# Bioinspired Hydrogel Scaffolds, Electronics and Machines

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MIT ILP Conference

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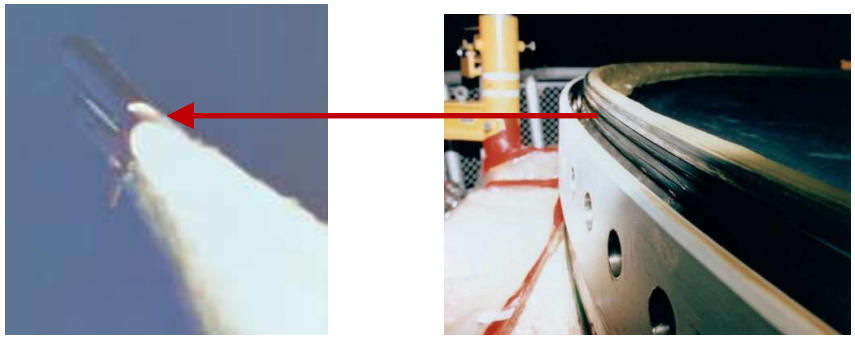
# Impact of Soft Materials: from engineering to health



- Failure of O-ring seal caused the Space Shuttle Challenger disaster in 1986.
- Approximately 17 percent of Americans witnessed the launch and disaster.

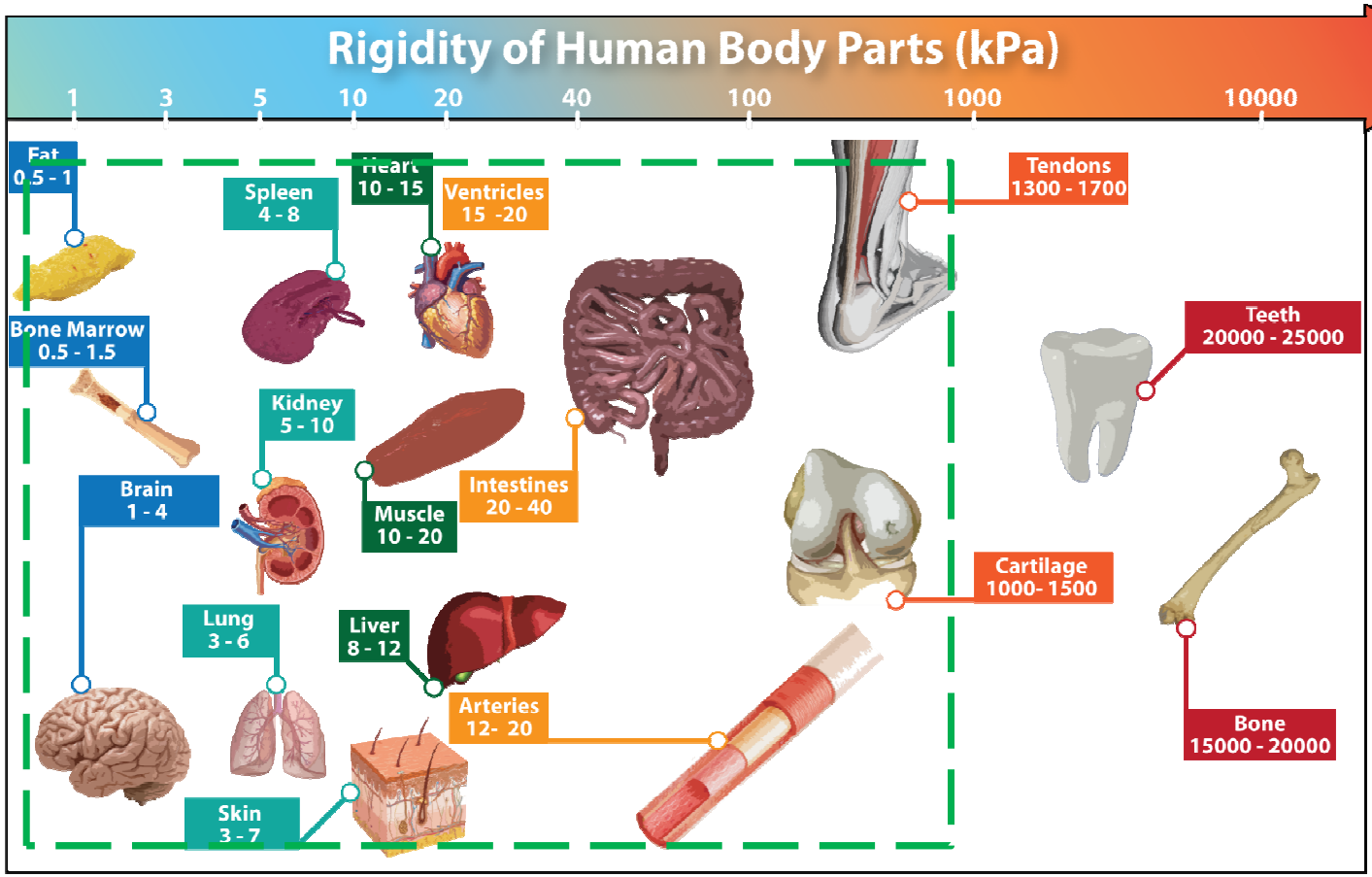
# Impact of Soft Materials: from engineering to health

**In collaboration with Samsung Display**



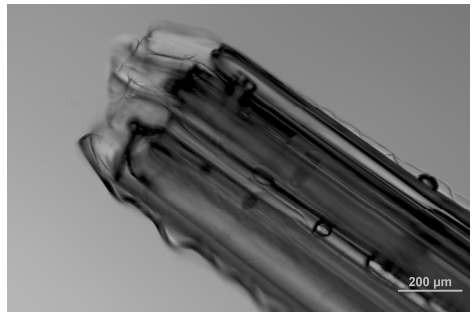
**Replace traditional rigid components with polymers and gels to enable flexible electronics and wearable devices.**

# Impact of Soft Materials: from engineering to health




- Modulus: 1kPa~10MPa;
- Water concentration: 70%
- Robust: under millions of cycles of loads.

Future biomedicine relies on soft materials with similar properties as body.

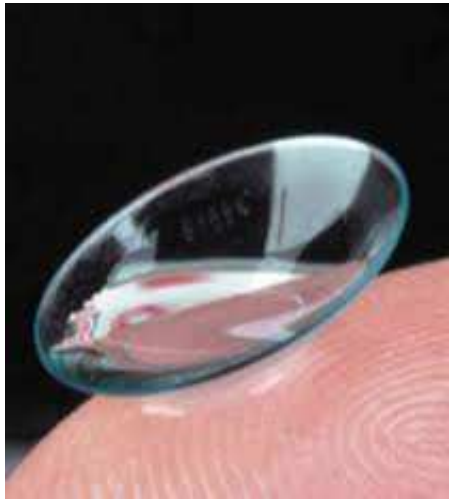



Hydrogel Neural Probe with Anikeeva

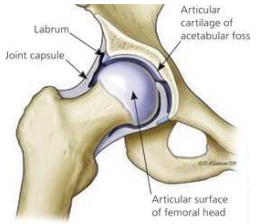
# Hydrogels

# Robust

# Conventional hydrogels are weak and brittle

Conventional hydrogel
Fracture toughness ~1 $Jm^{-2}$



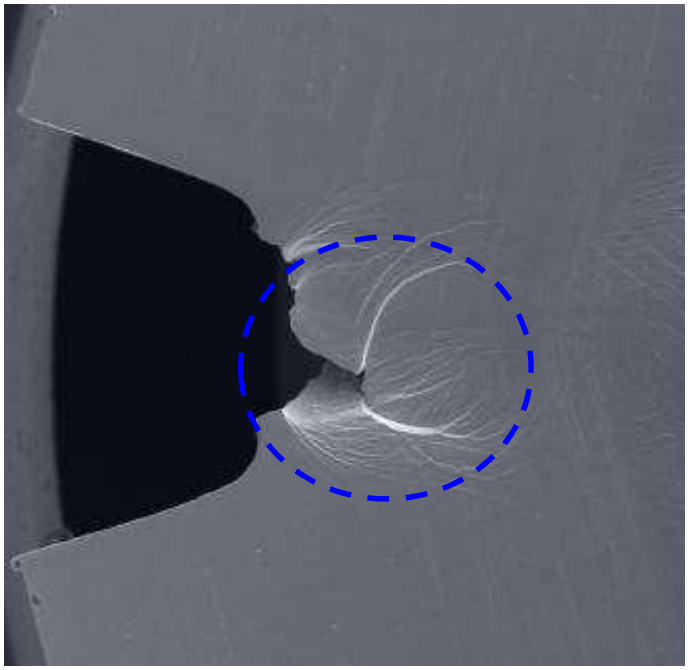

Cartilage
Fracture toughness
~1,000 $Jm^{-2}$

# Glass

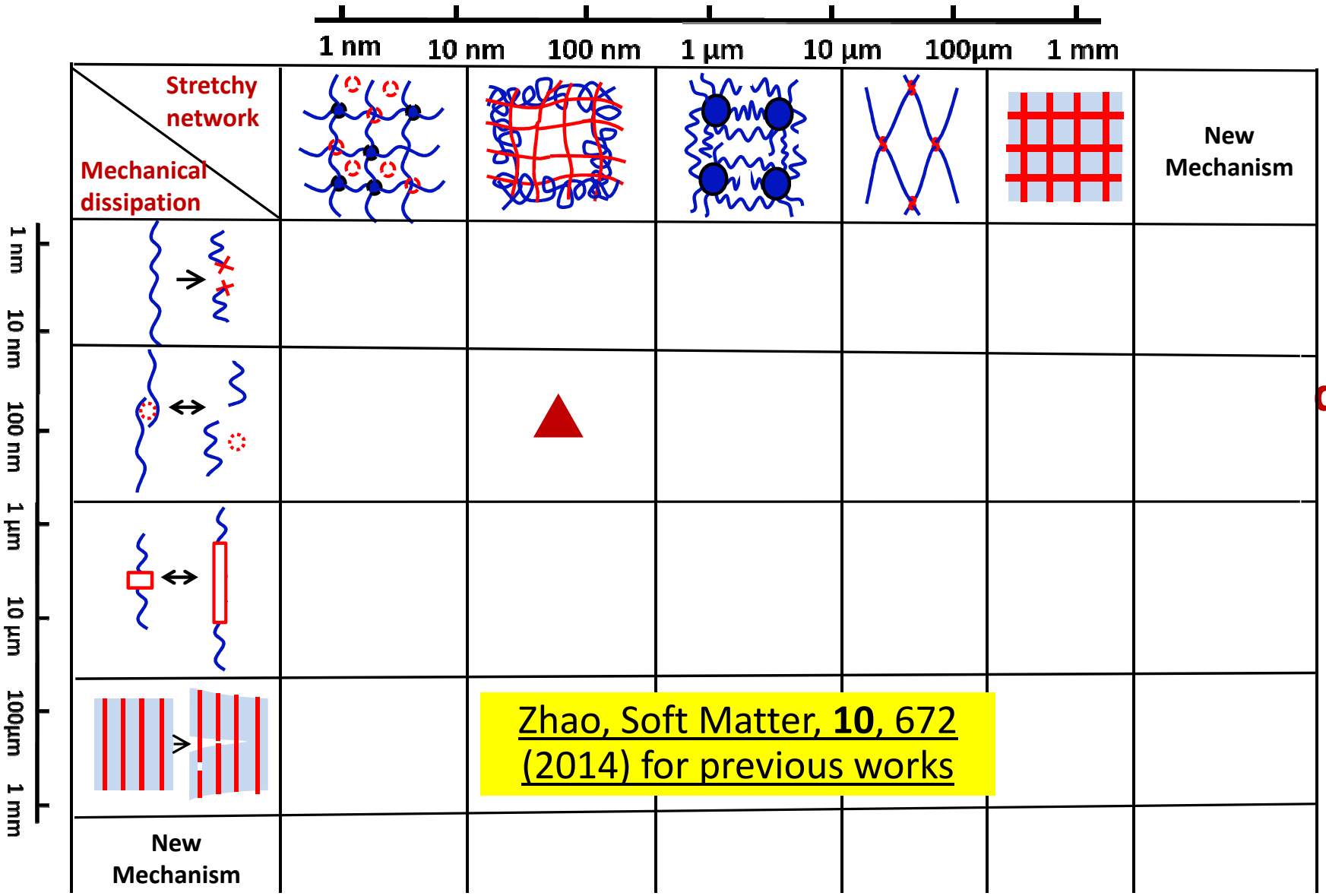
Fracture toughness
= Surface energy

Zhao, Soft Matter, 10, 672 (2014)

# Metal



Fracture toughness
= Surface energy +
Dissipation in a zone

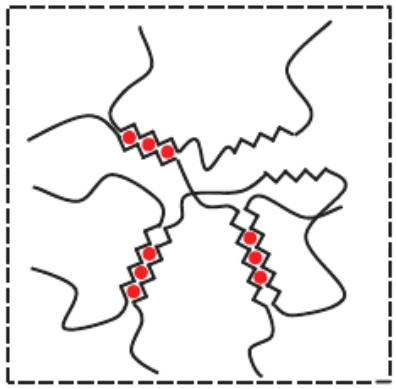



**Build dissipation into stretchy networks**

# Tough hydrogels: build dissipation into stretchy network

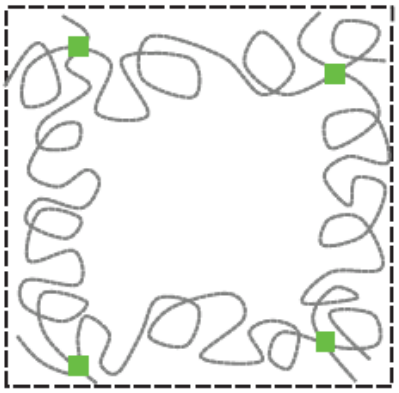
## Individual Components are very Brittle!

