

**Design strategy of metal alloys  
for robust bonding between hydrogels**

**Seoul National University  
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# Hybrids of soft and rigid components



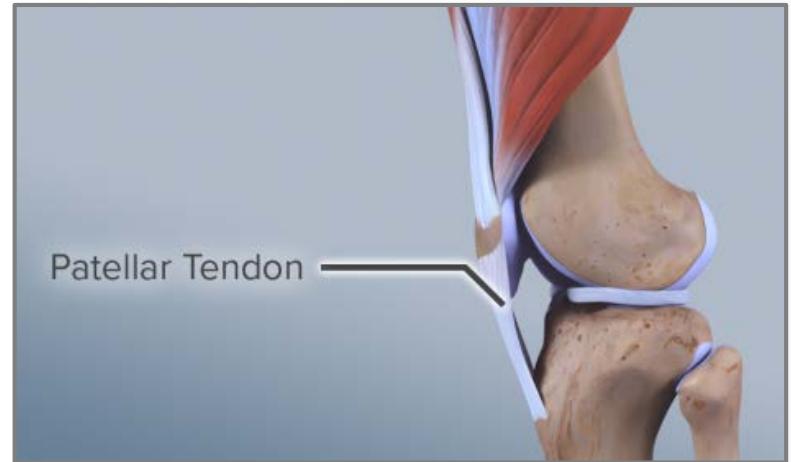
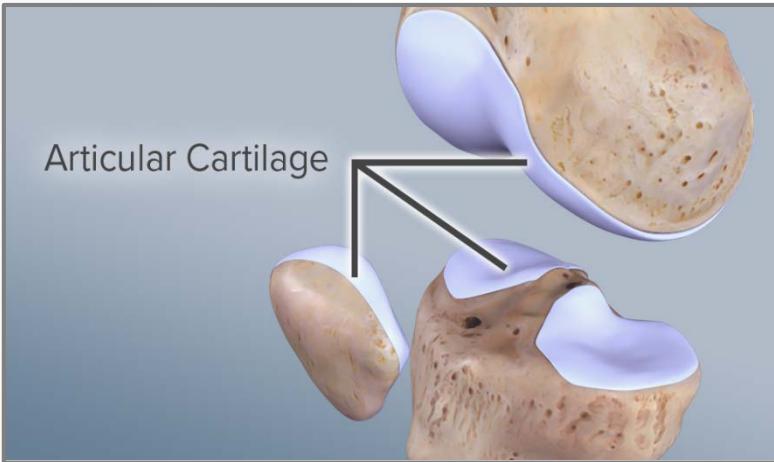
So far, **rigid-bodied robots** and **soft-bodied robots**  
have been actively researched

# Hybrids of soft and rigid components



However, little research has been done on **hybrid robots**

# Hybrids of soft and rigid components



In nature, hybrids of soft and rigid components generate synergy

# Tough interfaces between metals and hydrogels



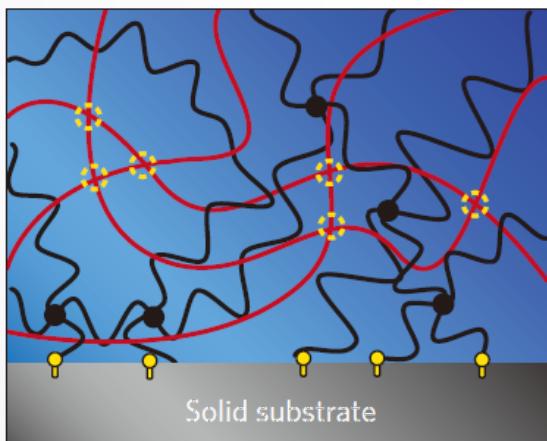
Metal



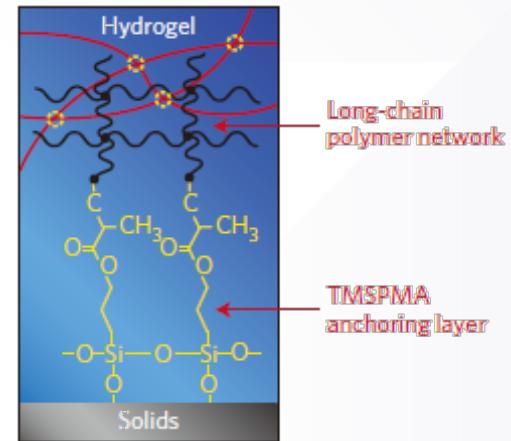
Hydrogel

For hybrid combinations of rigid and soft materials,  
achieving **tough interfaces** is most important

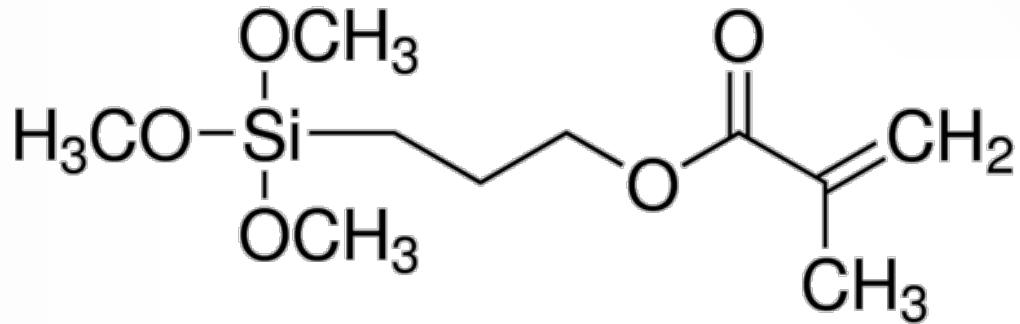
# Tough interfaces between metals and hydrogels



- Wavy line: Long-chain polymer network
- Straight line: Dissipative polymer network
- Black star-like symbol: Covalent crosslinks
- Red star-like symbol: Reversible crosslinks
- Yellow dot: Chemically anchored long-chain polymer network on solids



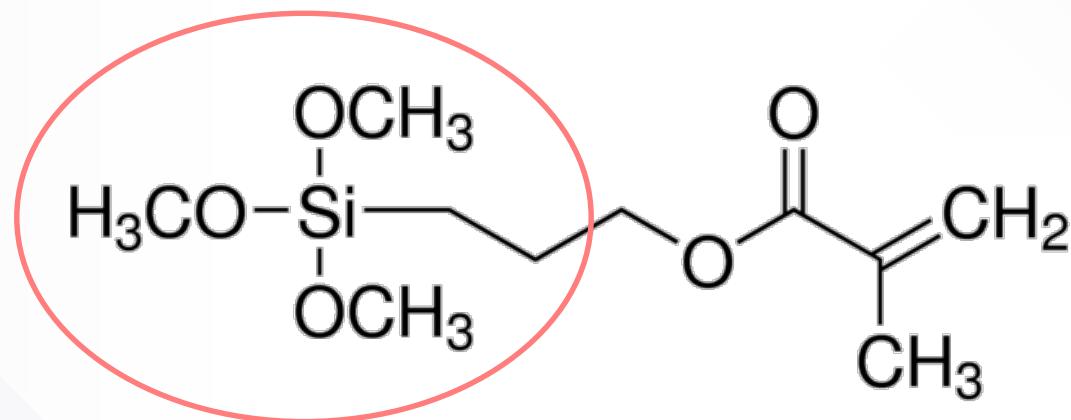
# Tough interfaces between metals and hydrogels



3-(trimethoxysilyl) propyl methacrylate (**TMSPMA**)

# Tough interfaces between metals and hydrogels

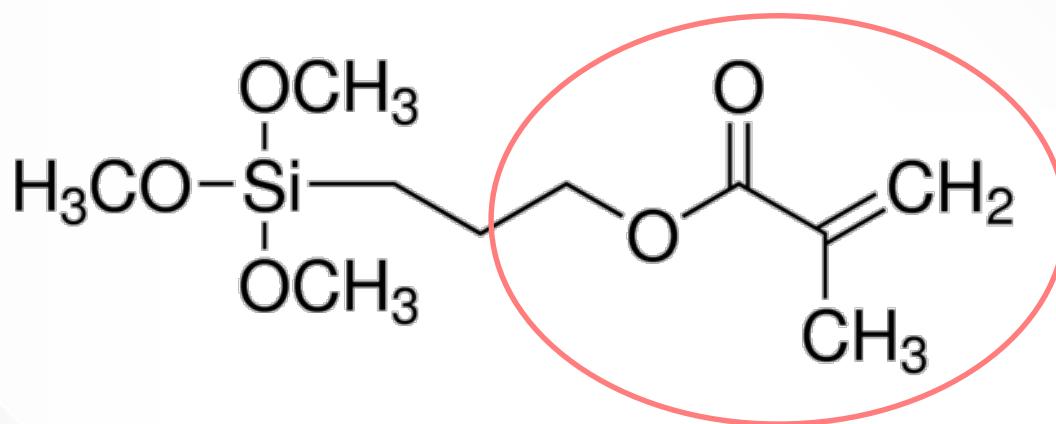
Covaletly anchored  
to solid substrate



3-(trimethoxysilyl) propyl methacrylate (**TMSPMA**)

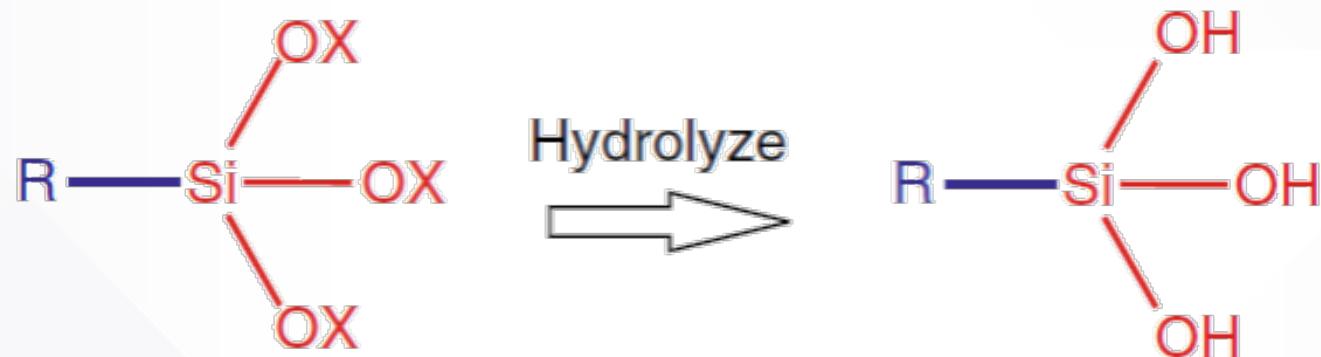
# Tough interfaces between metals and hydrogels

