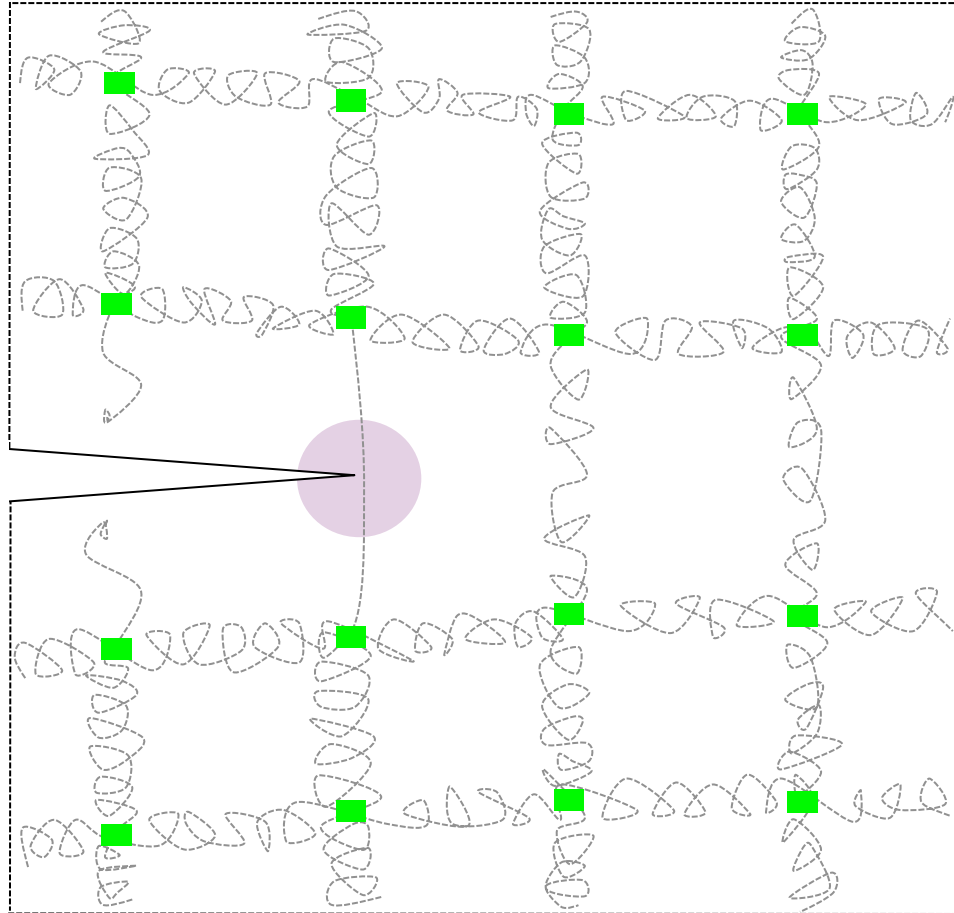


fatigue-resistant stretchable materials

Zhigang Suo

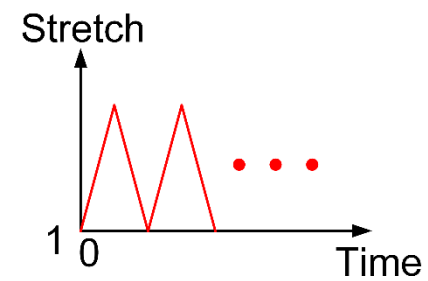
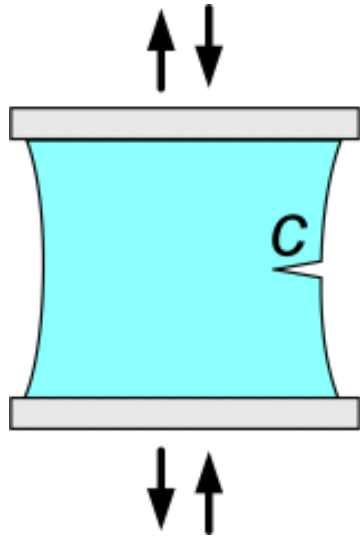
Harvard University

Hypothesis: an elastic network will *not* suffer fatigue



This hypothesis is wrong!

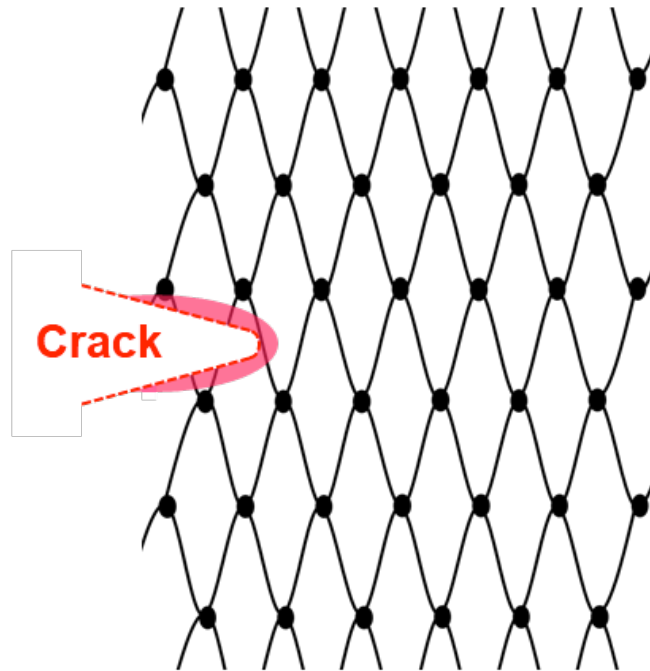
Fatigue fracture of hydrogel



A mechanism of fatigue in an elastic network

(a)

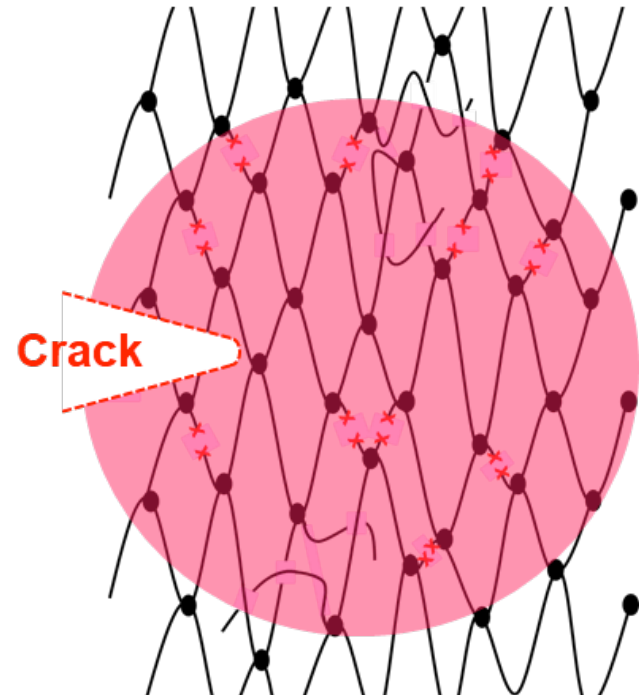
Perfect network



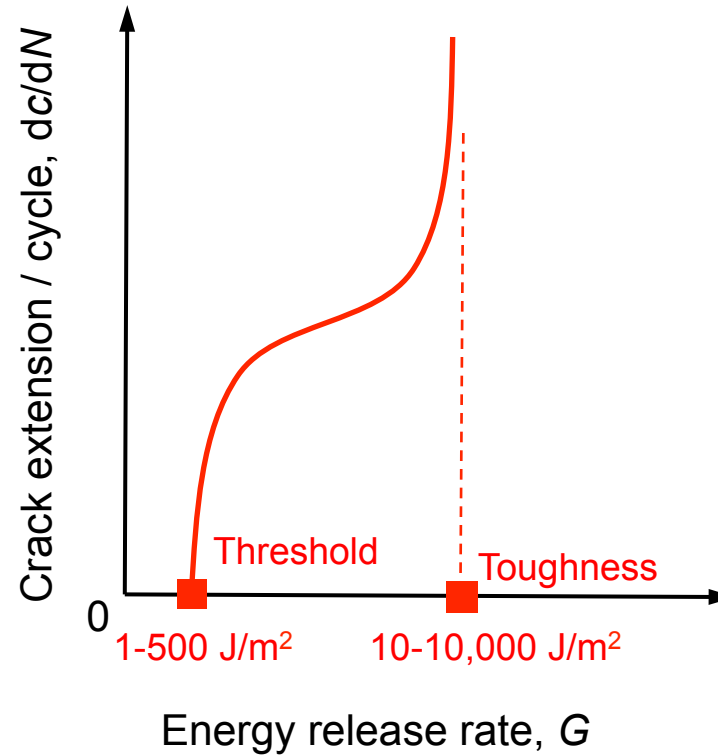
Lake-Thomas

(b)