**Dissipation**

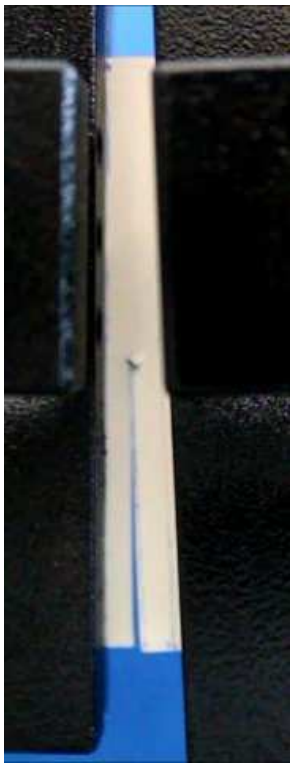


**Reversible crosslink:**
**Alginate $Ca^{2+}$**
**dissipates energy**

**Stretchy network**



**Long-chain network:**
**PAAm maintains high elasticity**



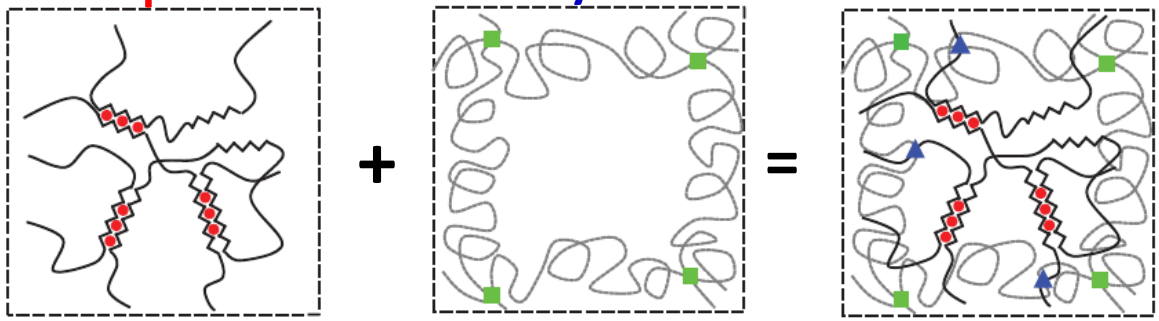
**Alginate**
**~50 $Jm^{-2}$;**

**PAAm**
**~50 $Jm^{-2}$**

# Tough hydrogels: Build dissipation into stretchy network

## Nanoscale Interpenetration gives Extremely High Toughness

**Dissipation** + **Stretchy network** =



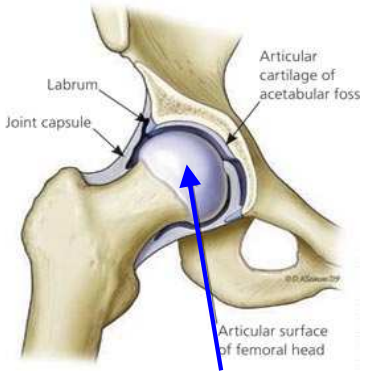
**Reversible crosslink:**
**Alginate+$Ca^{2+}$**

**Long-chain network:**
**PAAm**

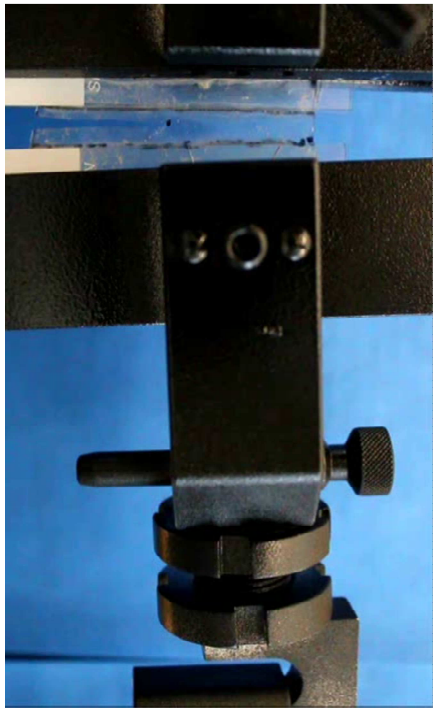
- **~90% water**
- **Fracture energy 9000 $Jm^{-2}$**
- **Stretchability 21 times**

Sun et al, Nature, **489**, 133 (2012)

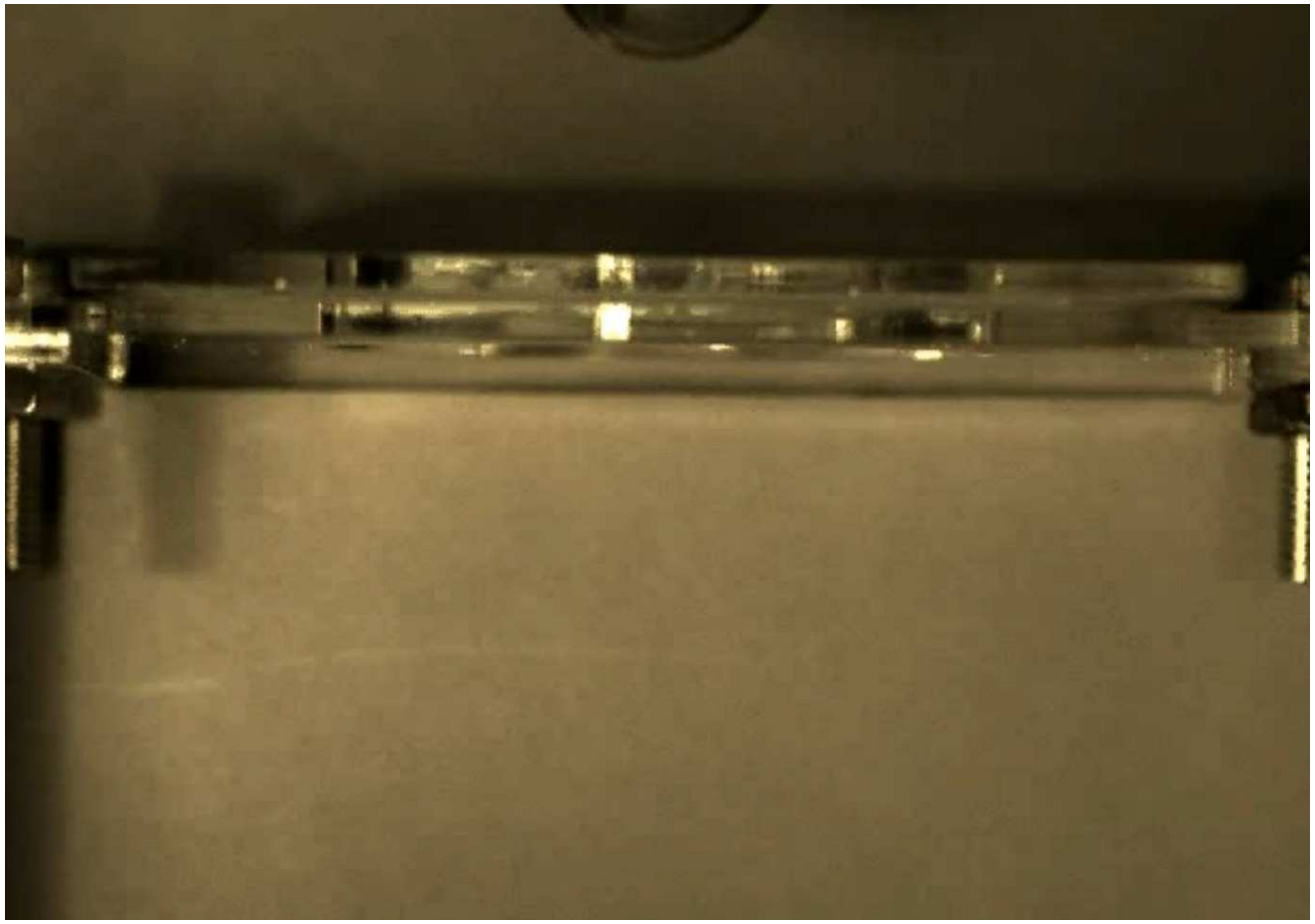
**in collaboration with Prof. Suo, Vlassak and Mooney**



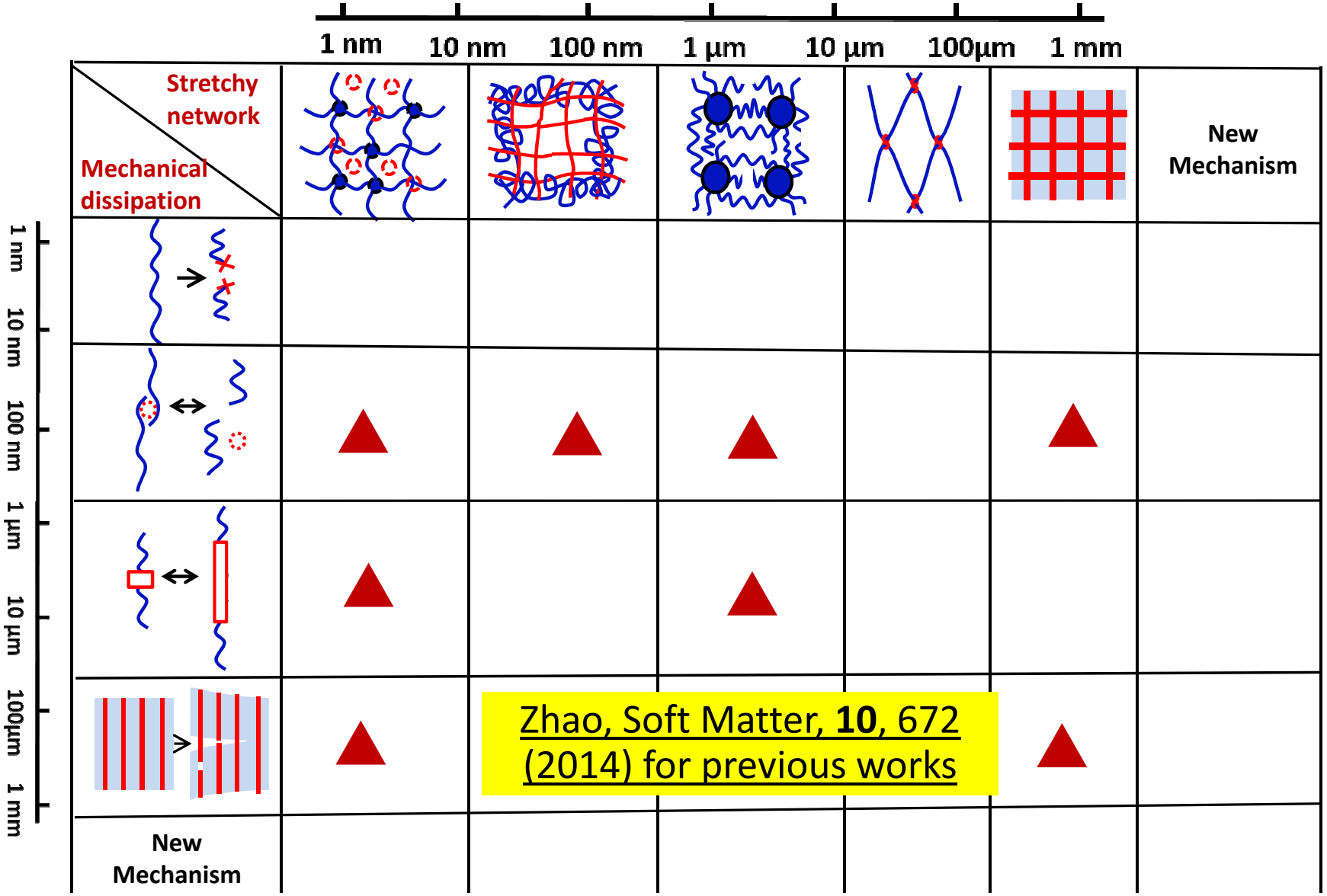

**>1000$Jm^{-2}$**



# Hydrogel film with 90% water and 1mm thickness



**Impact of a ball of 64 g at 6m/s**




**Polymer Candidates:**

- PAA
- PAAm
- PEG
- Alginate
- Chitosan
- ELP
- ….

**Nano-filler Candidates:**

- CNT
- Graphene
- Graphene oxide
- Nanoclay
- ….

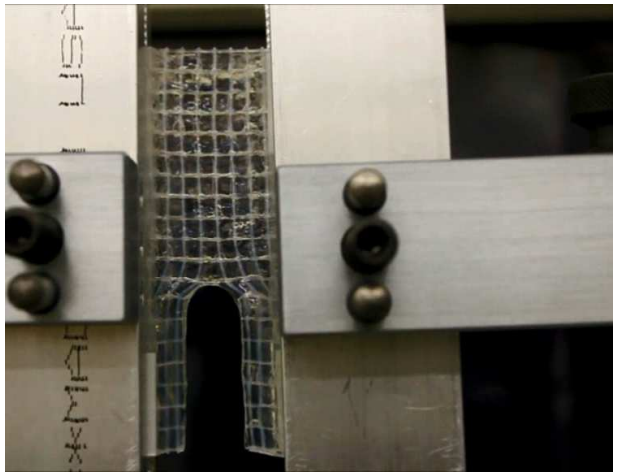
**Macro-fiber Candidates:**

- PLG
- PCL
- PGA
- …..

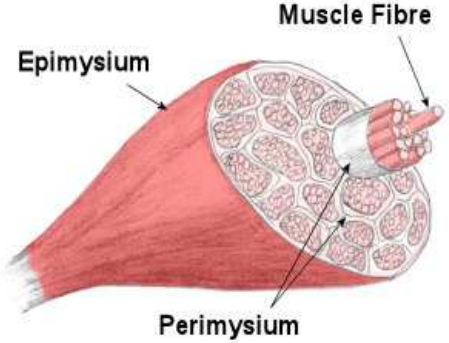
# Tough Hydrogels -- A Wide Range of Rigidity with Diverse Polymers


**1~5 kPa;**
**1,000 $Jm^{-2}$**

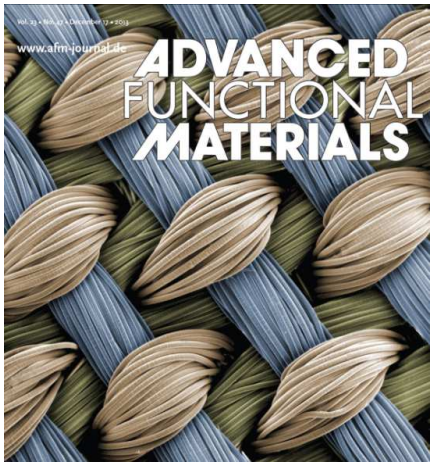
Lin et al, Extreme Mechanics Letters, 1, 70 (2014)



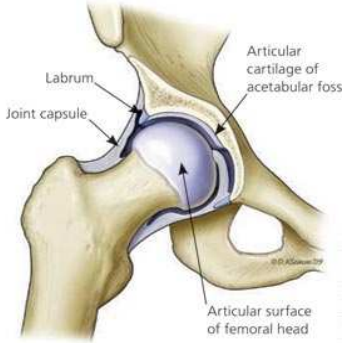
**10~100 kPa;**
**1~10 $kJm^{-2}$**




Sun et al, Nature, **489**, 133 (2012)
Lin et al Soft Matter, 10, 7519, (2014)

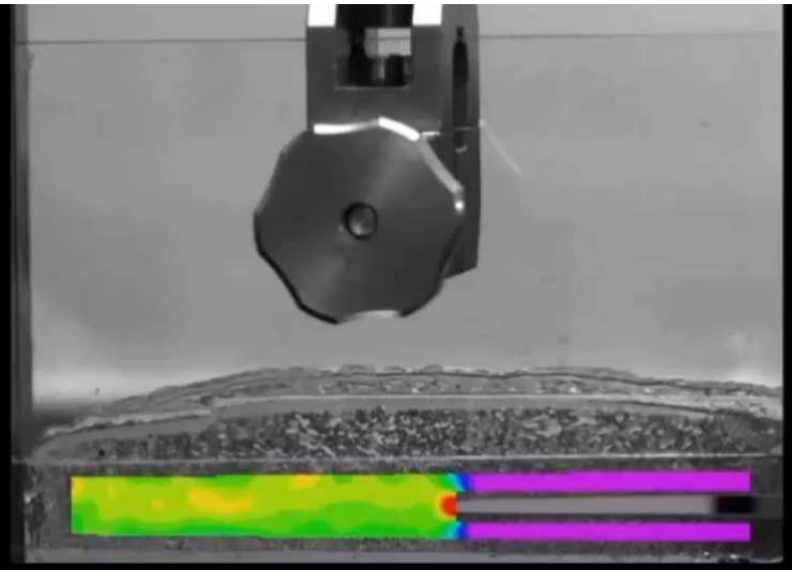


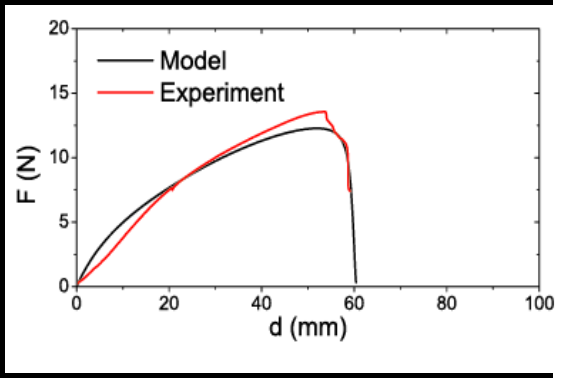

**1~10 MPa**
**10~50 $kJm^{-2}$**



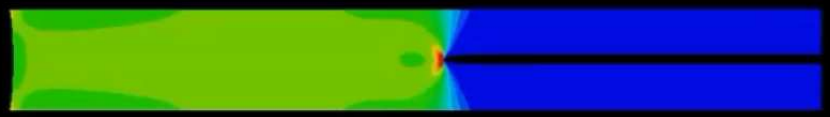

Liao et al, Advanced Functional Materials, **47**, 5833 (2013);