Covalently cross-linked  
to long chain polymer network

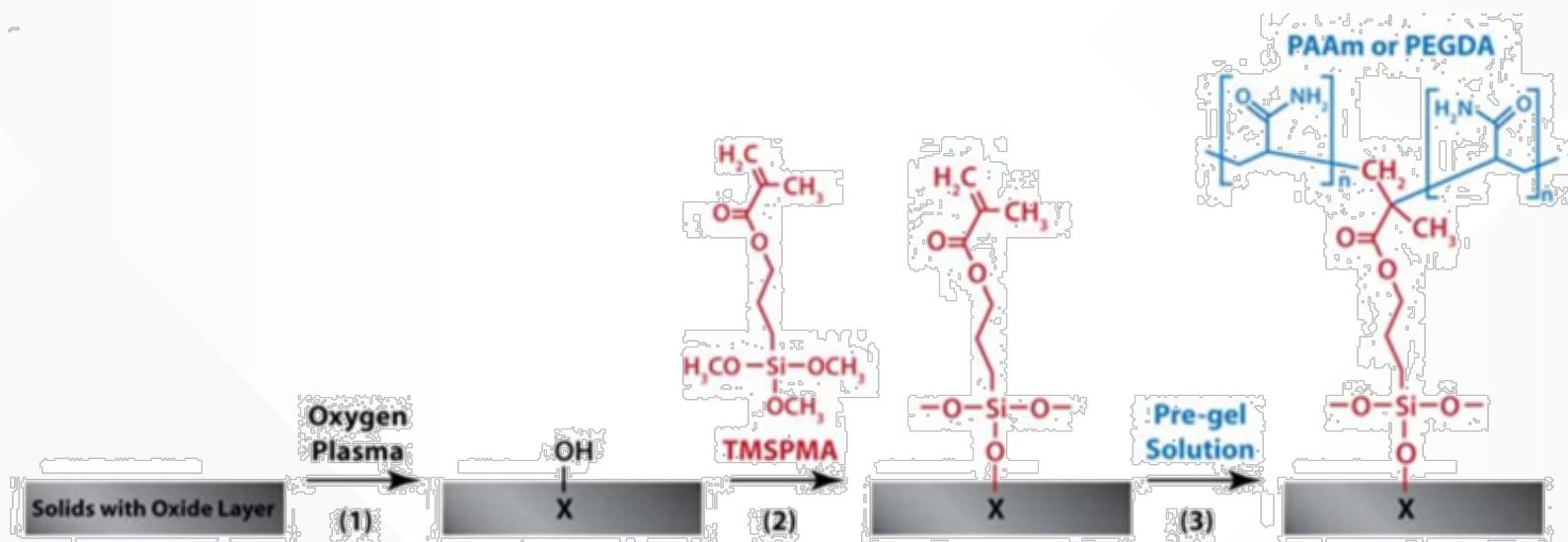


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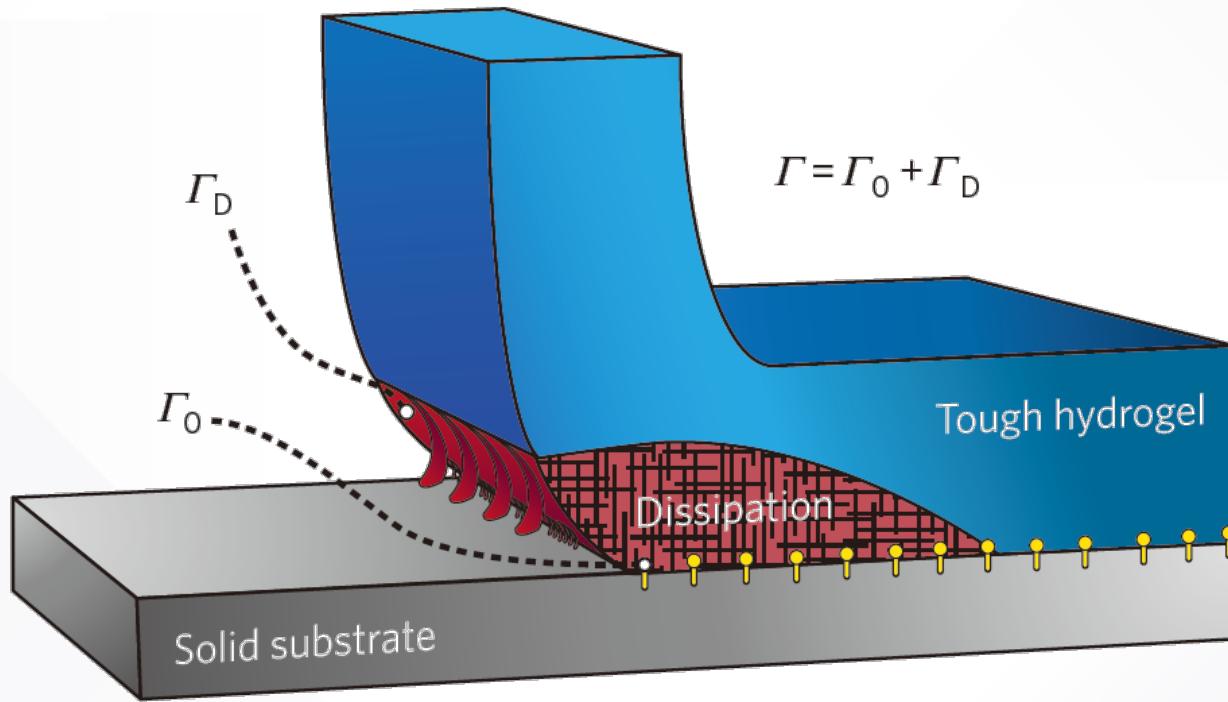
# Tough interfaces between metals and hydrogels



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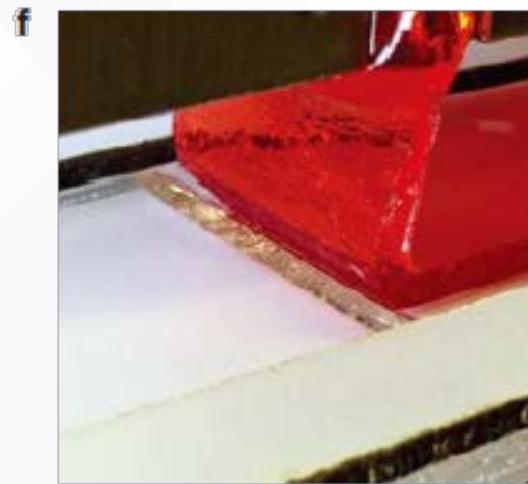
# Tough interfaces between metals and hydrogels



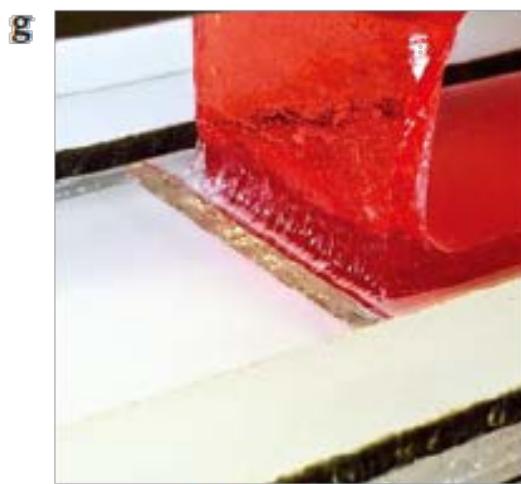
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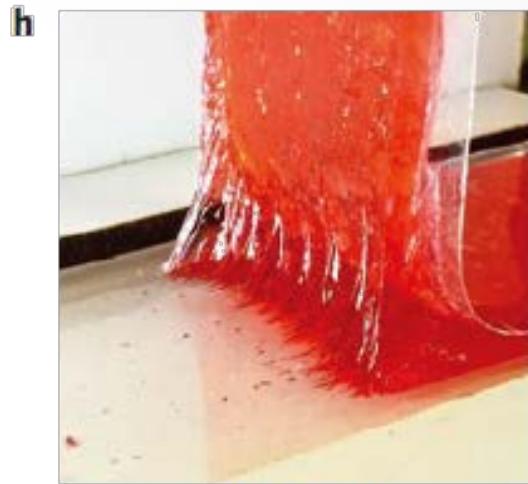
# Tough interfaces between metals and hydrogels



