

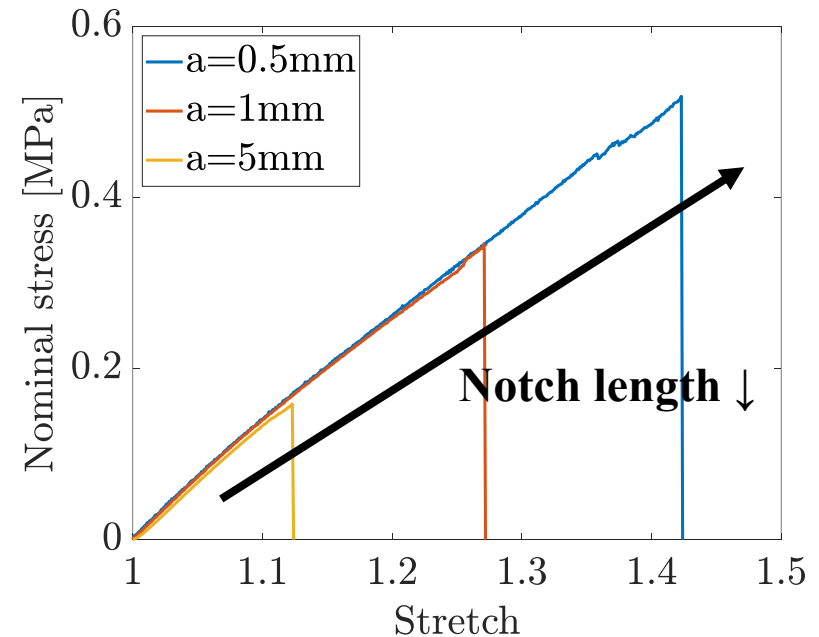
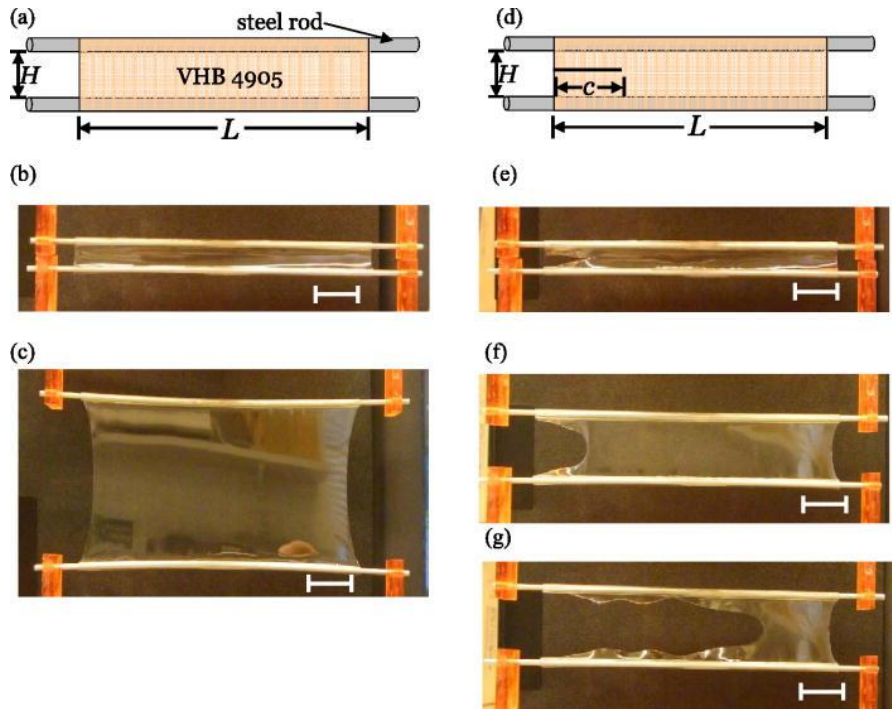
Experiments and nonlocal continuum modeling of the size-dependent fracture in elastomers^[1,2]

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Fracture in elastomers

- Extreme, nonlinear deformation \rightarrow fracture
- Influenced by the size of flaws; **the size-dependent fracture**^[1,3]
 - Rupture stretch increases as the specimen size decreases



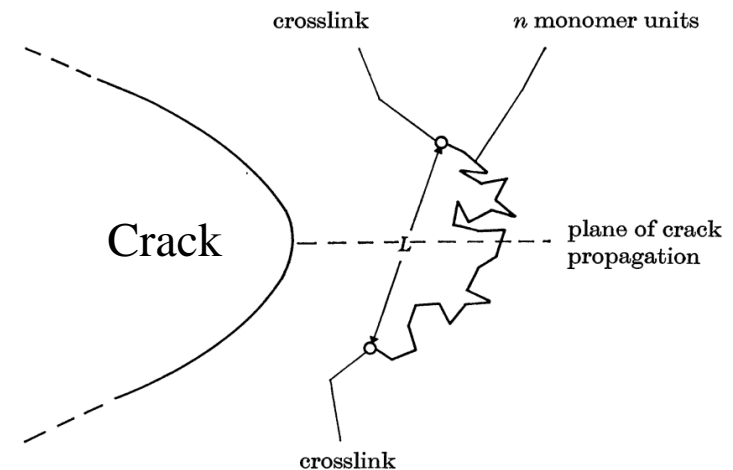
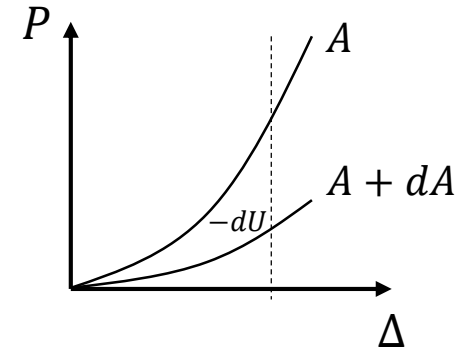
a) The presence of flaws impacts the fracture behavior^[4]

b) Size-dependent fracture in polydimethylsiloxane (PDMS) specimens

Fracture in elastomers

- Occurs when ...
 - Macroscopically, **G reaches Γ**
 - Griffith theory^[5,6]
 - G: Energy release rate
 - Γ : Fracture energy
 - Microscopically, **ε_R reaches ε_R^f**
 - Lake-Thomas theory^[7-9]
 - ε_R : Internal energy
 - ε_R^f : critical internal energy;
bond dissociation energy
- These approaches are compatible (Lake and Thomas ^[7])

