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

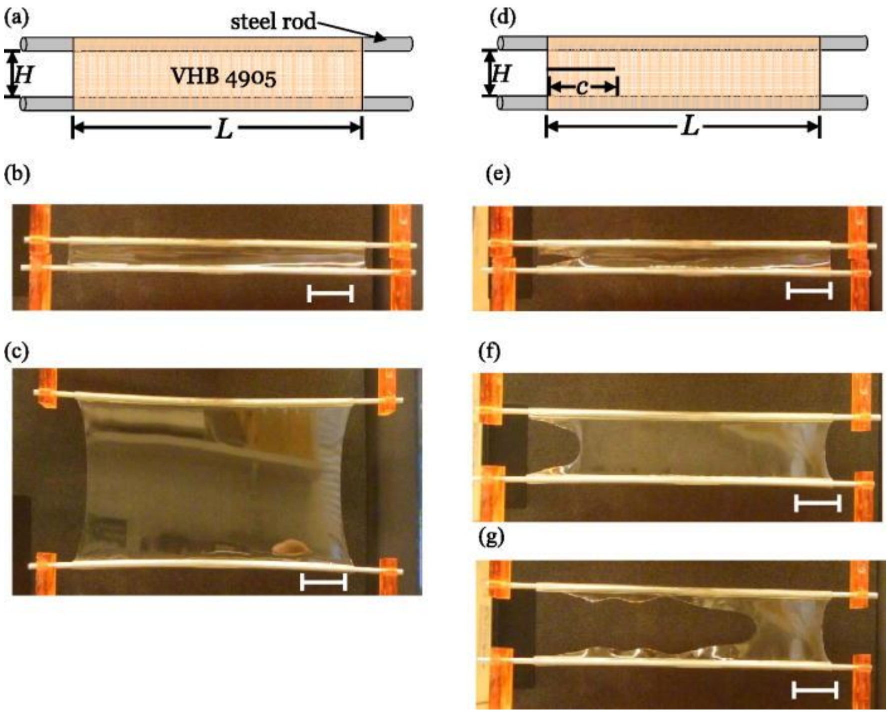
Jeongun Lee, Jaehee Lee and Hansohl Cho*

Korea Advanced Institute of Science and Technology

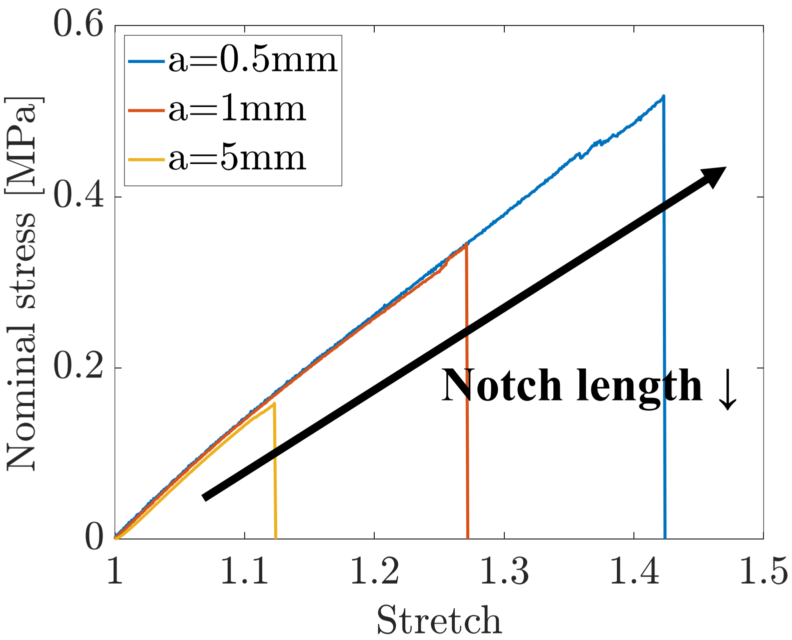
* hansohl@kaist.ac.kr

Fracture in elastomers

- Extreme, nonlinear deformation → 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]

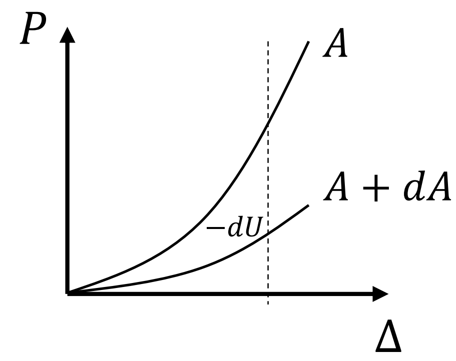


b) Size-dependent fracture in polydimethylsiloxane (PDMS) specimens

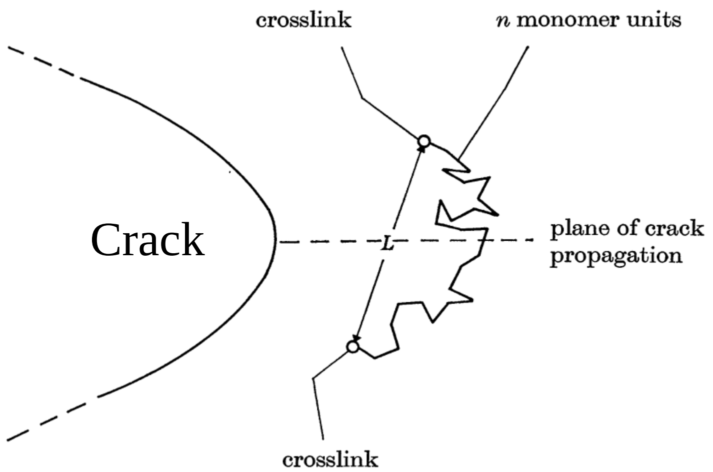
Fracture in elastomers

- Occurs when …
  - Macroscopically, **G reaches Γ**
    - Griffith theory[5,6]
    - G: Energy release rate
    - Γ: Fracture energy

