APS March Meeting 2024
Minneapolis, Minnesota, US

Micromechanics of thermoplastic elastomers with random microstructures[1]

Hansohl Cho*, Jaehee Lee, Jehoon Moon

Korea Advanced Institute of Science and Technology

Gregory C Rutledge

Massachusetts Institute of Technology

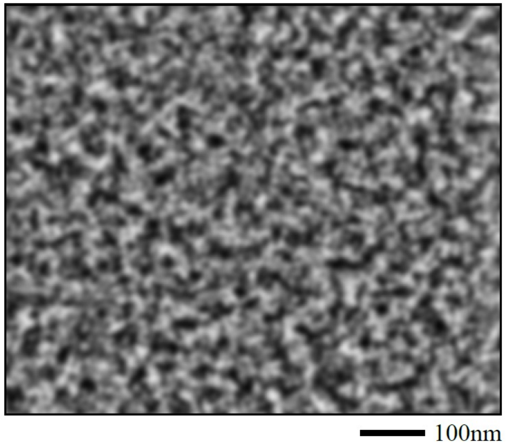
Mary C Boyce

Columbia University

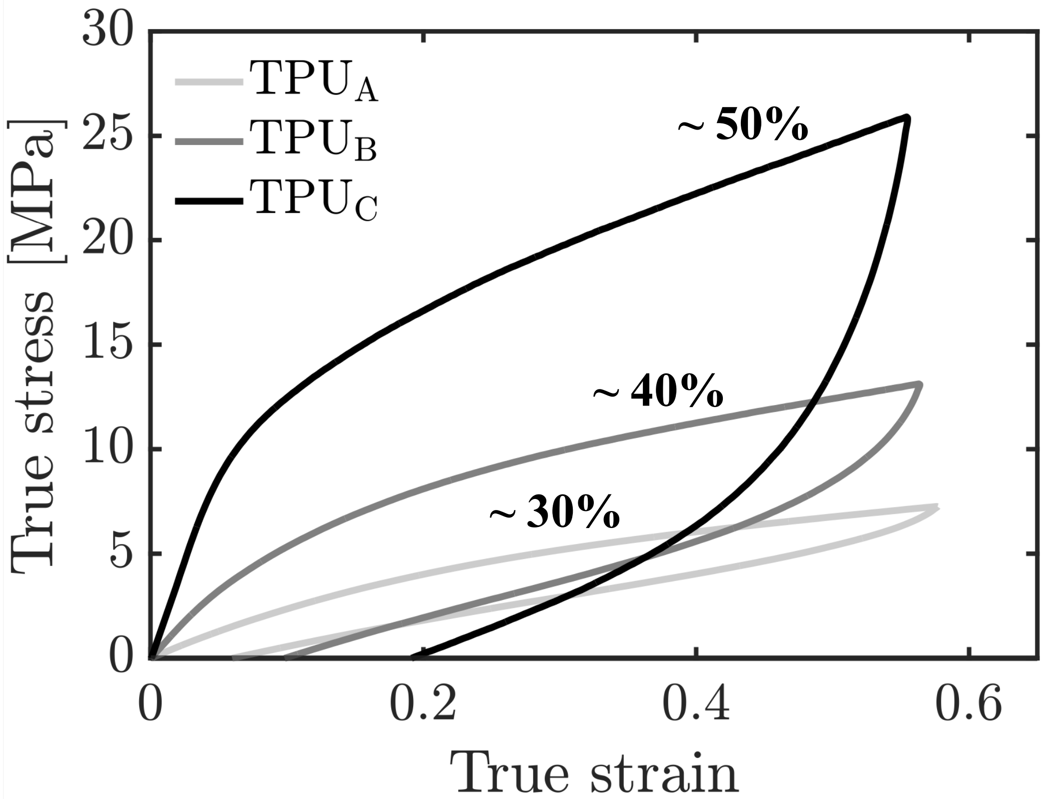
* hansohl@kaist.ac.kr

Mechanics & Extremes
@ KAIST

Thermoplastic polyurethanes (TPU)

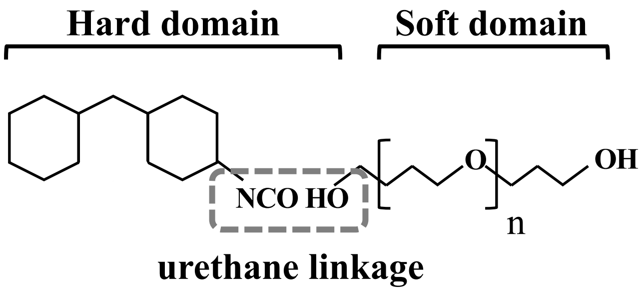


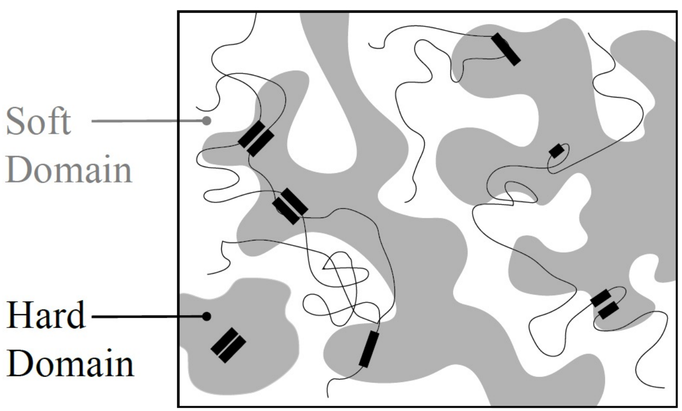
TEM image[2]

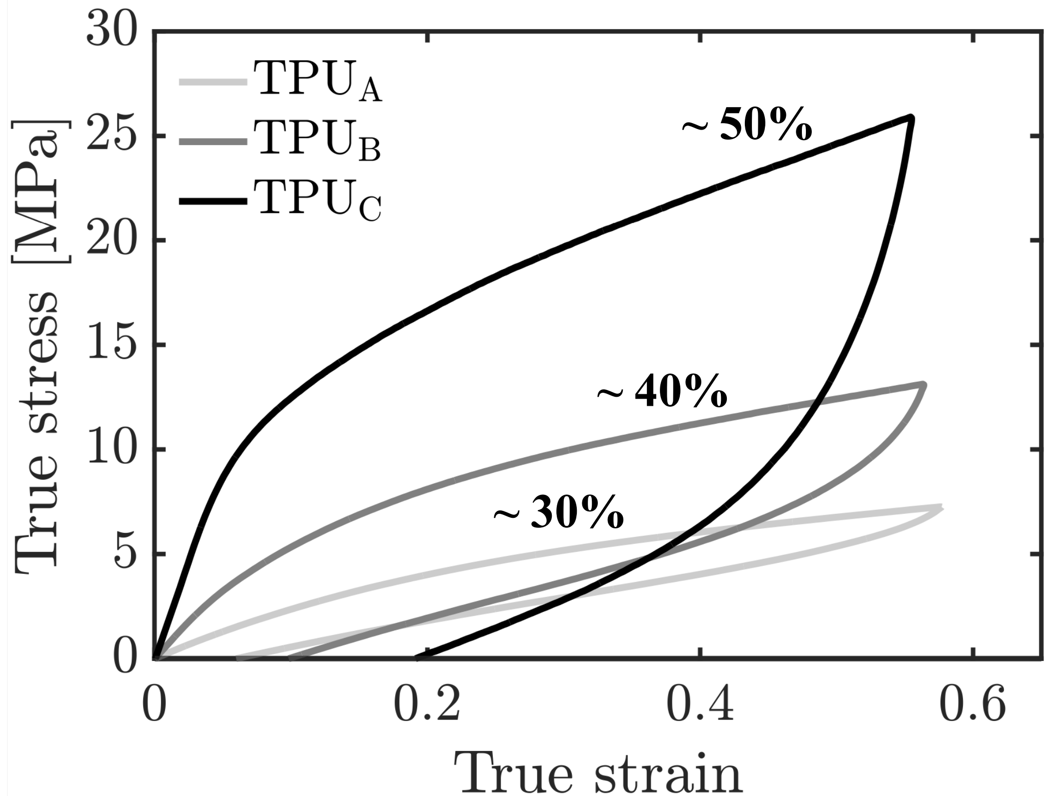


- Block copolymeric materials composed of hard and soft domains[3-5]
- Macro- and micromechanics of "large strain" behavior of thermoplastic polyurethanes (TPU)[1,2]