$$G = -\frac{dU(P, \Delta)}{dA}$$



Objectives

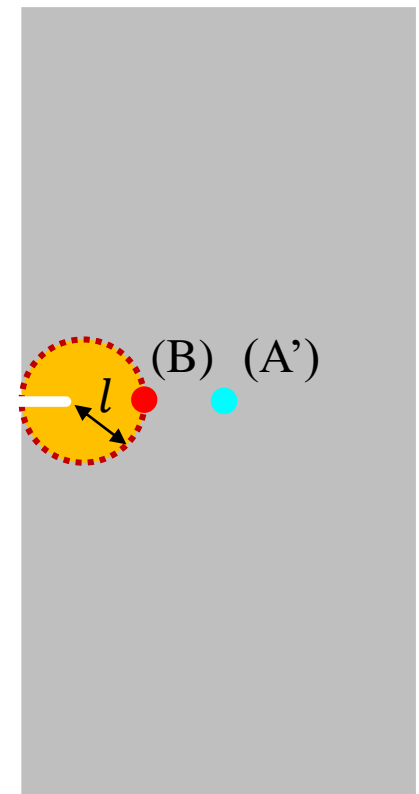
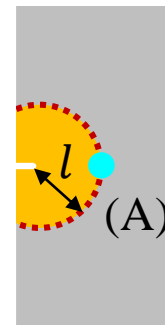
- Predicting the **size-dependent fracture** in elastomers^[1]
 - Experiments and numerical simulations^[2] were carried out
- **Internal energy-driven** fracture criterion; inspired by the Lake-Thomas model^[7-9]
- Using the **phase-field model** rooted in the gradient-damage theory^[2,9-13]
 - Mesh-insensitive crack propagation process
 - The internal energy-driven fracture criterion
 - Thermodynamics of the damage and fracture

Size-dependent fracture & Fracture process zone

- Fracture process zone
 - Where the polymer chains rupture = Where the dissipation mainly occurs
- Stress at point (B) is larger than those at (A) and (A')
 - $\sigma_A = \sigma_{A'} < \sigma_B$
- → Free energy at point (B) is larger than those at (A) and (A')
 - $\psi_A = \psi_{A'} < \psi_B$
- → ψ_B reaches the critical energy earlier than ψ_A
- → **The larger specimen ruptures earlier**

The size of fracture process zone^[1,3,14,15]:

$$l = \frac{\Gamma}{W^*} = \frac{\text{Fracture energy}}{\text{Critical deformation energy}}$$



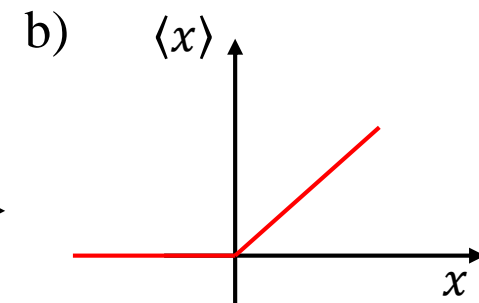
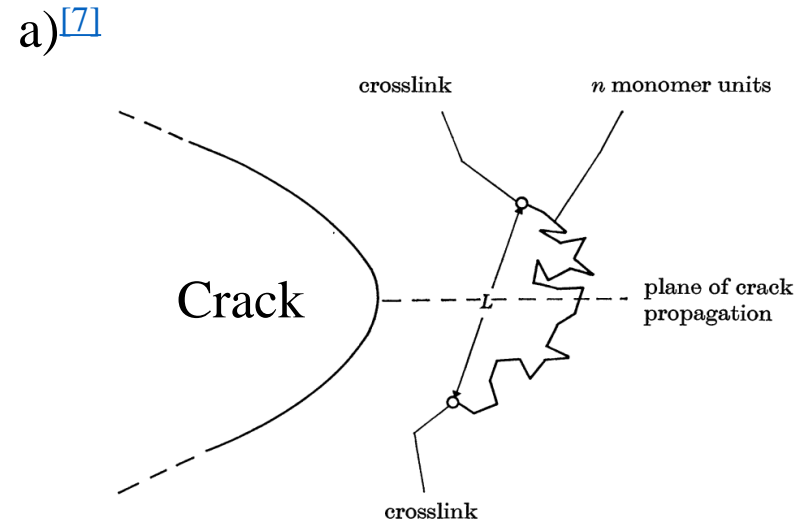
Nonlocal continuum modeling

- 1. The damage $d \in [0,1]$
 - $d=0$: intact
 - $d=1$: fully damaged
- Internal energy-driven** fracture criterion
 - Inspired by the Lake-Thomas model^[5]
 - Fracture = **Scission of polymer chains**
- Governing equations^[9]
 - Macroforce balance $\text{Div } \mathbf{T}_R = 0$
 - Microforce balance $\zeta \dot{d} = 2(1 - d)\mathcal{H}_R - \hat{\varepsilon}_R^f (d - l'^2 \Delta d)$

History function;
the fracture criterion

$$\mathcal{H}_R = \left\langle \underline{\varepsilon}_R^0 - \varepsilon_R^f / 2 \right\rangle, \quad \text{where } \langle x \rangle = \begin{cases} x & \text{if } x > 0 \\ 0 & \text{if } x \leq 0 \end{cases}$$

Internal energy



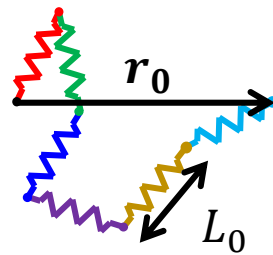
Nonlocal continuum modeling

- **Internal energy** should be considered → **Bond stretch**^[8,9]

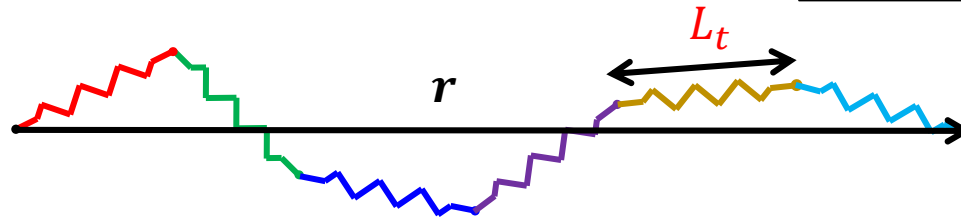
- Deformation = Chain configuration change + stretching of molecular bonds

$$\psi_R = \underbrace{(1-d)^2 \left[\frac{1}{2} N n E_b (\lambda_b - 1)^2 + \frac{1}{2} K (J - 1)^2 \right]}_{(1-d)^2 \varepsilon_R^0; \text{ Damage acts on the internal energy only}} + \underbrace{N k_b \theta n \left[\frac{\bar{\lambda} \lambda_b^{-1}}{\sqrt{n}} \beta + \ln \left(\frac{\beta}{\sinh \beta} \right) \right]}_{-\theta \eta_R; \text{ Entropic energy}} + \underbrace{\frac{1}{2} \varepsilon_R^f l^2 |\nabla d|^2}_{\text{Nonlocal energy}^{[9]}}$$

a) Reference configuration



a) Deformed configuration



$$\bar{\lambda} = \frac{|r|}{|r_0|}$$

$$\lambda_b = \frac{L_t}{L_0}$$

$$\mathcal{L}(x) = \coth x - \frac{1}{x}$$

$$\beta = \mathcal{L}^{-1} \left(\frac{\bar{\lambda} \lambda_b^{-1}}{\sqrt{n}} \right)$$