No force

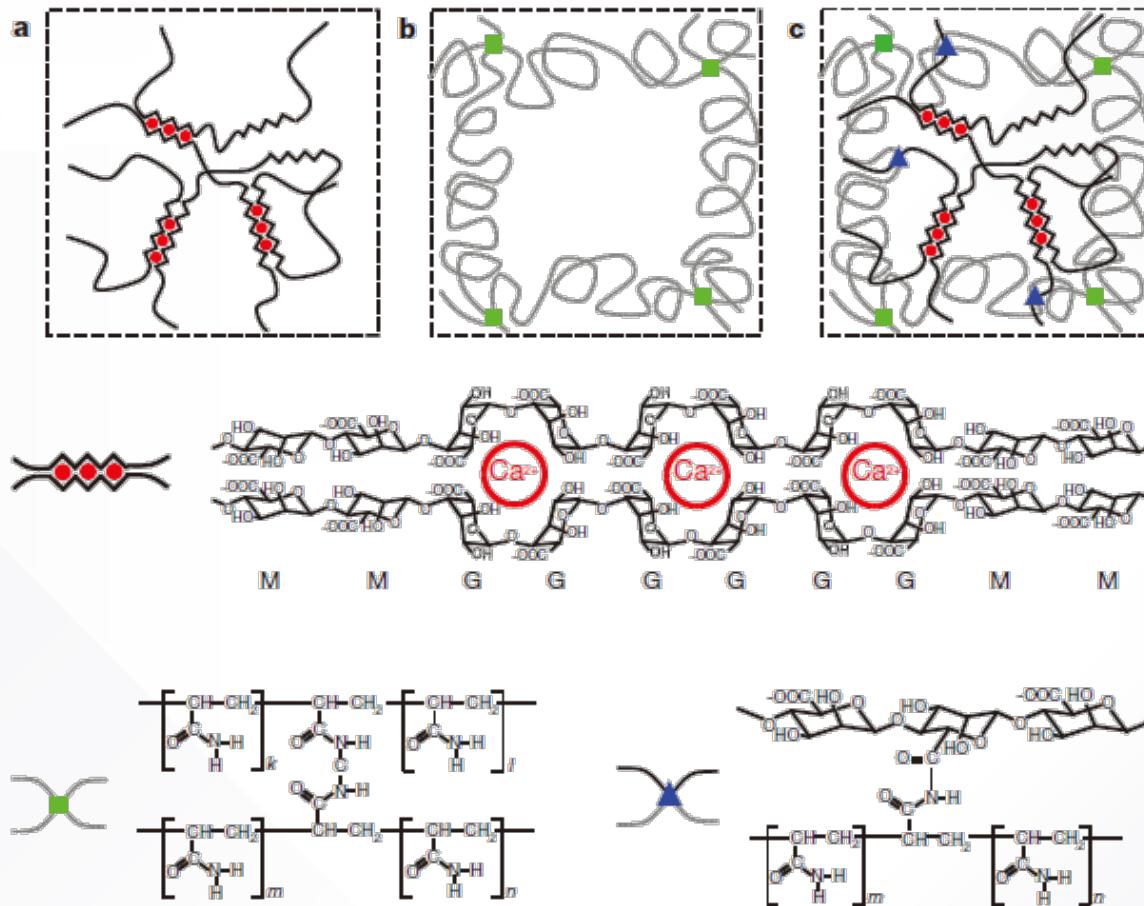


Crack initiation

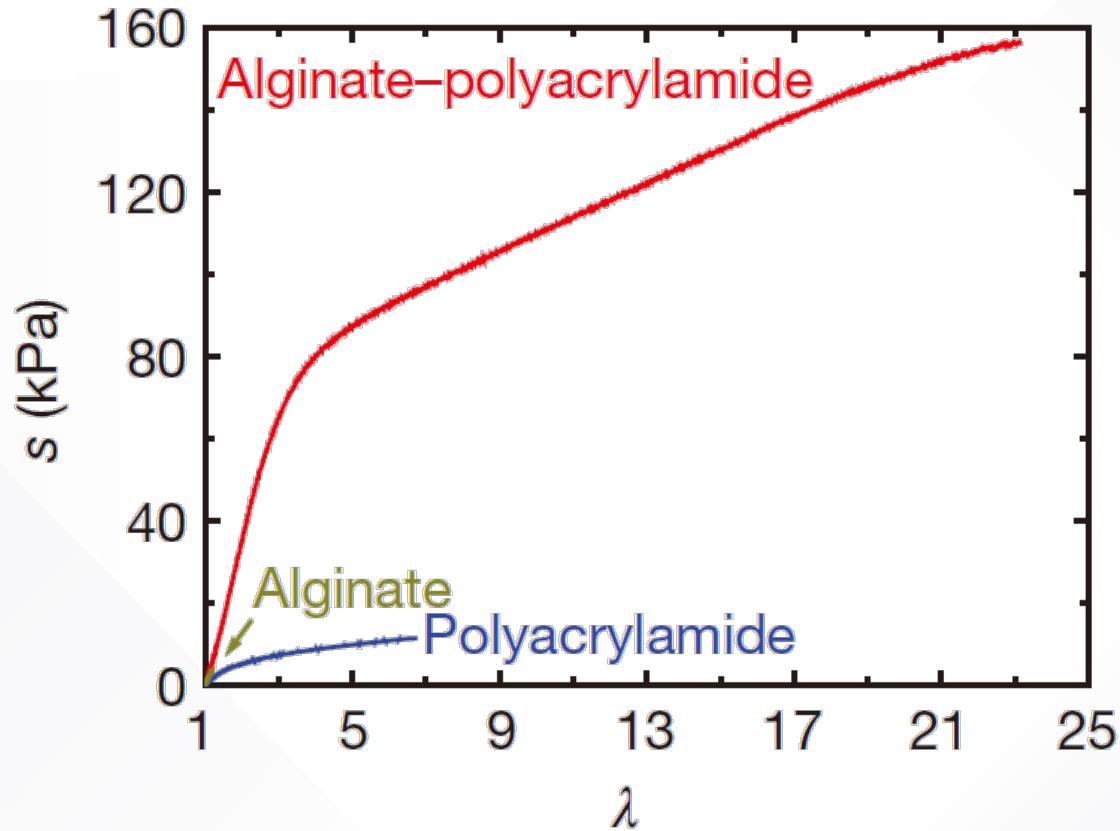


Steady state

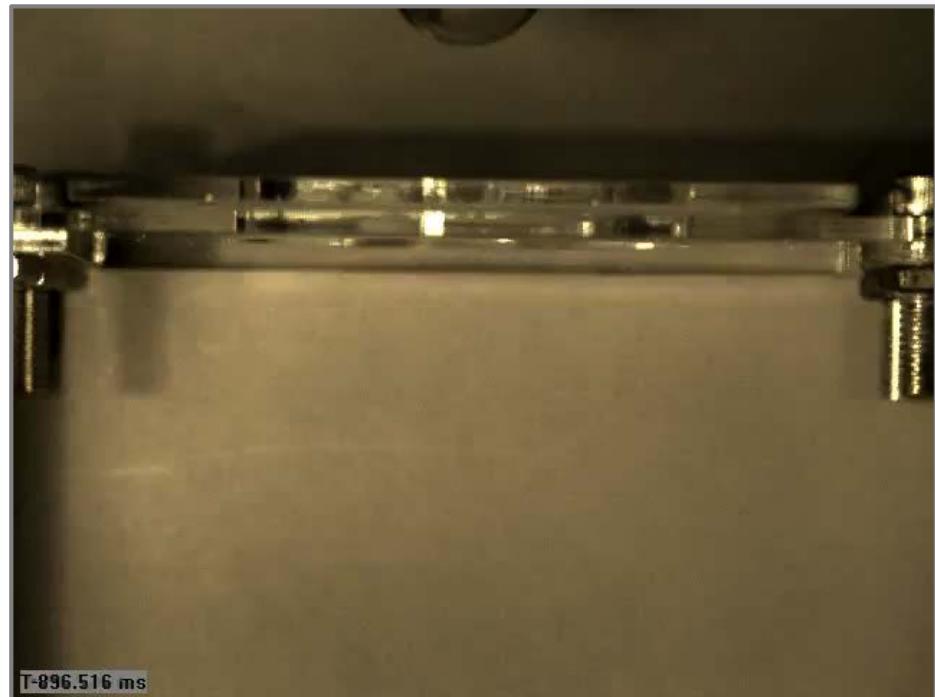
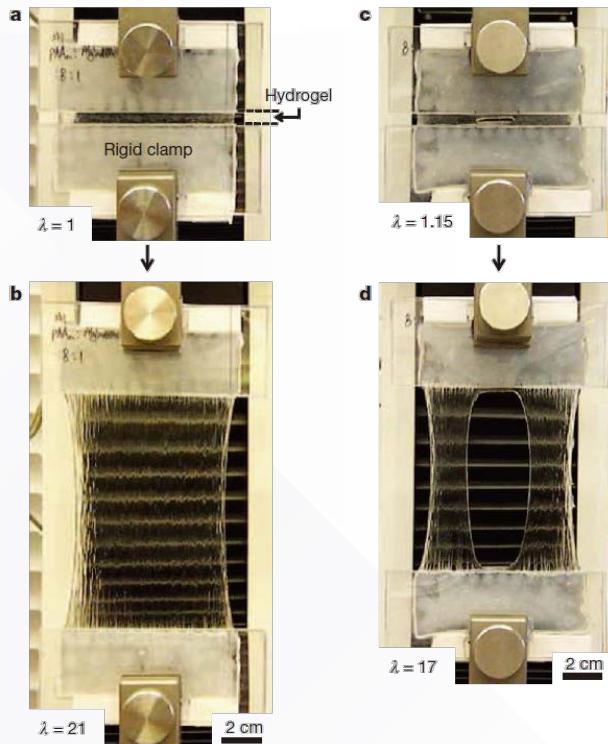
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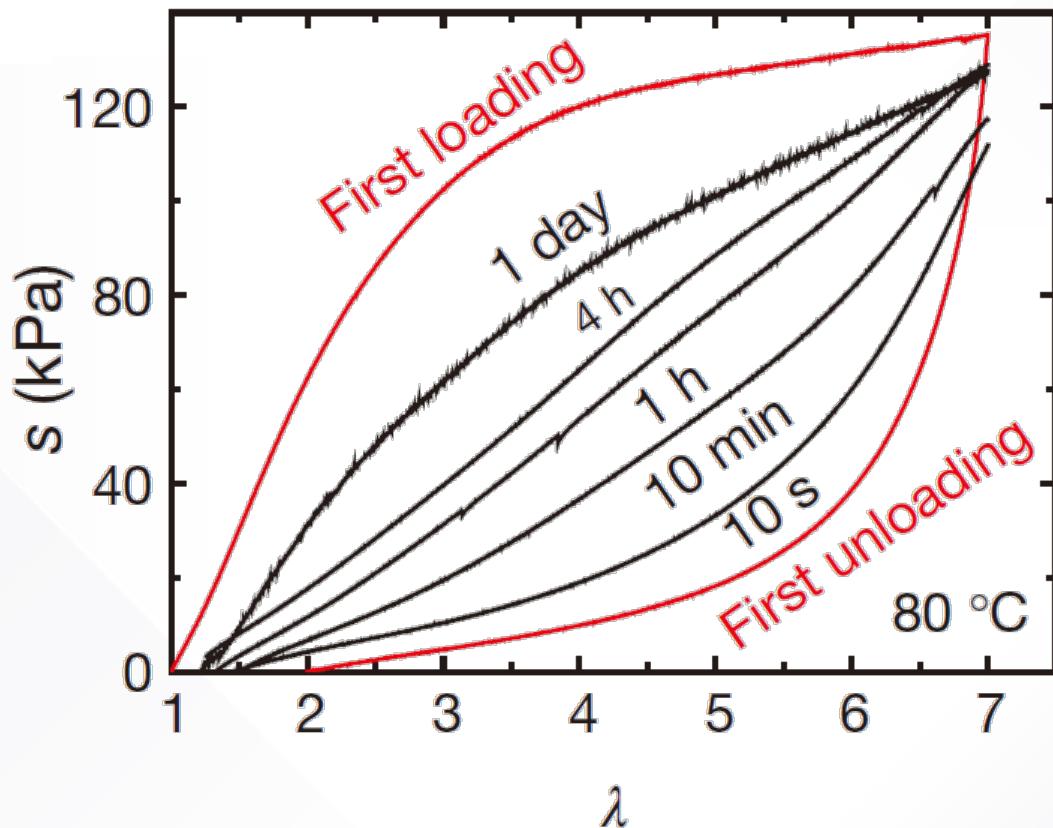
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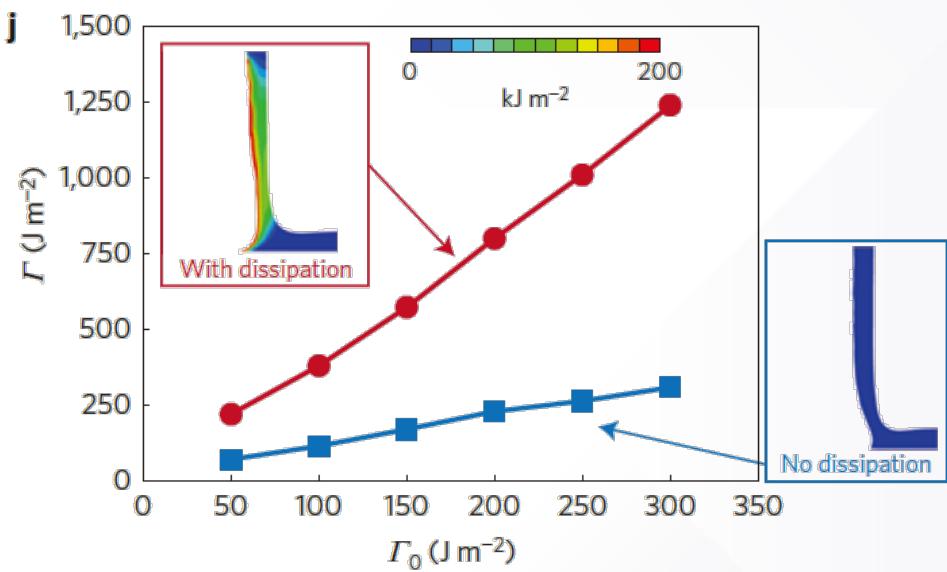
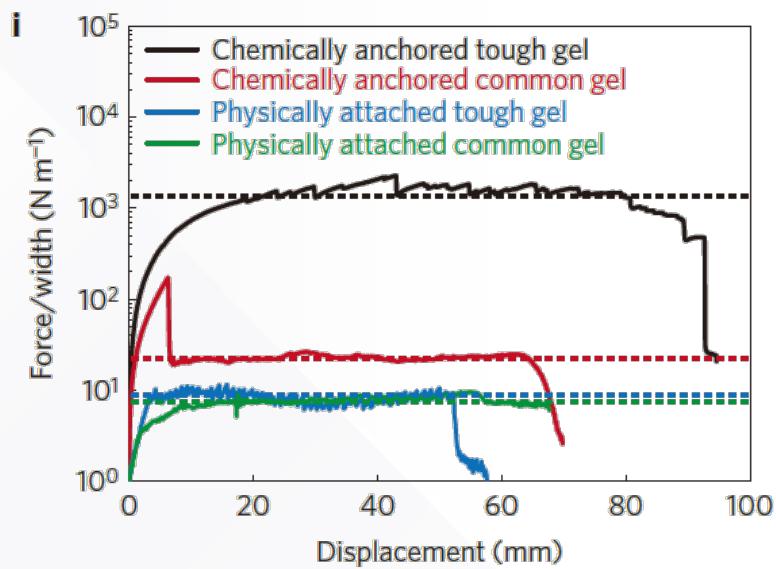
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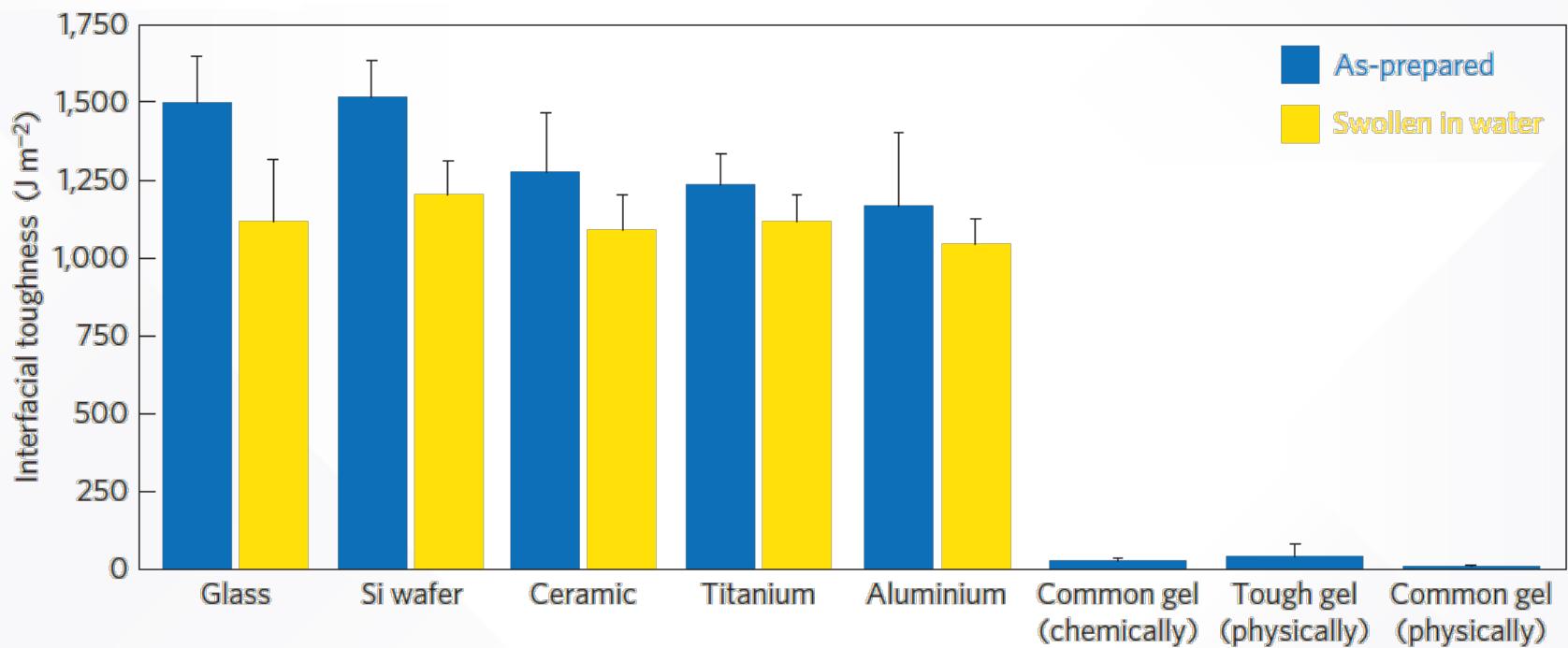
# Tough interfaces between metals and hydrogels



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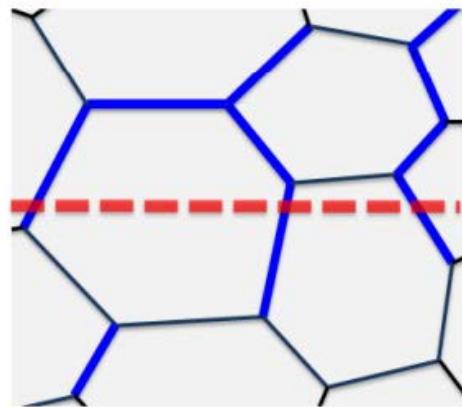