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



  - Microscopically, $\boldsymbol{\varepsilon_R}$ **reaches** $\boldsymbol{\varepsilon_R^f}$
    - 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])

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

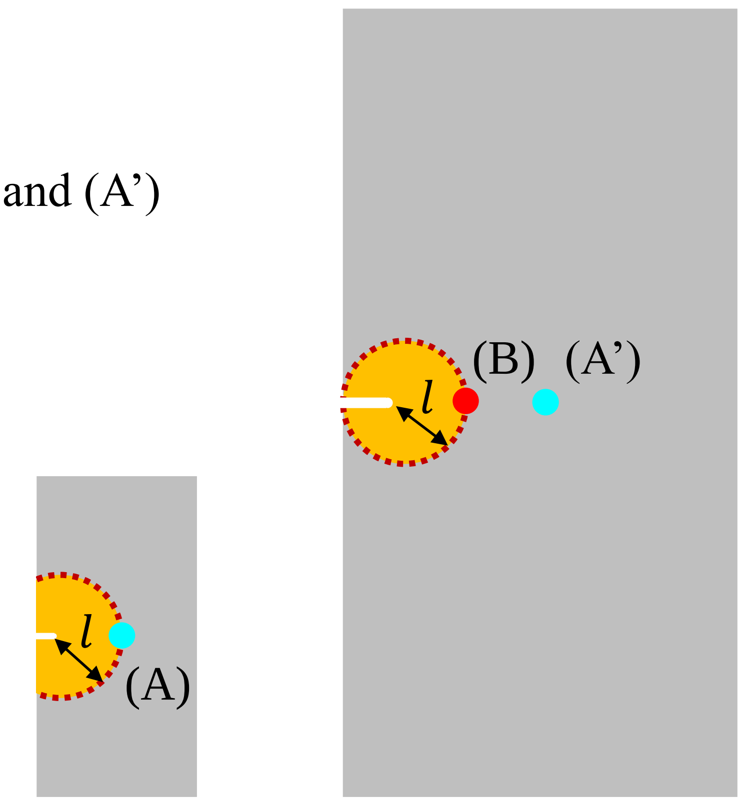
Size-dependent fracture & Fracture process zone

- 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')
  - $\psi_A = \psi_{A'} < \psi_B$
- → **$\boldsymbol{\psi_B}$ reaches the critical energy earlier than $\boldsymbol{\psi_A}$**
- → **The larger specimen ruptures earlier**

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

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



Nonlocal continuum modeling

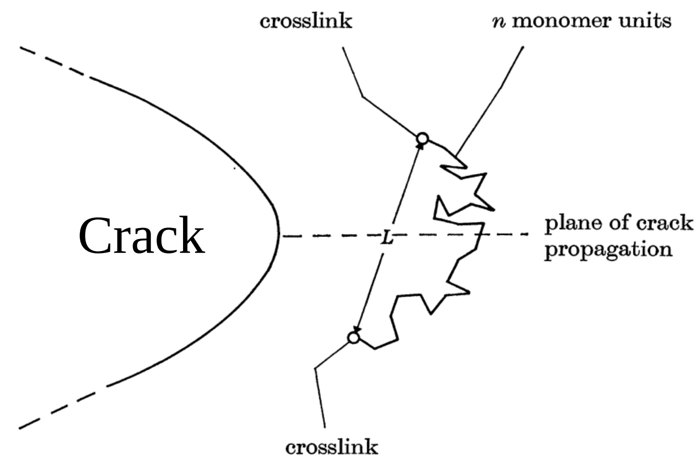
- 1. The damage $d \in [0,1]$
  - d=0: intact
  - d=1: fully damaged
- **Internal energy-driven** fracture criterion
  - Inspired by the Lake-Thomas model[5]
  - Fracture = **Scission of polymer chains**
- Governing equations[9]
  - Macroforce balance Div $\mathbf{T}_{\mathrm{R}}=0$
  - Microforce balance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

History function;
the fracture criterion

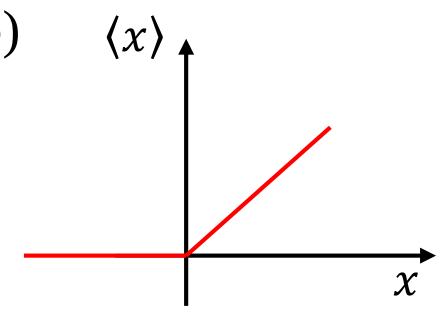
$$\mathcal{H}_R = \left\langle \underline{\varepsilon^0_R} - \varepsilon^f_R/2 \right\rangle, \quad \text{where } \langle x\rangle = \begin{cases} x & if\ x > 0 \\ 0 & if\ x \le 0 \end{cases}$$

Internal energy

a)[7]



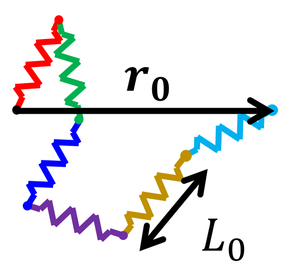
b)



