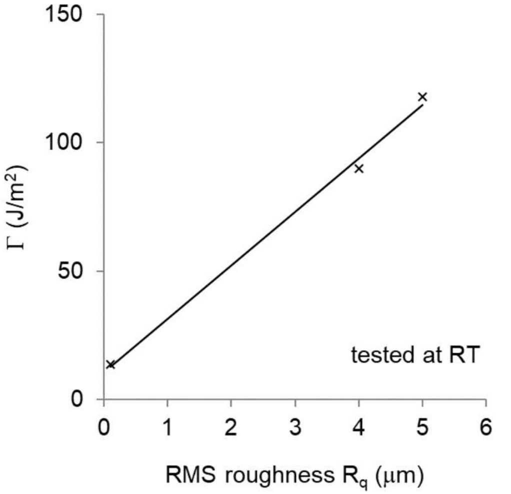
- In previous work, tested an EPON 828/T403 epoxy (100:43 pbw mix ratio)-to-6061 aluminum interface[1] (i.e., cured with a different hardening agent).
  - less comprehensive data set.
  - 6061-T6 aluminum surfaces sonicated and cleaned with isopropyl alcohol prior to bonding (i.e., same clean) .
- Dependence similar to that measured for the EPON 828/DEA epoxy to 6061 aluminum interface



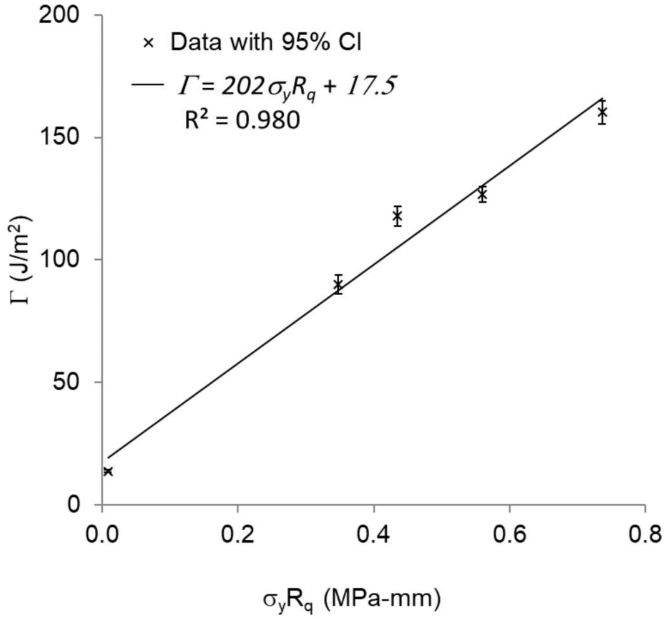
~80 % increase in Γ when test temperature is decreased from 22°C to -65°C (R_q = 4 μm)



> factor of 9 increase in Γ when R_q is increased from 0.1 μm to 5 μm (tested at 22°C)

[1] See Unlimited Release Report for further details: Reedy, E.D., Jr., et al., A Process and Environment Aware Sierra/SolidMechanics Cohesive Zone Modeling Capability for Polymer/Solid Interfaces, SAND2015-8066. 2015, Sandia National Laboratories: Albuquerque, NM.