Thermoplastic polyurethanes (TPU)

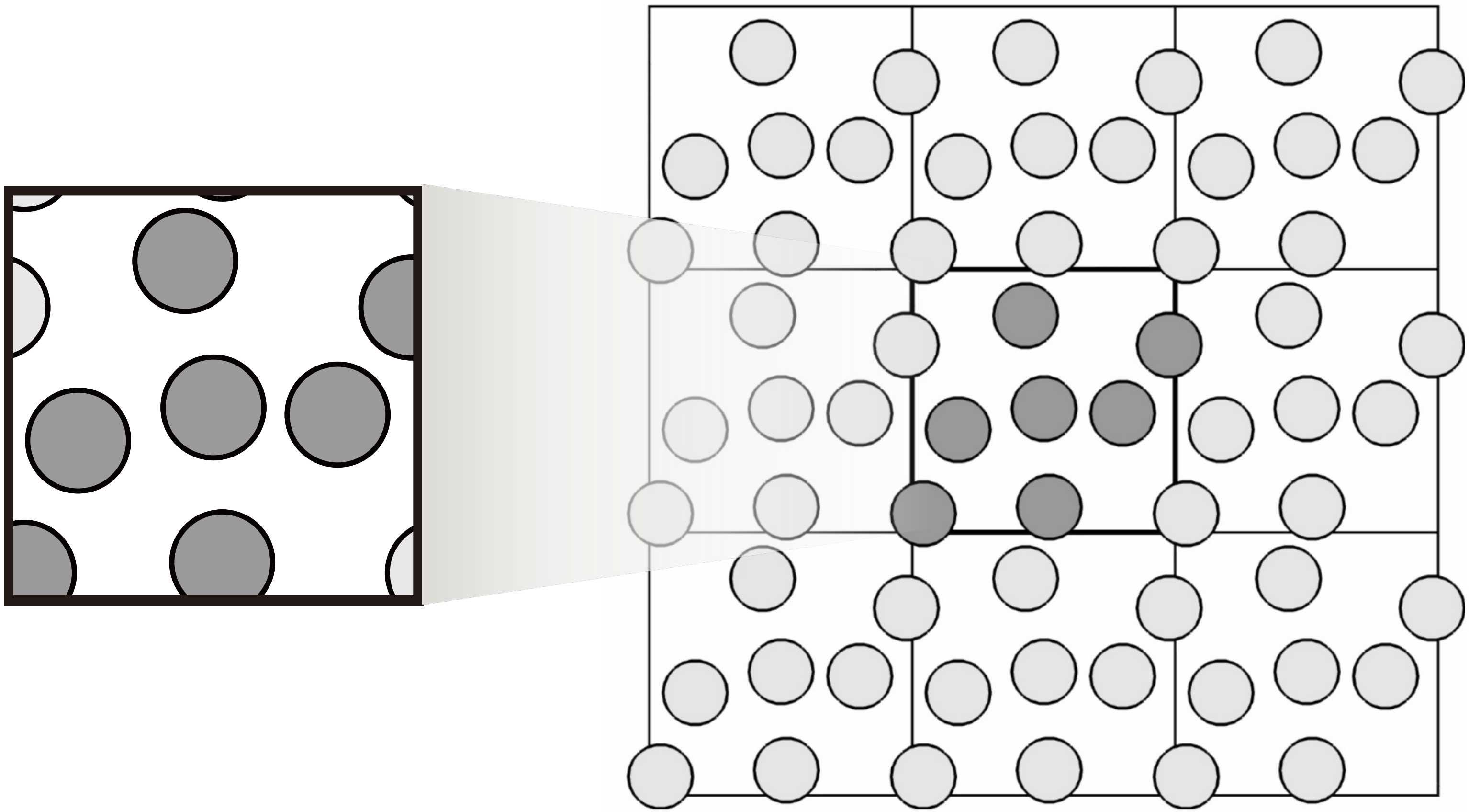






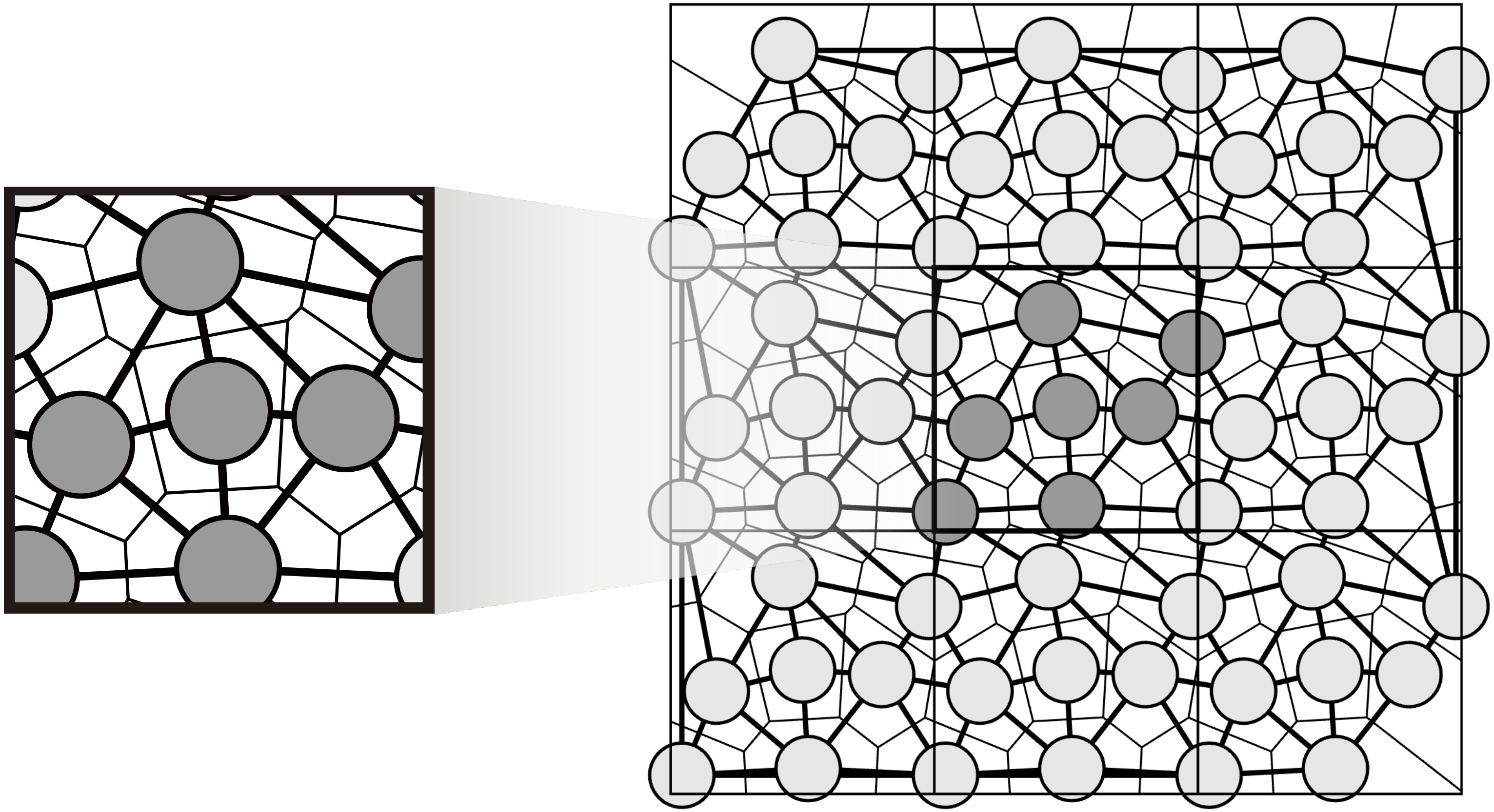
- Block copolymeric materials composed of hard and soft domains[3-5]
- Macro- and micromechanics of "large strain" behavior of thermoplastic polyurethanes (TPU)[1,2]