Nonlocal continuum modeling

- **Internal energy** should be considered → **Bond stretch**[8,9]
  - Deformation = Chain configuration change + stretching of molecular bonds
- $$\psi_R = \underbrace{(1-d)^2\left[\frac{1}{2}NnE_b(\lambda_b-1)^2+\frac{1}{2}K(J-1)^2\right]}_{(1-d)^2\varepsilon_R^0;\ \textbf{Damage acts on the internal energy only}} + \underbrace{Nk_b\theta n\left[\frac{\bar{\lambda}\lambda_b^{-1}}{\sqrt{n}}\beta+\ln\left(\frac{\beta}{\sinh\beta}\right)\right]}_{-\theta\eta_R;\ \text{Entropic energy}} + \underbrace{\frac{1}{2}\varepsilon_R^f l^2|\nabla d|^2}_{\text{Nonlocal energy[9]}}$$

a) Reference configuration



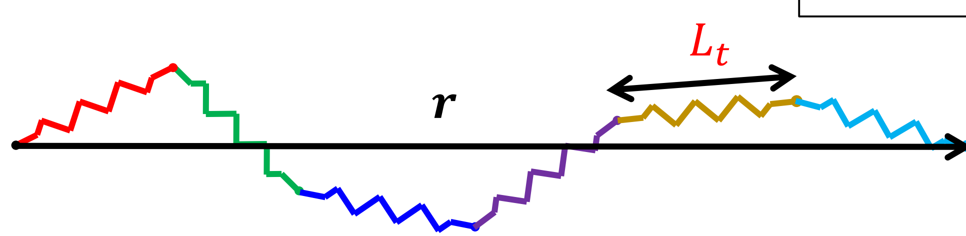
$$\bar{\lambda} = \frac{|\boldsymbol{r}|}{|\boldsymbol{r}_0|}$$

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

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

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

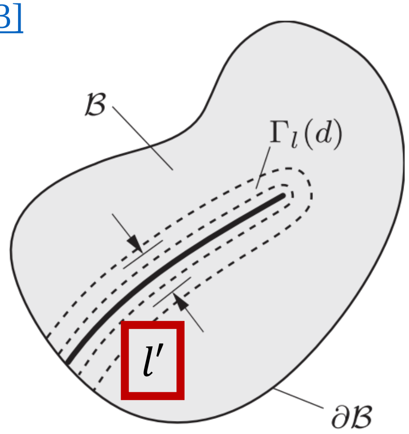
a) Deformed configuration



Nonlocal continuum modeling

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

a)[13]



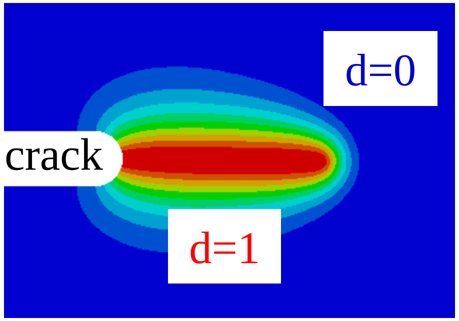
Intrinsic length scale l'

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

History function;
the fracture criterion

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

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

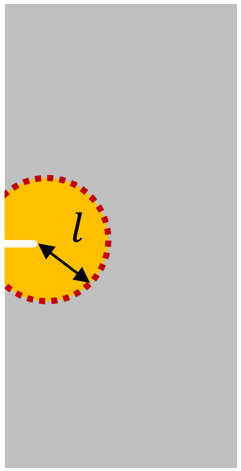
$$= \frac{\Gamma}{W^*} = \frac{Fracture\ energy}{Critical\ deformation\ energy}$$ → **Intrinsic length scale**