Experiment





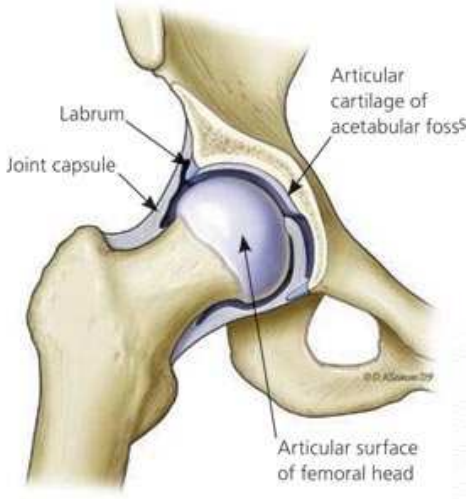

Model
(No fitting parameter)

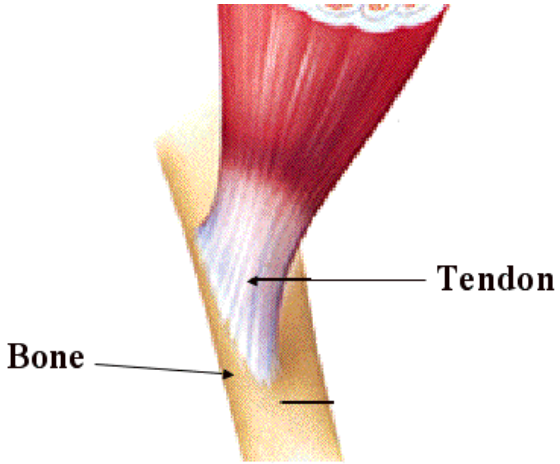


# Hydrogels

# Adhesive

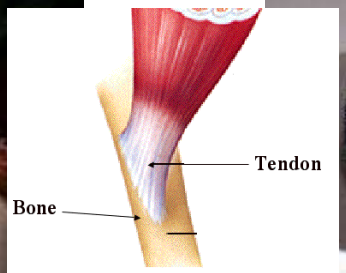
# Adhesive Hydrogels in Nature

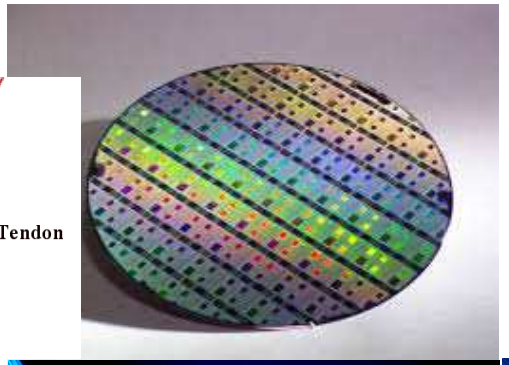


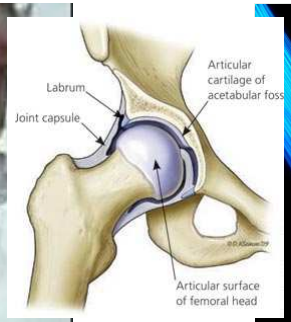



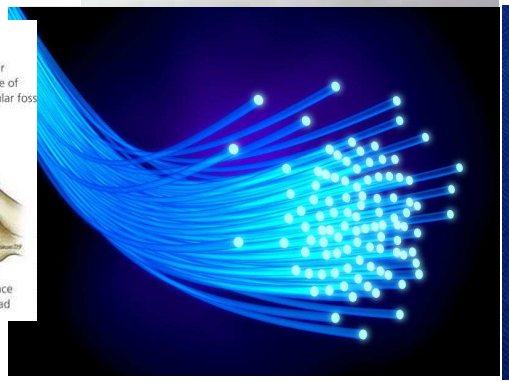

**Interfacial toughness ~800Jm$^{-2}$**

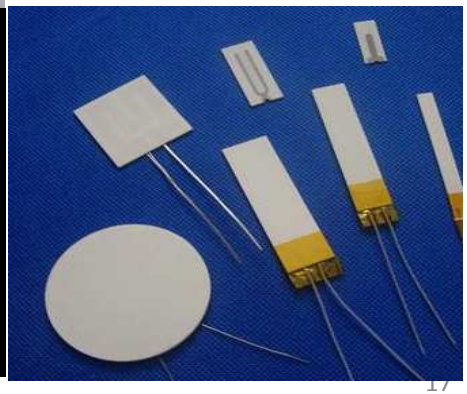
# Design Adhesive Hydrogels





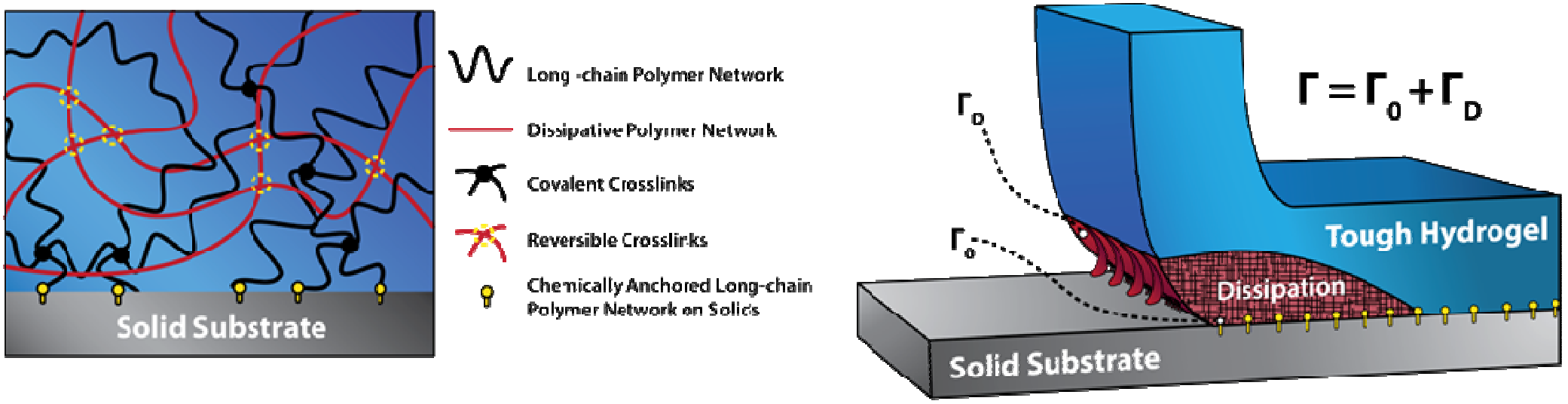


90% water; ~kPa

Diverse engineering solids; MPa~GPa

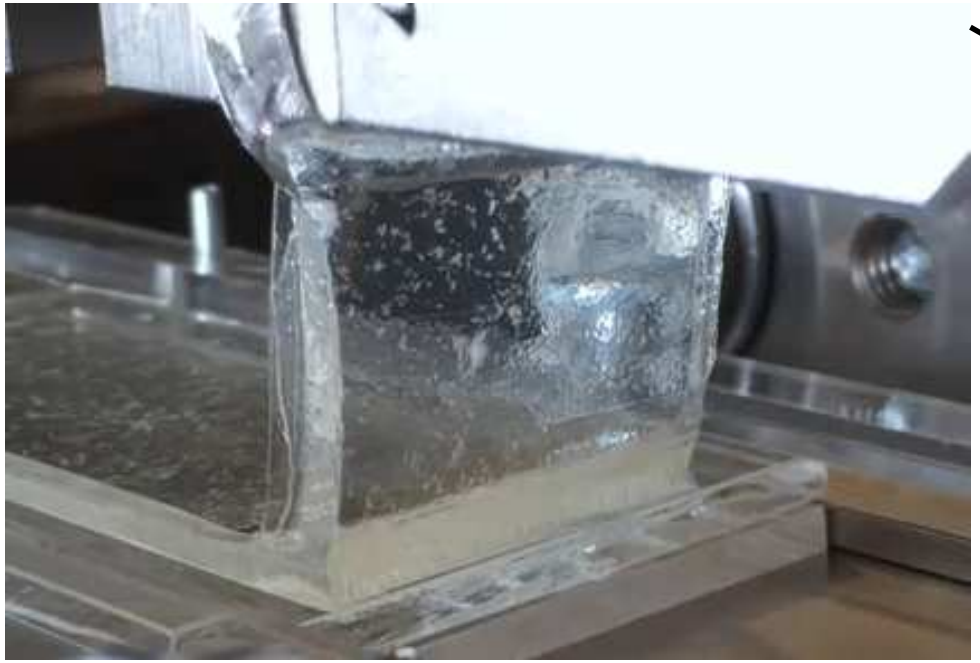
# Tough Hydrogels: Build dissipation into stretchy network.

## Adhesive Hydrogels: Anchor stretchy network on hard material.

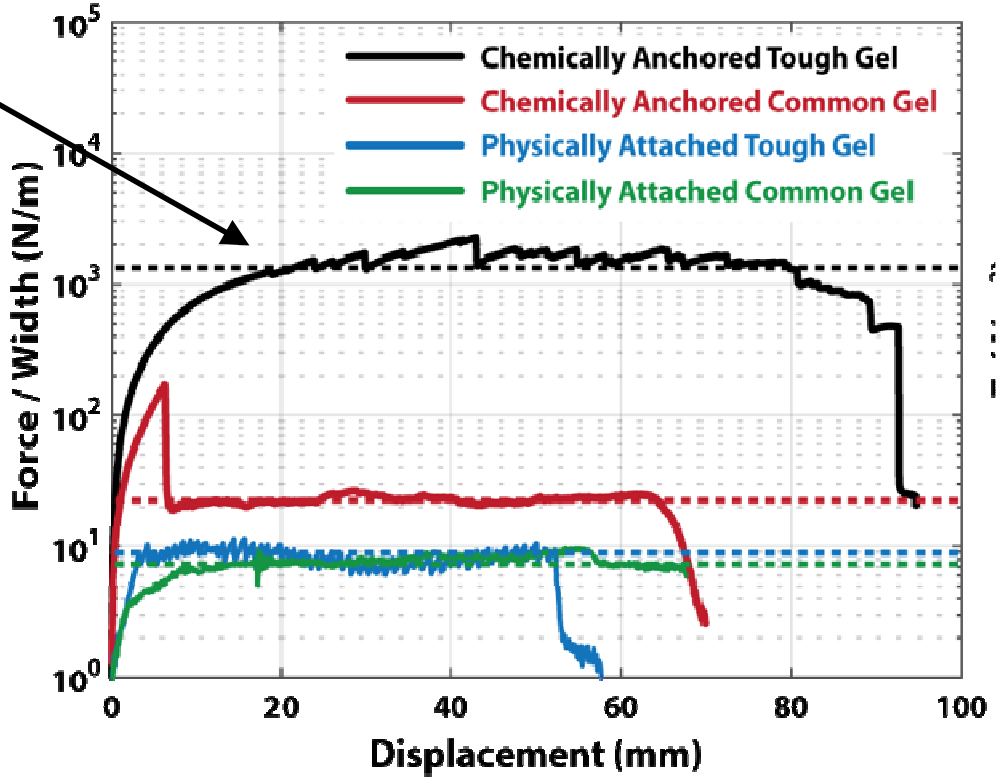



Yuk et al, Nature Materials, 15, 190-196 (2016);
Yuk et al, Nature Communications, 7, 12028 (2016)

# Hydrogels with 90% water bonded on Si, $SiO_2$, glass, ceramics, Ti, Al, Fe et al

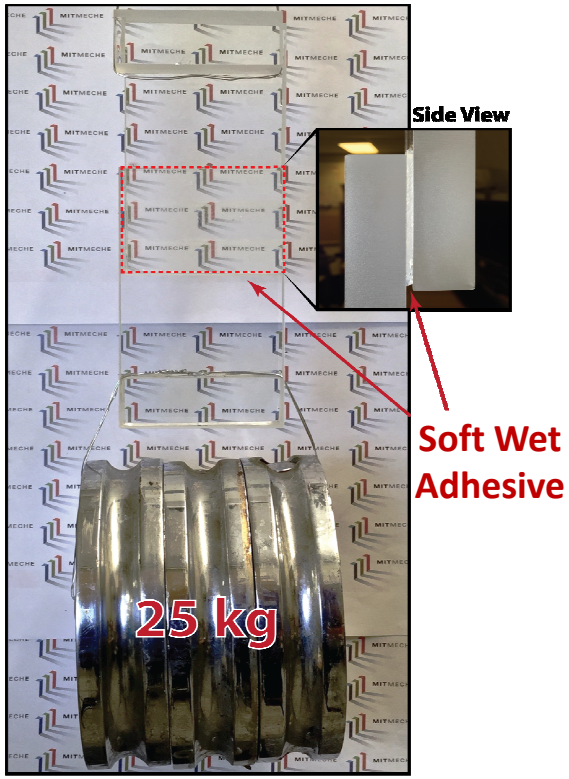


Detachment and Finger Instability



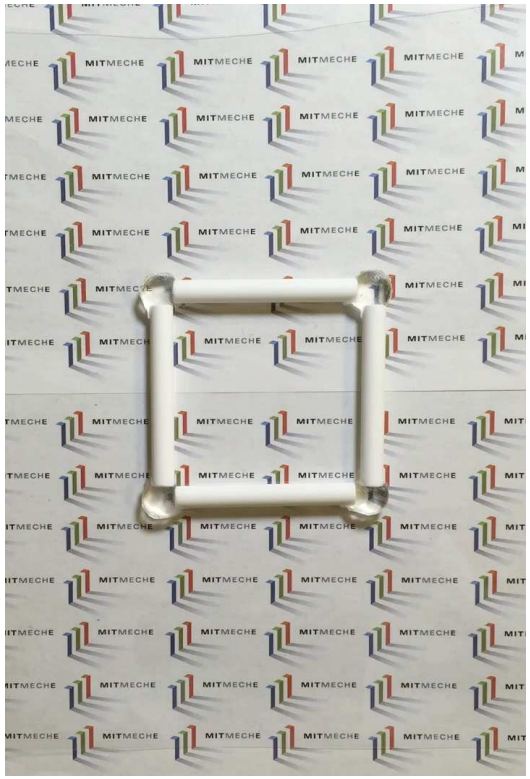

Yuk et al, Nature Materials, 15, 190-196 (2016);