Nonlocal continuum modeling

- 2. Phase-field model rooted in the gradient-damage theory^[9-13]
 - “Diffusive damage zone”

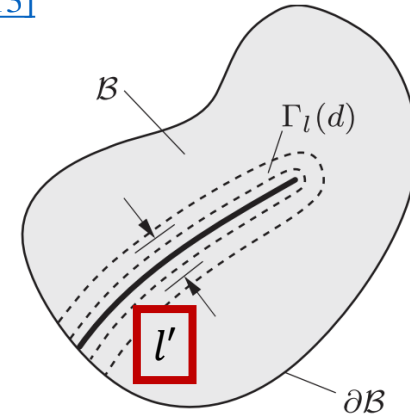
Intrinsic length scale l'

- Microforce balance $\zeta \dot{d} = 2(1 - d)\mathcal{H}_R - \hat{\varepsilon}_R^f (d - l'^2 \Delta d)$

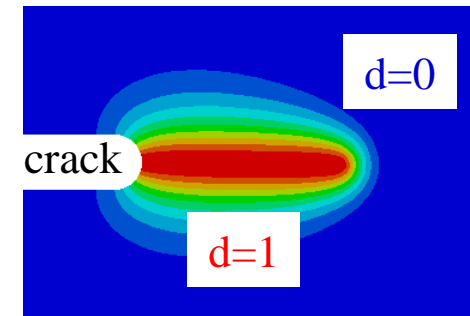
History function;
the fracture criterion

- The **intrinsic length scale l'** \rightarrow the size of diffusive damage zone
 - A numerical parameter; ambiguous physical meaning

a)^[13]



b)



Crack propagation;
at reference configuration

Nonlocal continuum modeling

- Assumption^[1]: Diffusive damage zone = Fracture process zone
 - Regions of the damage evolution and the dissipation

- The size of fracture process zone

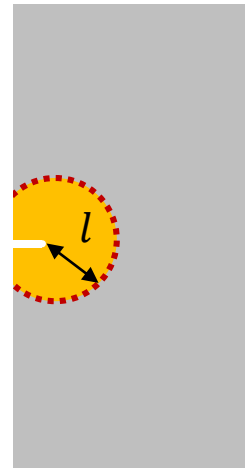
$$= \frac{\Gamma}{W^*} = \frac{\text{Fracture energy}}{\text{Critical deformation energy}} \rightarrow \text{Intrinsic length scale}$$

→ Identify the intrinsic length scale l **from experiments**

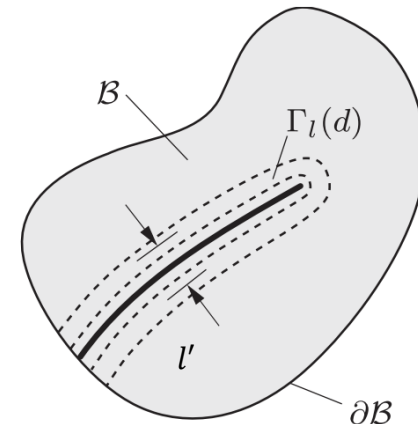
→ Apply to the phase field model

→ Predict the **size-dependent fracture** by numerical simulations^[1]

a) Fracture process zone

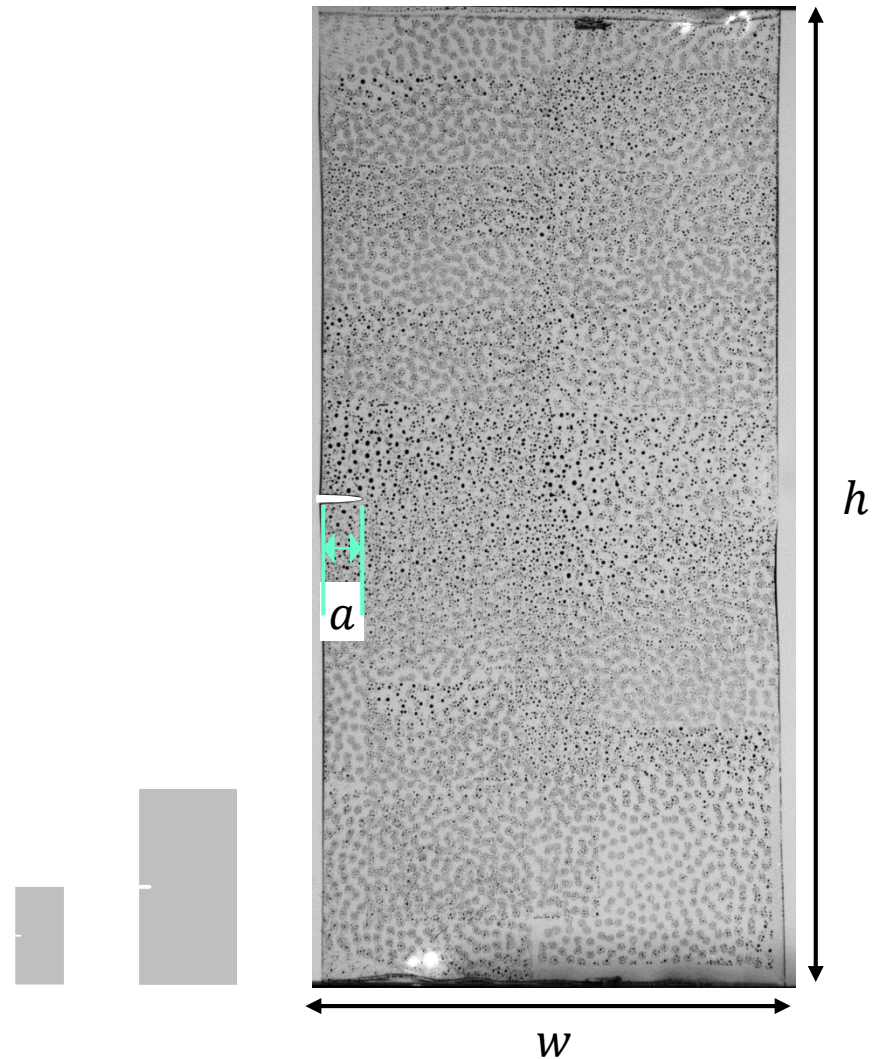


b)^[13] Diffusive damage zone



Experimental procedures^[1]

- Geometries
 - $a = \{0.5, 1, 5\}$ mm
 - $w = 10a$, $h = 20a$, specimen thickness: 0.5mm
 - $w = \{5, 10, 50\}$ mm
 - $h = \{10, 20, 100\}$ mm
- Materials
 - PDMS
 - TangoPlus (3D-printed elastomer)
- Strain rate 0.01 s^{-1} , temperature $\sim 21^\circ\text{C}$
- Digital image correlation (DIC) analysis
 - Strain fields from experiments



The intrinsic length scale l

- $l = \frac{\Gamma}{W^*} \rightarrow$ **Experimentally identified intrinsic length scale**^[1]
- Γ : Fracture energy
 - from **notched** specimens
- W^* : Critical deformation energy
 - from **unnotched** specimens