Construct "Random" microstructures



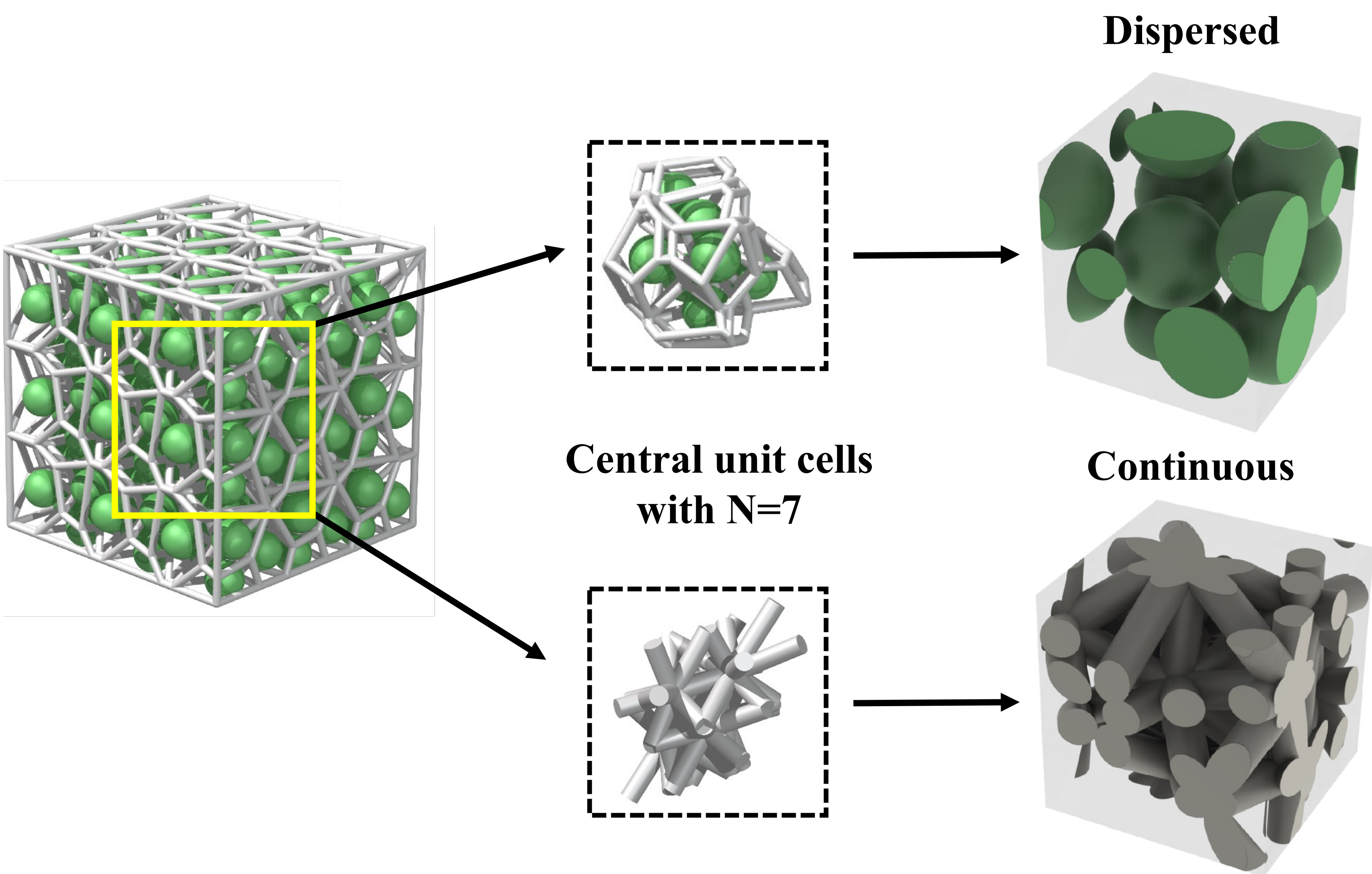
- Random packing of monodispersed spheres in periodic boundary conditions[6,7]

Random spatial points + tessellations



- Identification of the neighbors via Voronoi tessellations[8]
- By connecting with the neighbors → continuous, disordered microstructures available

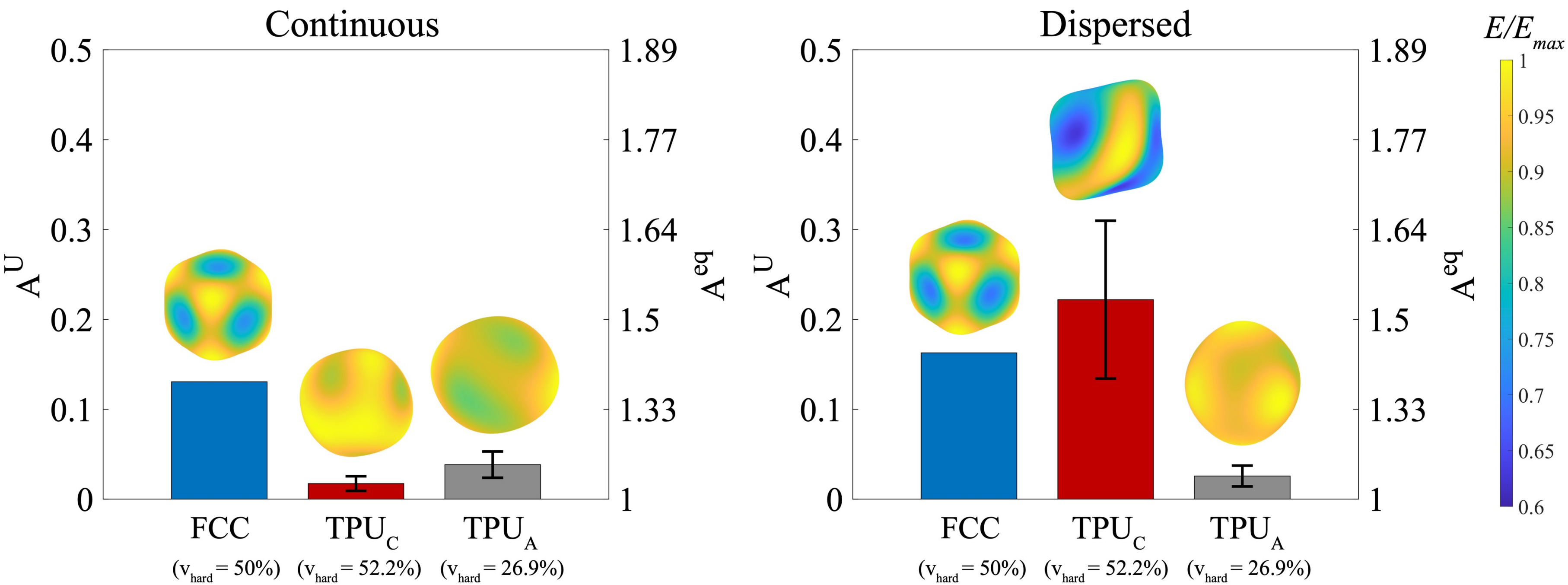
Proposed microstructures: Dispersed vs. continuous