Imperfect network



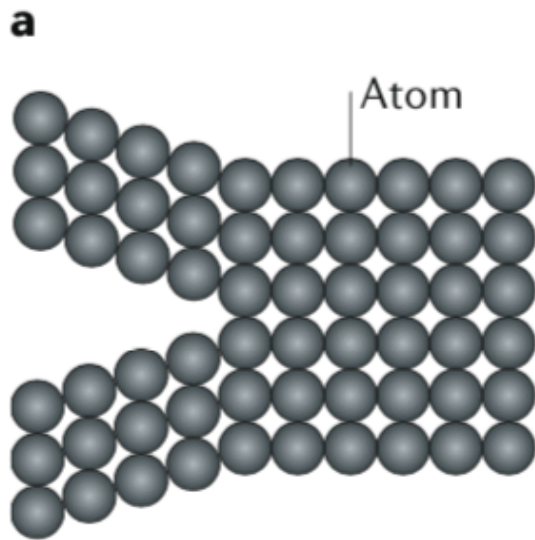
Irwin-Orowan



- **Toughness** of hydrogels has been extensively studied in last 16 years.
- **Threshold** of hydrogels is a new game in last 2 years.

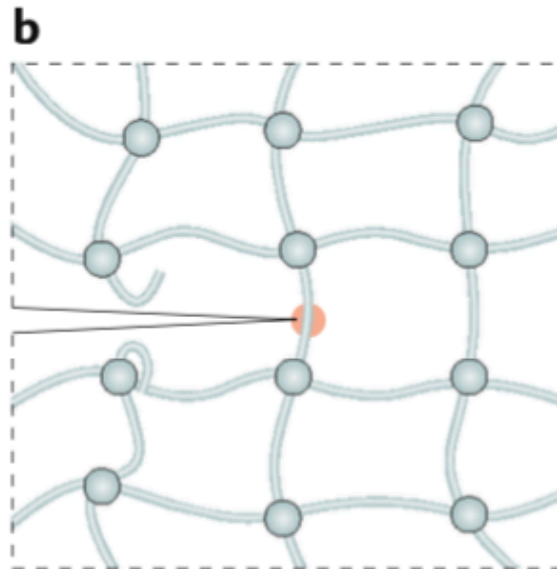
Toughness

Griffith 1921



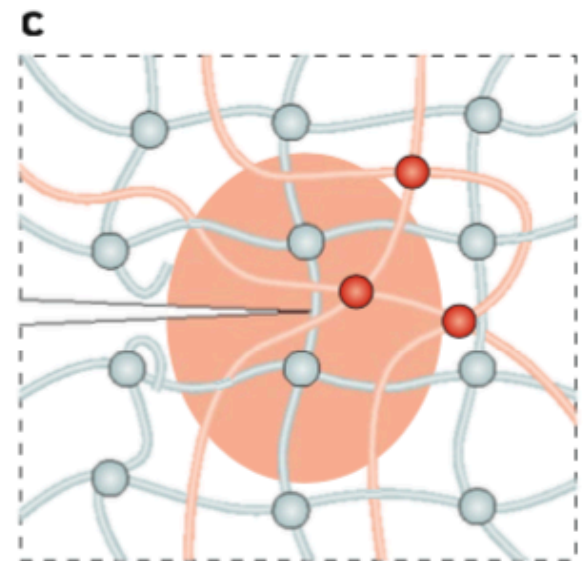
Break a layer of bonds
Toughness $\sim 1 \text{ J/m}^2$

Lake-Thomas 1967



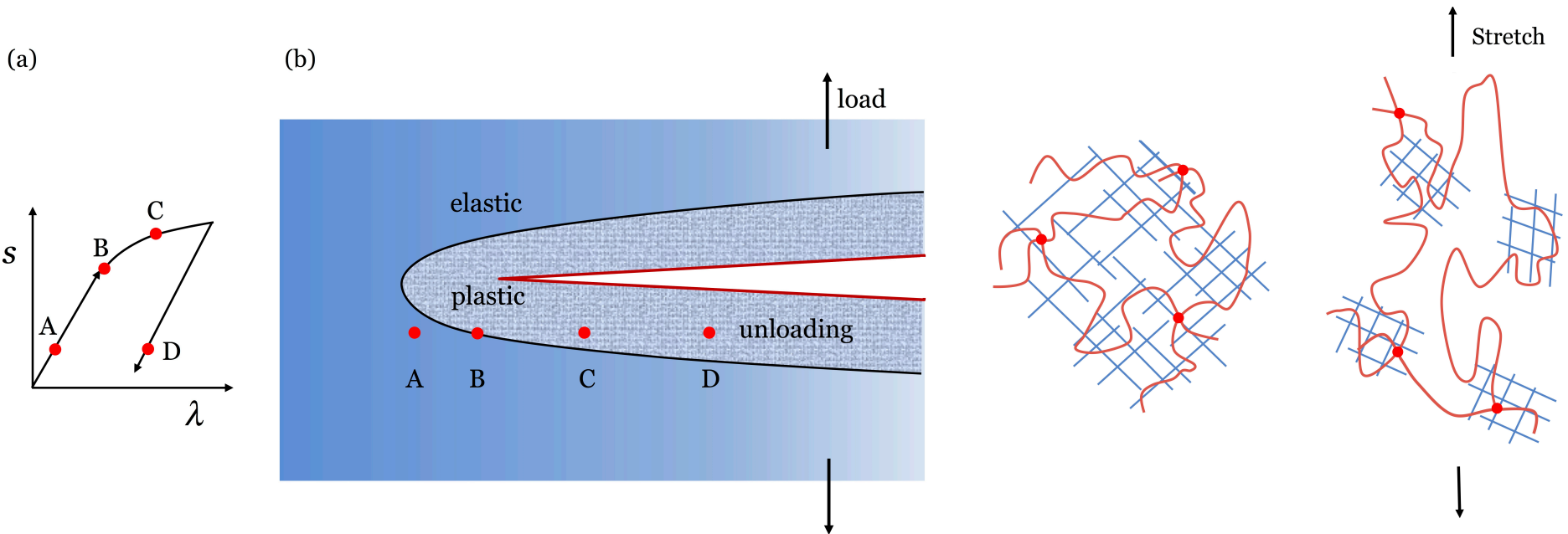
Snap a layer of chains
Toughness $\sim 10\text{-}1000 \text{ J/m}^2$