## Segregation engineering

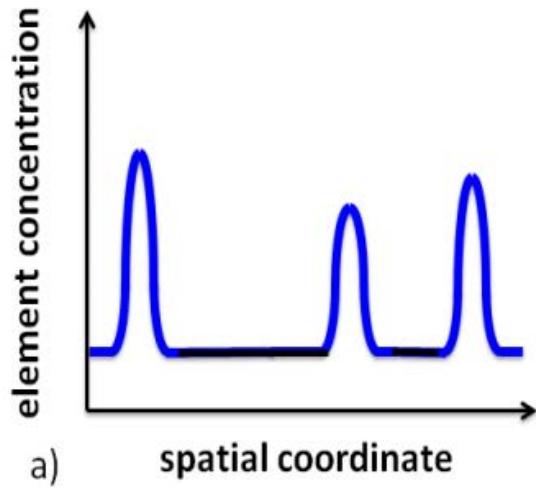
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**Segregation engineering**

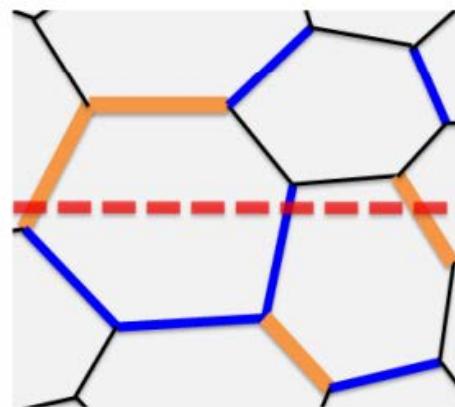
# Segregation engineering



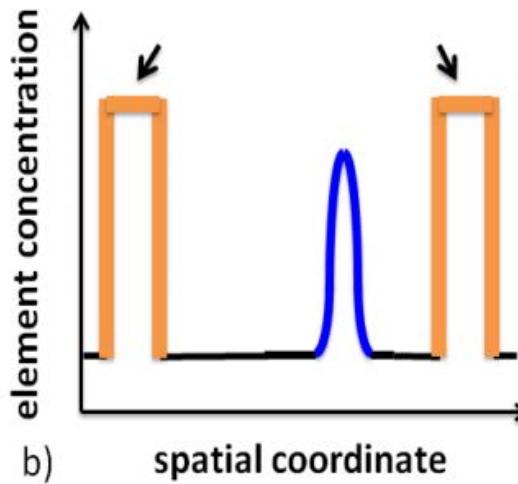
*Grain boundary segregation*



Heat  
treat-  
ment



*Confined ordering*



# Alloy design

$(Fe_{68.76}Cr_{10.73}Co_{7.67}Ni_{6.71}Al_{3.76}Mo_{1.62}W_{0.75})_{98}X_2$  (at. %)

## Low Ni Maraging Steel

$X = \text{Cu, Ti, Mn}$  ————— **High tendency to go to free surface**

$\Delta H_{segregation}$ (J/mol)		Solvent			
		Fe	Cr	Co	Ni
Solute	Cu	18932	16041	15989	12325
	Ti	22337	16737	17815	11767
	Mn	16605	15095	13781	10005