- Only hard domains shown; we constructed two-phase materials

Identification of the N for RVEs

- Elastic anisotropy of both continuous and dispersed morphologies with N=7

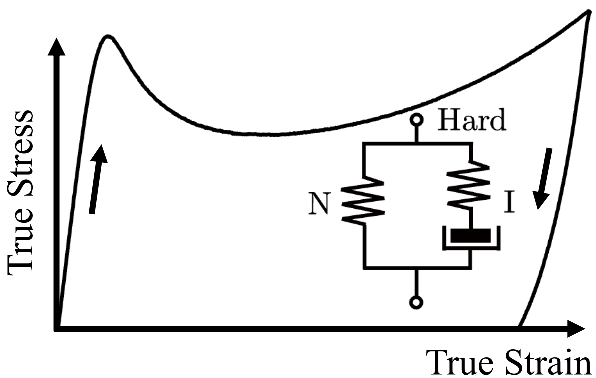


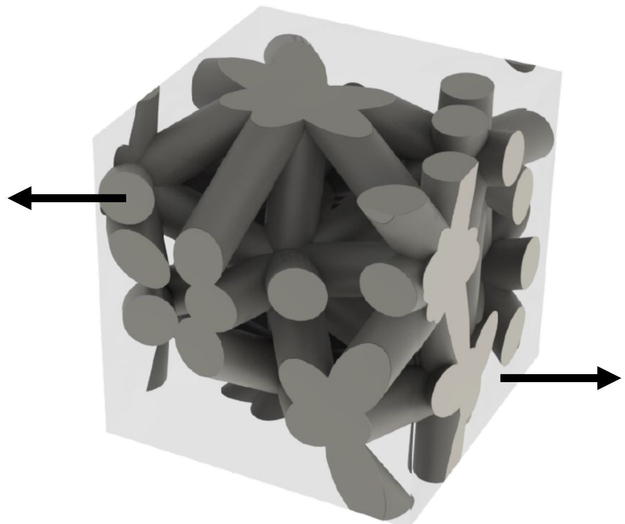
- **Universal anisotropy index**[9,10]

$$A^U = \mathbf{C}^V : \mathbf{S}^R - 6 = 5\frac{G^V}{G^R} + \frac{K^V}{K^R} - 6 \geq 0 \qquad (\text{Isotropic} : A^U = 0)$$

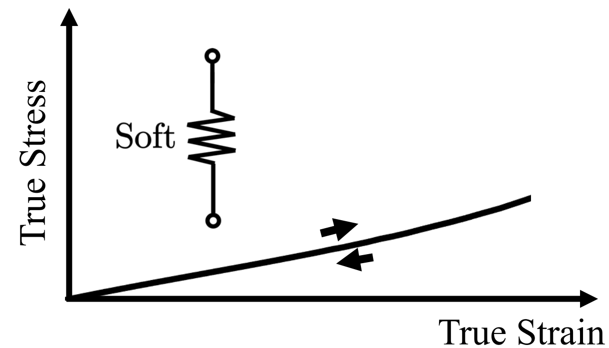
Constitutive behavior of hard and soft domains