Irwin-Orowan 1940s

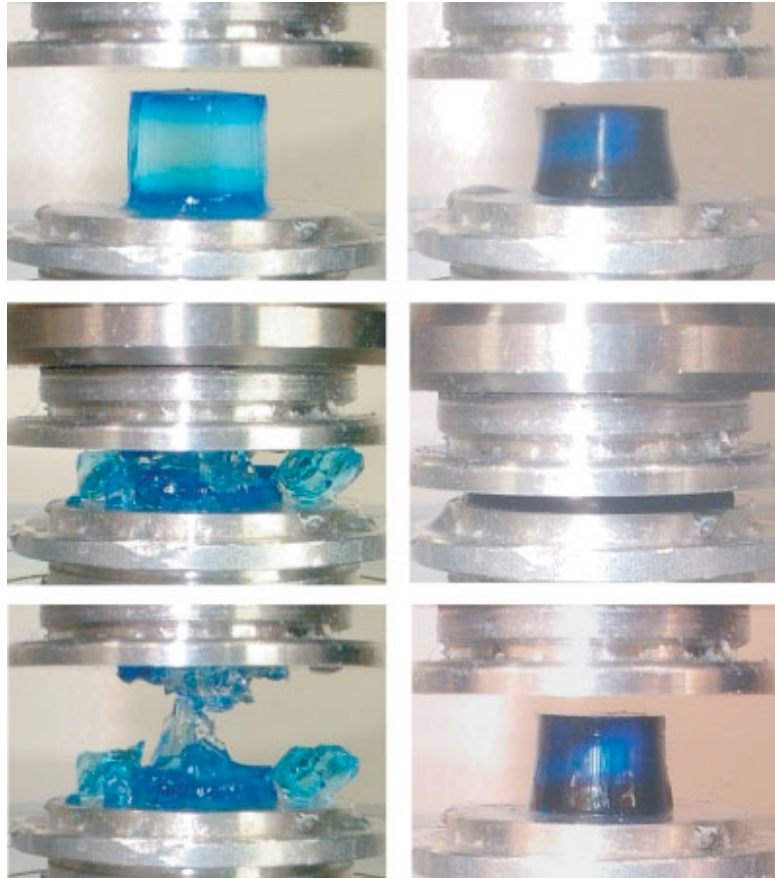


Dissipate energy in the bulk
Toughness $\sim 100\text{-}10000 \text{ J/m}^2$

Double-network hydrogel



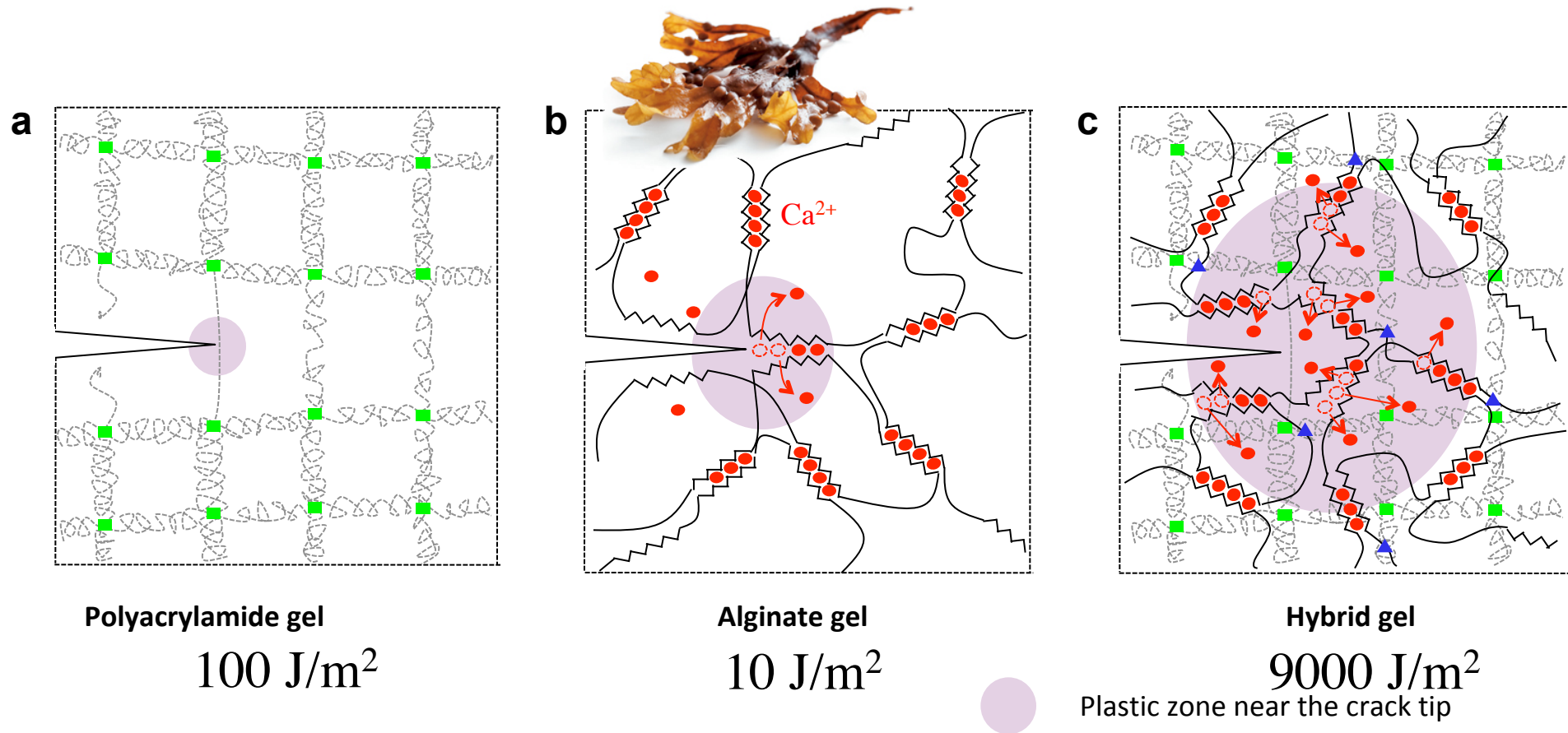
Double-network gels



Single-network gel

Double-network gel

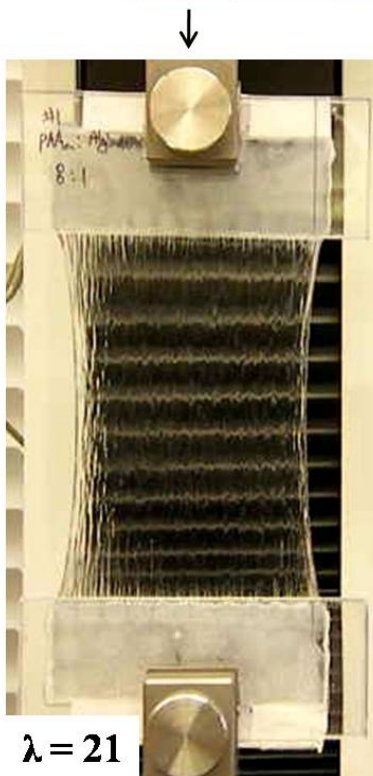
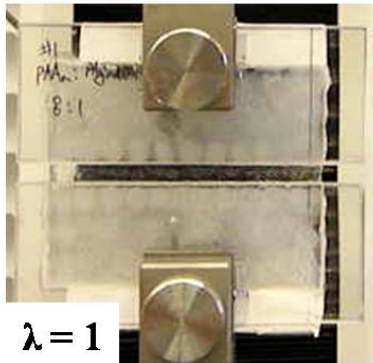
brittle + brittle = tough



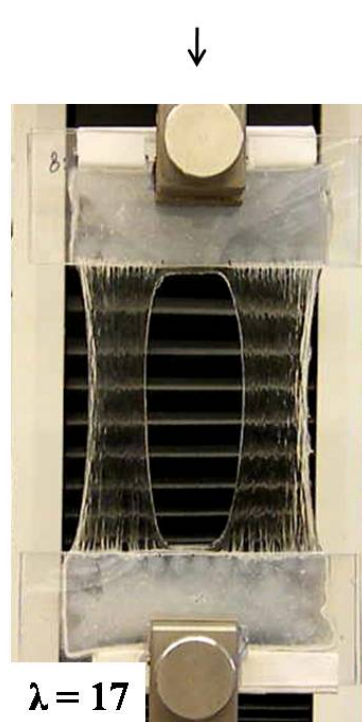
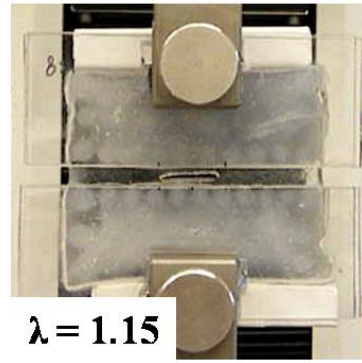
Sun, Zhao, Illeperuma, Chaudhuri, Oh, Mooney, Vlassak, Suo. Nature 489, 133 (2012)

Highly stretchable and tough hydrogels

(a)