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

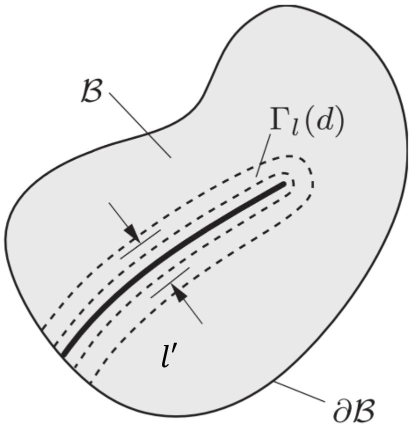
→ Apply to the phase field model

→ 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

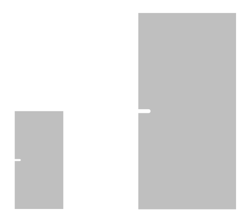
    → h = {10, 20, 100} mm

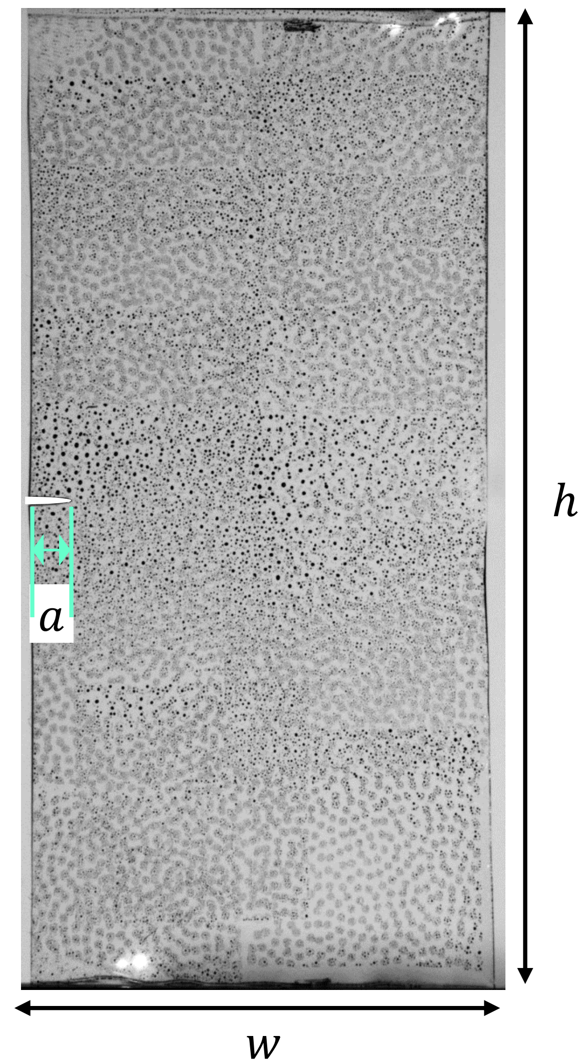
- Materials
  - PDMS
  - TangoPlus (3D-printed elastomer)

- Strain rate 0.01 s^{-1}, temperature ~21℃

- Digital image correlation (DIC) analysis

  → Strain fields from experiments





The intrinsic length scale $\boldsymbol{l}$

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

PDMS

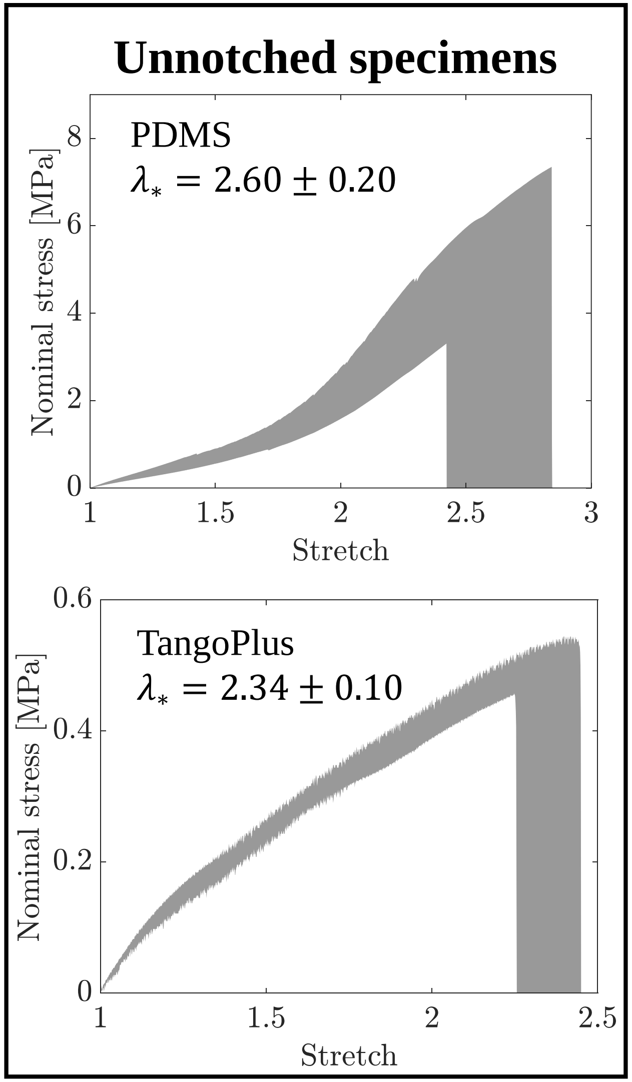
$\Gamma \approx 0.25 \mathrm{mJ/mm^2}$, $W^* \approx 2.7 \mathrm{mJ/mm^3}$