# Hydrogel-Engineering Material Hybrids

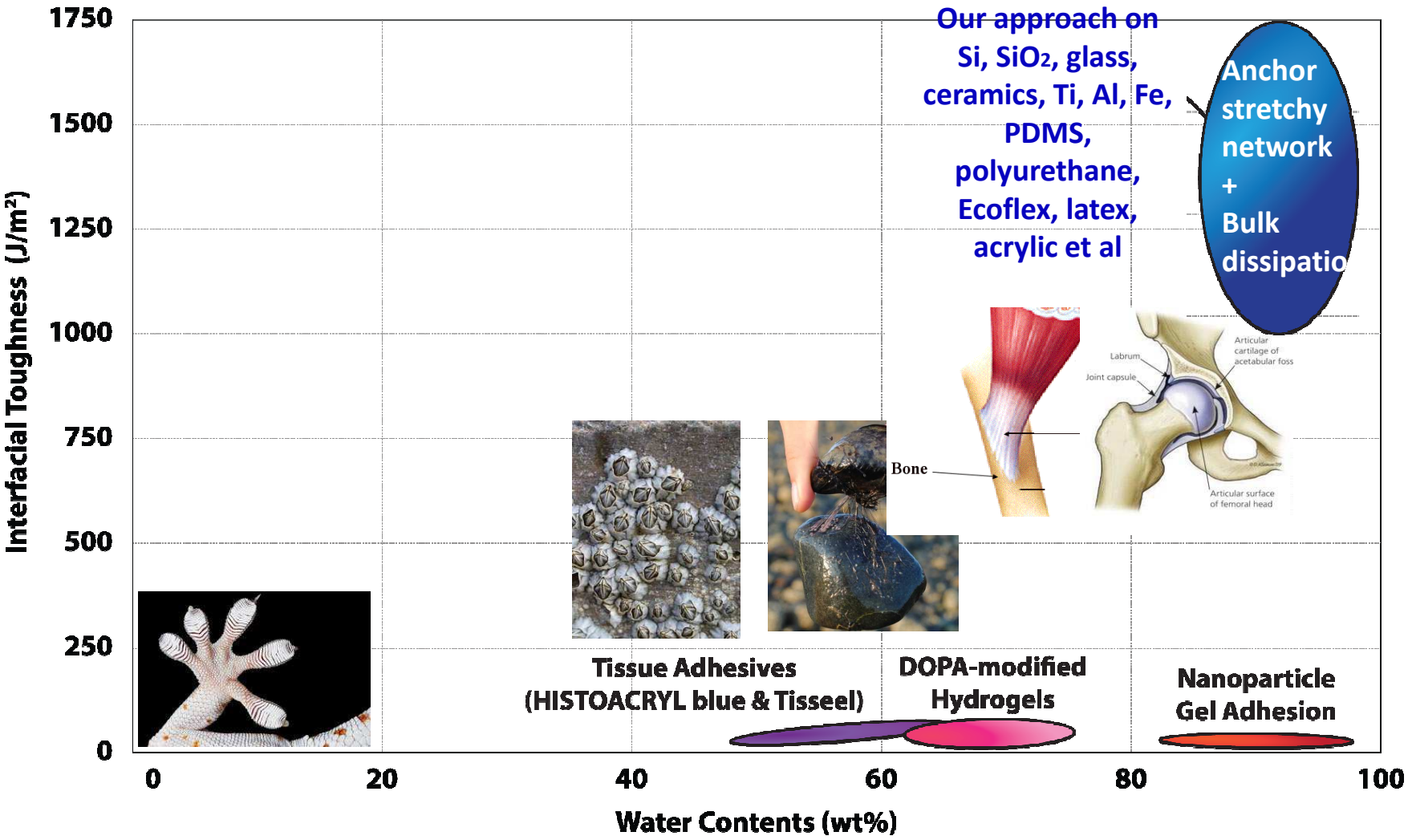



With glass

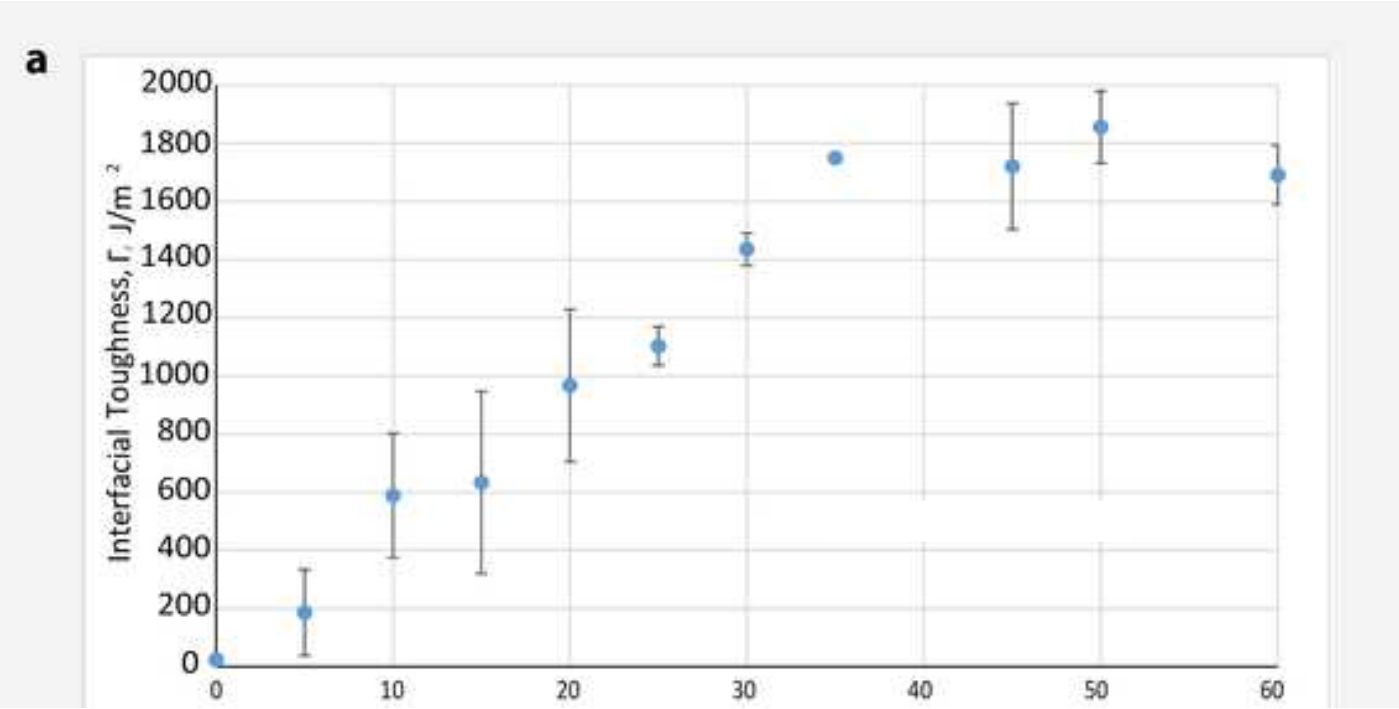
With silicon



With ceramics


Interfacial Toughness (J/m²)
1750
1500
1250
1000
750
500
250
0
0
20
40
60
80
100
Water Contents (wt%)
Our approach on
Si, SiO2, glass,
ceramics, Ti, Al, Fe,
PDMS,
polyurethane,
Ecoflex, latex,
acrylic et al
Anchor
stretchy
network
+
Bulk
dissipatio
Bone
Labrum
Joint capsule
Articular
cartilage of
acetabular foss
Articular surface
of femoral head
Tissue Adhesives
(HISTOACRYL blue & Tisseel)
DOPA-modified
Hydrogels
Nanoparticle
Gel Adhesion

# Tunable Adhesion from 1 to 1000 Jm-2




100 Jm-2

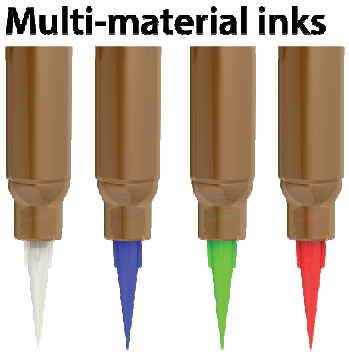
1000 Jm-2

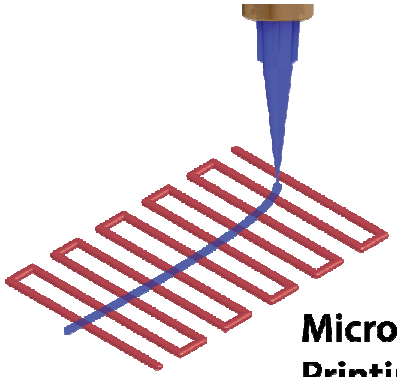
Parada et al, In submission (2016)

# Hydrogels

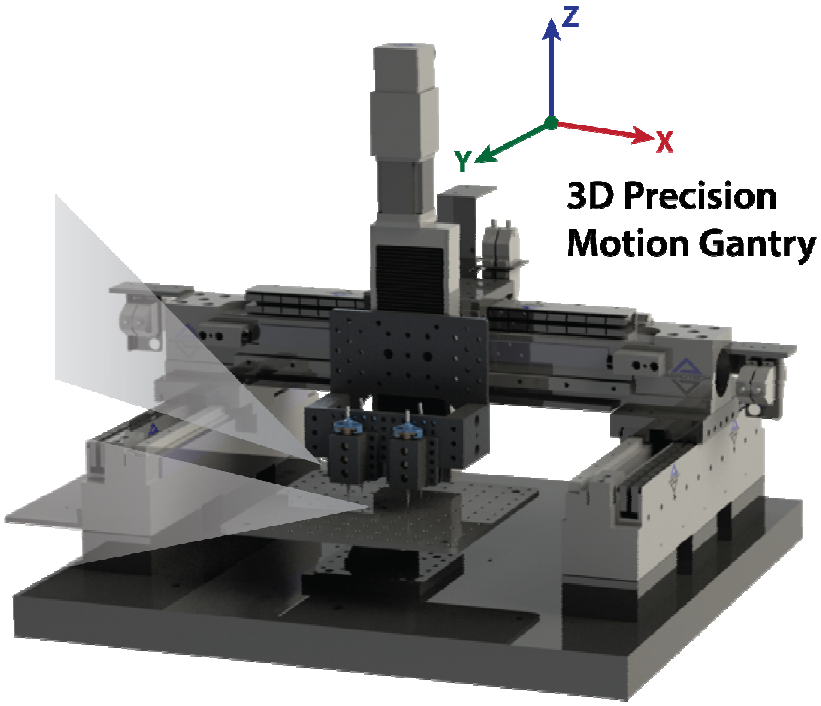
# Manufacturing

# Multi-material 3D Printer



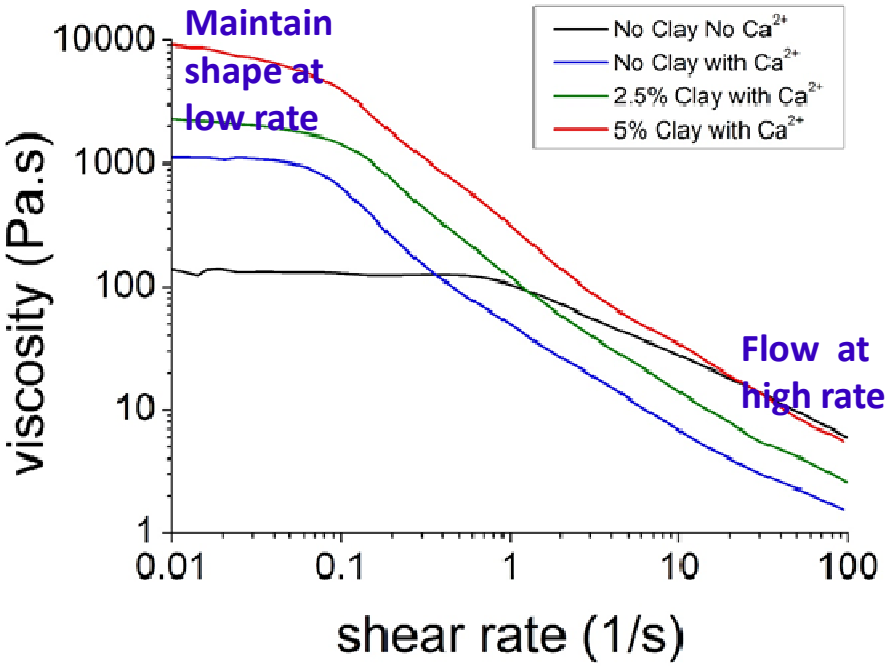



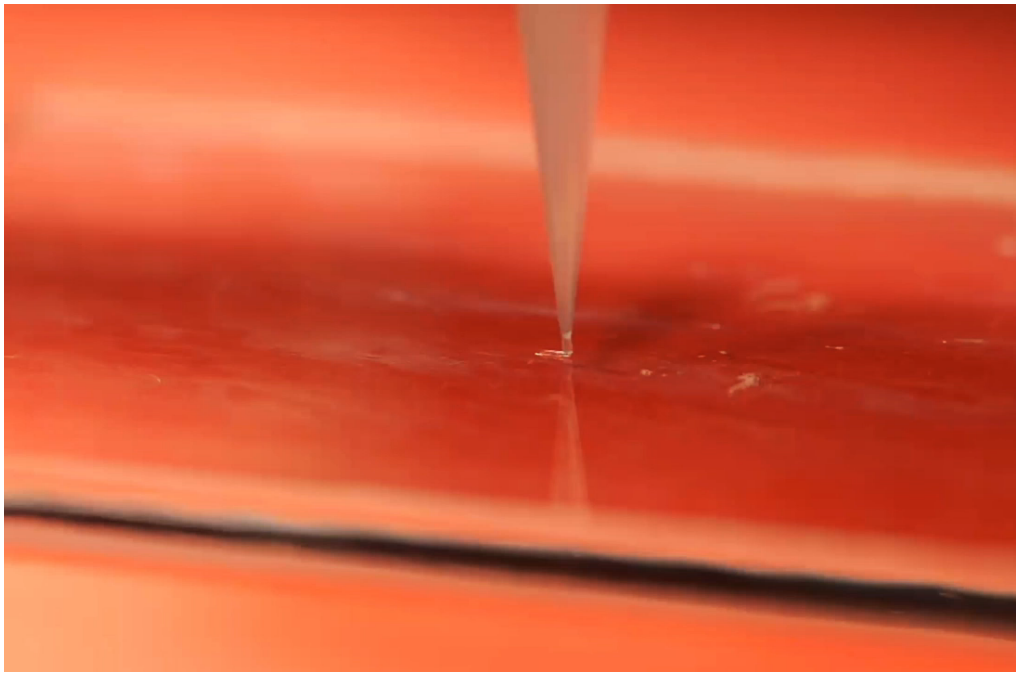
Microscale
Printing of Diverse Materials




- Micro-nozzle based
- Resolution up to 1~5um.
- Printing multiple materials in one structure
- Particularly suitable for soft materials and biomaterials.