PDMS

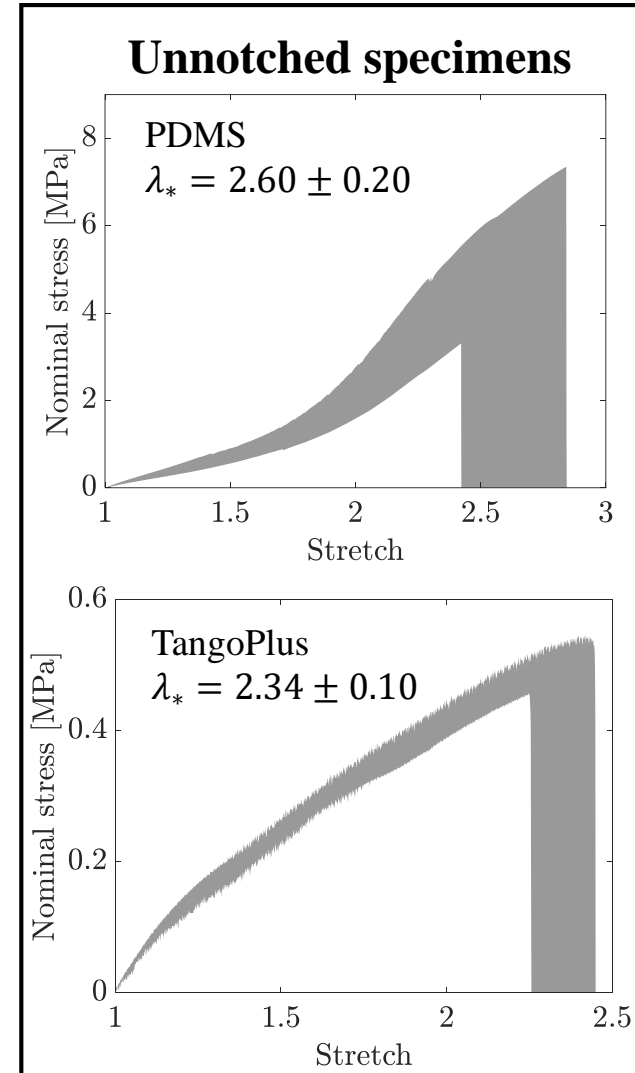
$$\Gamma \approx 0.25 \text{mJ/mm}^2, W^* \approx 2.7 \text{mJ/mm}^3$$

$$\rightarrow l \approx 0.08 \text{mm}$$

TangoPlus

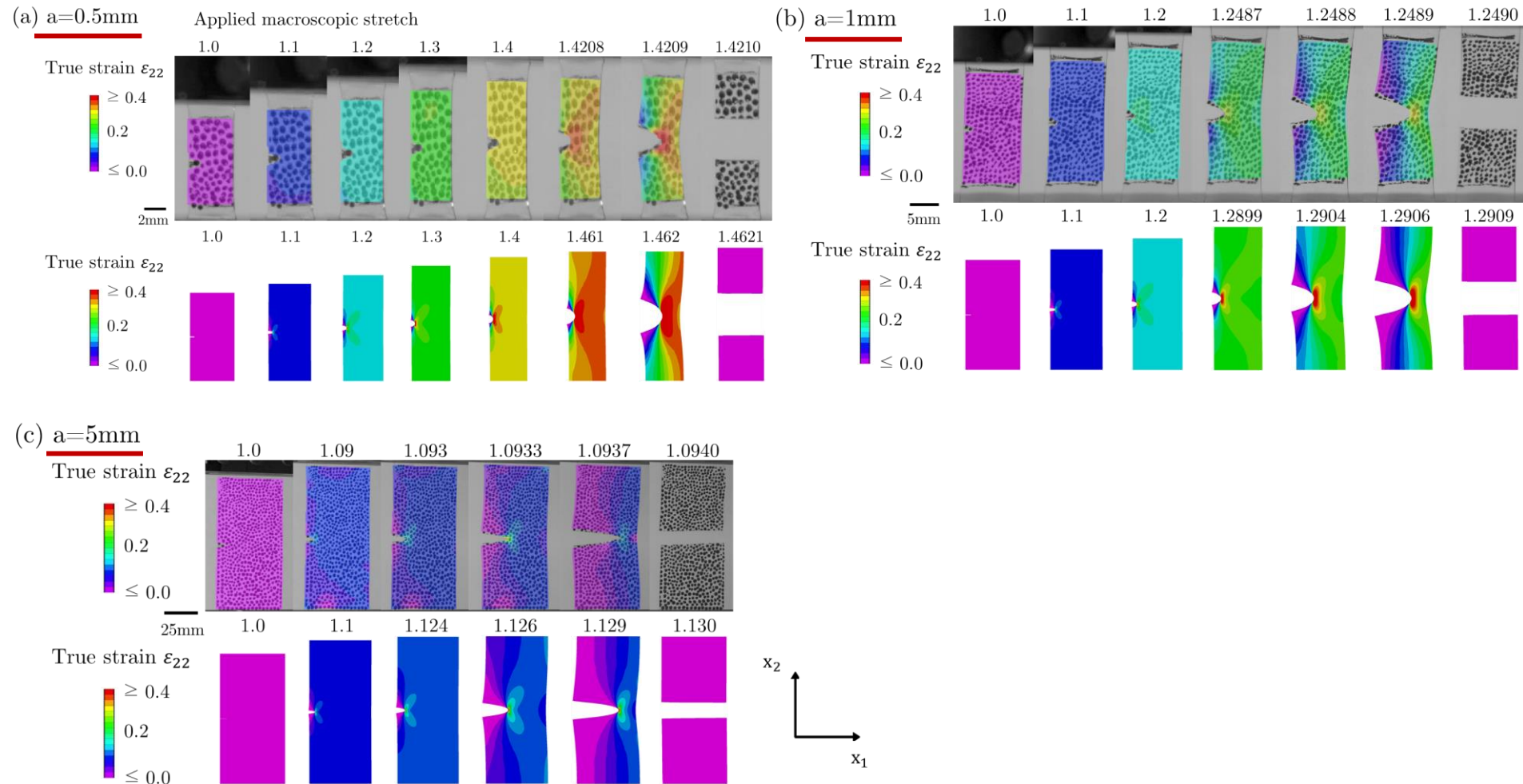
$$\Gamma = 0.5 \text{mJ/mm}^2, W^* \approx 0.45 \text{mJ/mm}^3$$