→ $\boldsymbol{l \approx 0.08mm}$

TangoPlus

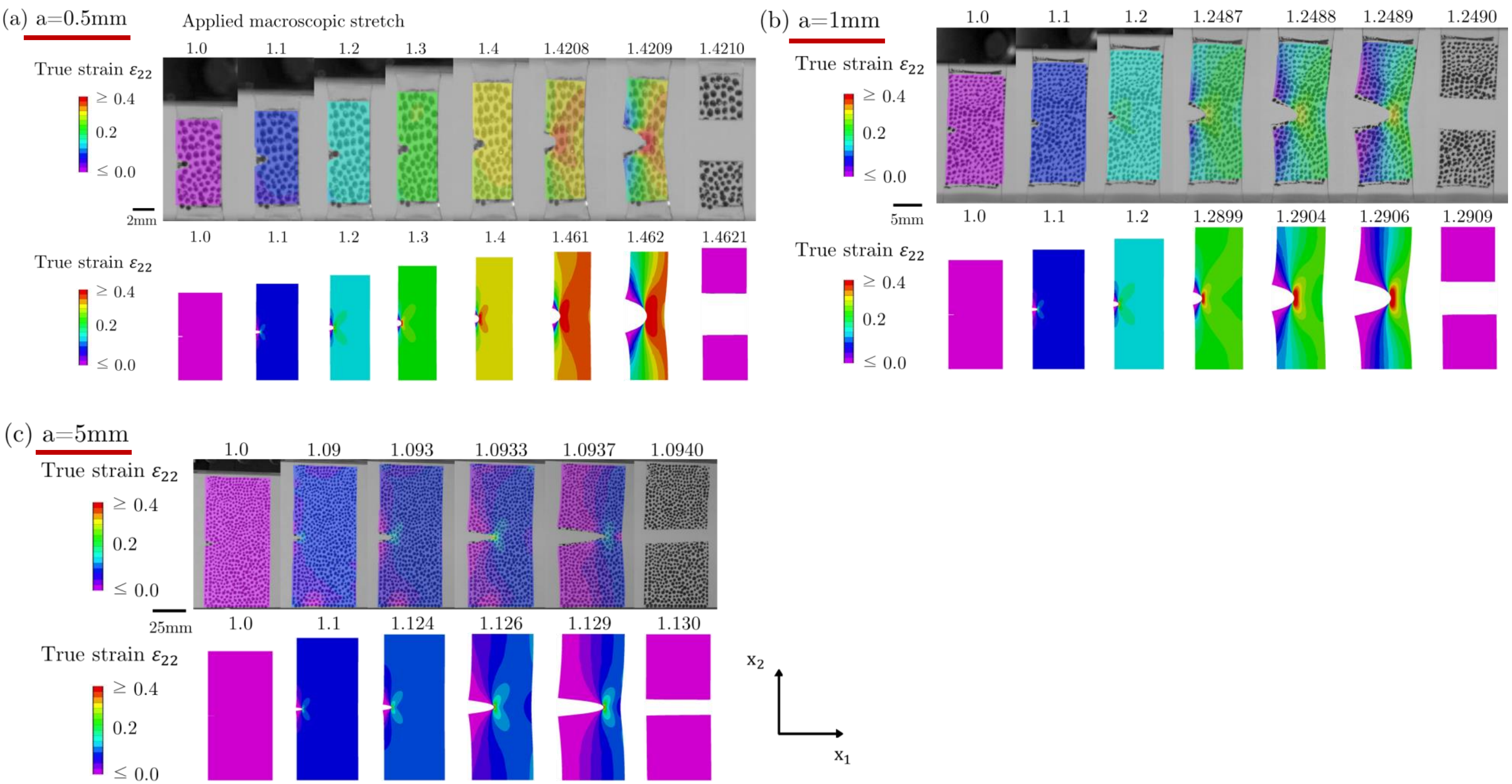
$\Gamma = 0.5 \mathrm{mJ/mm^2}$, $W^* \approx 0.45 \mathrm{mJ/mm^3}$

→ $\boldsymbol{l \approx 1mm}$



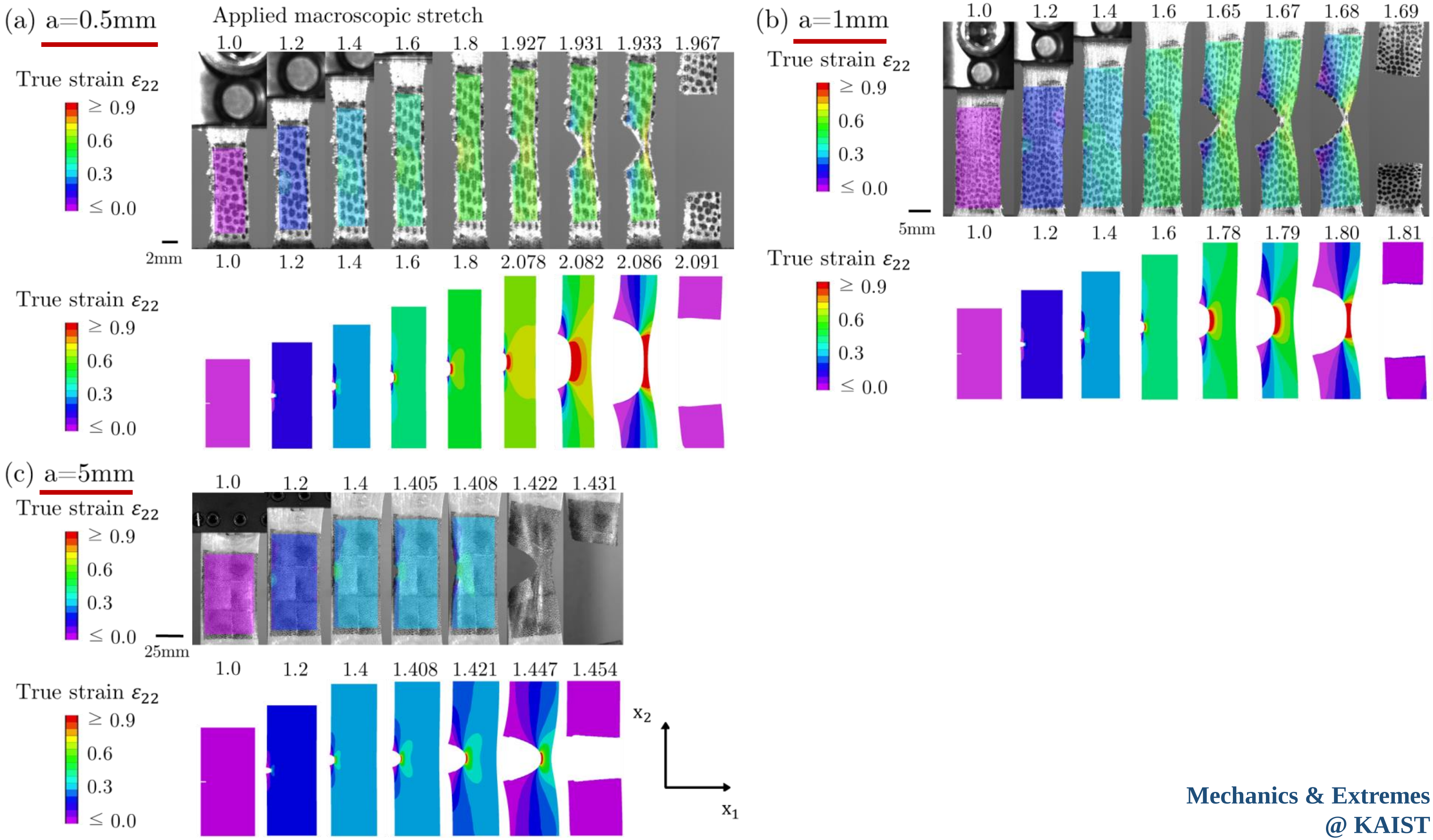
Results: Experiment vs. Numerical simulation[1]