# Hydrogel Microstructures by 3D Multi-material Printing

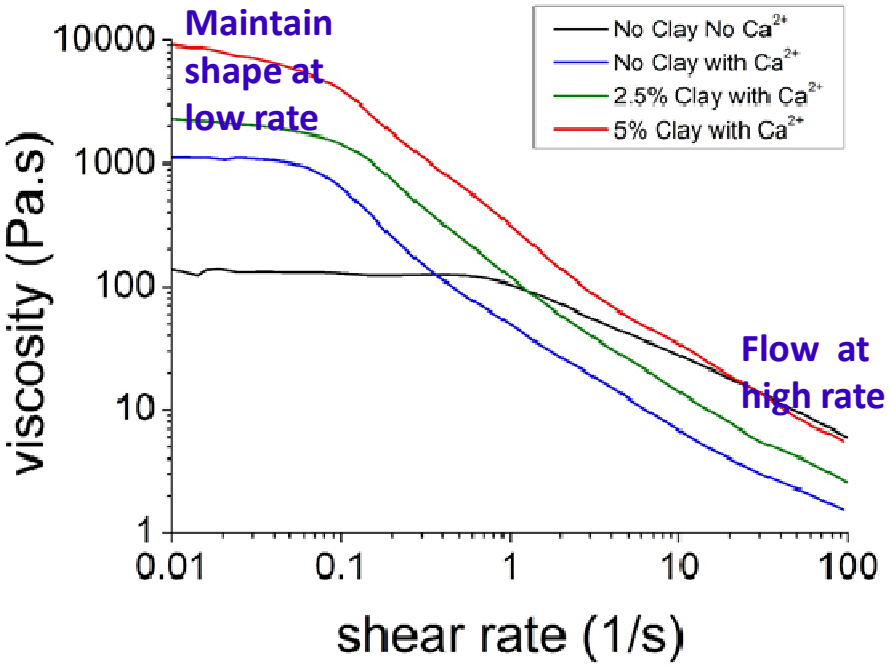


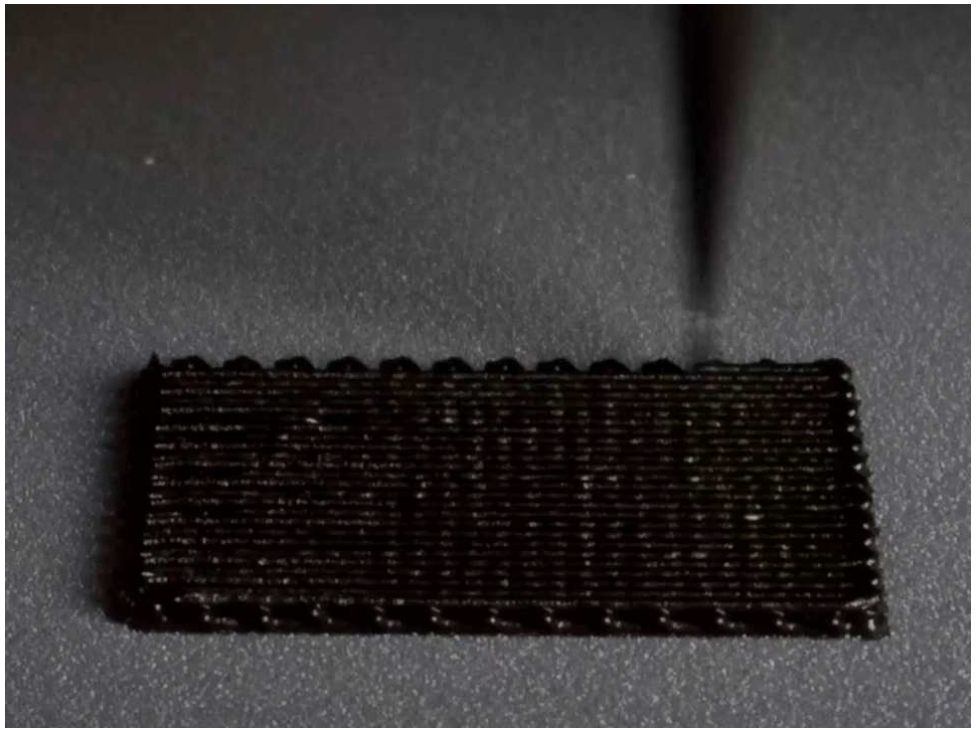



**Ink: Shear-thinned polymer solution & Robust hydrogel formula**

Hong et al Advance Materials, 27, 4035 (2015)

# Hydrogel Microstructures by 3D Multi-material Printing

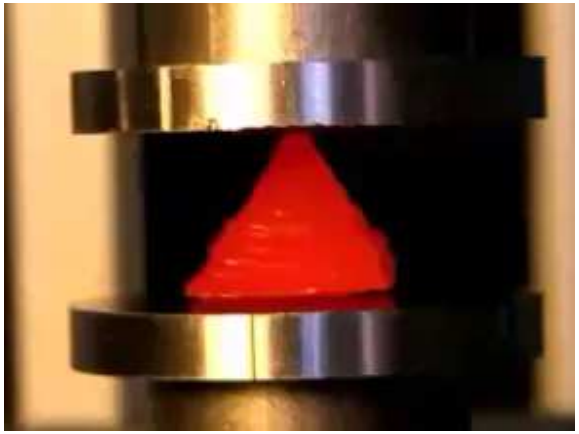


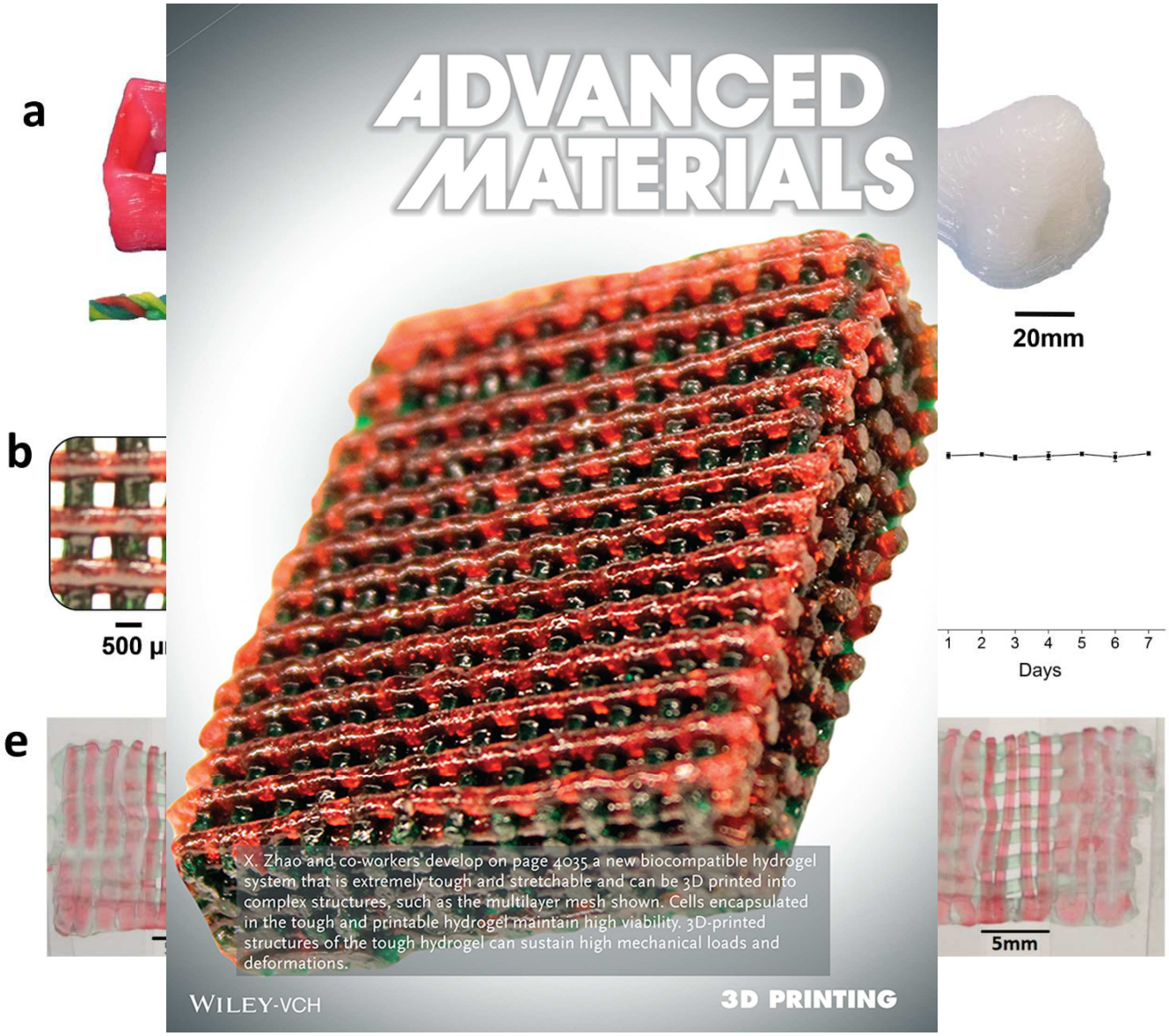
**Ink: Shear-thinned polymer solution & Robust hydrogel formula**

Hong et al Advance Materials, 27, 4035 (2015)

# Tough Soft Microstructures by 3D Printing



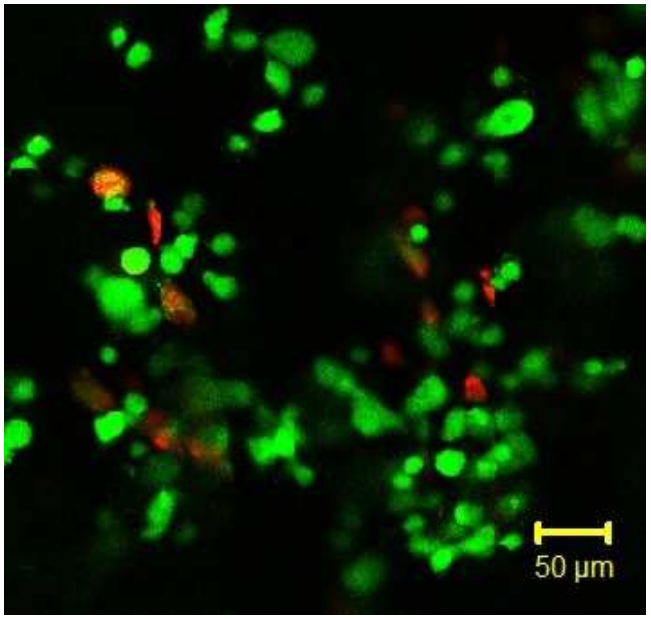
Hong et al Advance Materials, 27, 4035 (2015)



# Hydrogels

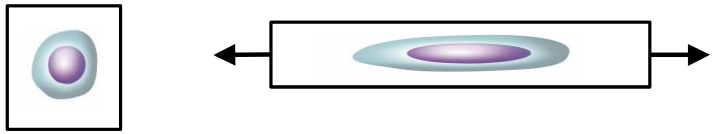
## Bio-scaffolds, Electronics, Machines and Robots

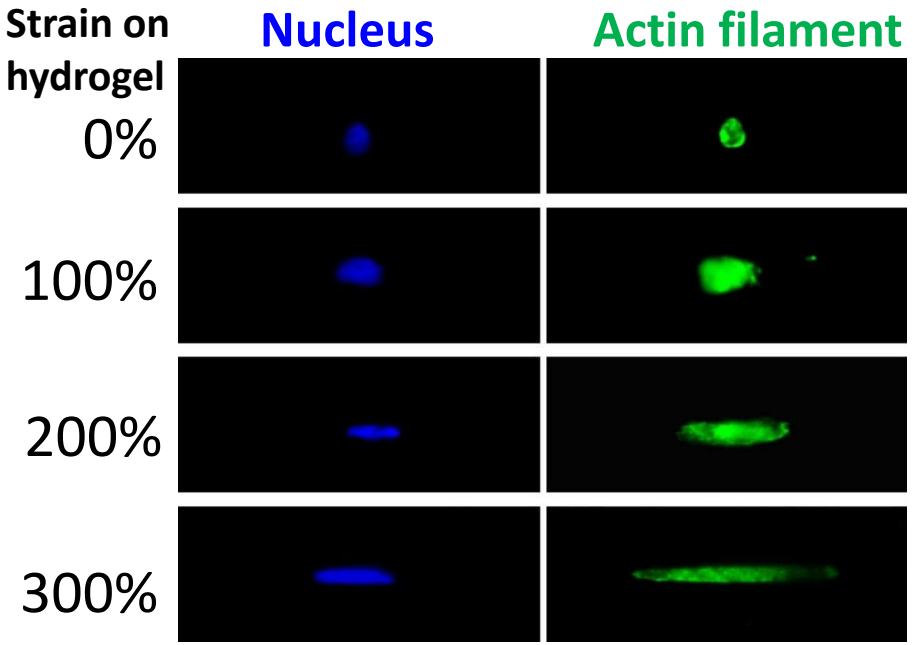
# Growing Cells and Tissues in Hydrogel Scaffolds




**Human mesenchymal stem cells (hMSC)**

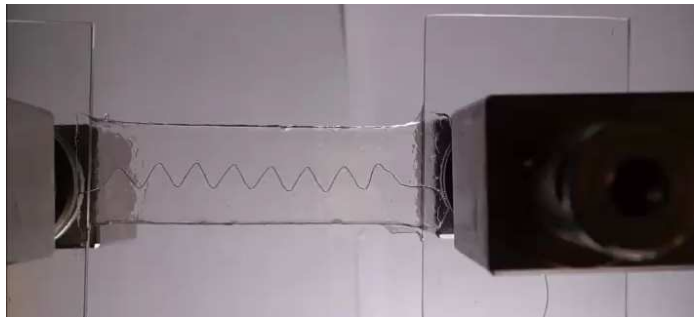
**live dead assay**



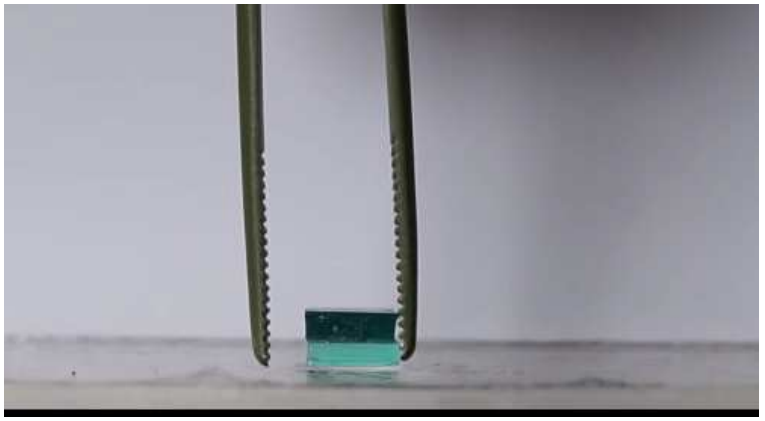



Hong et al Advance Materials, 27, 4035 (2015)

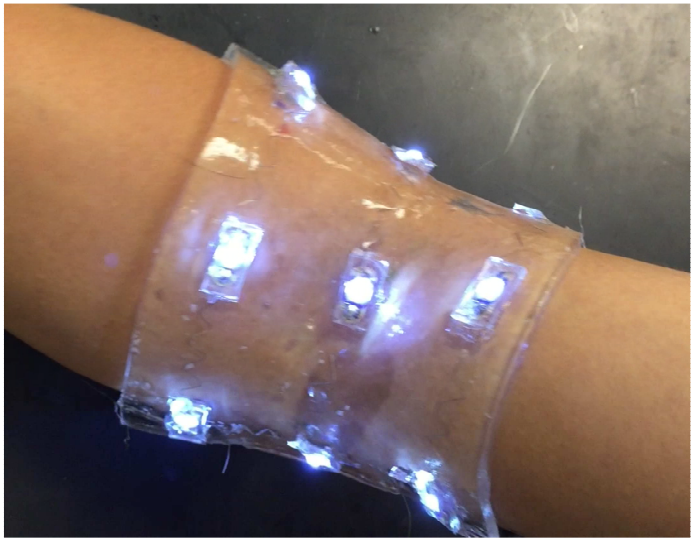
# Integrating Electronics with hydrogels (70~90% water)