$$\rightarrow l \approx 1 \text{mm}$$



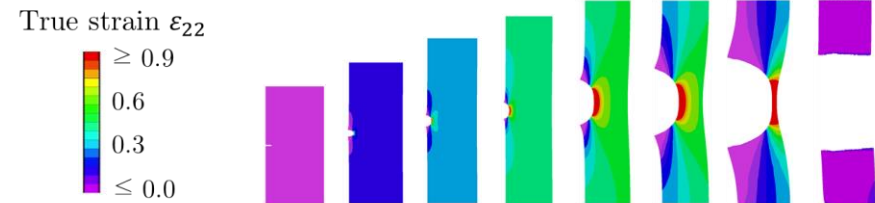
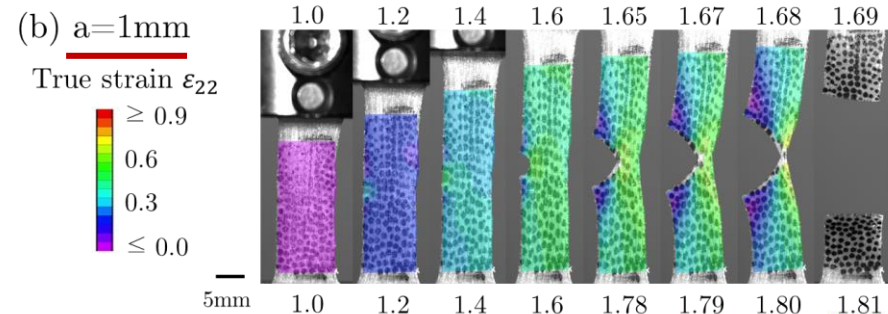
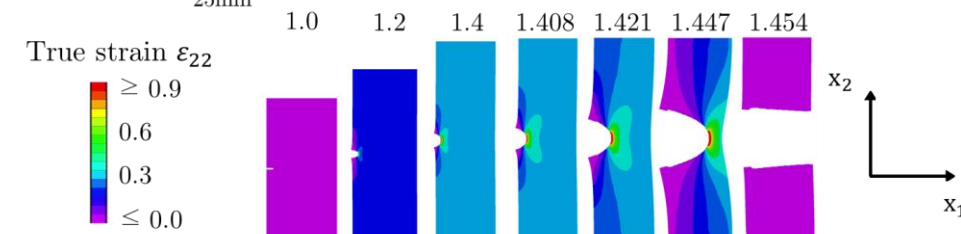
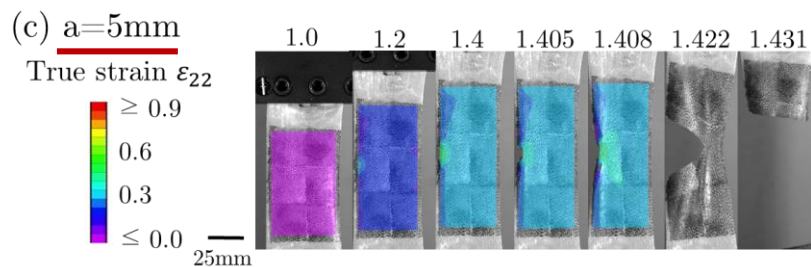
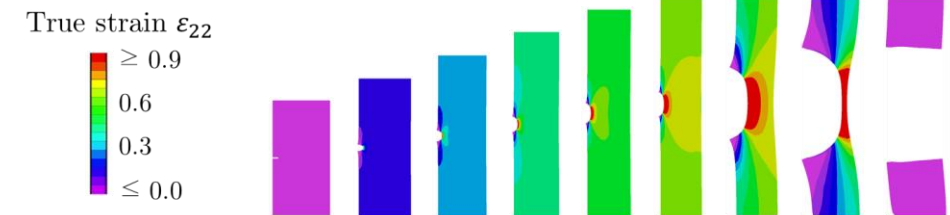
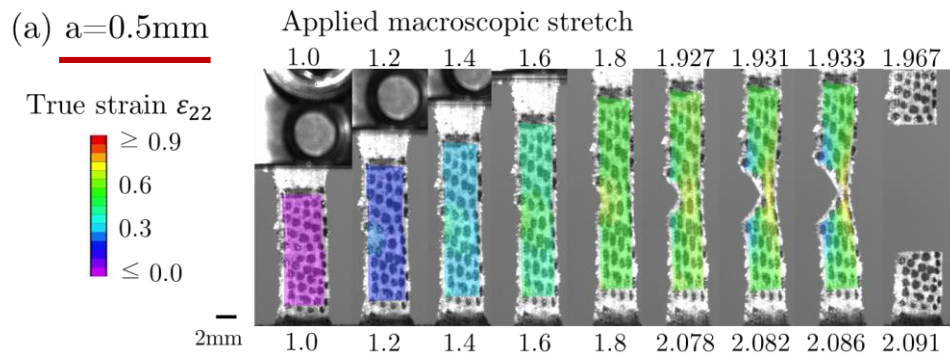
Results: Experiment vs. Numerical simulation^[1]

- Strain fields in **PDMS** specimens ($l = 0.08mm$)
 - Larger specimen ruptures earlier



Results: Experiment vs. Numerical simulation^[1]

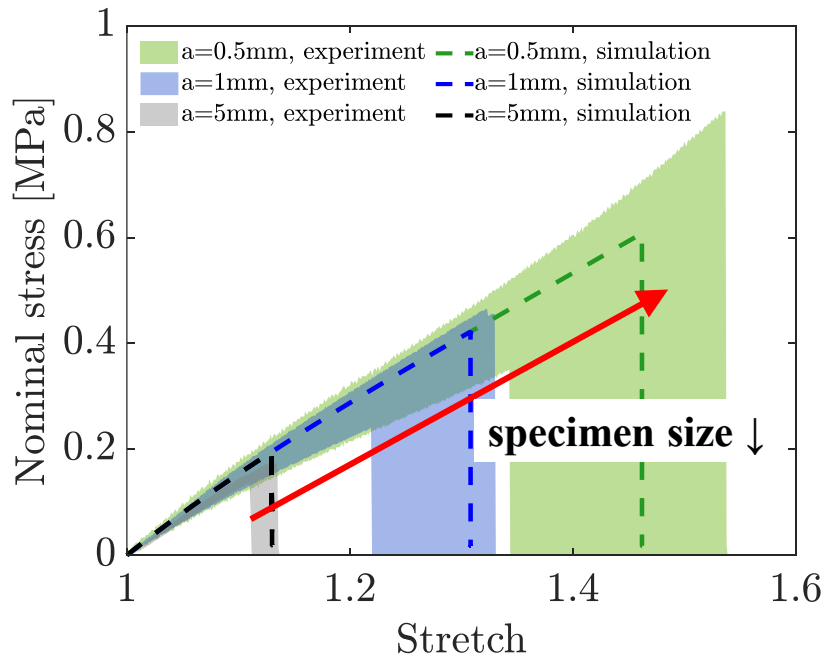
- Strain fields in **TangoPlus** specimens ($l = 1mm$)
 - Larger specimen ruptures earlier