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



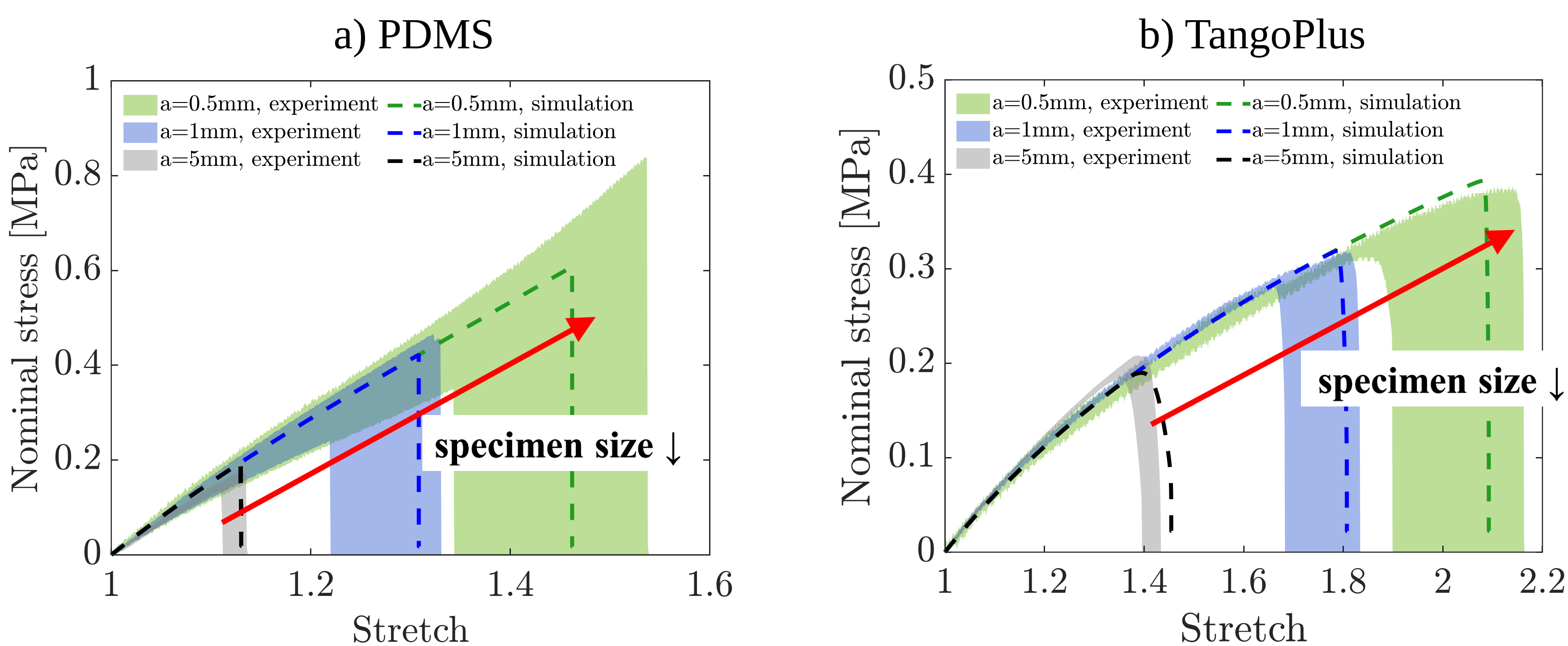
Results: Experiment vs. Numerical simulation[1]

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



Results: Experiment vs. Numerical simulation[1]

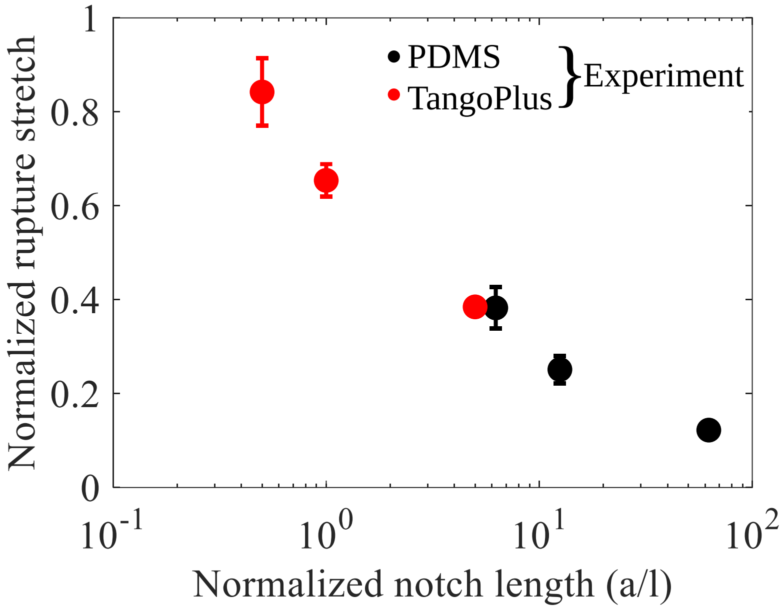
- Notch lengths a = {0.5, 1, 5} mm
- Geometric similarity → **Identical initial stress-stretch response**
- Smaller notch length → Higher rupture stretch