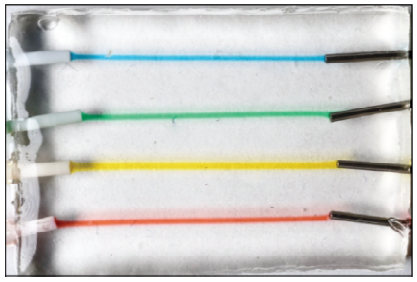
Conductive wires



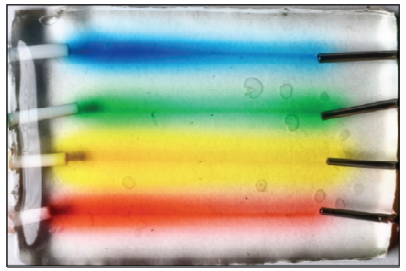
Functional islands



LED arrays



t = 0 min, λ = 1

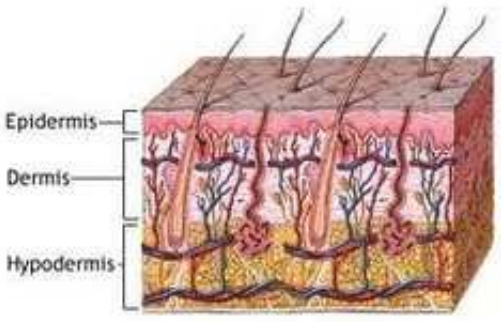


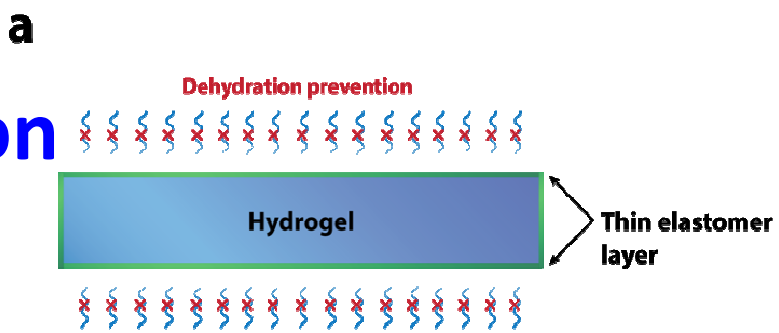
t = 120 min, λ = 1

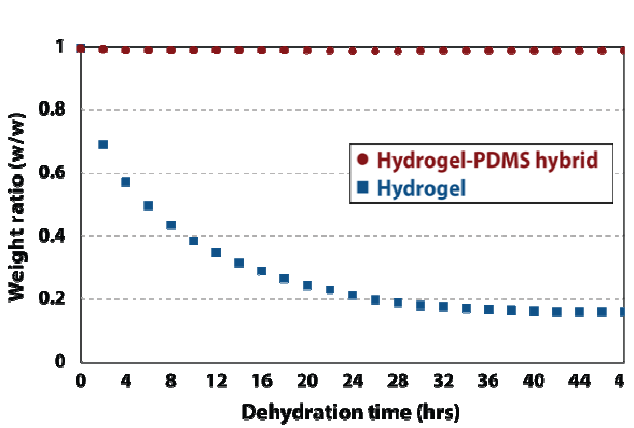
Drug delivery channels

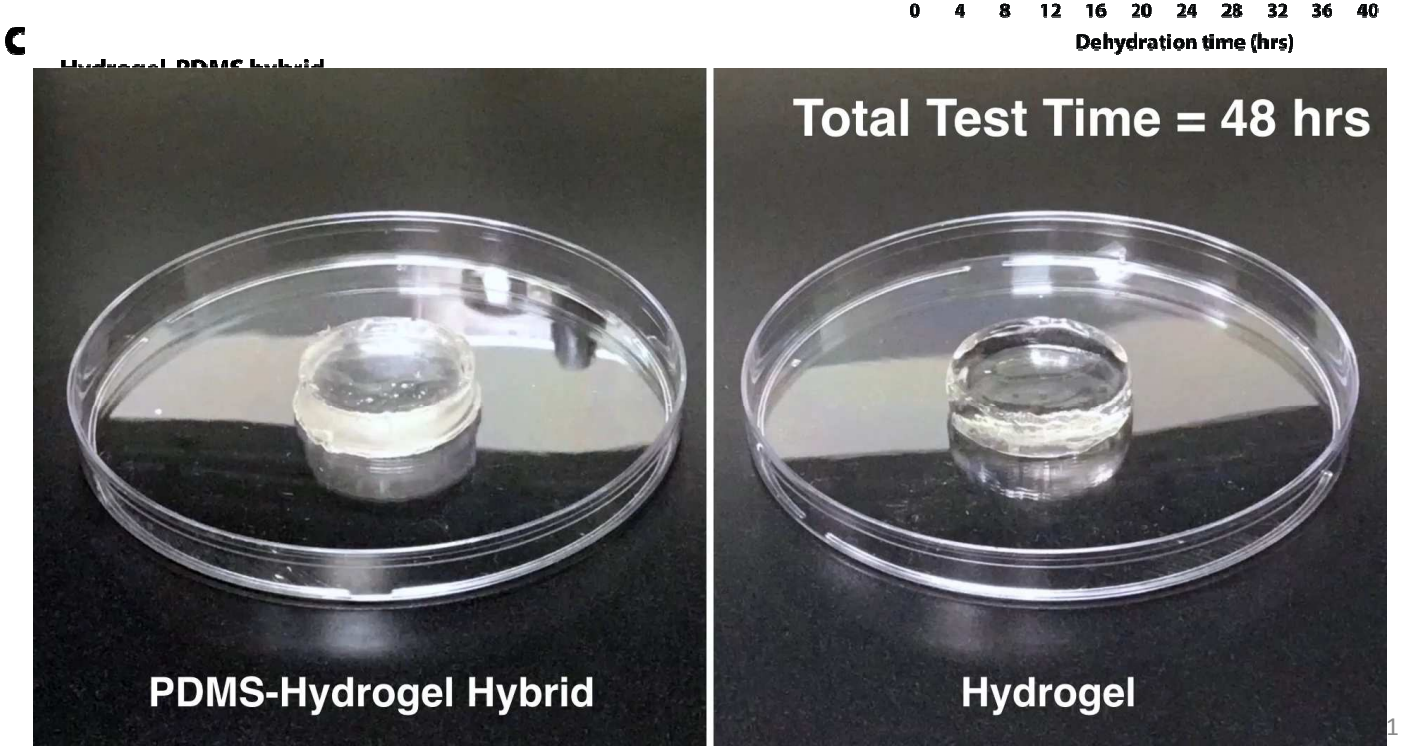
Lin, et al, Advanced Materials, 28, 4497–4505(2016)

# Anti-dehydration hydrogel



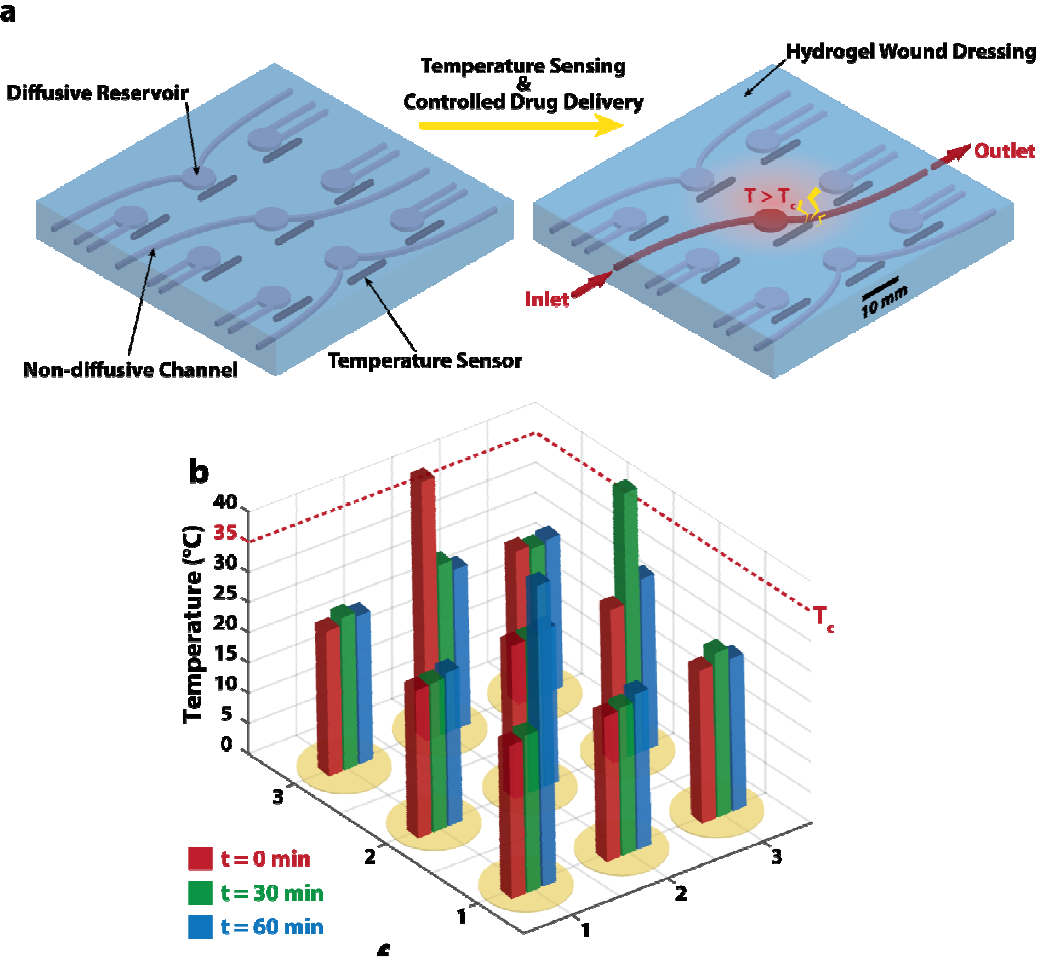



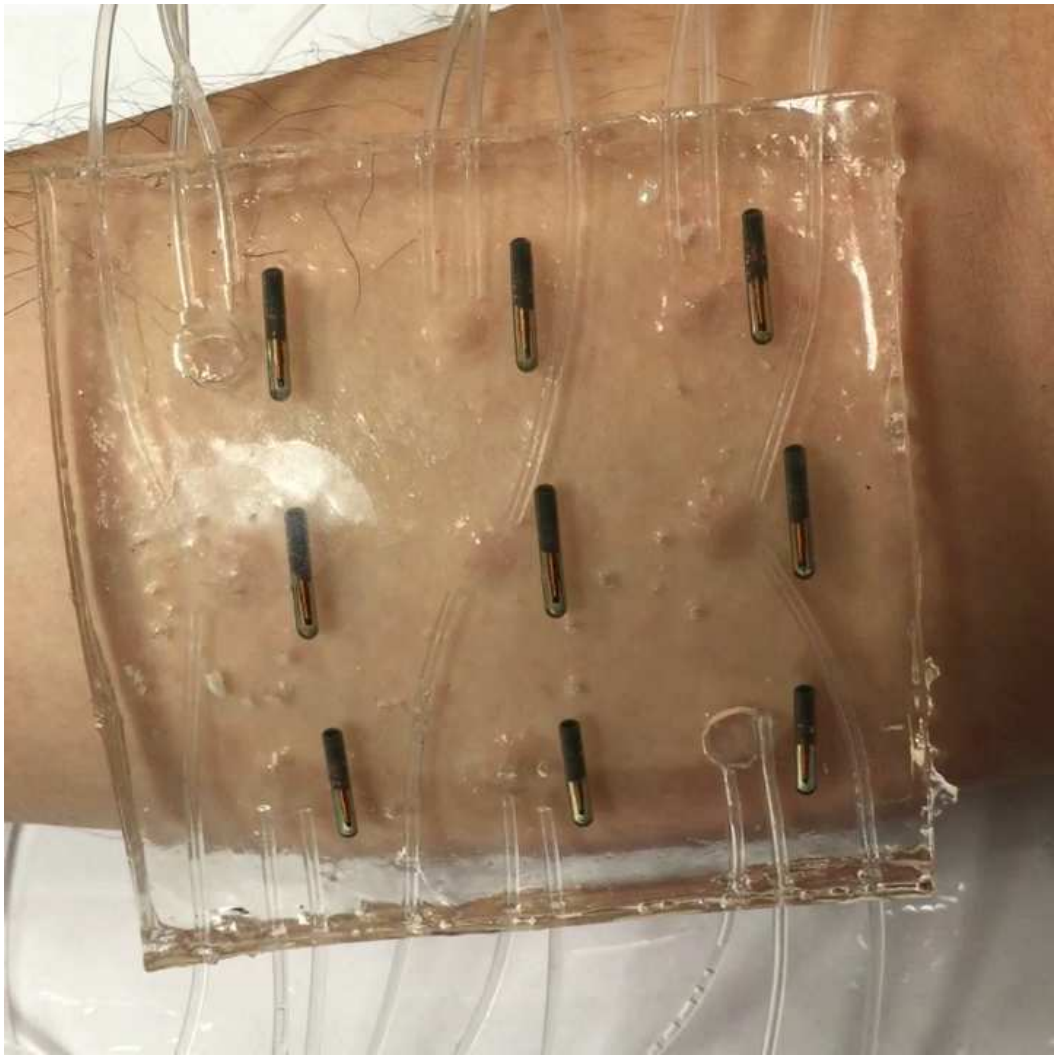




Yuk et al, Nature Communications, In press (2016)

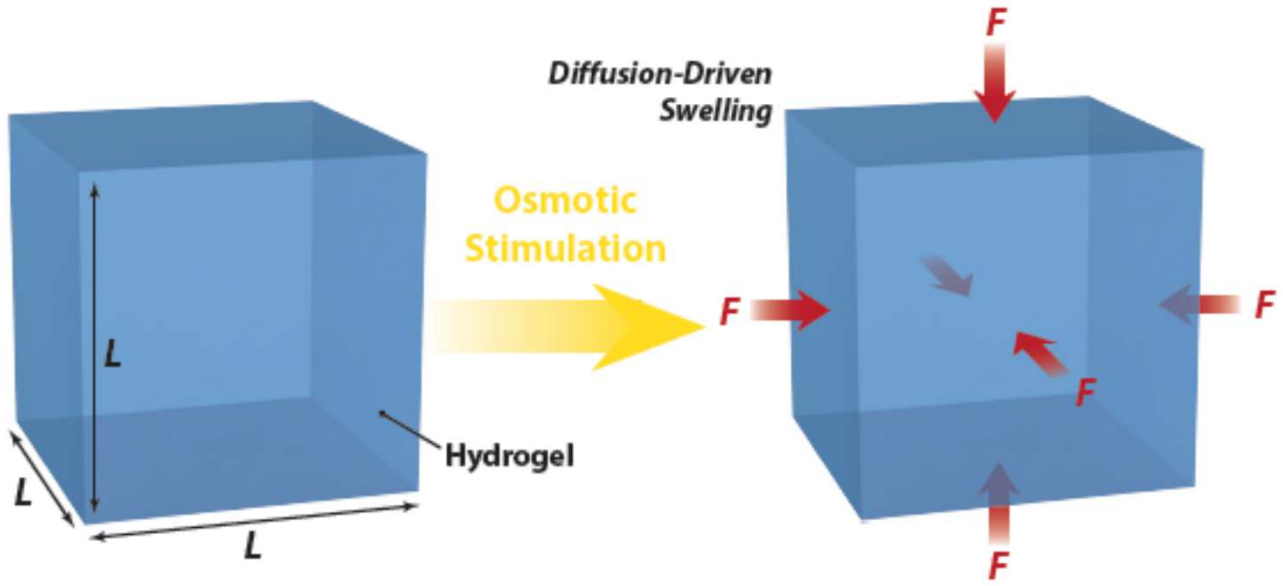
# Smart Hydrogel Band-Aid



Lin, et al, Advanced Materials, 28, 4497–4505(2016)