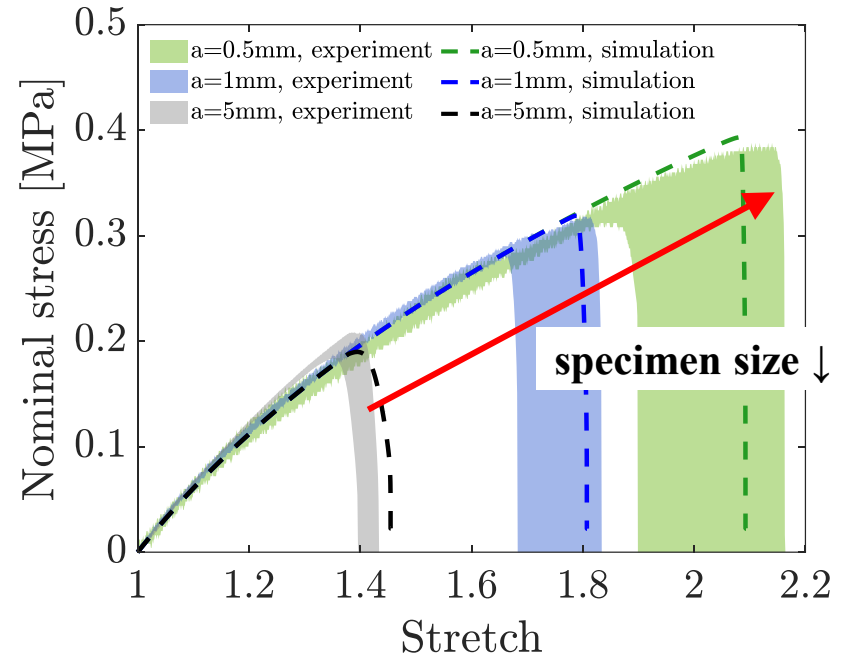
Results: Experiment vs. Numerical simulation^[1]

- Notch lengths $a = \{0.5, 1, 5\}$ mm
- Geometric similarity \rightarrow **Identical initial stress-stretch response**
- Smaller notch length \rightarrow Higher rupture stretch

a) PDMS



b) TangoPlus



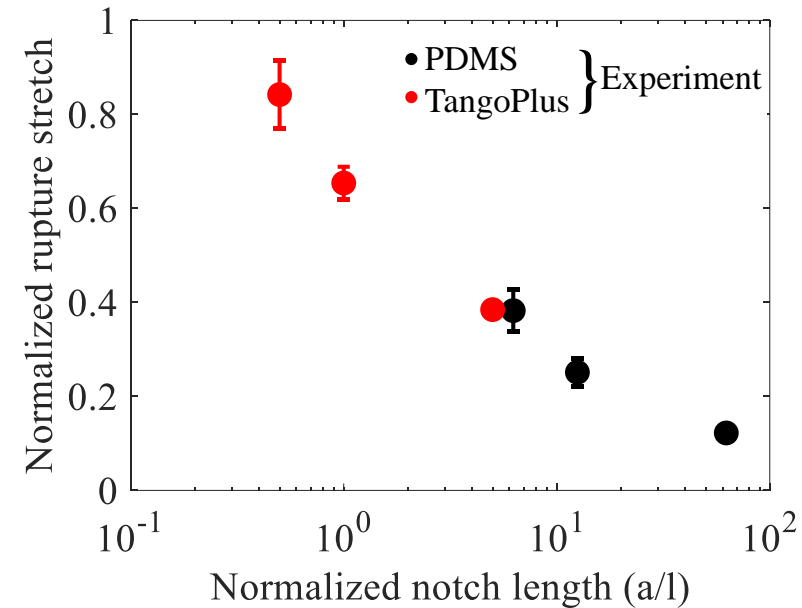
Notch-length sensitivity^[1]

- PDMS vs. TangoPlus; same specimen sizes
 - PDMS: $l = 0.08\text{mm}$
 - TangoPlus: $l = 1\text{mm}$
-) **More than 10 times**

- Normalized rupture stretch

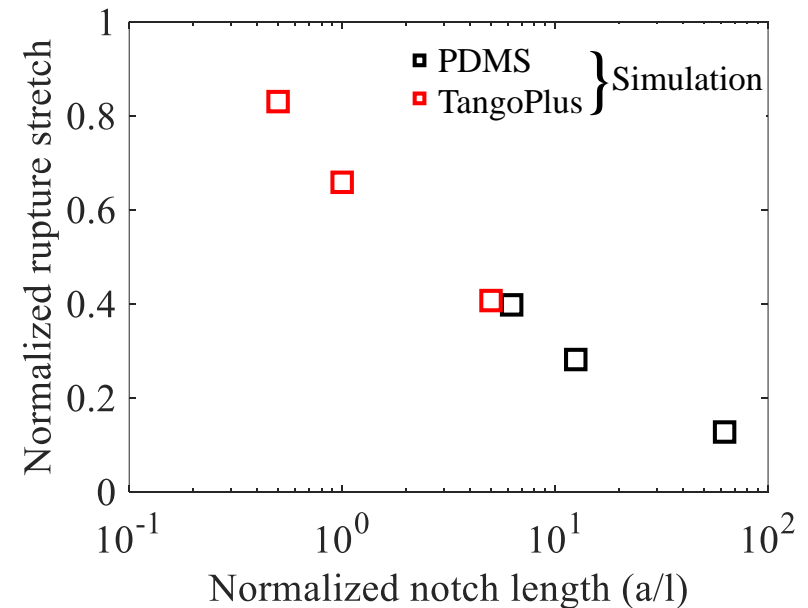
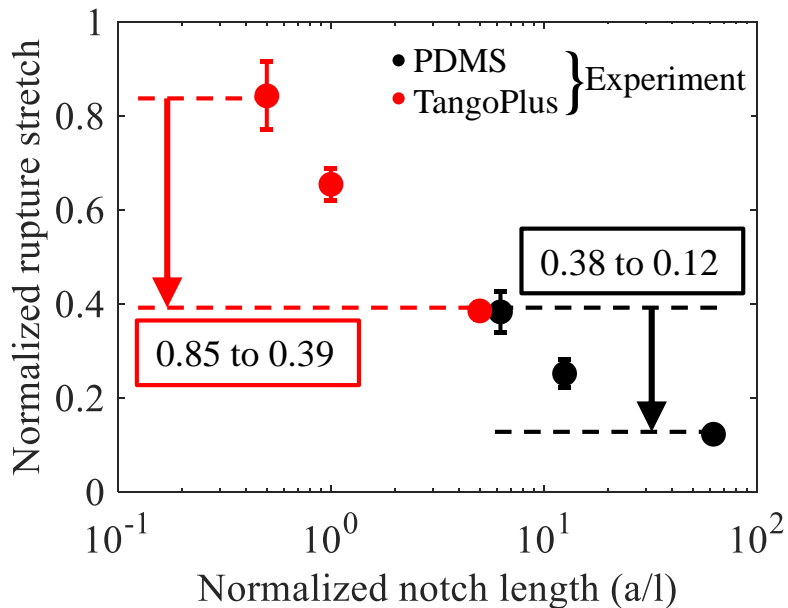
$$= \frac{\text{Rupture stretch of **notched** specimens}}{\text{Rupture stretch of **unnotched** specimens}}$$

- Normalized notch length = $\frac{\text{Notch length (a)}}{\text{Intrinsic length scale (l)}}$