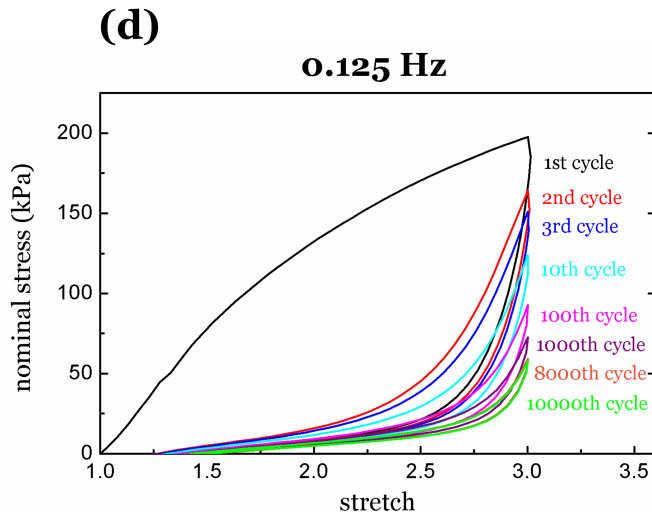
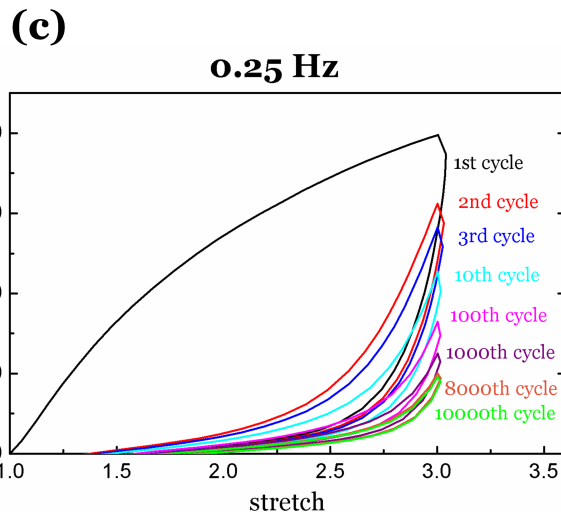
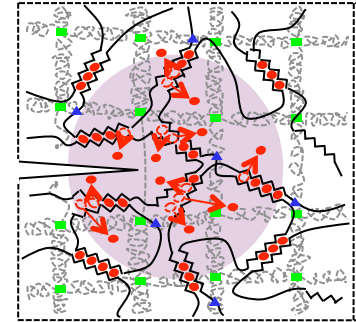
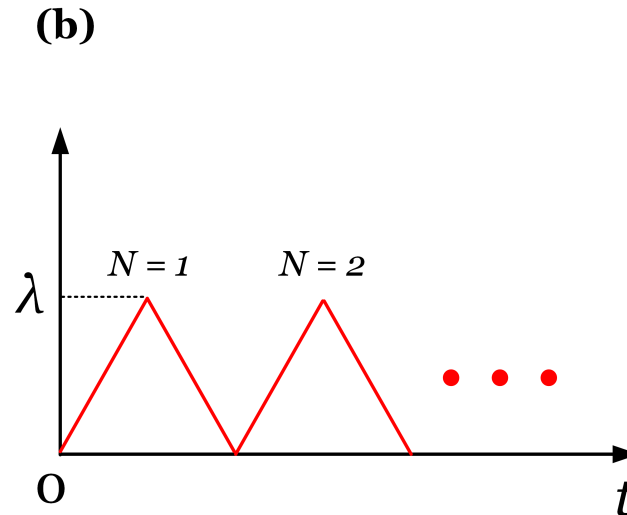
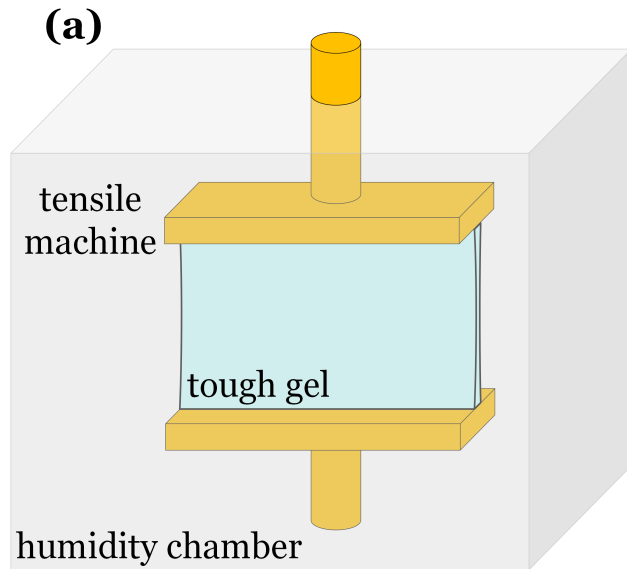
(b)



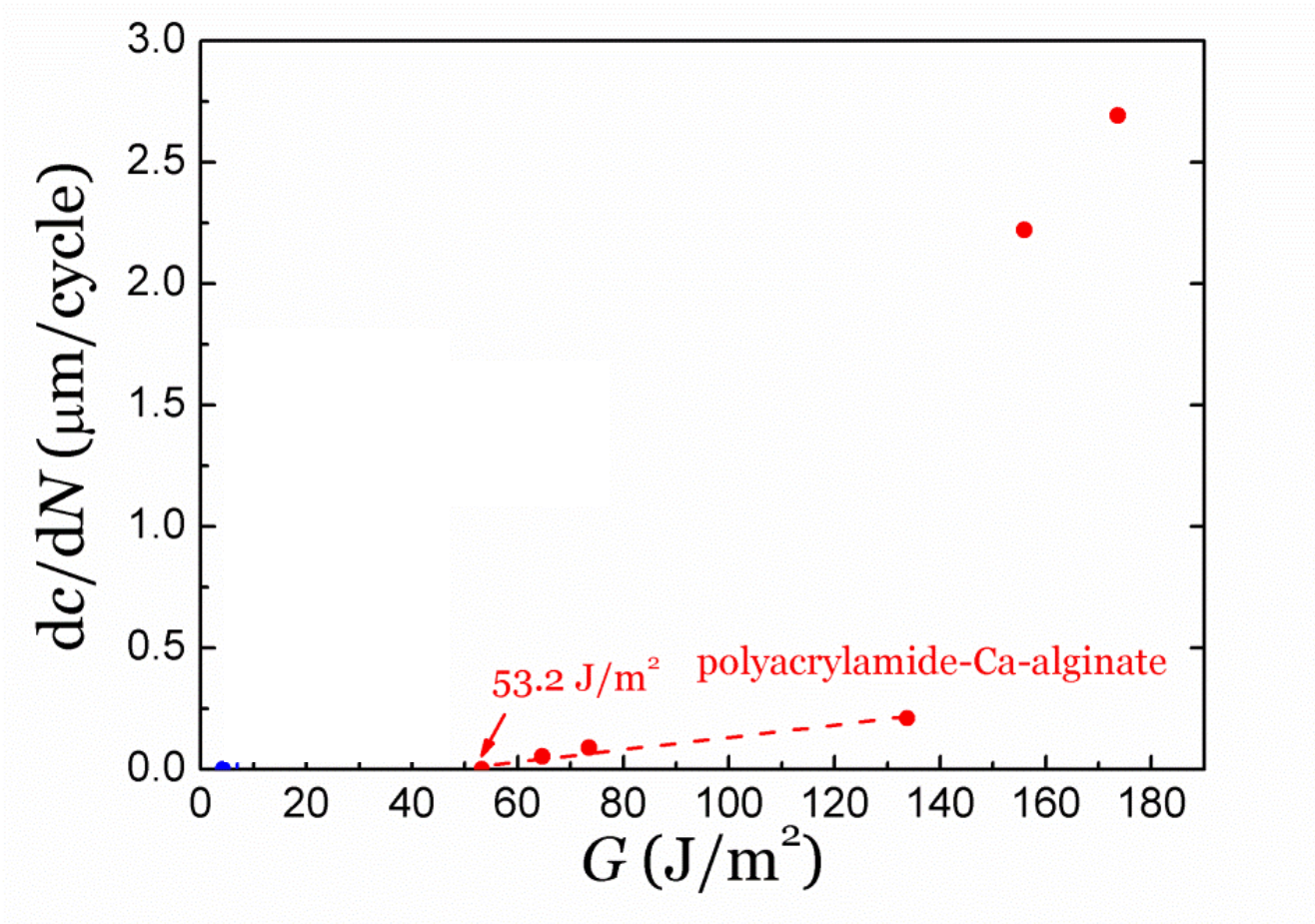
- Water: 90%
- Stretch: 21
- Fracture energy: 9000 J/m²

- ~10 J/m², tofu, jello
- ~10 J/m², most gels
- ~100 J/m², contact lenses
- ~1000 J/m², cartilage
- ~1000 J/m², double network gels (Gong 2003)
- ~1000 J/m², VHB
- ~10,000 J/m², natural rubber