# Existing Hydrogel Actuators are Mostly Osmotic-Driven




$$F \propto \Delta\Pi L^2 \sim 10N$$

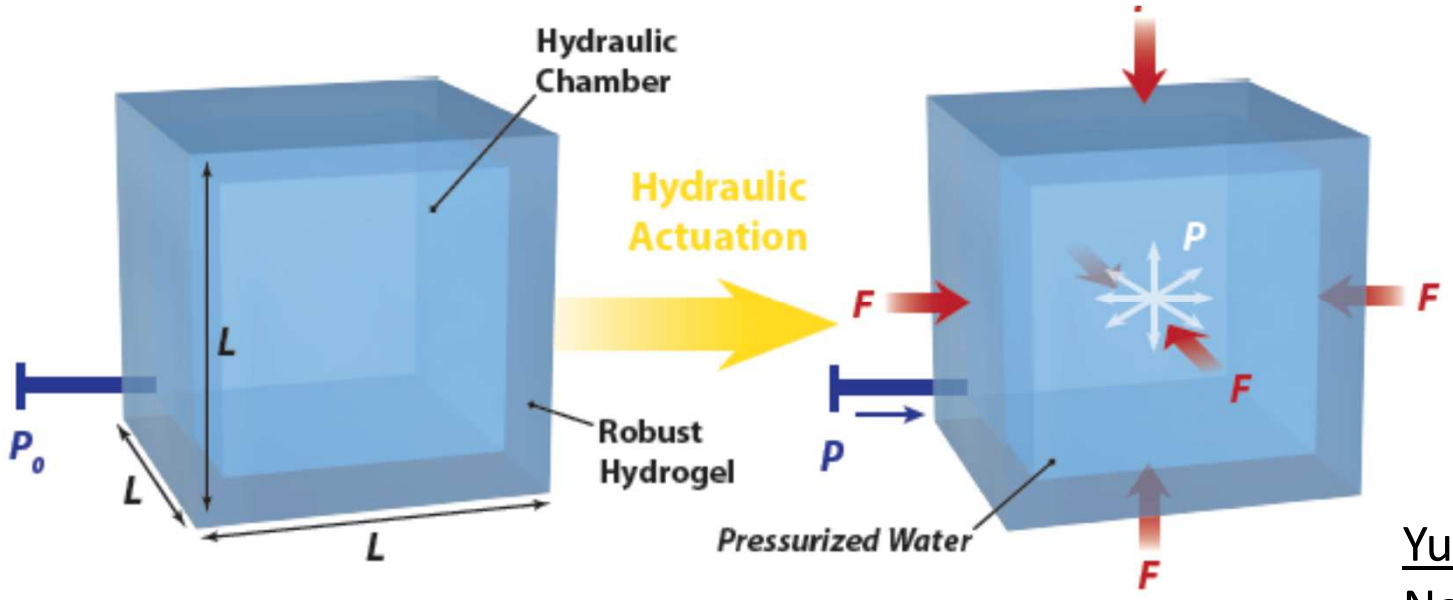
$$t \propto L^2 / D \sim 10^6 s$$

$$\Delta\Pi \sim 100kPa$$

$$L \sim cm$$

$$D \sim 10^{-10} m^2 / s$$

# Hydraulic Actuation of Tough Hydrogel Structures




$$F \propto \Delta P \cdot L^2 \sim 10N$$

$$t = t_{external} < 1s$$

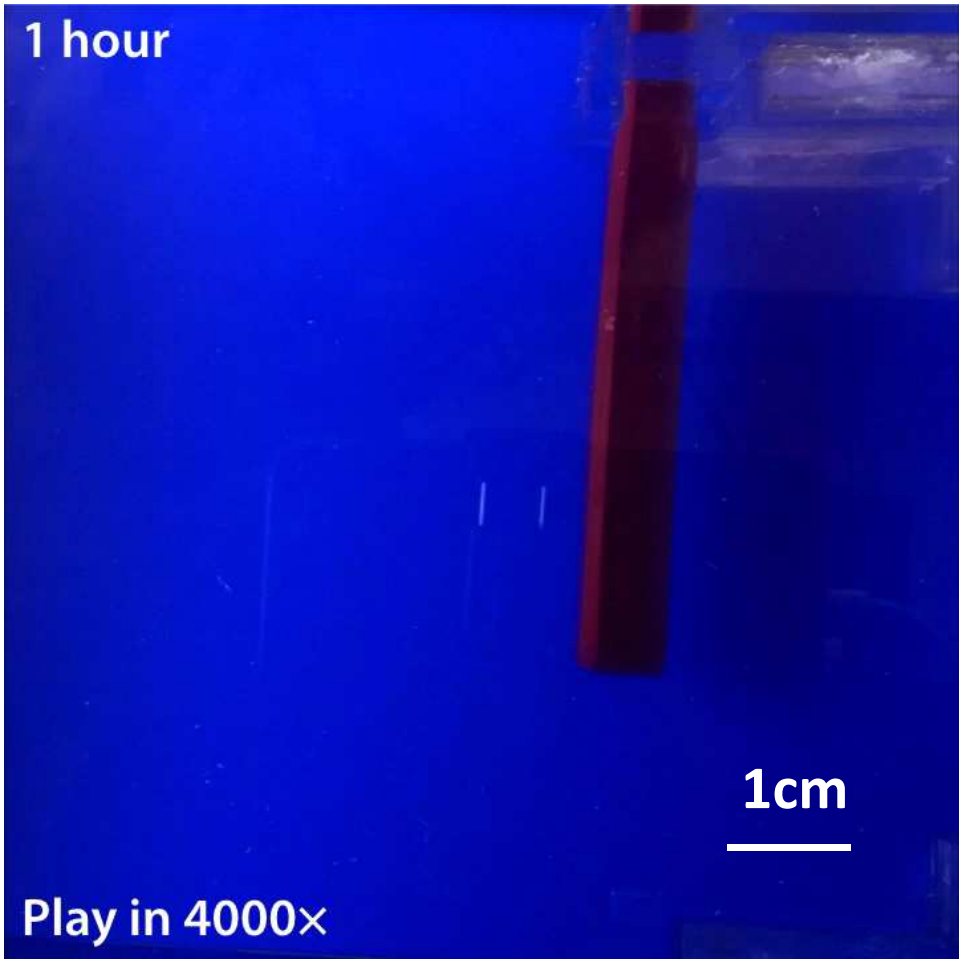
$$\Delta P \sim 100kPa$$

$$L \sim cm$$

$$t_{external} < 1s$$

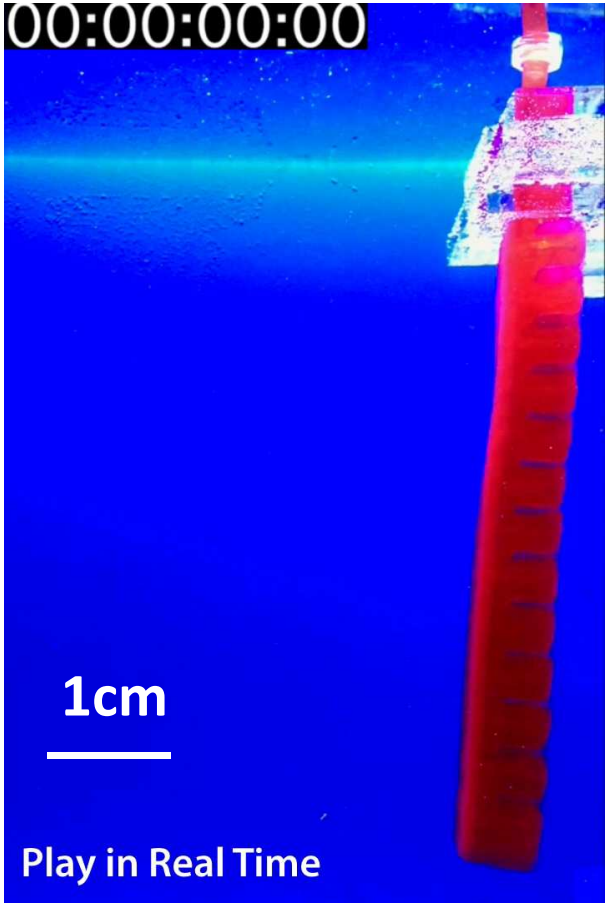
Yuk et al, Nature Communications, (under revision)

## Osmotic Actuation



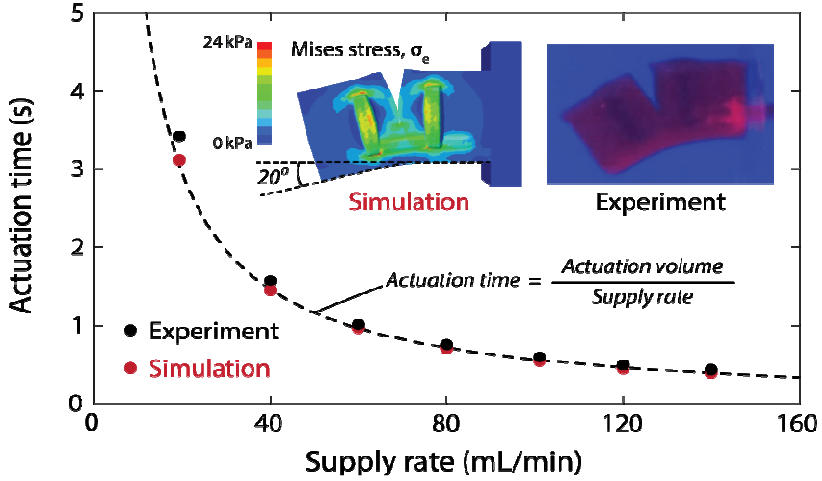

Speed: 4000 X

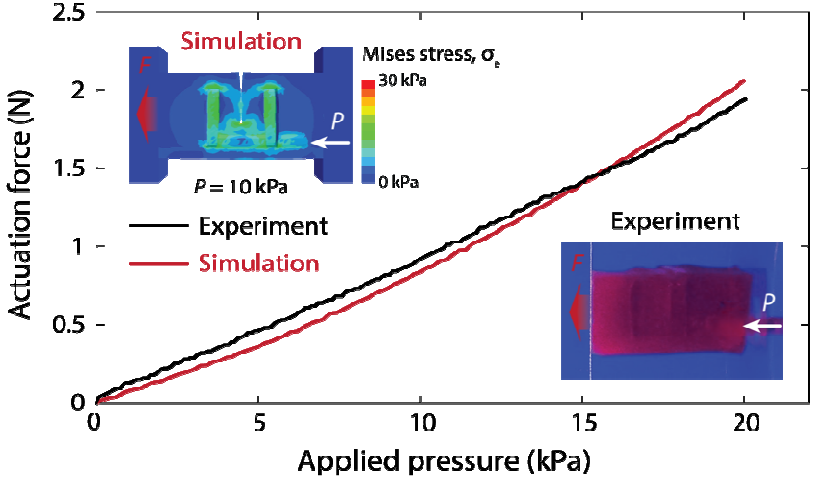
## Hydraulic Actuation

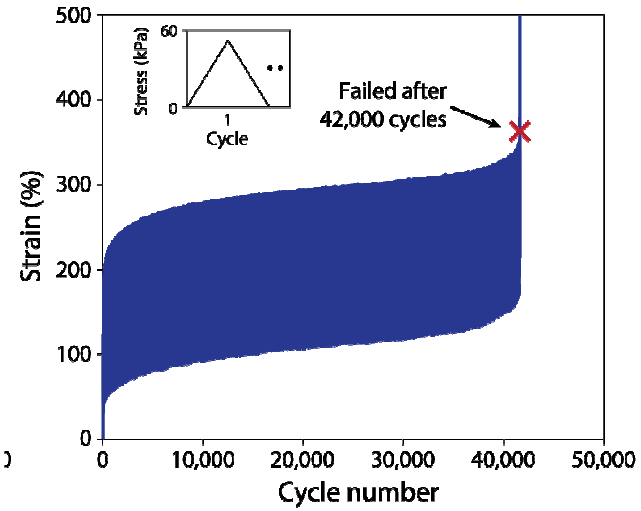


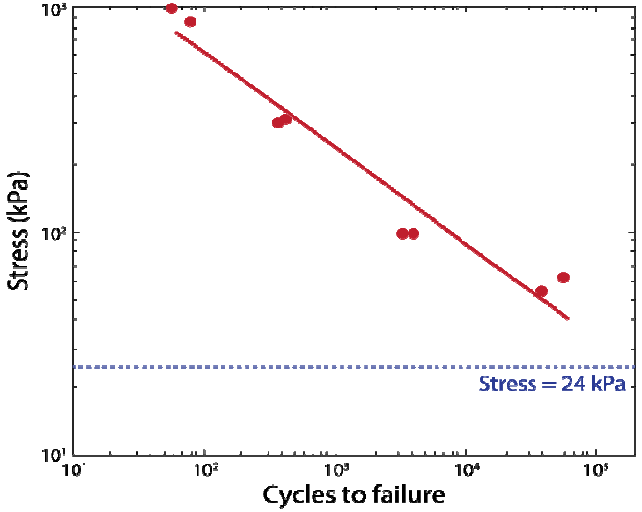

Speed: Real Time

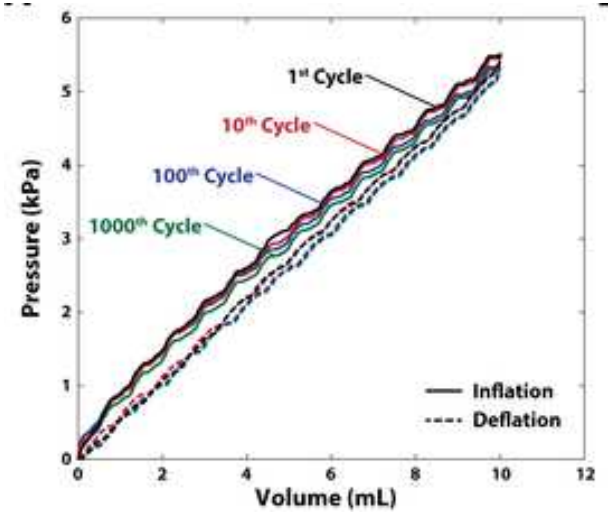
# High-Speed, High-Force, Anti-fatigue Actuations for Hydrogels