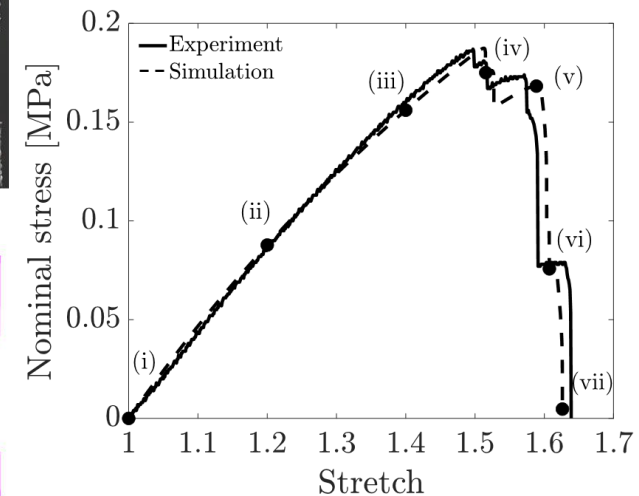
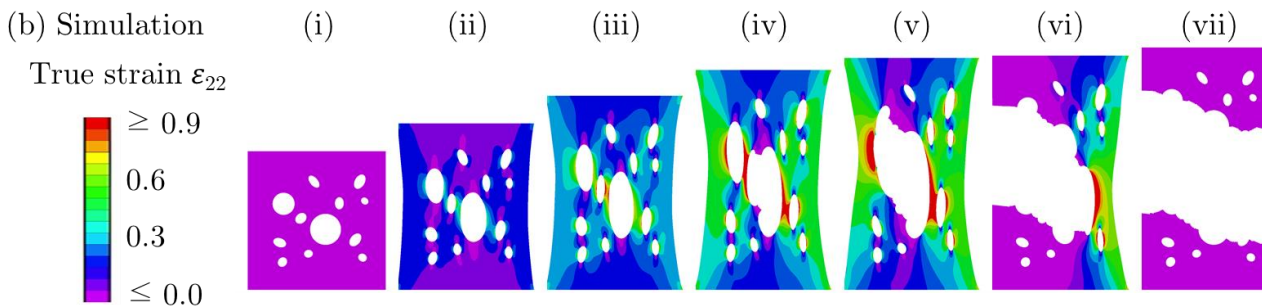
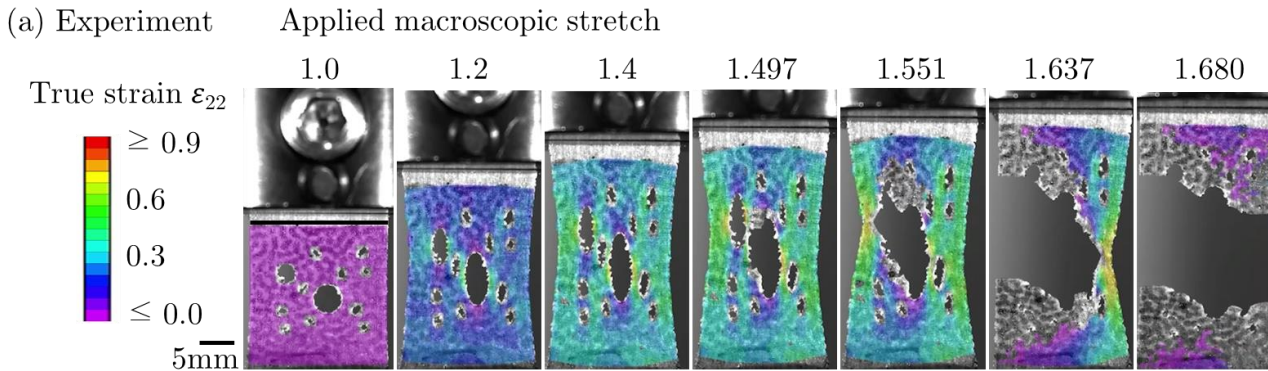
Notch-length sensitivity^[1]

- PDMS vs. TangoPlus; same specimen sizes
 - PDMS: $l = 0.08\text{mm}$
 - TangoPlus: $l = 1\text{mm}$
) **More than 10 times**
- $a/l: 0.5 \sim 5$ (TangoPlus; $l = 1\text{mm}$) \rightarrow Highly notch length-sensitive
- $a/l: 5 \sim 50$ (PDMS; $l = 0.08\text{mm}$) \rightarrow Less notch length-sensitive



Randomly perforated specimen (TangoPlus)^[1]

- Nicely predicted the response **without modification of parameters**
 - Progressive fracture of ligaments