Hard: Thermoplastic behavior





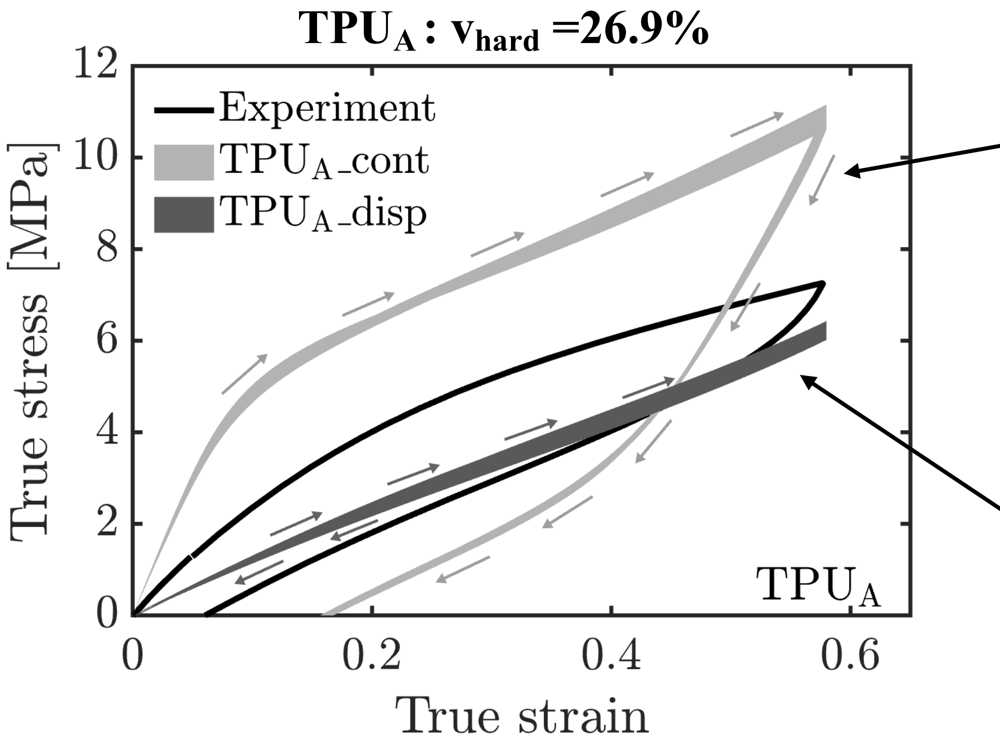
Soft: Elastomeric behavior

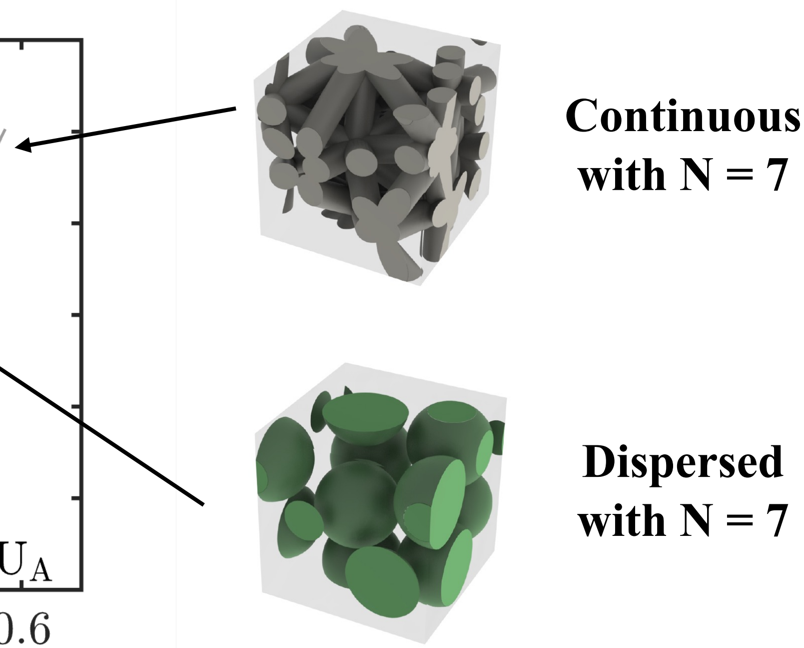


- **High initial stiffness**
- **Energy dissipation**
- **Residual strain**

- **Rubbery-like behavior**
- **Compliance**
- **Resilience**

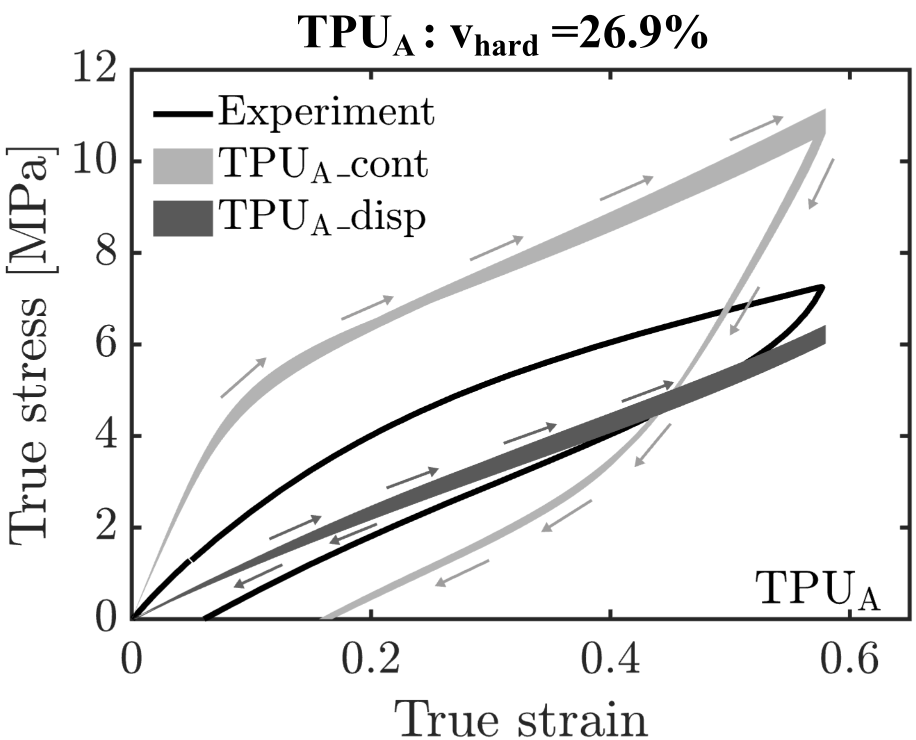
Dispersed vs. continuous: Role of connectivity

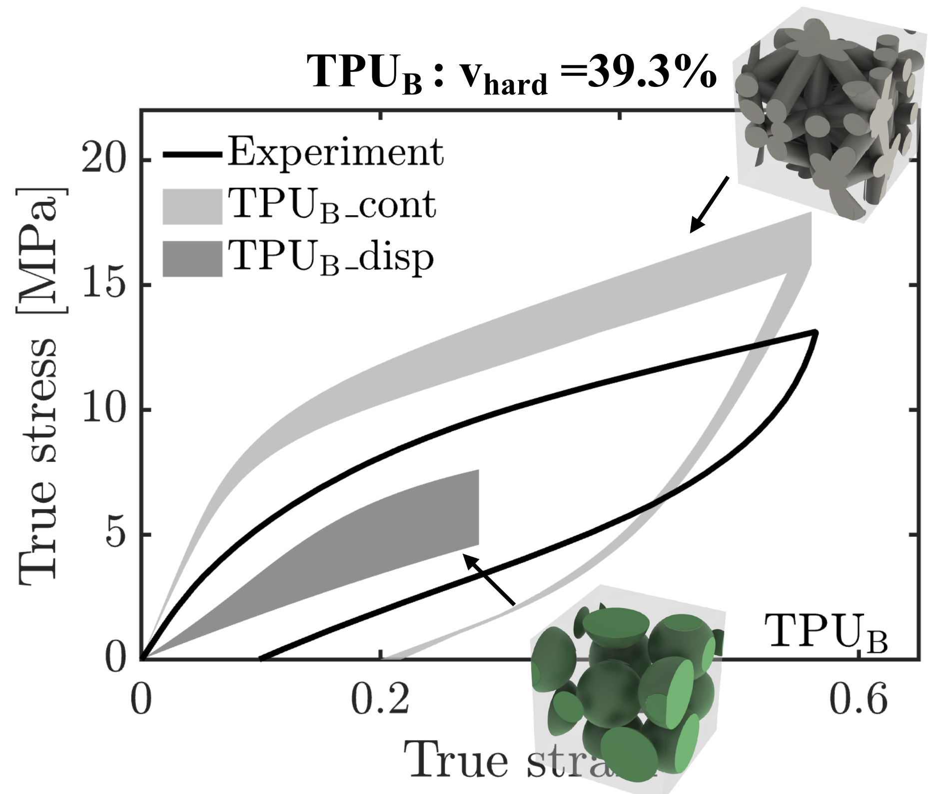




- Greater stress response, stiffer initial modulus, significant energy dissipation in the RVE with continuous hard domain [11,12]
- Numerical simulation results with five statistical realizations