Shakedown of tough gel



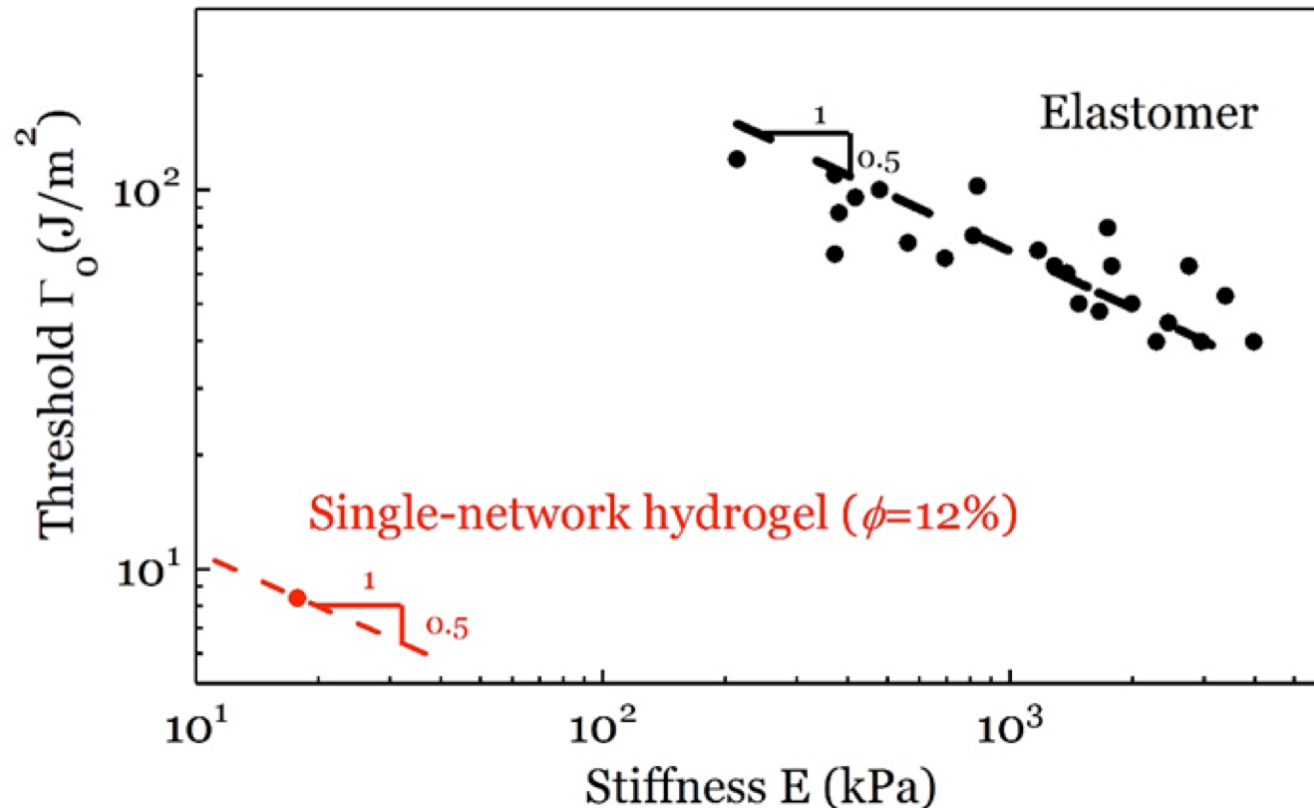
Alginate-polyacrylamide hydrogel



Tough for the first loading.
But suffers fatigue fracture.

Stiffness-threshold conflict

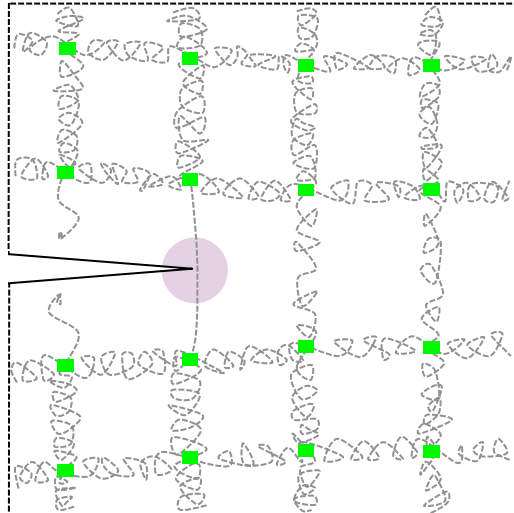
$$\mu \sim \frac{1}{n}, \quad \Gamma_{th} \sim \sqrt{n}$$



Remove the stiffness-threshold conflict

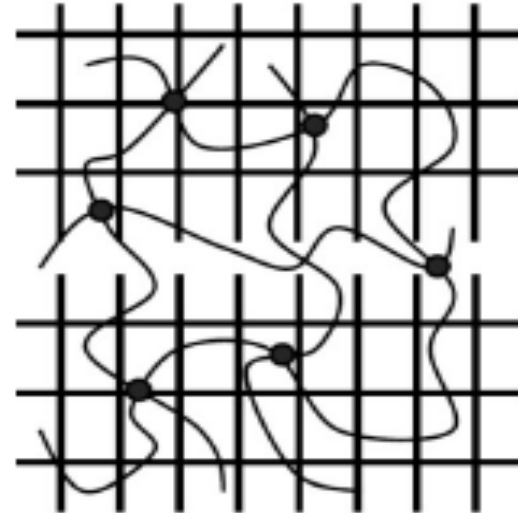
Single network

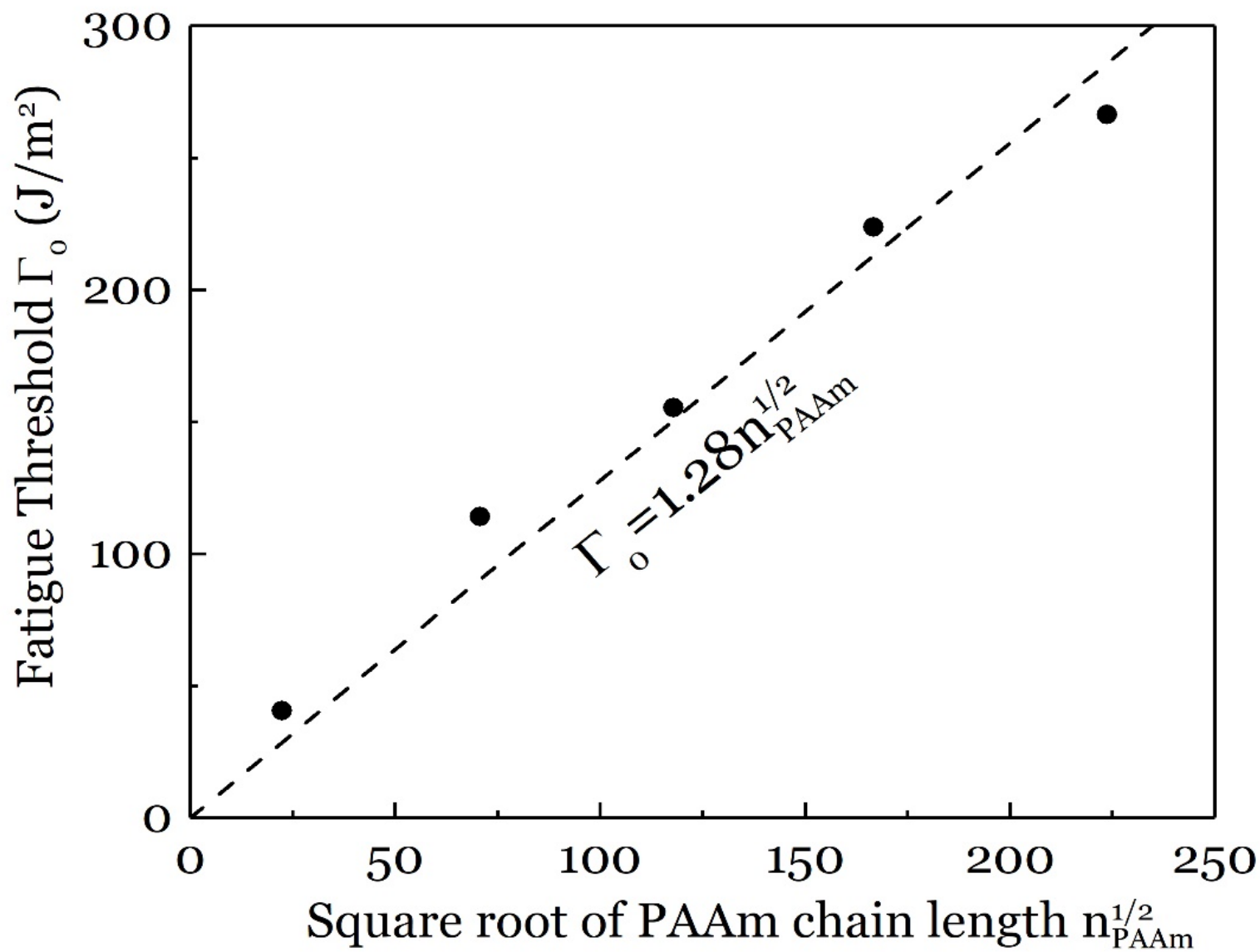
$$\mu \sim \frac{1}{n}, \quad \Gamma_{th} \sim \sqrt{n}$$