# Alloy design

$\Delta H_{segregation}$ (J/mol)		Solvent			
		Fe	Cr	Co	Ni
Solute	Cu	18932	16041	15989	12325
	Ti	22337	16737	17815	11767
	Mn	16605	15095	13781	10005
	Fe	0	-1676	-527	-4765
	Cr	3737	0	2720	-728

## Question

- 1. Is the solute concentration sufficient for observing segregation effect?**
- 2. GB segregation → Free surface segregation?**

# Alloy design

Segregation engineering enables nanoscale martensite to austenite phase transformation at grain boundaries:  
A pathway to ductile martensite

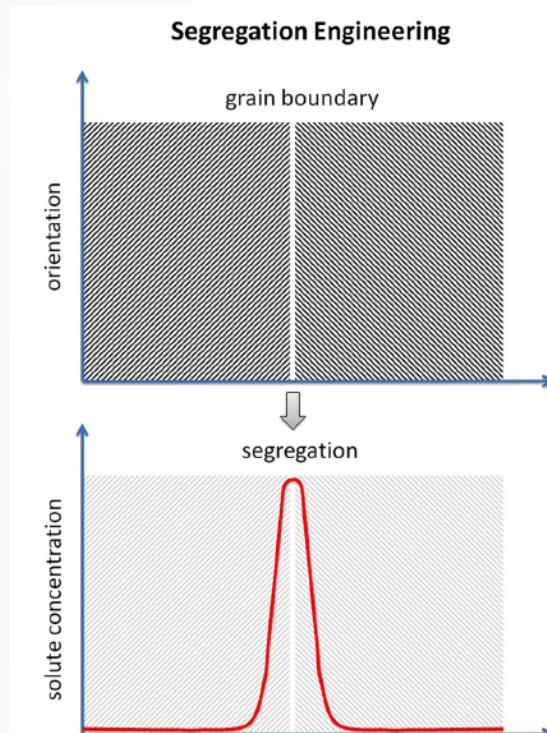
D. Raabe\*, S. Sandlöbes, J. Millán, D. Ponge, H. Assadi, M. Herbig, P.-P. Choi

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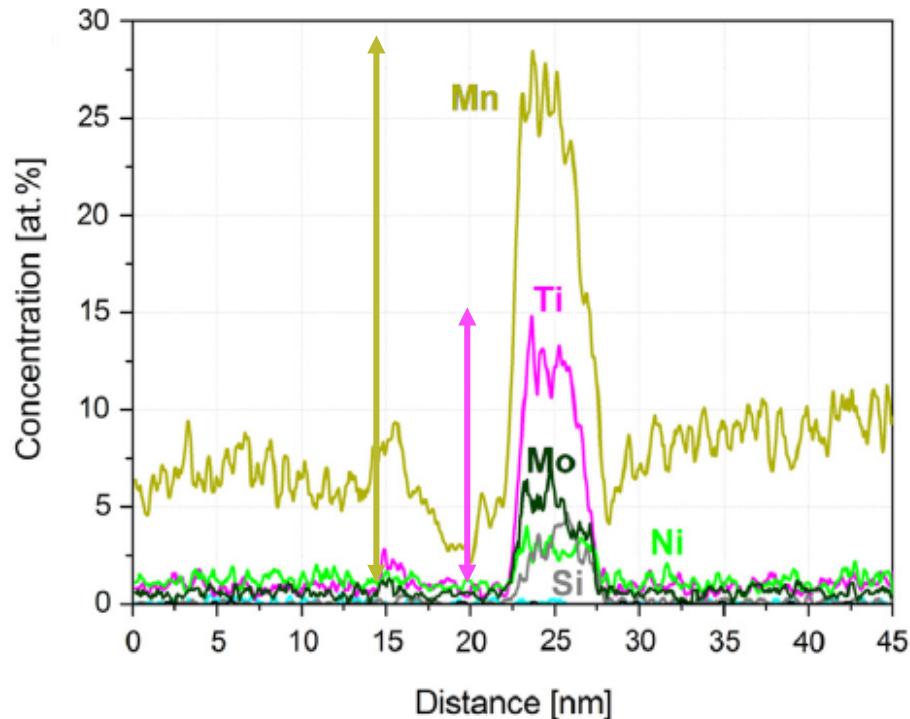
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$(Fe_{68.76}Cr_{10.73}Co_{7.67}Ni_{6.71}Al_{3.76}Mo_{1.62}W_{0.75})_{98}X_2$  (at. %)



$Fe - Mn_{12.2} Ni_{1.9} Mo_{0.6} Ti_{1.2} Al_{0.3}$ ,  
450 C, 48 h

## Question

1. Is the solute concentration sufficient for observing segregation effect?
2. GB segregation → Free surface segregation?

# Alloy design

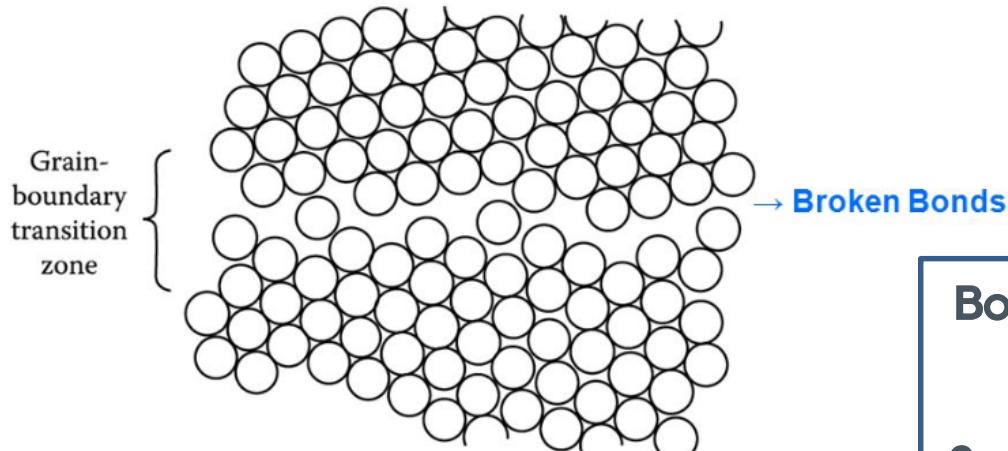
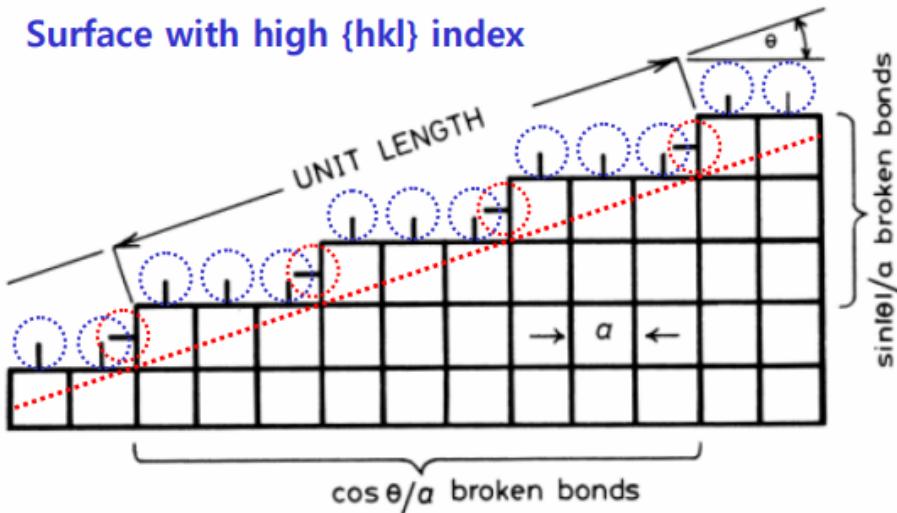


Fig. 3.10 Disordered grain boundary structure (schematic).

Surface with high {hkl} index



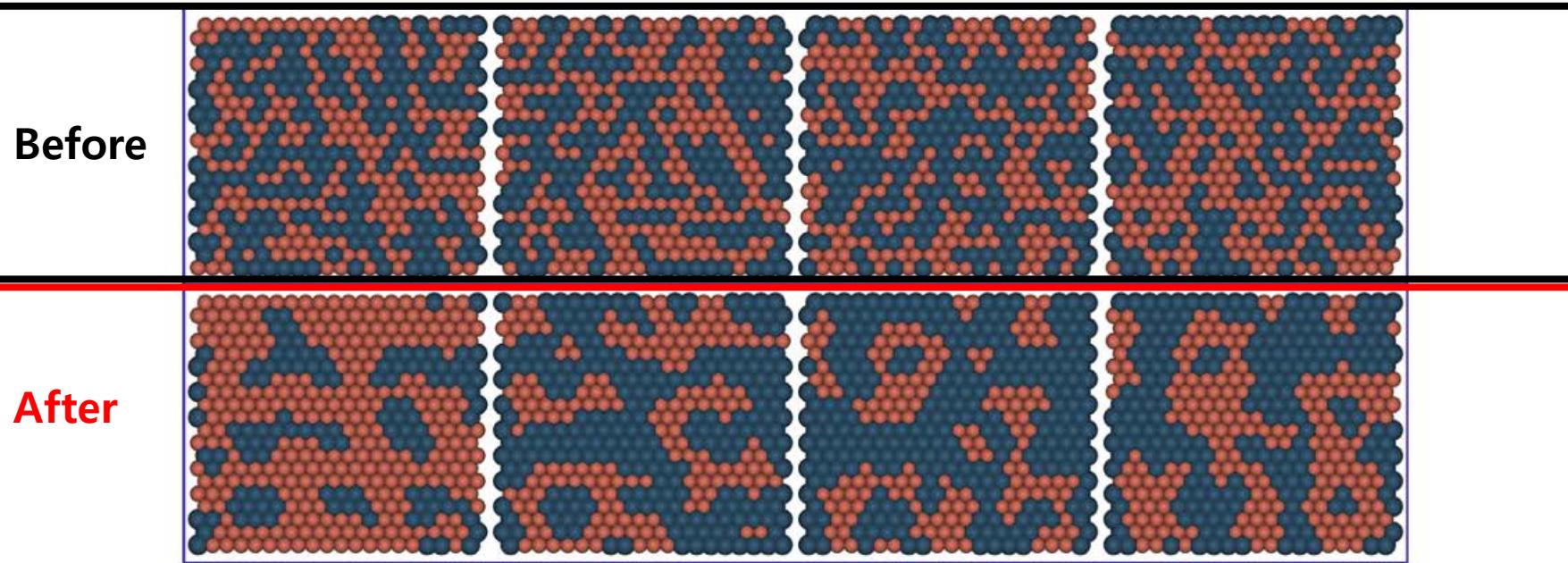
Both boundary energy comes from broken bonds