Dispersed vs. continuous: Role of connectivity

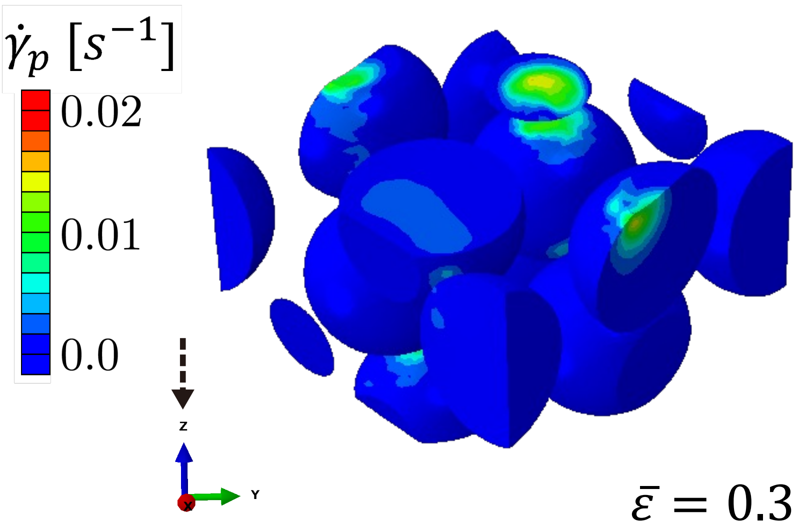




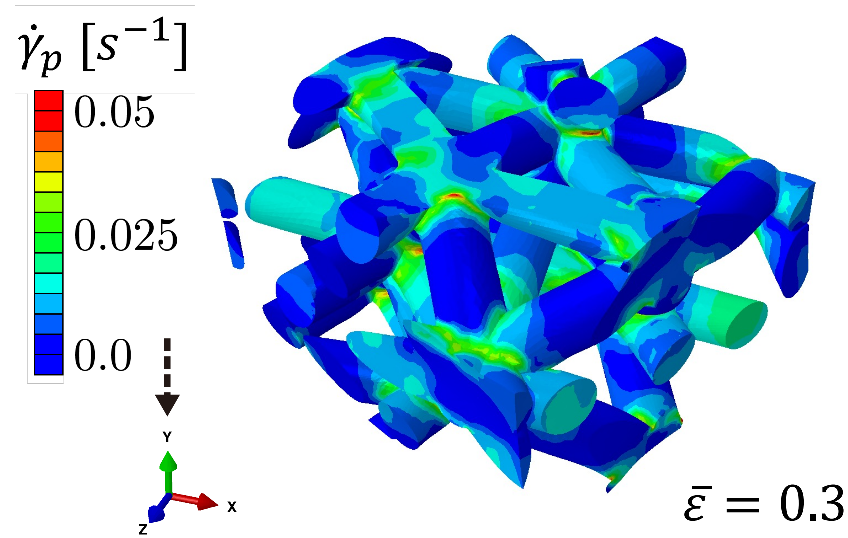
- In case of TPU$_B$ → closer to the stress-strain response with continuous hard domain
- TPU$_B$ (higher volume fraction; **39.3%**) **is likely to possess more "connected" domains**

Dispersed vs. continuous: Role of connectivity

Dispersed

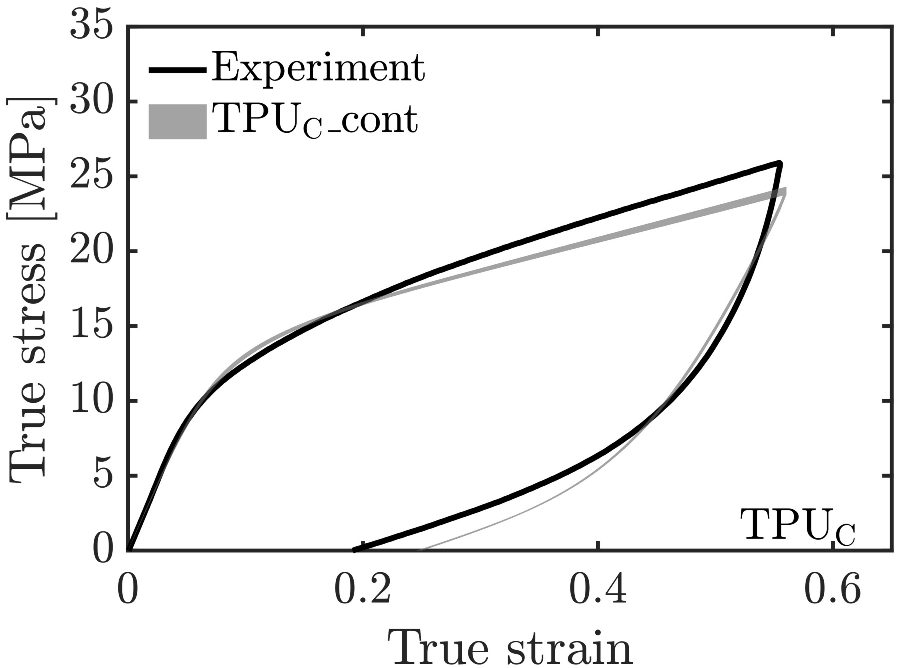


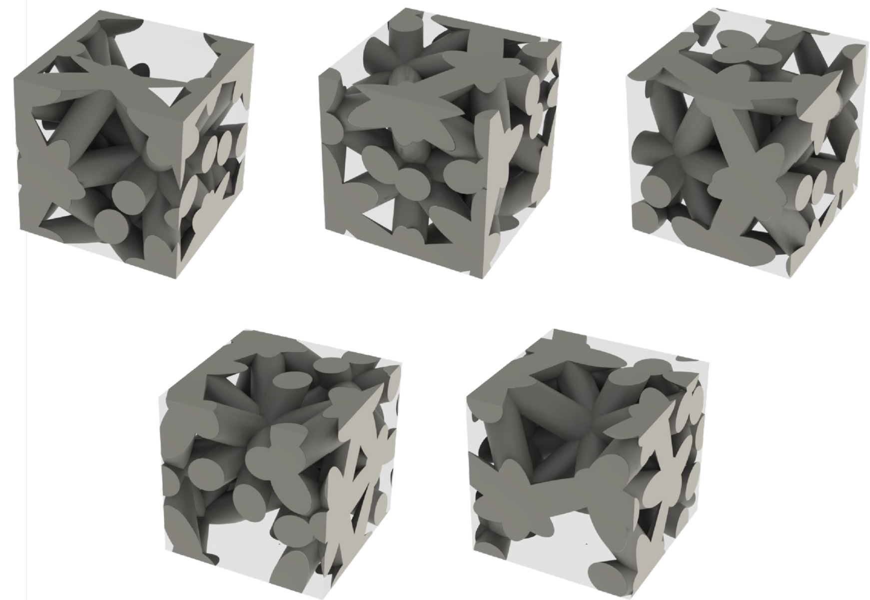
Continuous



- Contours of plastic flow rates in dispersed and continuous RVEs of **TPU_B (v_{hard} =39.3%)**
- **Plastic flow** developed throughout the hard ligament, which results in the **stress-rollover** in the RVEs **with continuous hard domain**

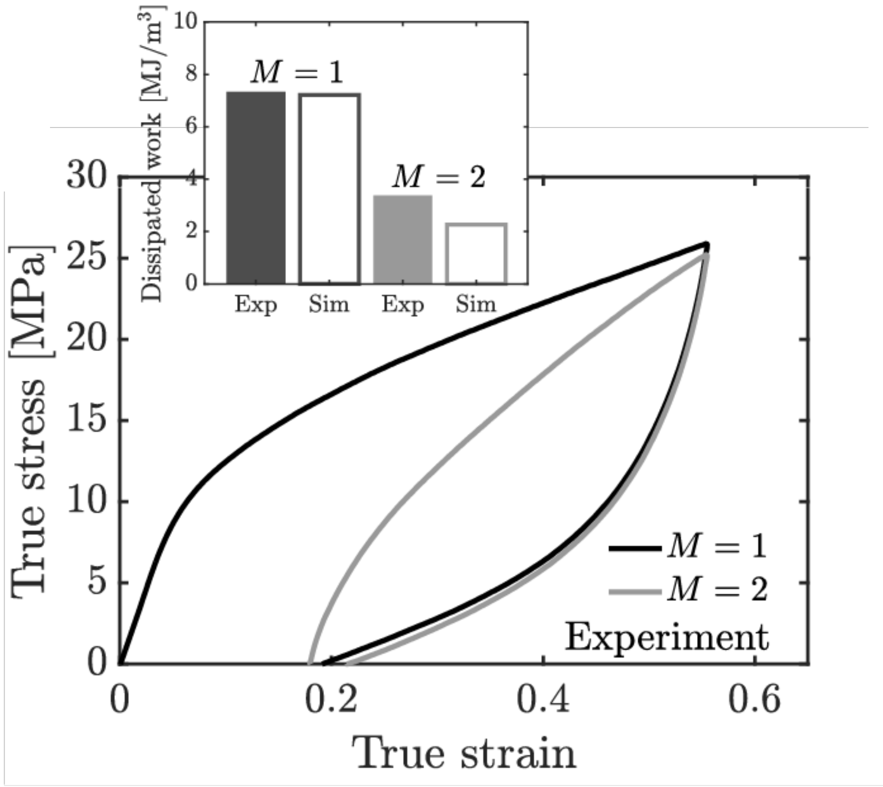
Dispersed vs. continuous: Role of connectivity