Notch-length sensitivity[1]

- PDMS vs. TangoPlus; same specimen sizes
  - PDMS: $l = 0.08mm$
  - TangoPlus: $l = 1mm$

More than 10 times



- Normalized rupture stretch

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

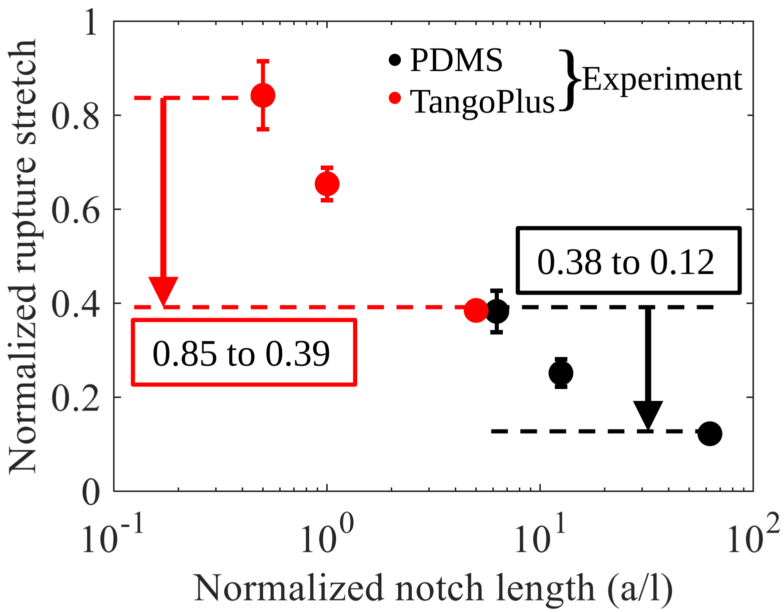
- Normalized notch length $= \dfrac{\text{Notch length (a)}}{\text{Intrinsic length scale } (l)}$

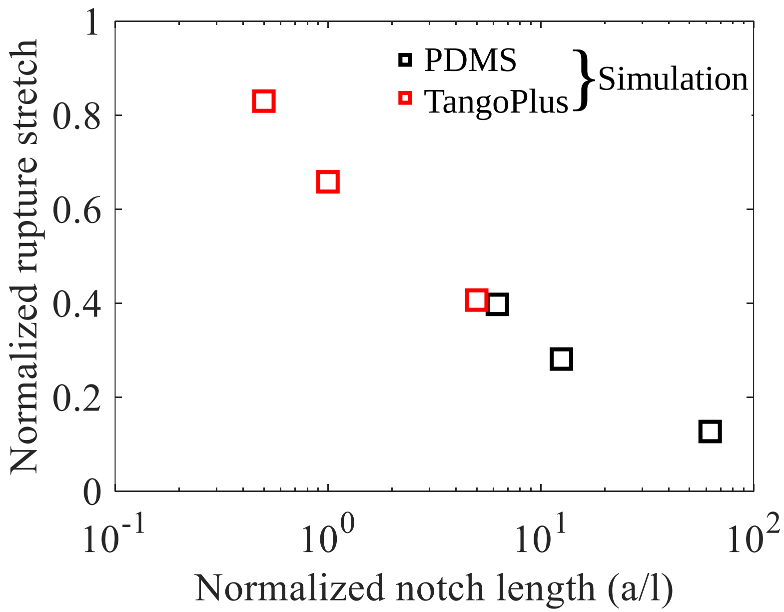
Notch-length sensitivity[1]

- PDMS vs. TangoPlus; same specimen sizes
  - PDMS: $l = 0.08mm$
  - TangoPlus: $l = 1mm$