Segregation phenomenon occurs to lower the boundary energy

High GB segregation tendency  
→ High free surface segregation tendency (in general)

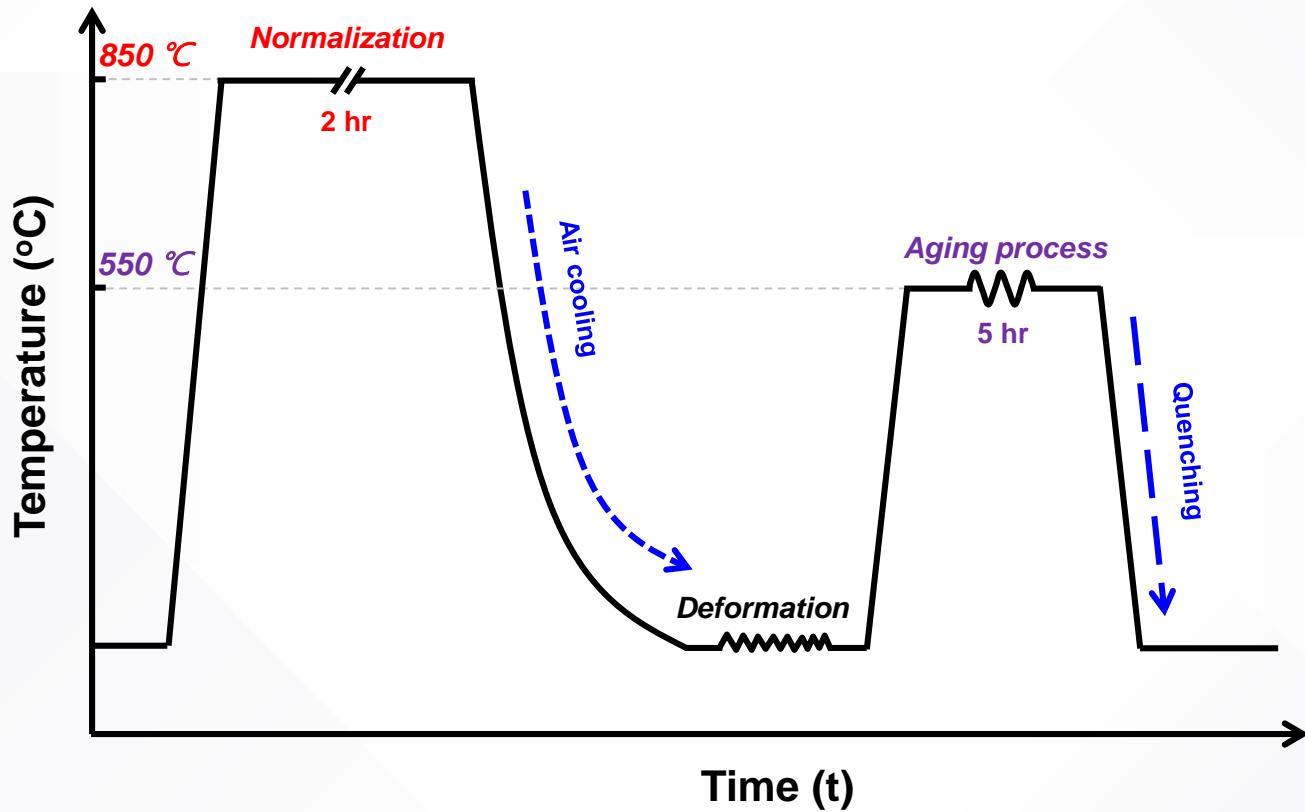
# Alloy design

$\Delta H_{segregation}$ (J/mol)		Solvent	
		Cu	Pd
Solute	Cu	0	10747
	Pd	-12850	0



**Fig. 4.** Atomic arrangements of the top four layers (from left to right: 1st to 4th layer) of 52% Pd–Cu alloy. Upper panels are initial configurations and lower ones are annealed after 20 min at 500 K. Red spheres: Cu; dark blue: Pd. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

# Processing design



# Tough interfaces between metals and hydrogels

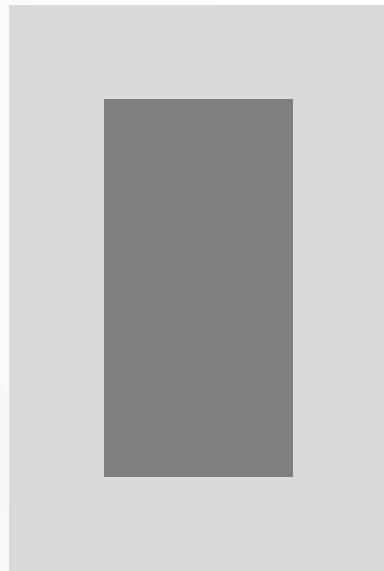


Top view



Cross section

# Tough interfaces between metals and hydrogels

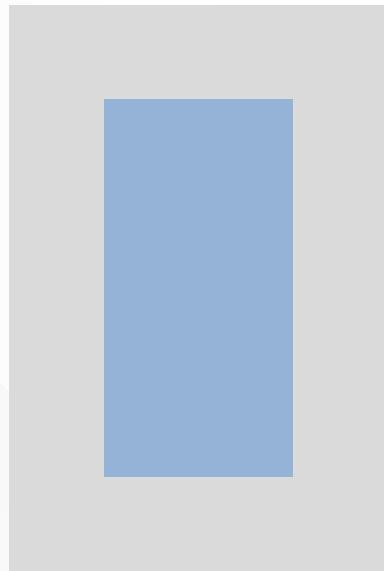


Top view



Cross section

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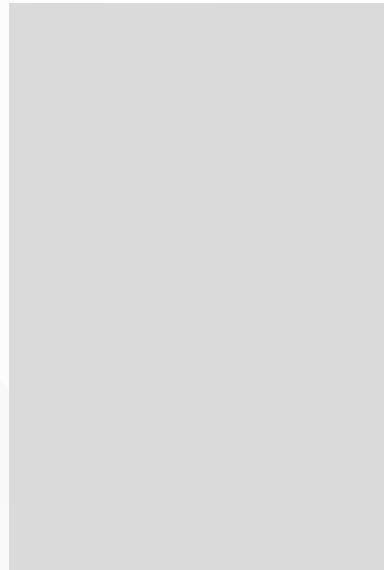


Top view



Cross section

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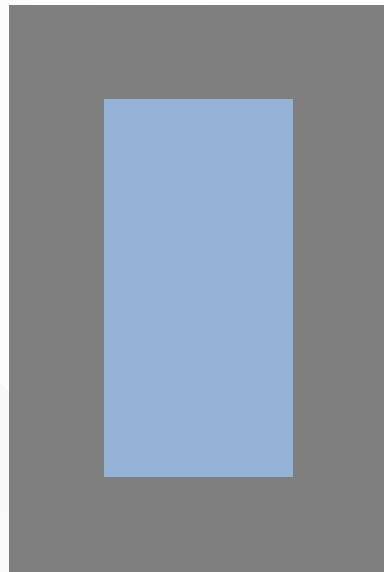