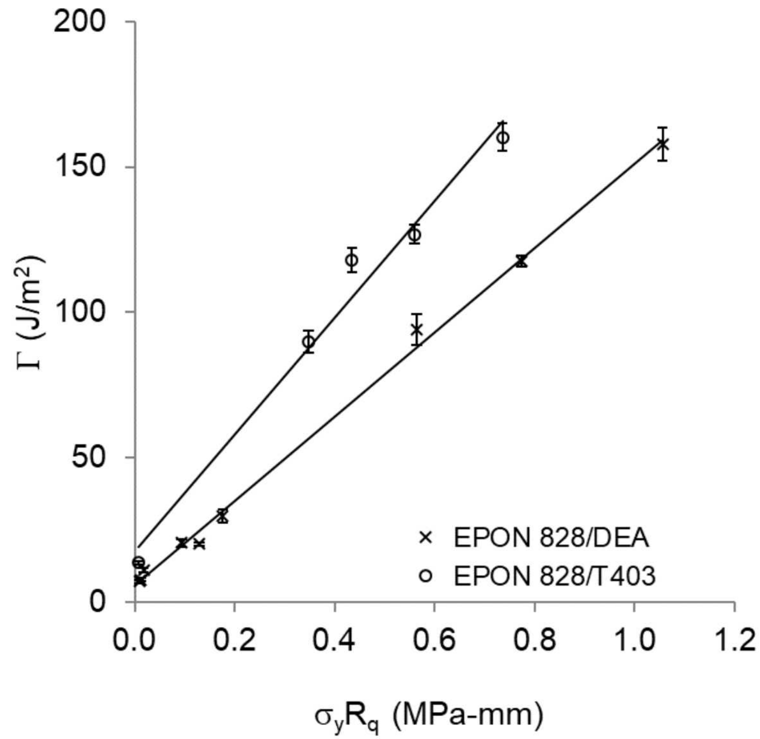
Dependence on surface roughness and test temperature EPON 828/T403 epoxy-to-6061 aluminum interface



T (°C)	σ_y (MPa)	R_q (µm)	$\sigma_y R_q$ (N/mm)	Γ (J/m²)	St Dev Γ (J/m²)	# data pts
23	87	0.1	0.01	13.7	0.9	13
23	87	4.0	0.35	89.9	4.6	8
23	87	5.0	0.44	117.9	7.3	15
-25	140	4.0	0.56	126.8	4.4	10
-65	184	4.0	0.74	160.3	7.6	12

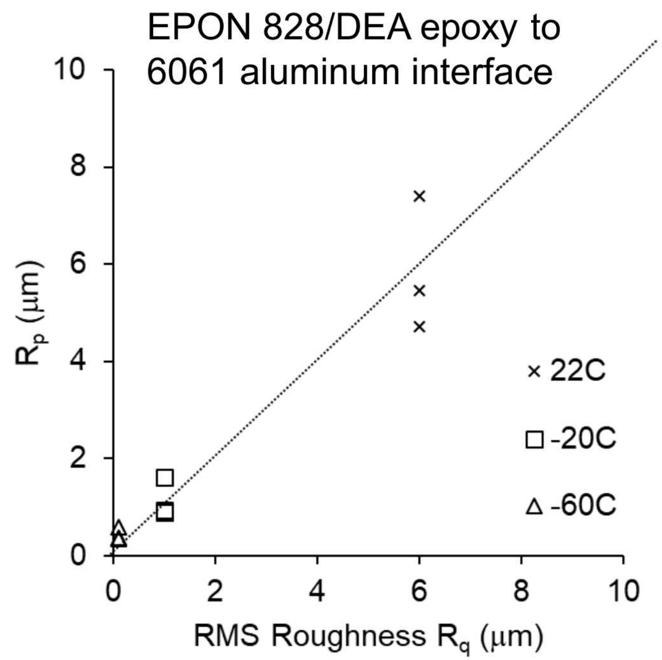
Plot mean with 95% confidence interval

Comparison of aluminum interfaces with either an EPON828/DEA or EPON828/T403 epoxy



- Slope of the Γ vs. $\sigma_y R_q$ line differs with the choice of epoxy adhesive.

- **Is there a basis for observed Γ, R_q, and σ_y scaling?**



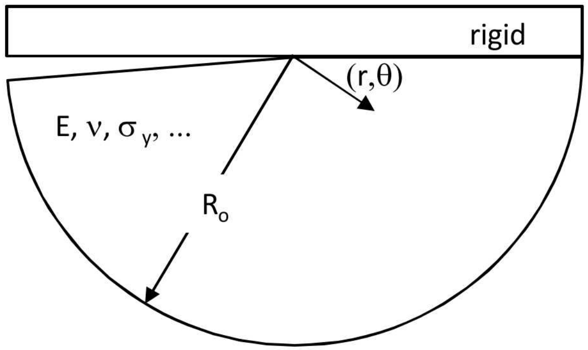
- Estimated plane strain crack-tip plastic zone size R_p for case of a rigid adherend

$$R_p = \frac{1}{3\pi}\left[\frac{2\bar{E}\Gamma}{(1-\beta^2)\sigma_y^2}\right] \quad \text{where} \quad \beta = \frac{(1-2\nu)}{2(1-\nu)} \quad \text{and} \quad \bar{E} = \frac{E}{(1-\nu^2)}$$

- R_p is roughly commensurate with surface roughness R_q and much smaller than the 500 µm ADCB bond thickness, etc. (i.e., SCY applies).