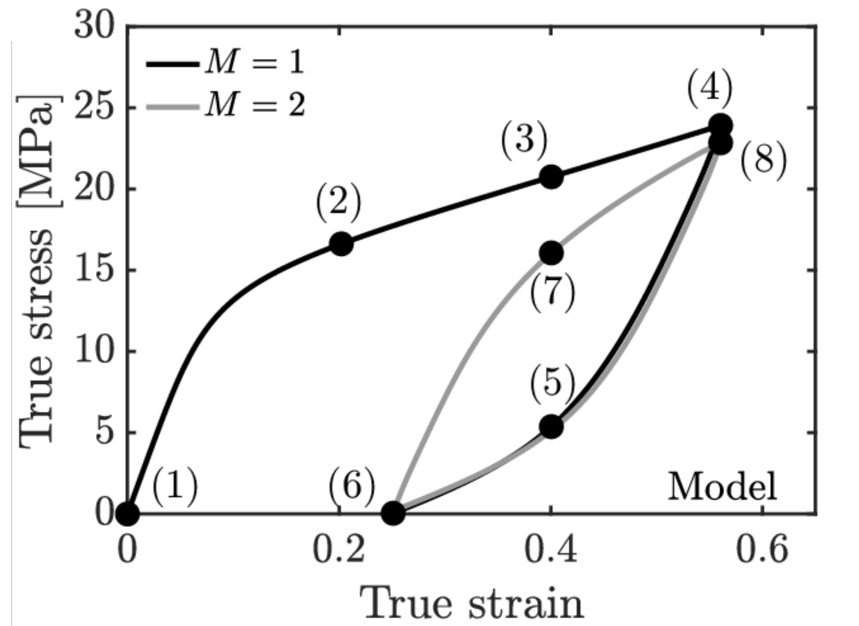




- Micromechanical model with **continuous hard domain** nicely captured the main features of TPU$_C$ with highest volume fraction ($\mathbf{v_{hard}}$ **= 52.2%**)

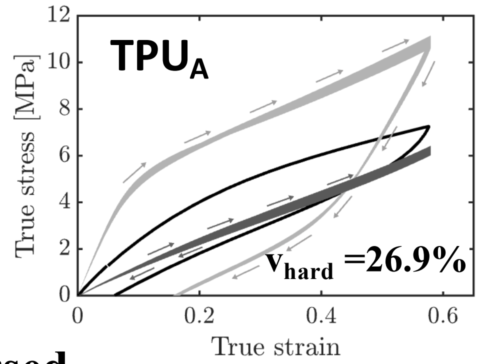
Cyclic loading behavior

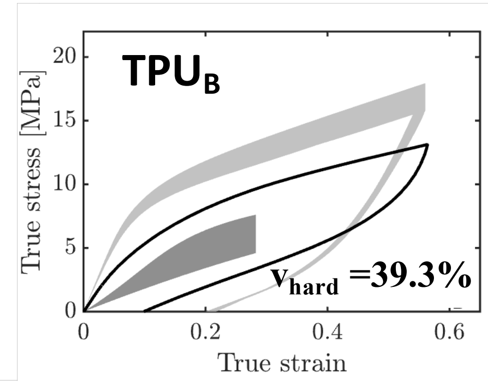




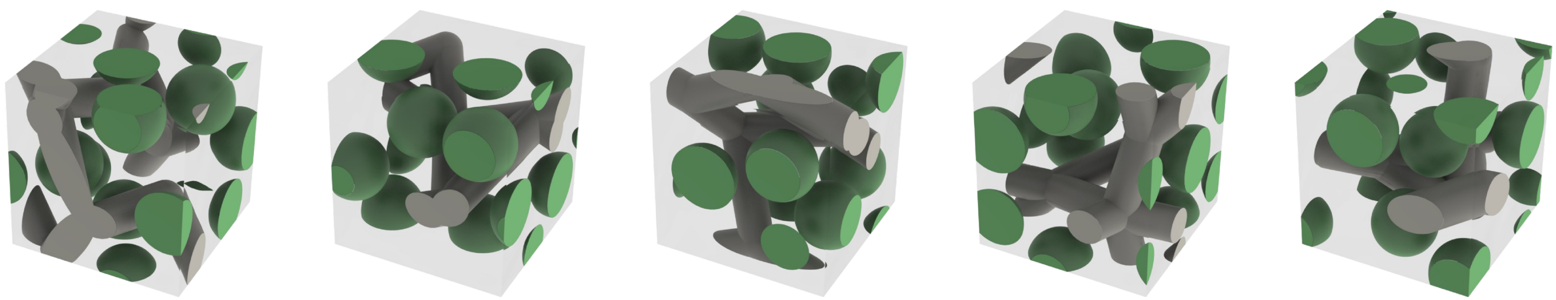
- Stretch-induced softening (Mullins' effect) was clearly manifested in the second cycle and was nicely captured by the micromechanical model[13,14]