Top view



Cross section

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Top view

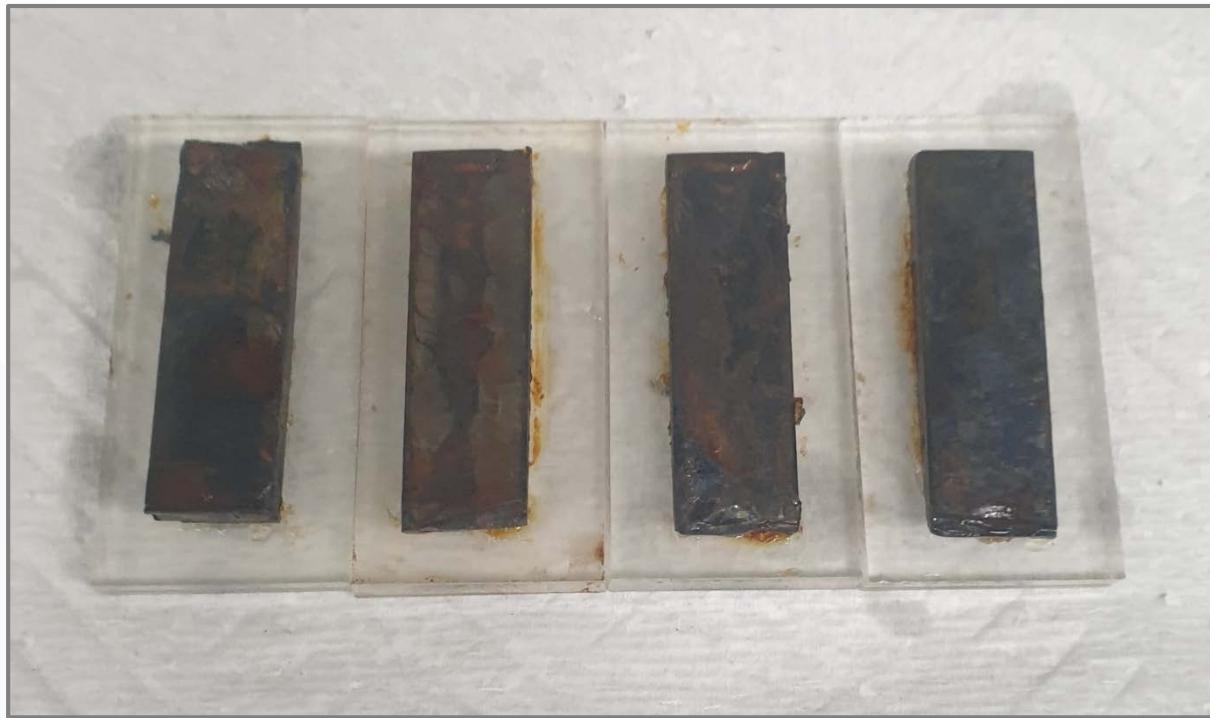


Cross section

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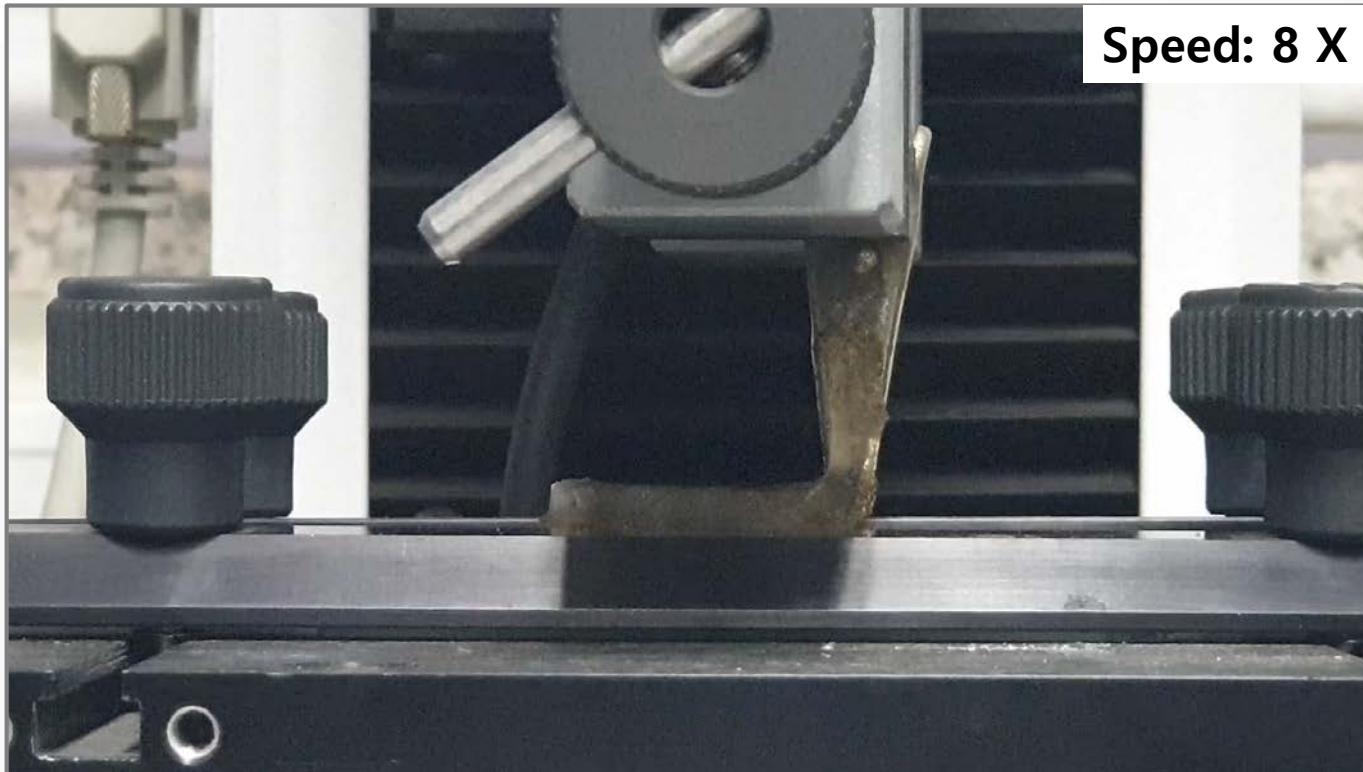
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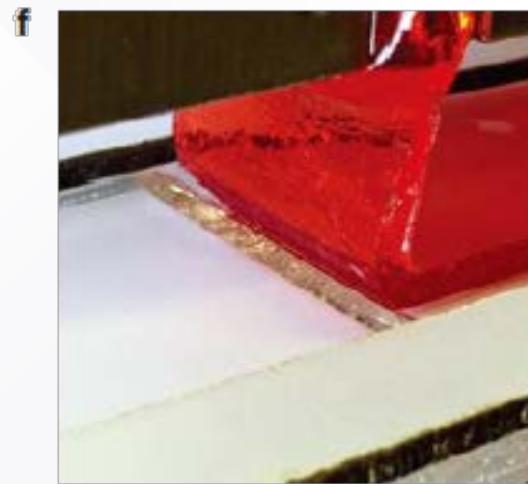


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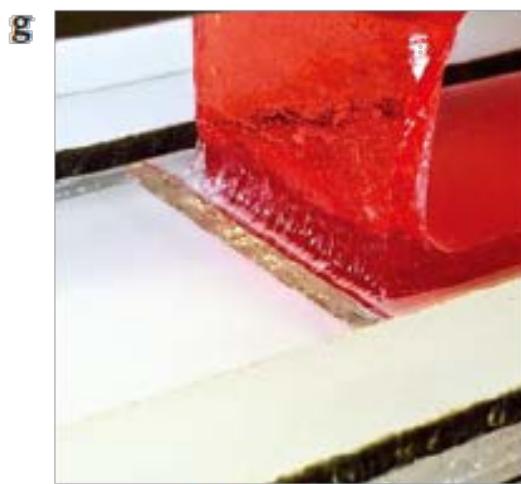


Strain rate: 1 cm/min

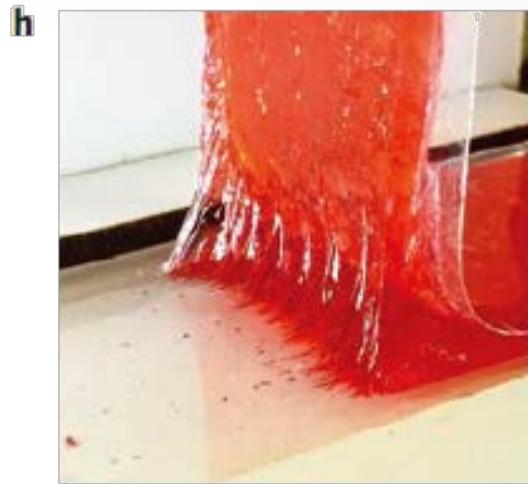
# Tough interfaces between metals and hydrogels