Is there a basis for observed Γ, R_q, and σ_y scaling?

- Consider the plane strain, elastic-plastic small scale yielding problem.
- Upper material is rigid while lower material is elastic-plastic.
  - plastic yielding will depend on the yield strength σ_y and other nondimensionalized properties (depends on the plasticity model and assume they are independent of σ_y).
- Desire a solution that determines ε at position r and θ.



Loading at $r = R_o$ defined by known linear elastic asymptotic crack tip fields so that it is consistent with a prescribed energy release rate G and crack-tip mode mixity $\psi_{r=Ro}$.

- Based on dimensional considerations, strain can be expressed as $\varepsilon = f\left(\frac{G}{Er}, \frac{G}{\sigma_y r}\psi_{r=R_o}, \nu, \ldots\right)$
- $\sqrt{\frac{G}{Er}} = \frac{1}{E}\sqrt{\frac{GE}{r}} = \frac{|K|}{E}\sqrt{\frac{1}{r}}$ is consistent with a LEFM prediction for crack-tip strain.
- $\frac{G}{\sigma_y}\frac{1}{r}$ is consistent with the PP limit of the power-law-hardening plasticity prediction for crack-tip strain.

Is there a basis for observed Γ, R_q, and σ_y scaling?

- <u>Hypothesize</u> interfacial separation initiates at the tip of an arrested interfacial crack when the localized strain at the crack tip exceeds ε_h over some characteristic distance that scales with R_q.
  - recall epoxy shows strain-softening followed by rapid hardening at large strains ($\varepsilon = \varepsilon_h$~0.4).
- If applied loading, and material parameters are fixed, then within the zone of very large plastic strains: $\epsilon = f\left(\frac{G}{\sigma_y r}\right)$
  - the proposed criterion is $G = \Gamma$ and the crack propagates when $\varepsilon = \varepsilon_h$ at $r = R_q$
  - since ε_h is a constant (does not depend on σ_y) this implies that

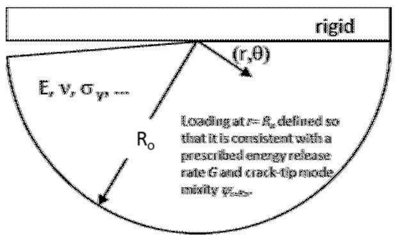
    $\frac{\Gamma}{\sigma_y R_q} = a\ constant$ => $\Gamma \sim \sigma_y R_q$
  - this result is consistent with the observed scaling of

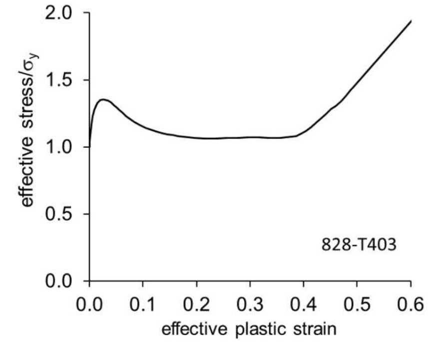
$$\Gamma - \Gamma_o = C\sigma_y R_q$$

- Also note that for SCY in a homogeneous, perfectly plastic material, COD $\delta_t \sim G/\sigma_y$.
  - the same scaling as above if the crack propagates at a critical δ_t that is commensurate with R_q.