More than 10 times

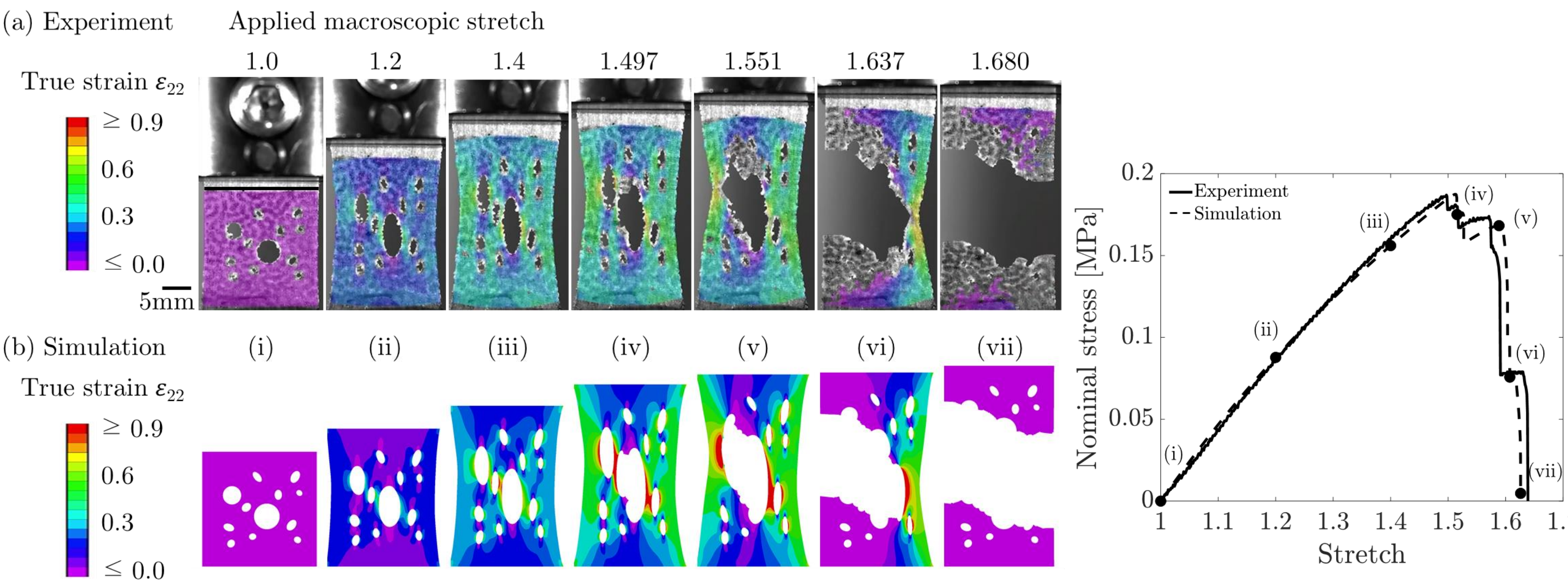
- a/l: 0.5~5 (TangoPlus; $l = 1mm$) → Highly notch length-sensitive
- a/l: 5~50 (PDMS; $l = 0.08mm$) → Less notch length-sensitive





Randomly perforated specimen (TangoPlus)[1]

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



Conclusion

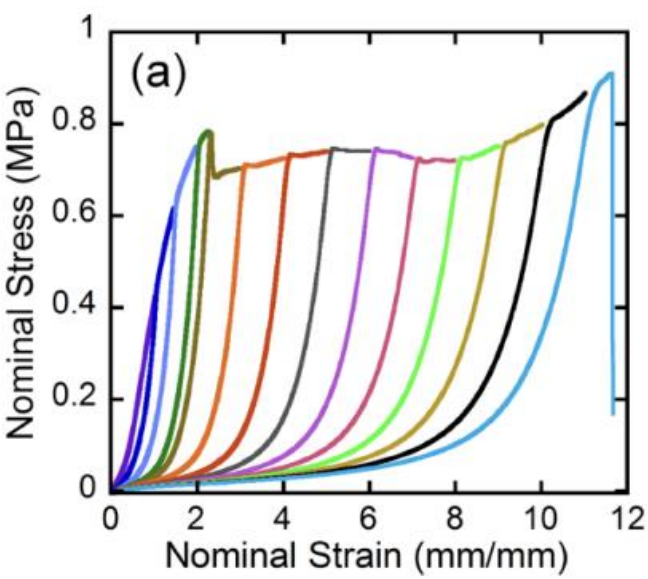
- **Size-dependent fracture** is clearly observed in experiments[1]
  - Rupture stretch increases as the notch length decreases
  - Size-dependence increases as the notch-root radius decreases
- **The intrinsic length scale** is central to account for the size-dependent behavior[1]
  - The intrinsic 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

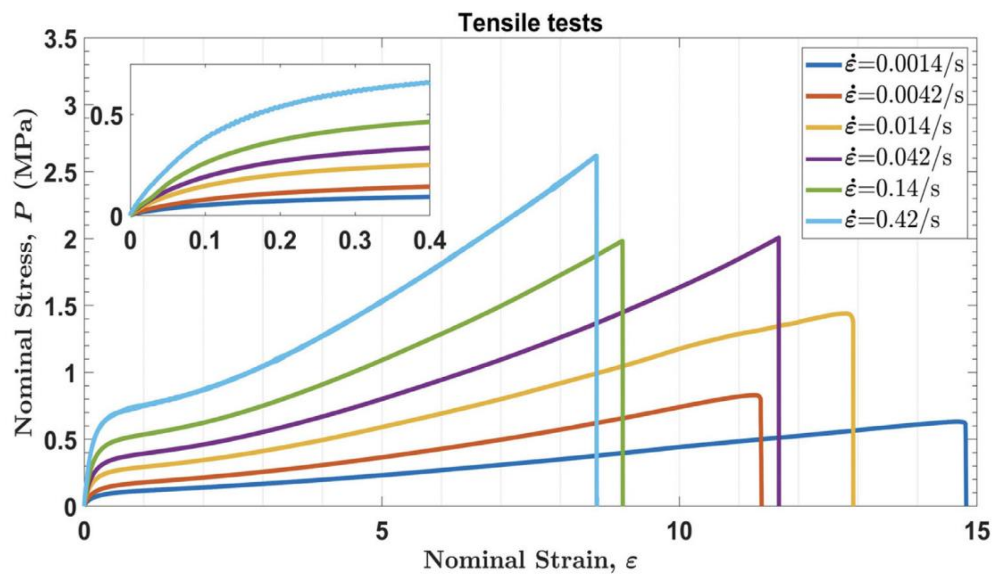
- Fracture involving non-trivial dissipation
  - Mullins effect[16-20]

    → 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., Chester, S. A., & Cho, H. (2024). Size-dependent fracture in elastomers: experiments and continuum modeling. *arXiv preprint arXiv:2403.19997.*

[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.