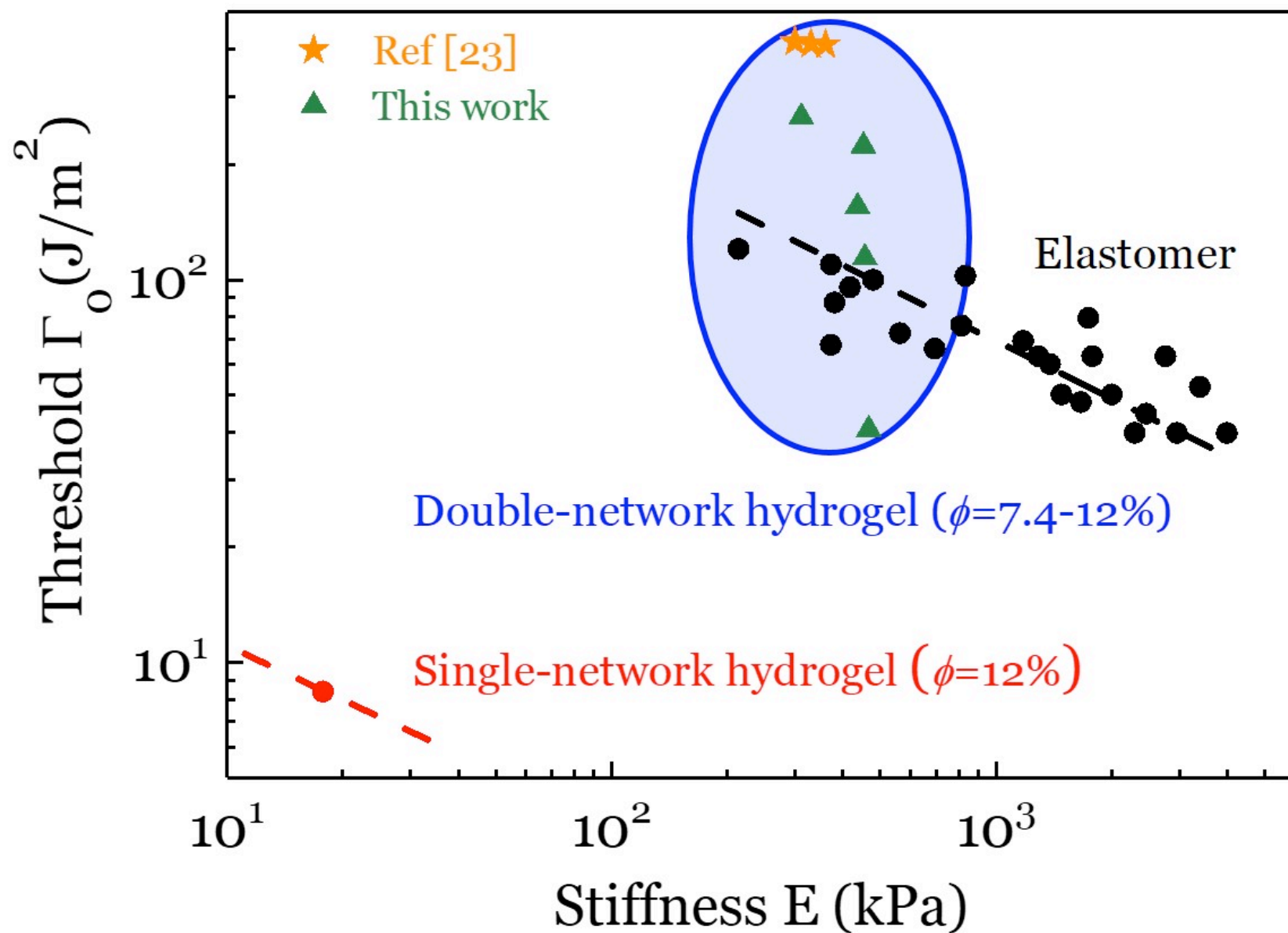


Double network

$$\mu \sim \frac{1}{n_A}, \quad \Gamma_{th} \sim \sqrt{n_B}$$





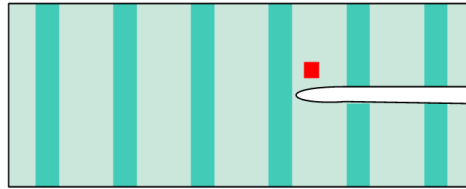


Elastic dissipater

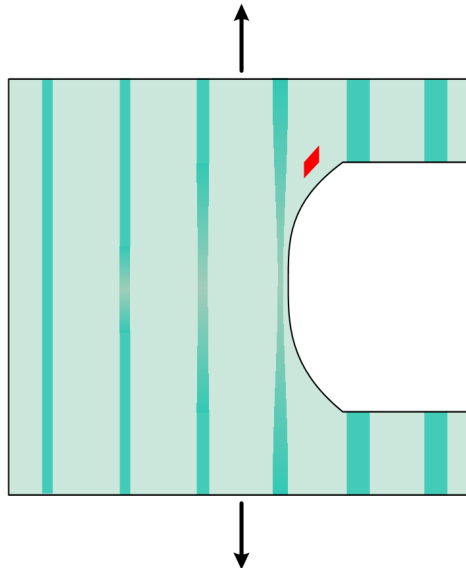


Elastic dissipater

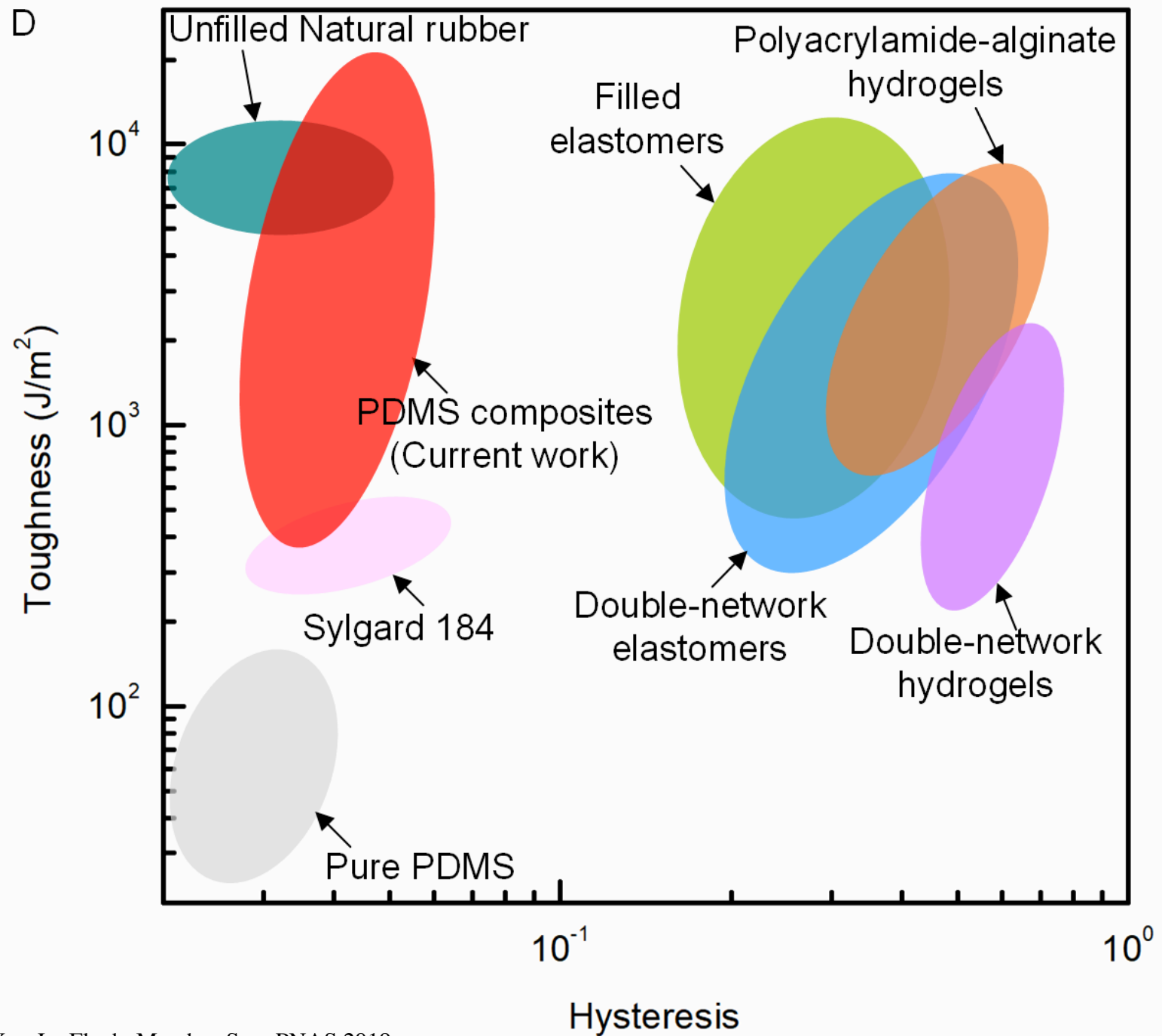
(a) Undeformed state



(b) Deformed state



Wang, Xiang, Yao, Le Floch, Mendez, Suo, PNAS 2019
Xiang, Wang, Yang, Yao, Wang, Suo, Materials Today 2019