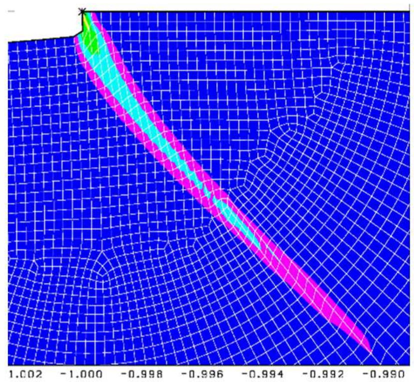
Is there a basis for observed Γ, R_q, and σ_y scaling?

- Performed a plane strain FEA of the small-scale yielding problem using a J2-plasticity model whose effective stress-strain relationship is based on the experimentally measured Epon 828/T403 epoxy data.
- Note: analysis does not include pressure-dependent yield (believed to be important).



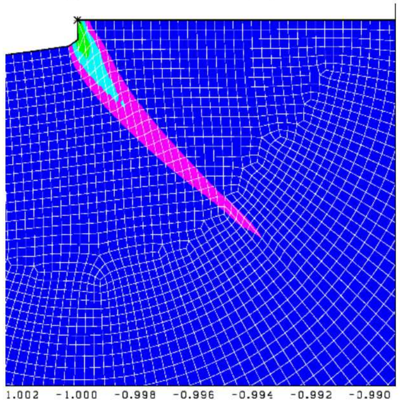


Crack tip ε_e = 0.39, COD = 0.64 μm
σ_e = 85 MPa, p = 185 MPa,



σ_y=65 MPa, Γ = 0.1 J/m^2

Crack tip ε_e = 0.38, COD = 0.67 μm
σ_e = 165 MPa, p = 353 MPa



σ_y=130 MPa, Γ = 0.2 J/m^2

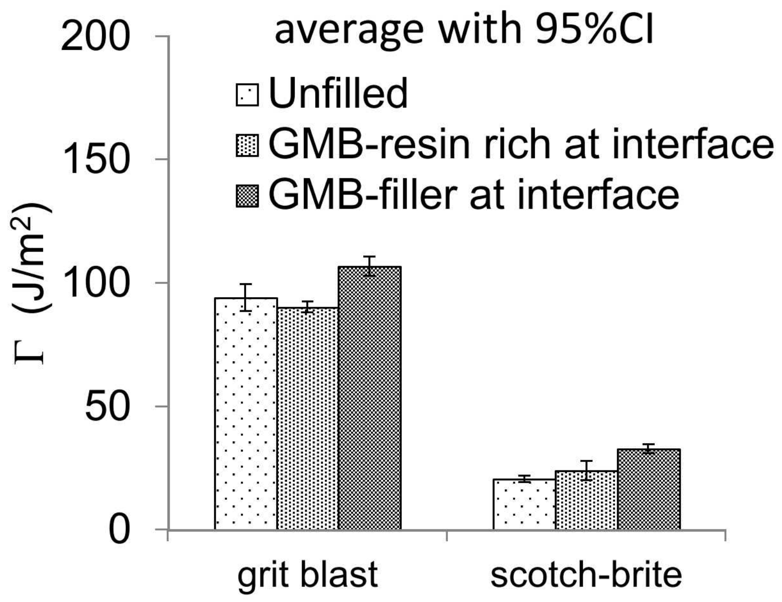
- FEA results that include softening are consistent with the previous scaling-based prediction that the plastic strain at the crack-tip is unchanged when Γ/σ_y is held fixed.

Comments

- Anticipate the observed dependence of interface toughness on test temperature can be related to its dependence on loading rate.
- Does this approach extend to epoxy that do not display post-yield softening followed by rapid hardening at large strains?

Effect of adding GMB filler to the EPON 828/DEA epoxy

- Measured Γ for a GMB-filled EPON 828/DEA epoxy-to -6061 aluminum interface.
- Aluminum interface either grit blasted or roughened with scotch-brite.
- Epoxy either unfilled or with ~30% by weight glass microballoons (GMB).
    - during cure sample oriented so that GMB either floats to the interface or away from the interface (resin rich).
- Tested at RT
- GMB filler has a modest effect on Γ when the GMB floats to the interface.



R_q~6 μm

R_q~1 μm