“Mixed” RVEs

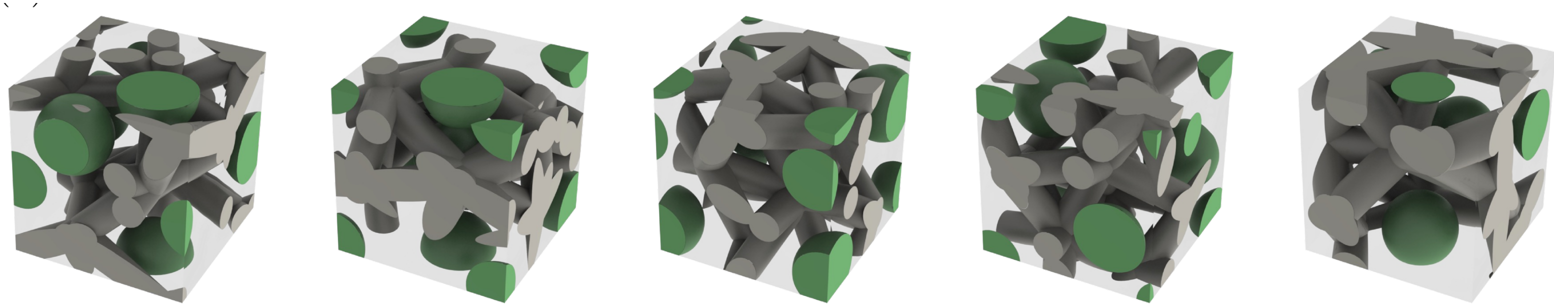




(1) TPU_A : 40% continuous / 60% dispersed



(2) TPU_B : 70% continuous / 30% dispersed

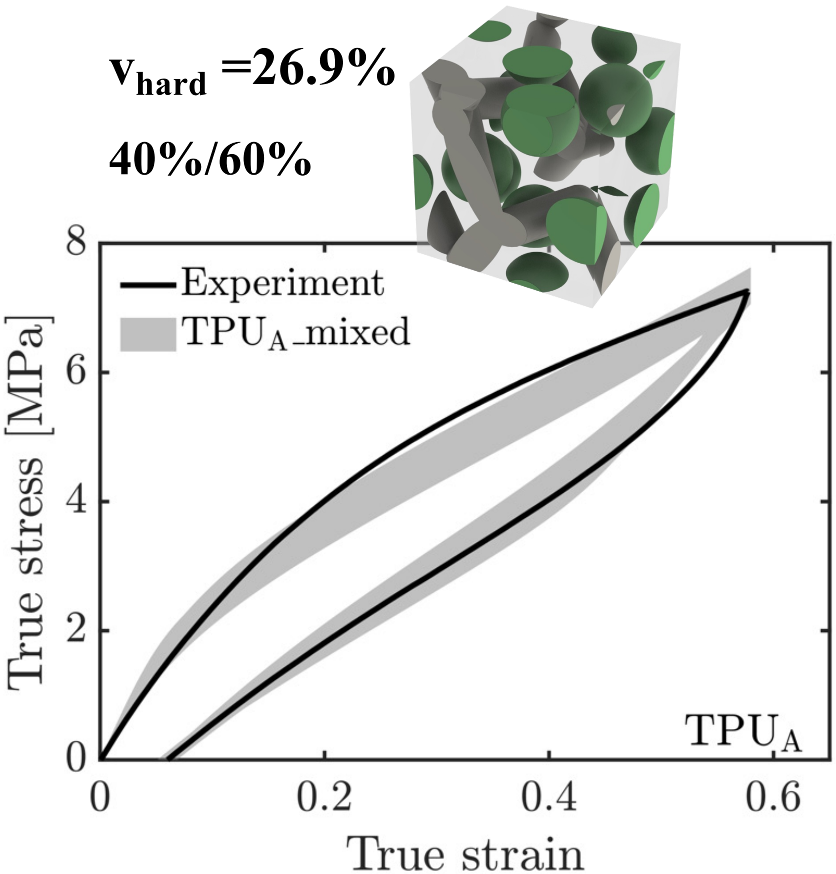


- Voronoi points N=10

"Mixed" RVEs

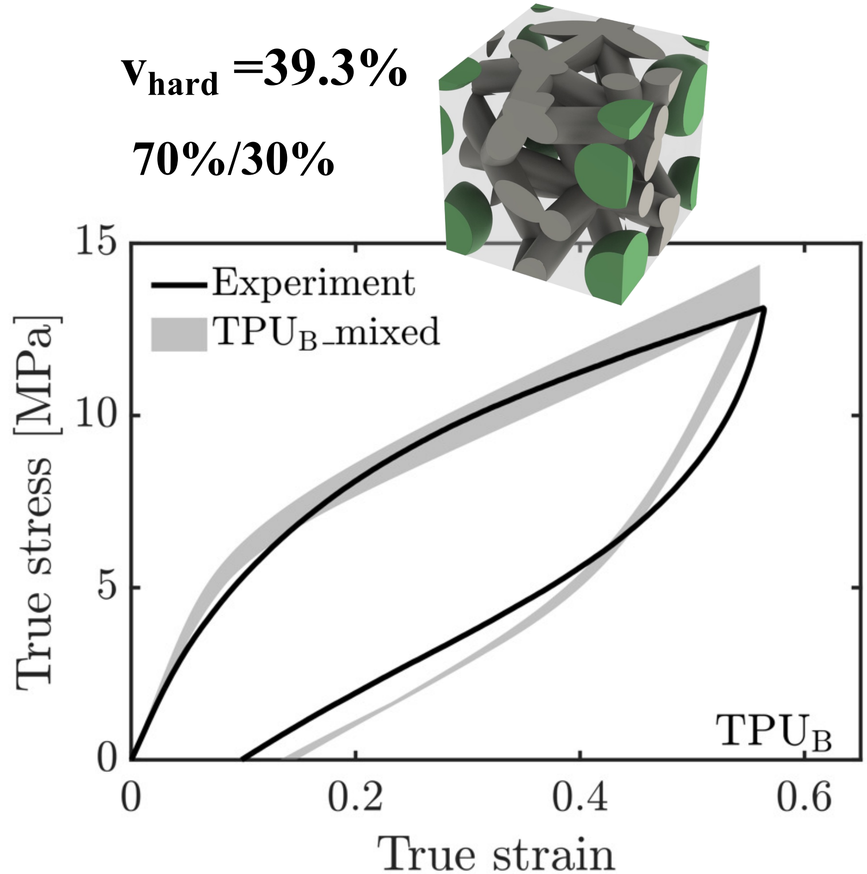
v_{hard} =26.9%

40%/60%



v_{hard} =39.3%

70%/30%



- Nicely captured the major features of the experimentally measured stress-strain response

Conclusion and Future works

- Micromechanical modeling of **"two-phase" elastomers** with two different disordered morphologies: **(1) dispersed** and **(2) continuous hard domains**

- **Connectivity** of hard domains impacts key elastic/inelastic features under cyclic loading

- Newly constructed **mixed RVEs** → co-existing dispersed and continuous morphologies

- Useful tool for micromechanical analysis of **"two-phase" materials with random microstructures**

→ Design of topological features for tailoring macroscopic mechanical properties[11,12]

→ Furthermore, explore the fracture behavior of **"two-phase" elastomers**[15-18]

Acknowledgement

• This work is supported by BASF and National Research Foundation of Korea (2021R1A4A103278312 and RS-2023-00279843)

References

[1] 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.

[2] 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.

[3] 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.

[4] Cho, H., Bartyczak, S., Mock Jr, W., & Boyce, M. C. (2013). Dissipation and resilience of elastomeric segmented copolymers under extreme strain rates. *Polymer, 54*(21), 5952-5964.

[5] 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.

[6] Danielsson, M., Parks, D. M., & Boyce, M. C. (2002). Three-dimensional micromechanical modeling of voided polymeric materials. *Journal of the Mechanics and Physics of Solids, 50*(2), 351-379.

[7] Danielsson, M., Parks, D. M., & Boyce, M. C. (2007). Micromechanics, macromechanics and constitutive modeling of the elasto-viscoplastic deformation of rubber-toughened glassy polymers. *Journal of the Mechanics and Physics of Solids, 55*(3), 533-561.

[8] Rycroft, C. (2009). *VORO++: A three-dimensional Voronoi cell library in C++.*

[9] Ranganathan, S. I., & Ostoja-Starzewski, M. (2008). Universal elastic anisotropy index. *Physical review letters, 101*(5), 055504.

References

[10] Nye, J. F. (1985). Physical properties of crystals: their representation by tensors and matrices. *Oxford University Press,*

[11] Cho, H., Weaver, J. C., Pöselt, E., in't Veld, P. J., Boyce, M. C., & Rutledge, G. C. (2016). Engineering the mechanics of heterogeneous soft crystals. *Advanced Functional Materials, 26*(38), 6938-6949.

[12] Lee, G., Lee, J., Lee, S., Rudykh, S., & Cho, H. (2024). Extreme resilience and dissipation in heterogeneous elasto-plastomeric crystals. *Soft Matter, 20*(2), 315-329.

[13] Qi, H. J., & Boyce, M. C. (2004). Constitutive model for stretch-induced softening of the stress–stretch behavior of elastomeric materials. *Journal of the Mechanics and Physics of Solids, 52*(10), 2187-2205.

[14] Diani, J., Fayolle, B., & Gilormini, P. (2009). A review on the Mullins effect. *European Polymer Journal, 45*(3), 601-612.

[15] Chen, C., Wang, Z., & Suo, Z. (2017). Flaw sensitivity of highly stretchable materials. *Extreme Mechanics Letters, 10,* 50-57.

[16] Mao, Y., Talamini, B., & Anand, L. (2017). Rupture of polymers by chain scission. *Extreme Mechanics Letters, 13,* 17-24.

[17] Narayan, S., & Anand, L. (2021). Fracture of amorphous polymers: A gradient-damage theory. *Journal of the Mechanics and Physics of Solids, 146,* 104164.

[18] 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.