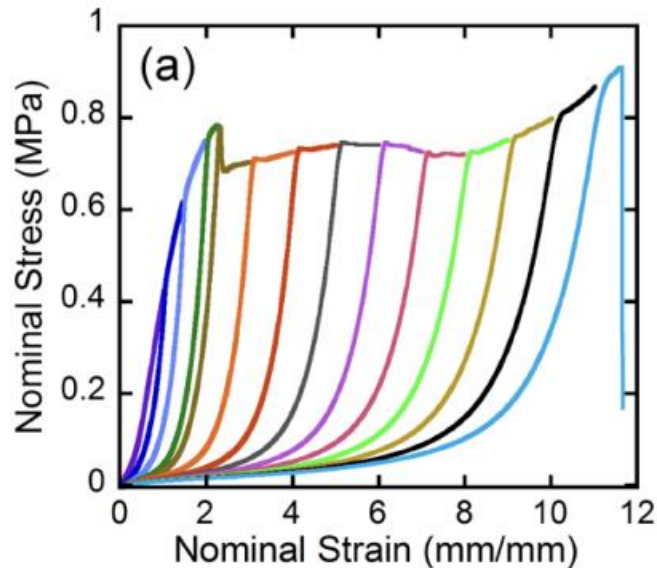


Conclusion

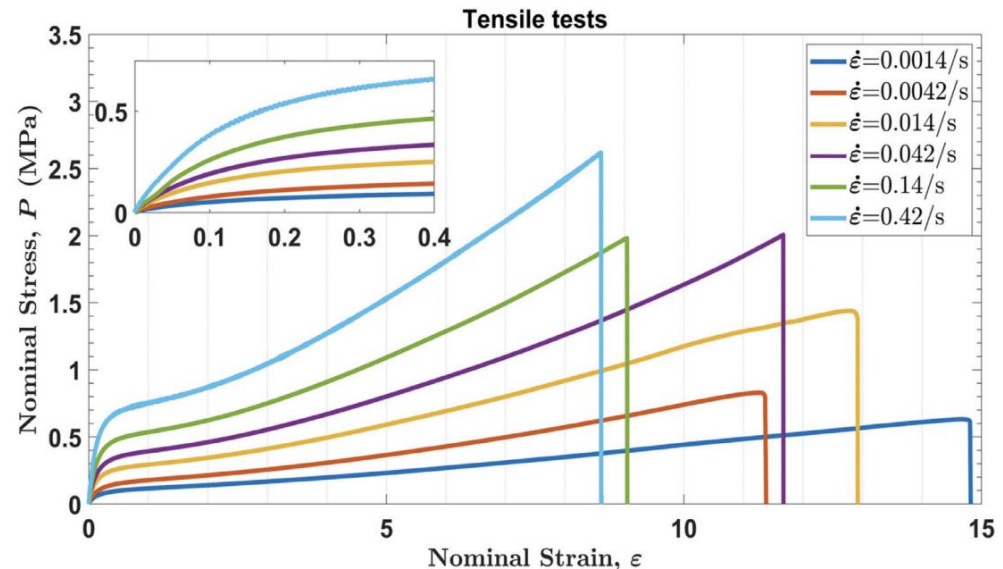
- **Size-dependent fracture** is clearly observed in experiments^[1]
 - Rupture stretch increases as the notch length decreases
 - Size-dependence increases as the notch-root radius decreases
- **The intrinsic length scale** is central to account for the size-dependent behavior^[1]
 - The intrinsic length scale l defines the size of diffusive damage zone / fracture process zone
 - The intrinsic length scales were identified from experiments
 - Normalized notch length (a/l) determines the size-dependence
- **Nonlocal continuum model**^[2,9] nicely predicted the fracture in elastomers^[1]
 - Nonlocal continuum model utilizes experimentally identified intrinsic length scales
 - The model captures the size-dependent fracture in elastomers
 - The model is capable of predicting the fracture of complex geometries

Future work

- Fracture involving non-trivial dissipation
 - Mullins effect [\[16-20\]](#)
 - Is the fracture behavior influenced by the rate-independent dissipation (e.g., the Mullins effect) ?
 - Viscous dissipation [\[16-19,21\]](#)
 - How to describe complicated deformation and fracture behaviors in polymers?



a) Fracture in double-network elastomers; the Mullins effect and fracture occur [\[20\]](#)



b) Rate-dependent deformation and fracture behaviors in a hydrogel (polyampholyte gel) [\[21\]](#)

References

- [1] Lee, J., Lee, J., Yun, S., Kim, S., Lee, H., Chester, S. A., & Cho, H. (2024). Size-dependent fracture in elastomers: Experiments and continuum modeling. [*Physical Review Materials*, 8\(11\), 115602.](#)
- [2] Lee, J., Lee, S., Chester, S. A., & Cho, H. (2023). Finite element implementation of a gradient-damage theory for fracture in elastomeric materials. [*International Journal of Solids and Structures*, 279, 112309.](#)
- [3] Chen, C., Wang, Z., & Suo, Z. (2017). Flaw sensitivity of highly stretchable materials. [*Extreme Mechanics Letters*, 10, 50-57.](#)
- [4] Pharr, M., Sun, J. Y., & Suo, Z. (2012). Rupture of a highly stretchable acrylic dielectric elastomer. [*Journal of Applied Physics*, 111\(10\).](#)
- [5] Griffith, A. A. (1921). VI. The phenomena of rupture and flow in solids. [*Philosophical transactions of the royal society of london. Series A, containing papers of a mathematical or physical character*, 221\(582-593\), 163-198.](#)
- [6] Rivlin, R. S., & Thomas, A. G. (1953). Rupture of rubber. I. Characteristic energy for tearing. [*Journal of polymer science*, 10\(3\), 291-318.](#)
- [7] Lake, G. J., & Thomas, A. G. (1967). The strength of highly elastic materials. [*Proceedings of the Royal Society of London. Series A. Mathematical and Physical Sciences*, 300\(1460\), 108-119.](#)
- [8] Mao, Y., Talamini, B., & Anand, L. (2017). Rupture of polymers by chain scission. [*Extreme Mechanics Letters*, 13, 17-24.](#)
- [9] Talamini, B., Mao, Y., & Anand, L. (2018). Progressive damage and rupture in polymers. [*Journal of the Mechanics and Physics of Solids*, 111, 434-457.](#)

References

- [10] Peerlings, R. H., de Borst, R., Brekelmans, W. M., & de Vree, J. (1996). Gradient enhanced damage for quasi-brittle materials. [*International Journal for numerical methods in engineering*, 39\(19\), 3391-3403.](#)
- [11] De Borst, R., Pamin, J., & Geers, M. G. (1999). On coupled gradient-dependent plasticity and damage theories with a view to localization analysis. [*European Journal of Mechanics-A/Solids*, 18\(6\), 939-962.](#)
- [12] Francfort, G. A., & Marigo, J. J. (1998). Revisiting brittle fracture as an energy minimization problem. [*Journal of the Mechanics and Physics of Solids*, 46\(8\), 1319-1342.](#)
- [13] Miehe, C., Welschinger, F., & Hofacker, M. (2010). Thermodynamically consistent phase-field models of fracture: Variational principles and multi-field FE implementations. [*International journal for numerical methods in engineering*, 83\(10\), 1273-1311.](#)
- [14] Bažant, Z. P. (1997). Scaling of quasibrittle fracture: asymptotic analysis. [*International Journal of Fracture*, 83, 19-40.](#)
- [15] Yang, C., Yin, T., & Suo, Z. (2019). Polyacrylamide hydrogels. I. Network imperfection. [*Journal of the Mechanics and Physics of Solids*, 131, 43-55.](#)
- [16] Cho, H., Rinaldi, R. G., & Boyce, M. C. (2013). Constitutive modeling of the rate-dependent resilient and dissipative large deformation behavior of a segmented copolymer polyurea. [*Soft Matter*, 9\(27\), 6319-6330.](#)
- [17] Cho, H., Mayer, S., Pöselt, E., Susoff, M., in't Veld, P. J., Rutledge, G. C., & Boyce, M. C. (2017). Deformation mechanisms of thermoplastic elastomers: Stress-strain behavior and constitutive modeling. [*Polymer*, 128, 87-99.](#)
- [18] Lee, J., Veysset, D., Hsieh, A. J., Rutledge, G. C., & Cho, H. (2023). A polyurethane-urea elastomer at low to extreme strain rates. [*International Journal of Solids and Structures*, 280, 112360.](#)

References

- [19] Cho, H., Lee, J., Moon, J., Pöselt, E., Rutledge, G. C., & Boyce, M. C. (2024). Large strain micromechanics of thermoplastic elastomers with random microstructures. [*Journal of the Mechanics and Physics of Solids*, 187, 105615.](#)
- [20] Nakajima, T., Fukuda, Y., Kurokawa, T., Sakai, T., Chung, U. I., & Gong, J. P. (2013). Synthesis and fracture process analysis of double network hydrogels with a well-defined first network. [*ACS Macro Letters*, 2\(6\), 518-521.](#)
- [21] Venkata, S. P., Cui, K., Guo, J., Zehnder, A. T., Gong, J. P., & Hui, C. Y. (2021). Constitutive modeling of bond breaking and healing kinetics of physical Polyampholyte (PA) gel. [*Extreme Mechanics Letters*, 43, 101184.](#)