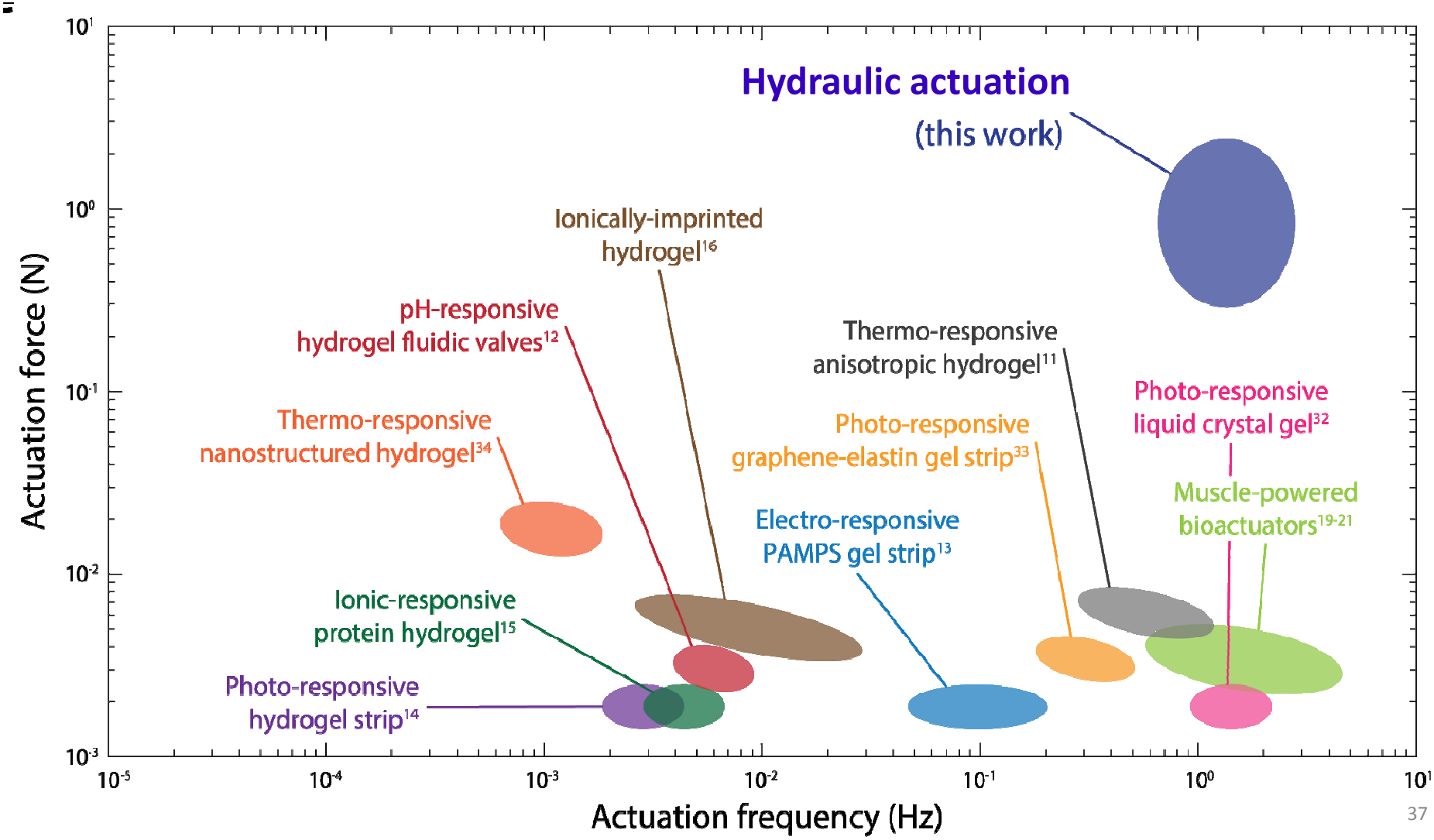


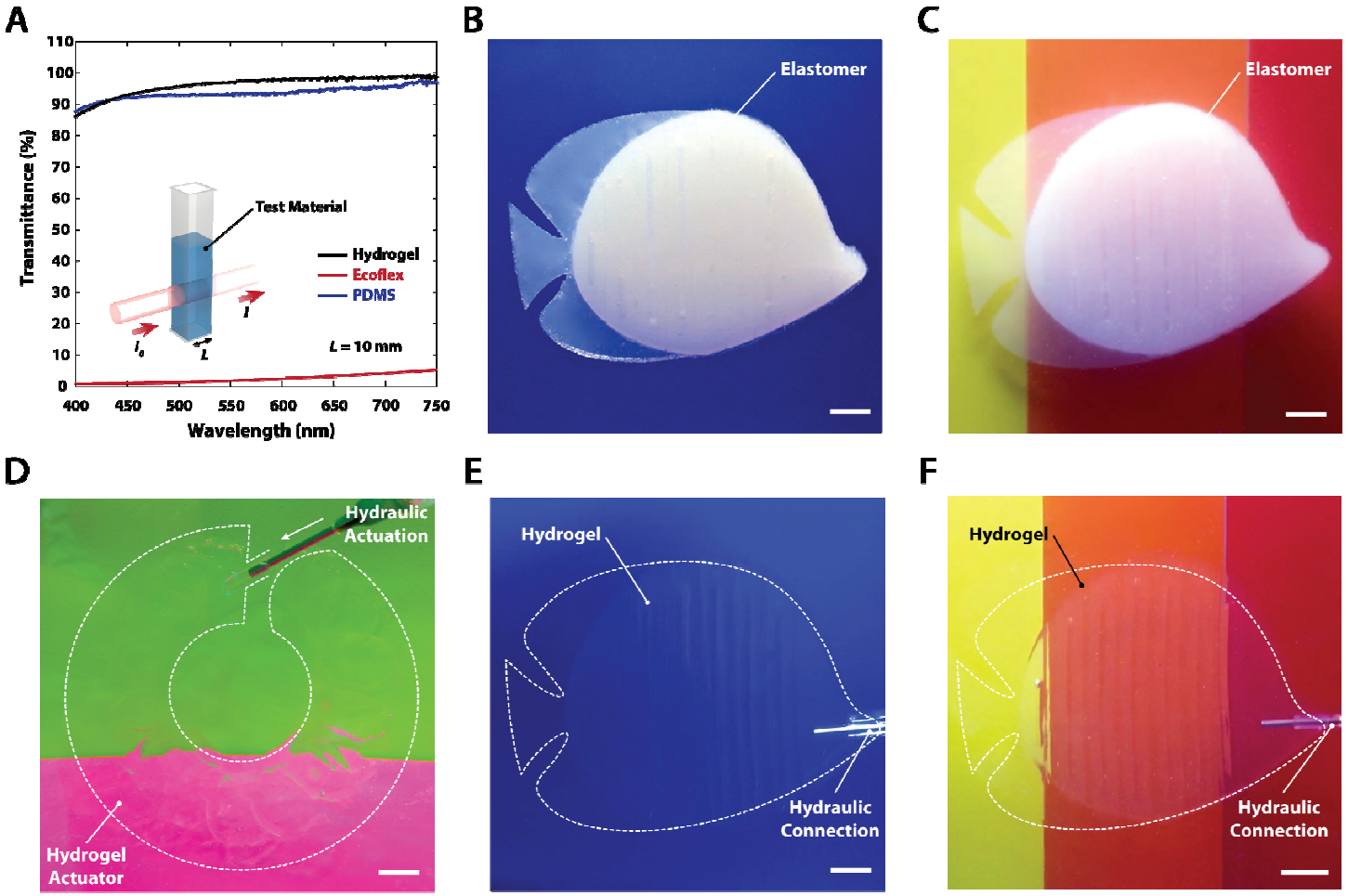




Hydraulic actuation
(this work)
Ionically-imprinted hydrogel[16]
pH-responsive hydrogel fluidic valves[12]
Thermo-responsive anisotropic hydrogel[11]
Photo-responsive liquid crystal gel[32]
Thermo-responsive nanostructured hydrogel[34]
Photo-responsive graphene-elastin gel strip[33]
Muscle-powered bioactuators[19-21]
Electro-responsive PAMPS gel strip[13]
Ionic-responsive protein hydrogel[15]
Photo-responsive hydrogel strip[14]
Actuation force (N)
Actuation frequency (Hz)
10^1
10^0
10^-1
10^-2
10^-3
10^-5
10^-4
10^-3
10^-2
10^-1
10^0
10^1

# Optically and Acoustically Invisible in Water

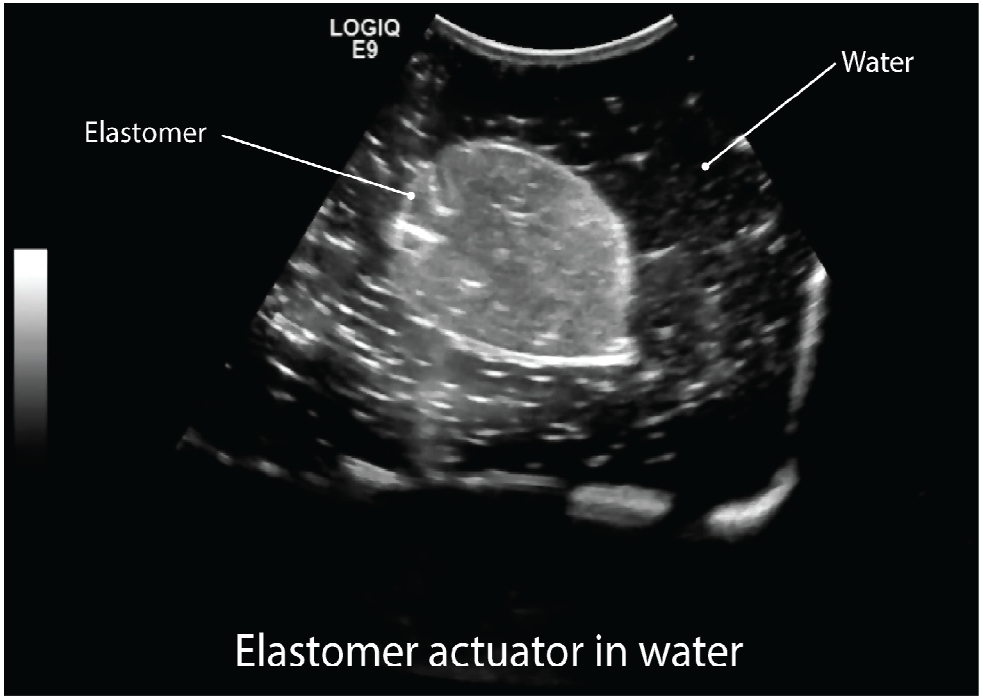
| | Water | Hydrogel | Ecoflex | Elastosil | Sylgard 184 |
|---|---|---|---|---|---|
| ***n*** **Refractive Index** | **1.3330** | **1.3365** | N/A† | N/A† | 1.4225 |
| $I/I_0$ **Transmittance (relative to water)** | **100 %** | **> 90 %** | < 5 % | < 0.1 % | > 90 % |
| $c^{\ddagger}$ **Speed of Sound [m·sec$^{-1}$]** | **1447.5** | **1485.7** | 983.4 | 979.6 | 1022.4 |
| $z_0$ **Acoustic Impedance [Pa·s·m$^{-1}$]** | **$1.448 \times 10^6$** | **$1.487 \times 10^6$** | $1.052 \times 10^6$ | $1.058 \times 10^6$ | $1.053 \times 10^6$ |
| ***R*** **Acoustical Reflection Coefficient** | **0** | **0.013** | 0.158 | 0.156 | 0.158 |

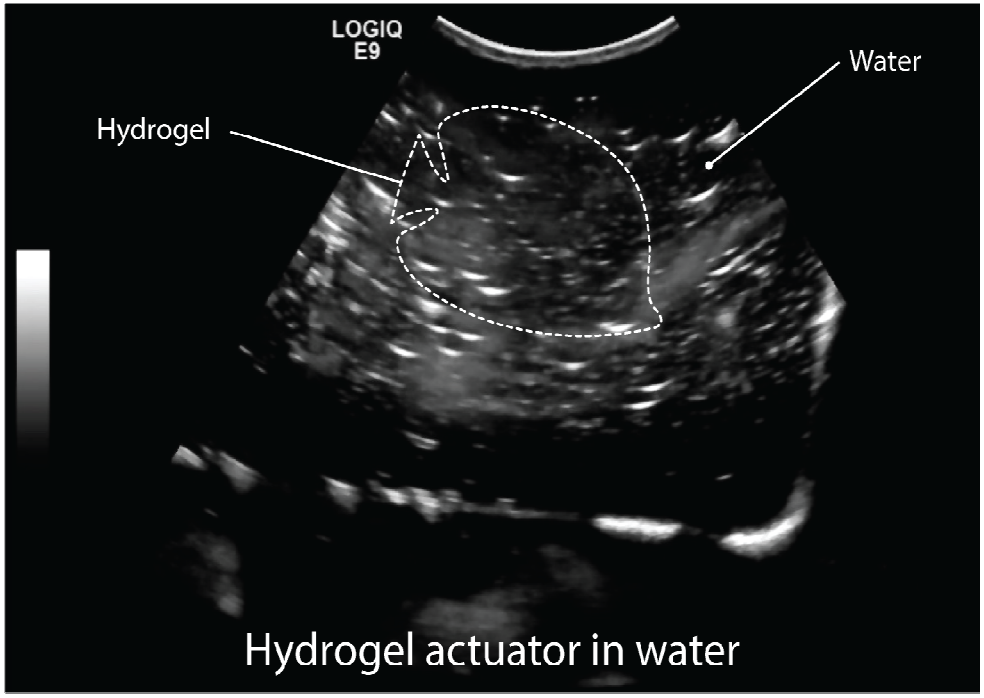
# Optically and Acoustically Invisible in Water



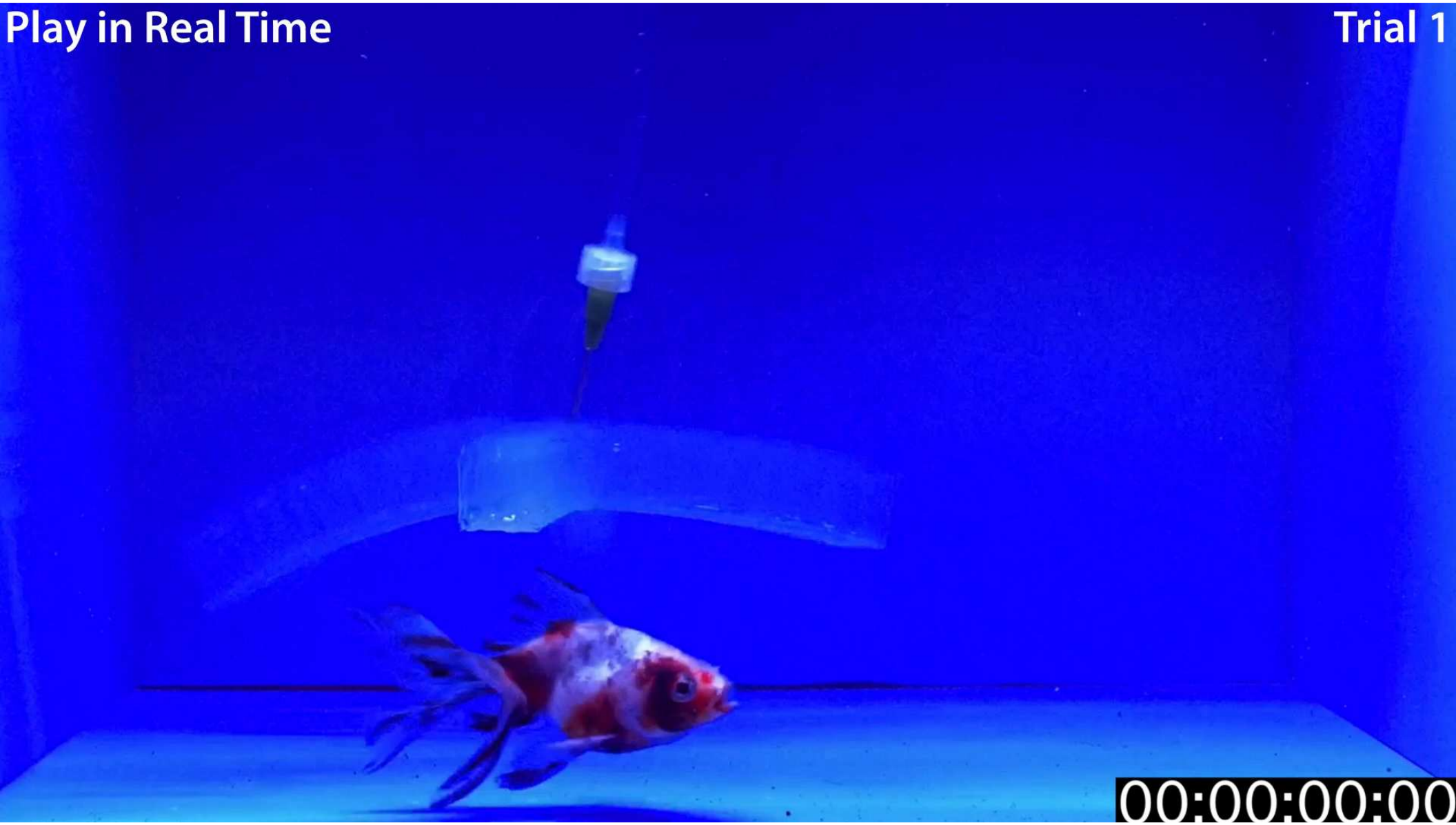

Yuk et al, Nature Communications, (under revision)

# Optically and Acoustically Invisible in Water




Yuk et al, Nature Communications, (under revision)


Play in Real Time
Trial 1
00:00:00:00

# Hydrogels

- **Tough Hydrogels: Build dissipation into stretchy network.**
- **Tough Bonding: Anchor stretchy network on surface.**
- **Manufacturing: 3D printing.**
- **Applications: Scaffolds, Hydrogel electronics, Hydrogel actuators**

- **More information: www.zhaox.org**

# Acknowledgement

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