(a) $\lambda_m = 2.303$, $G = 4,441 \text{ J/m}^2$, $N=1$



(b) $\lambda_m = 2.261$, $G = 4,092 \text{ J/m}^2$, $N=10$



(c) $\lambda_m = 2.107$, $G = 2,978 \text{ J/m}^2$, $N=565$



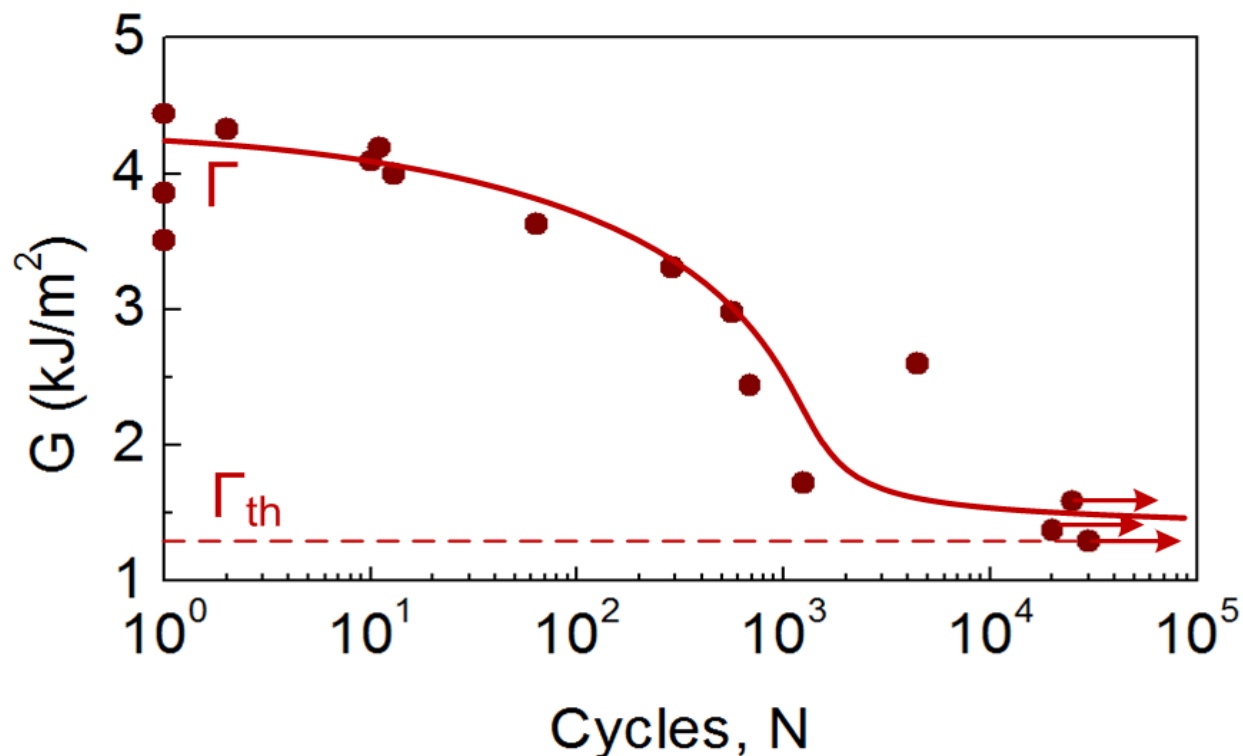
(d) $\lambda_m = 1.780$, $G = 1,372 \text{ J/m}^2$, $N=20,000$



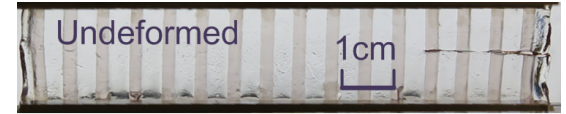
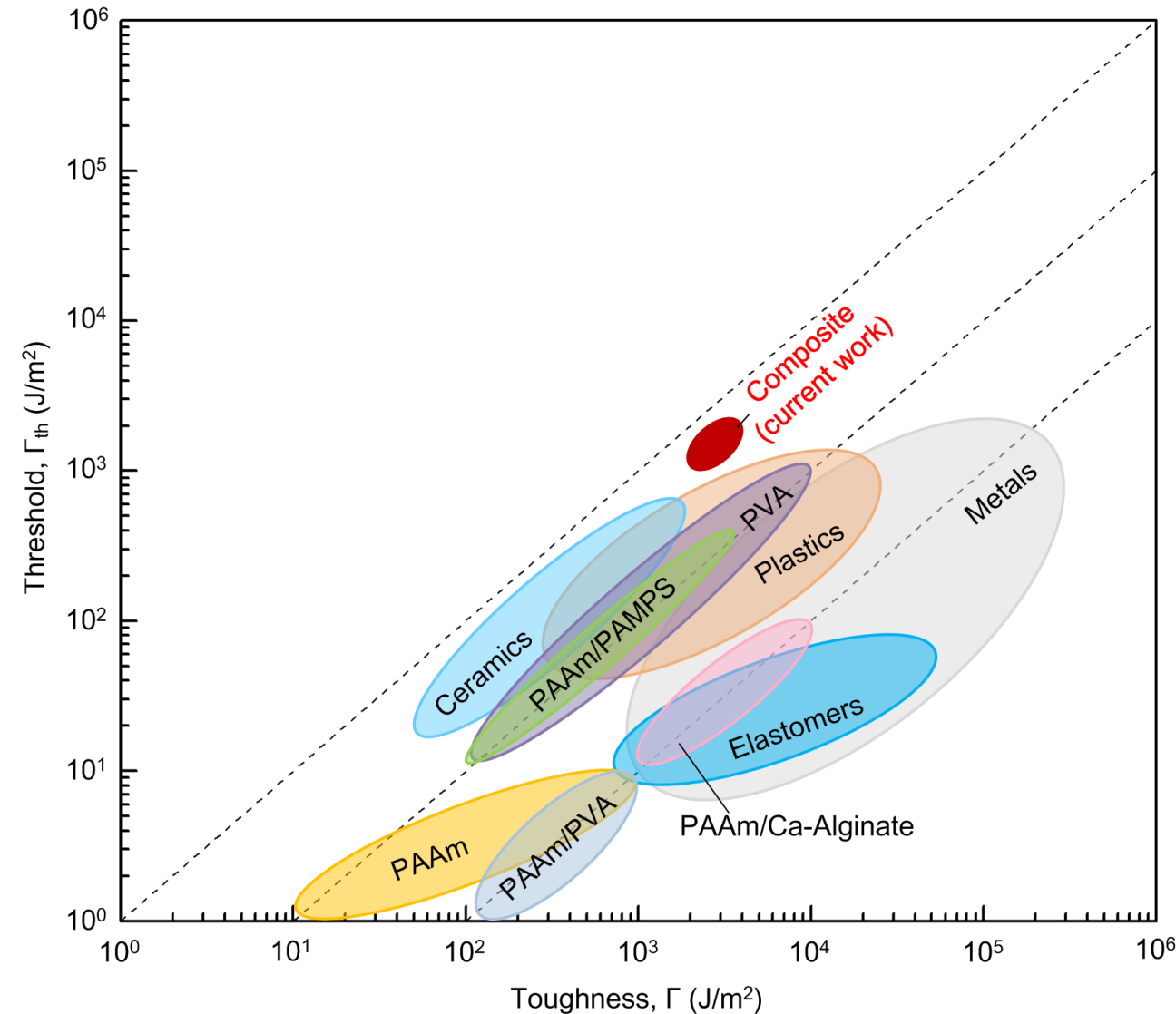
(e) $\lambda_m = 1.725$, $G = 1,290 \text{ J/m}^2$, $N=30,000$



- Matrix: PAAm hydrogel
- Fibers: PDMS elastomer

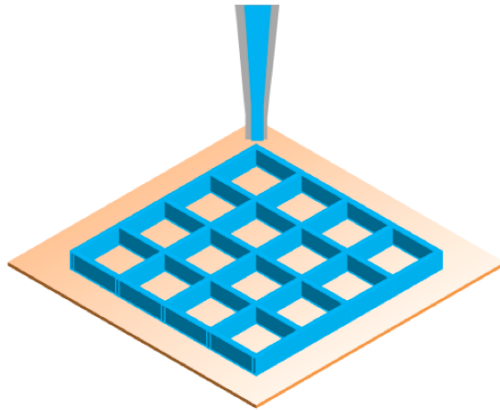


Fatigue-resistant hydrogel-elastomer hybrid

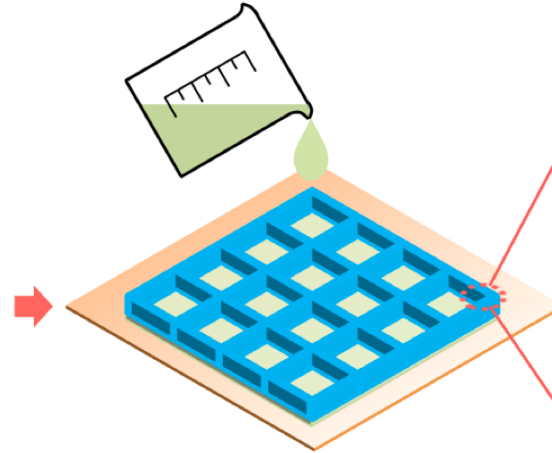


Elastic dissipater of any architecture

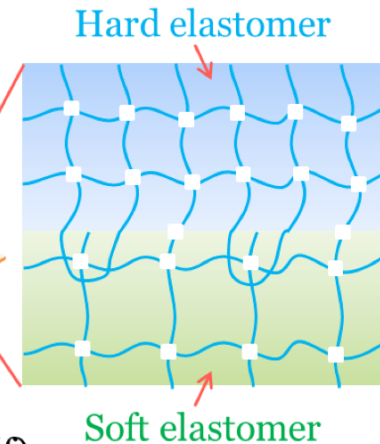
(a) Print hard elastomer



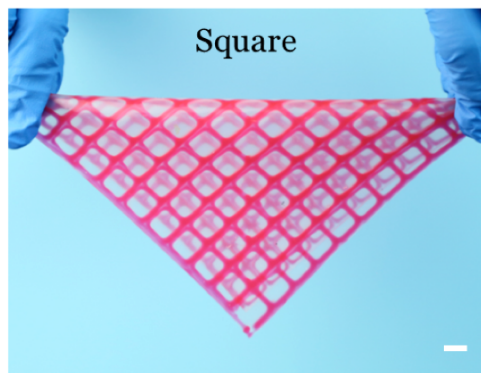
(b) Infuse precursor of soft elastomer



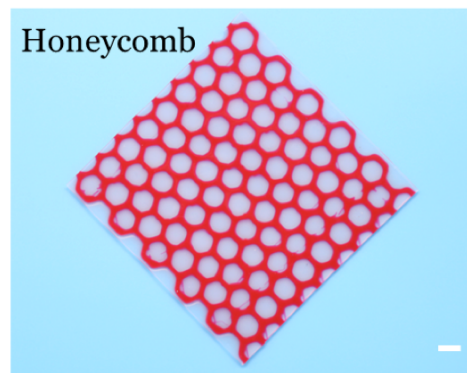
(c) Composite with strong adhesion



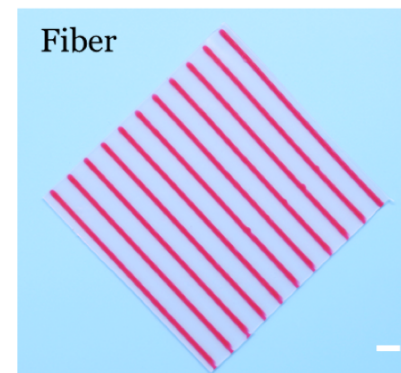
(d)



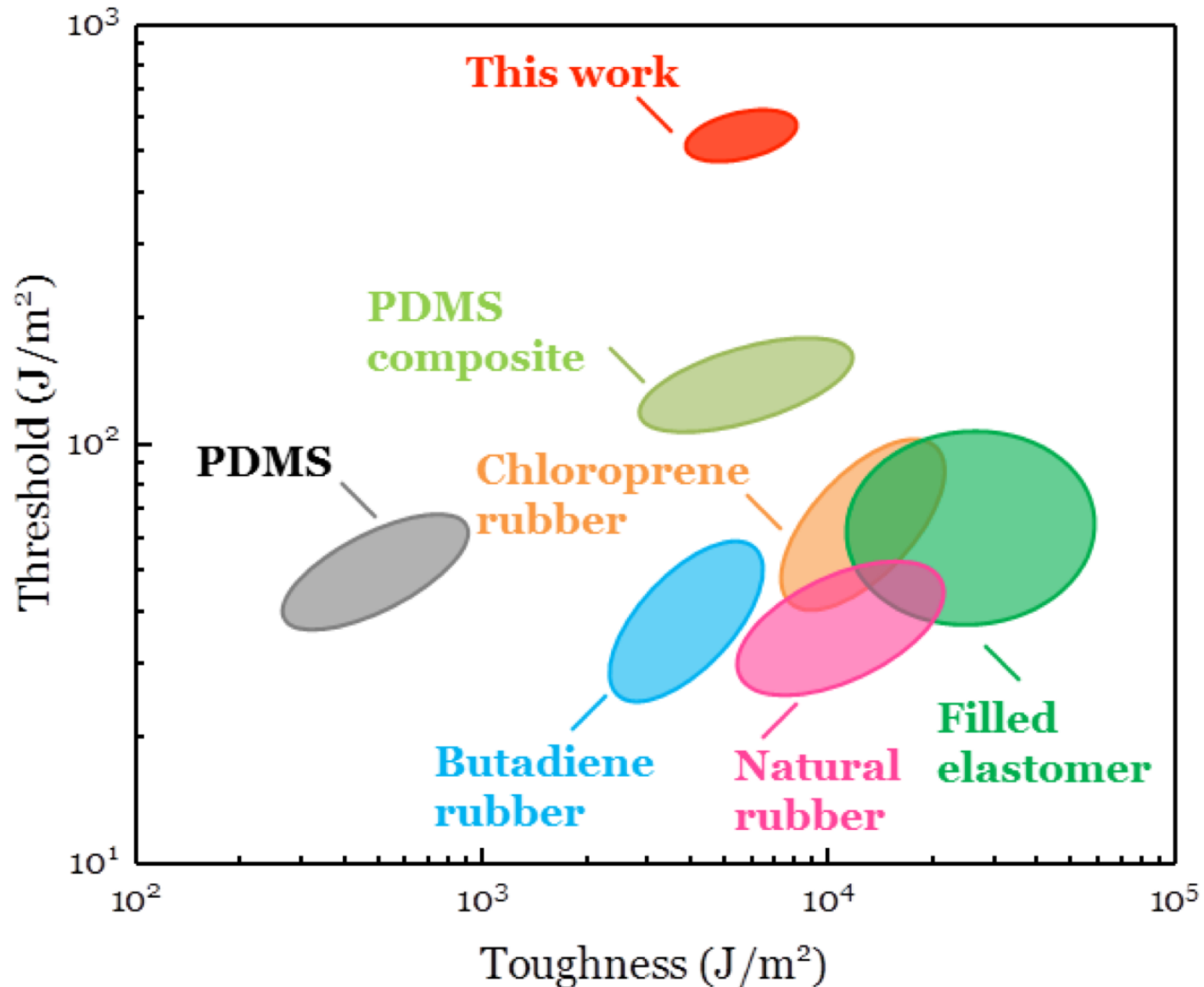
(e)



(f)



Fatigue-resistant elastomer



Summary

- **Inelastic dissipater** enhances toughness, but not threshold.
- Double networks **remove the stiffness-threshold conflict**.
- **Elastic dissipater**: low hysteresis, high toughness, high threshold.
- Winning the war on fatigue.