No force

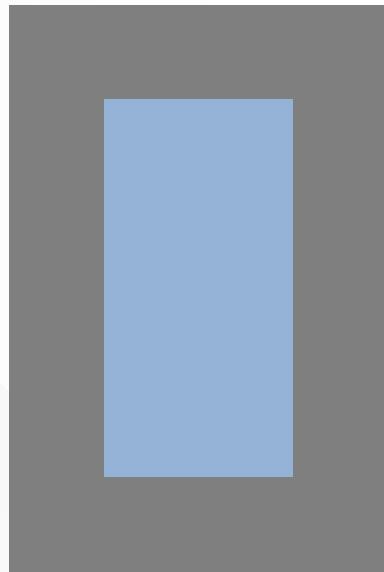


Crack initiation



Steady state

# Tough interfaces between metals and hydrogels

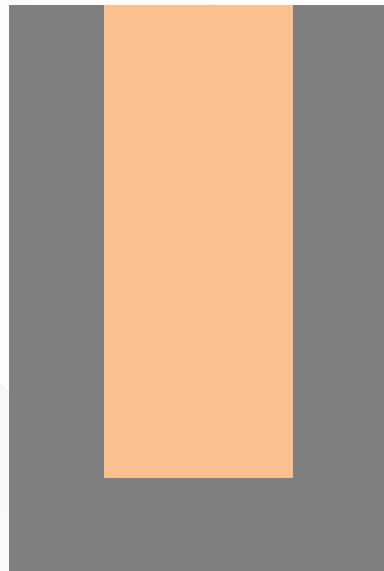


Top view



Cross section

# Tough interfaces between metals and hydrogels

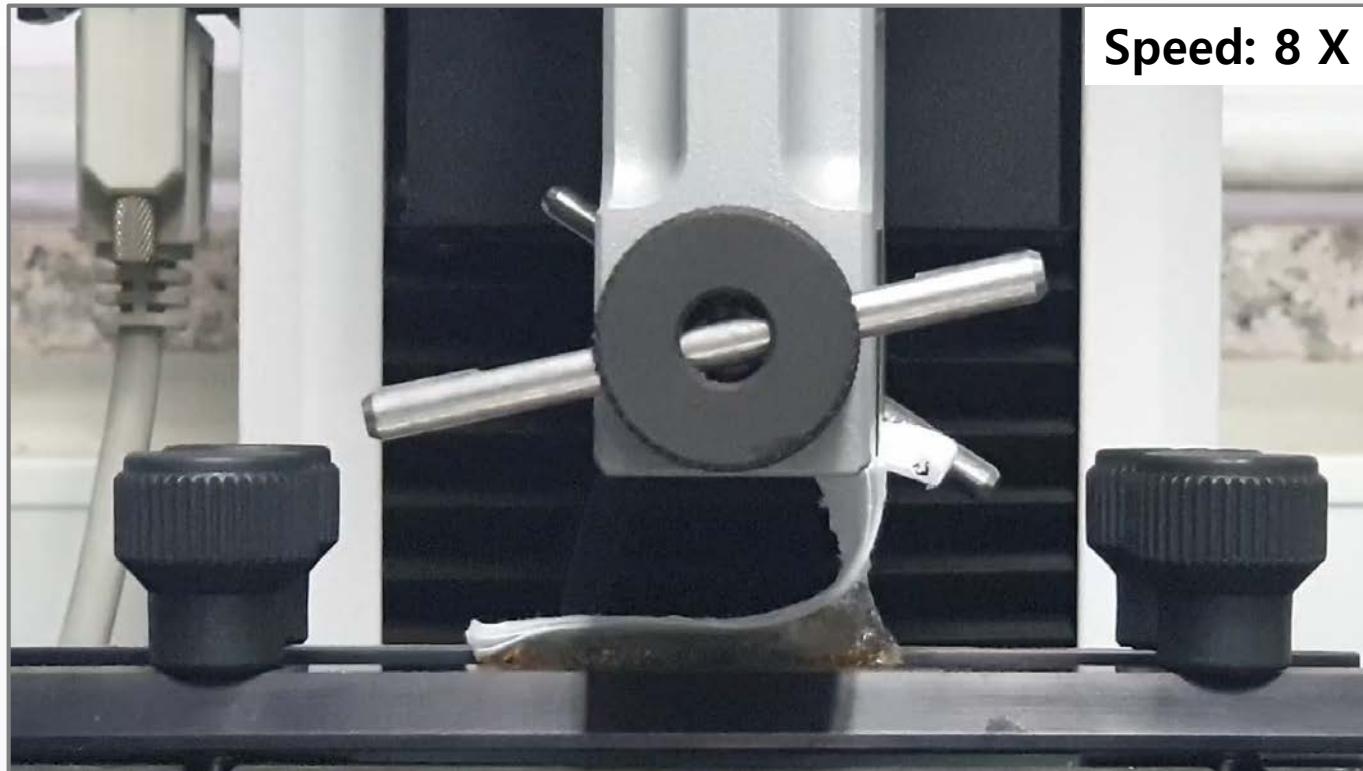


Top view



Cross section

# Tough interfaces between metals and hydrogels



Strain rate: 1 cm/min

# Tough interfaces between metals and hydrogels

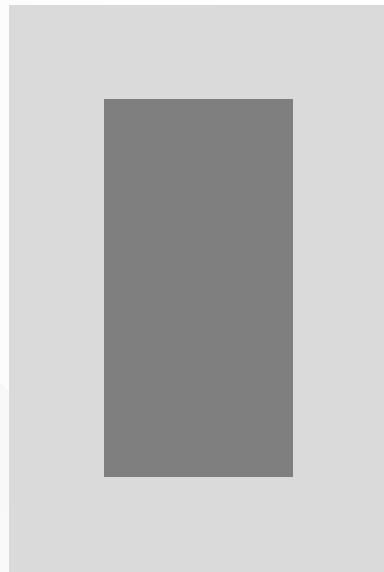


Top view



Cross section

# Tough interfaces between metals and hydrogels

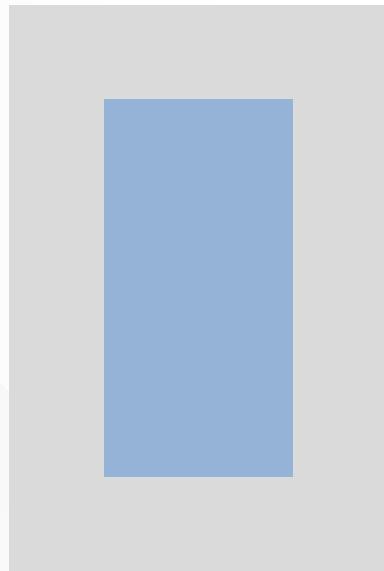


Top view



Cross section

# Tough interfaces between metals and hydrogels

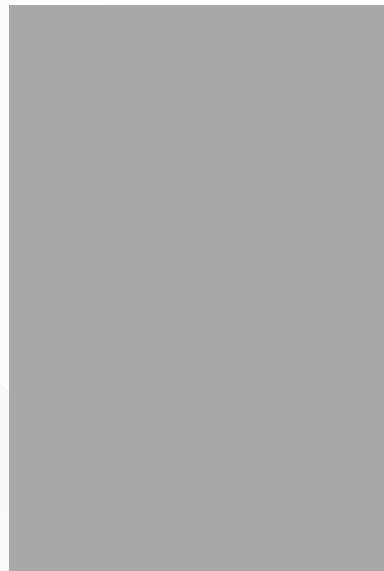


Top view



Cross section

# Tough interfaces between metals and hydrogels

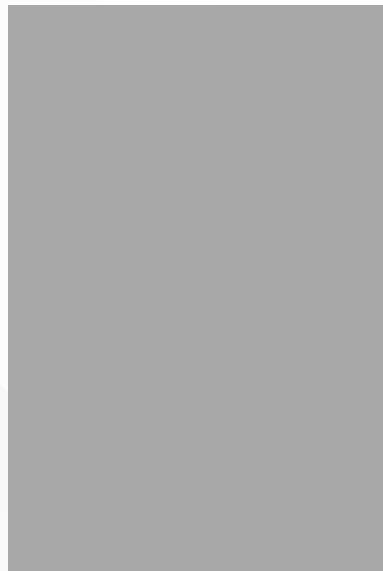


Top view



Cross section

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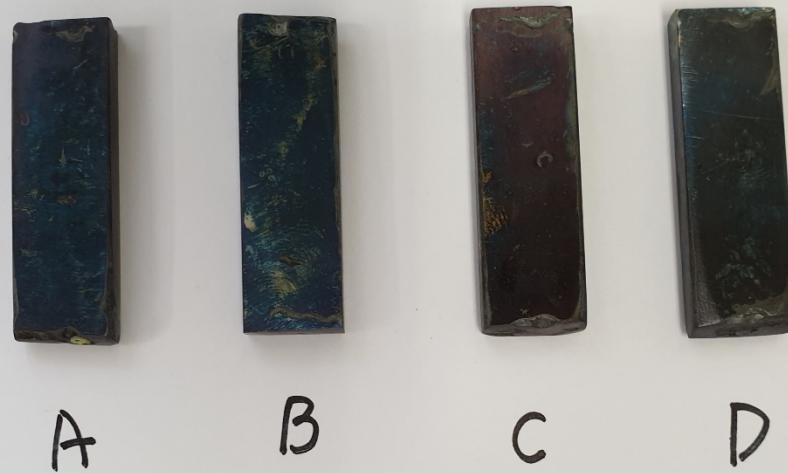


Top view



Cross section

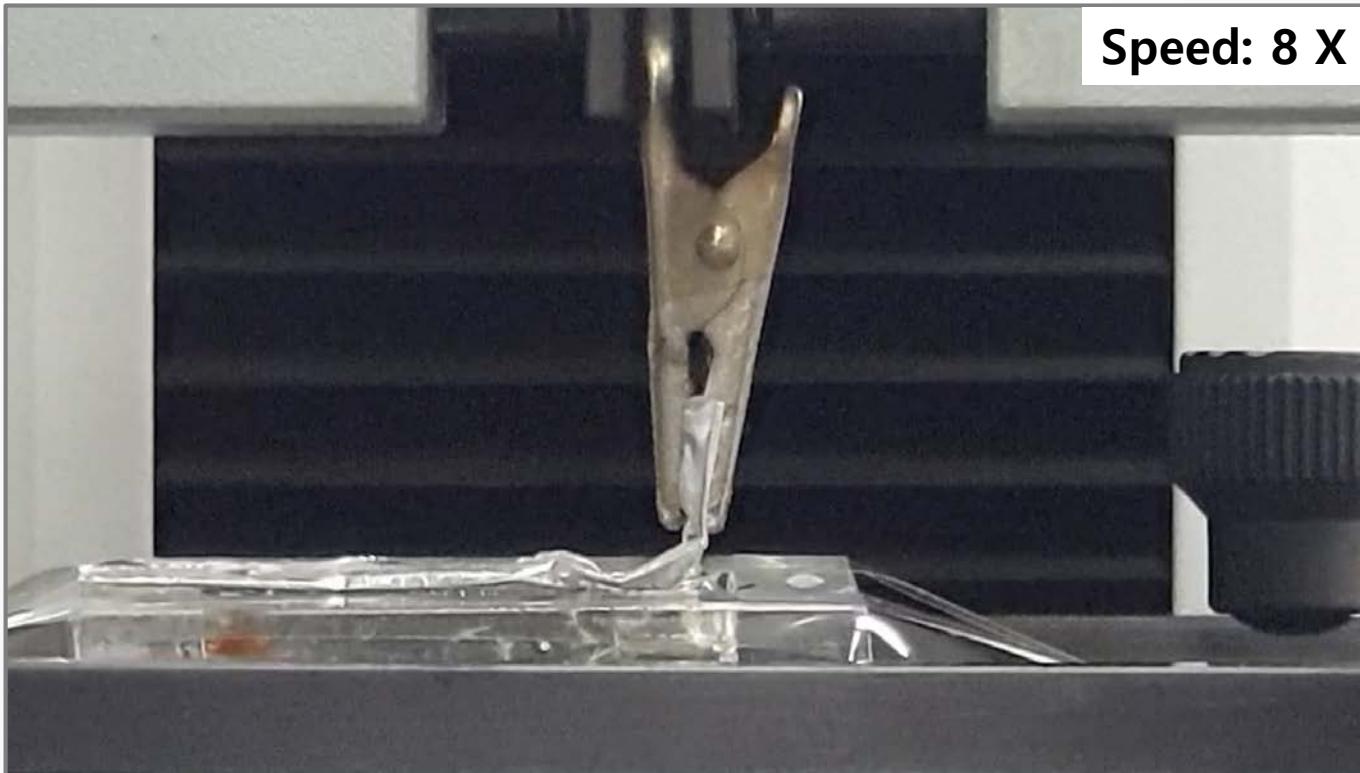
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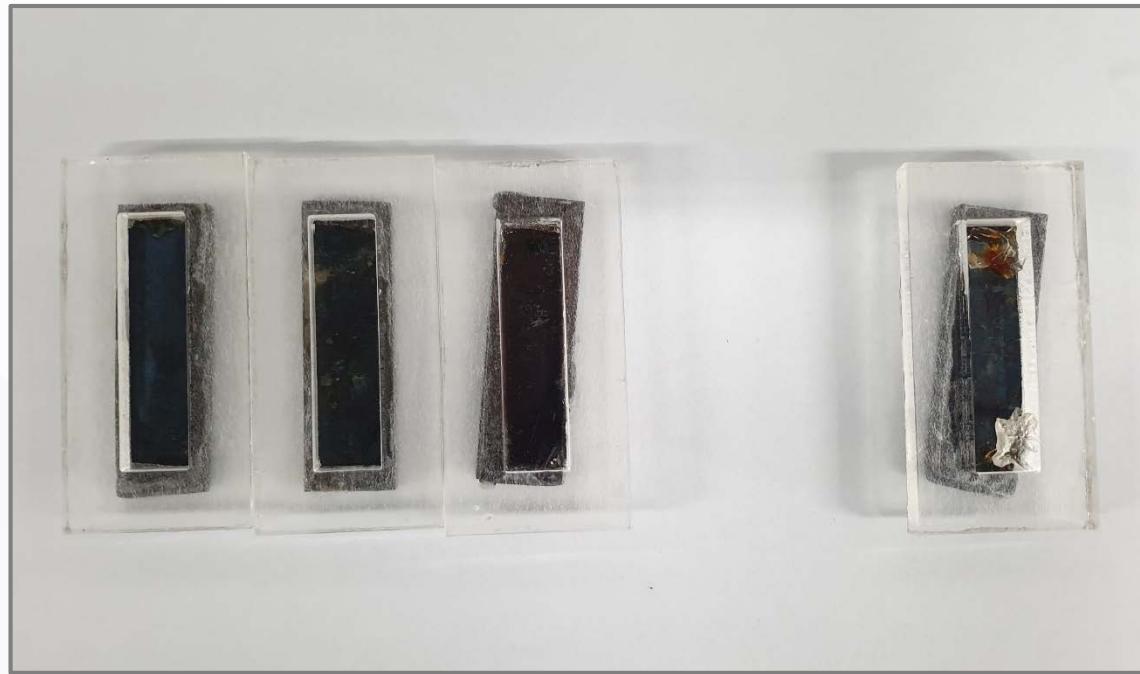
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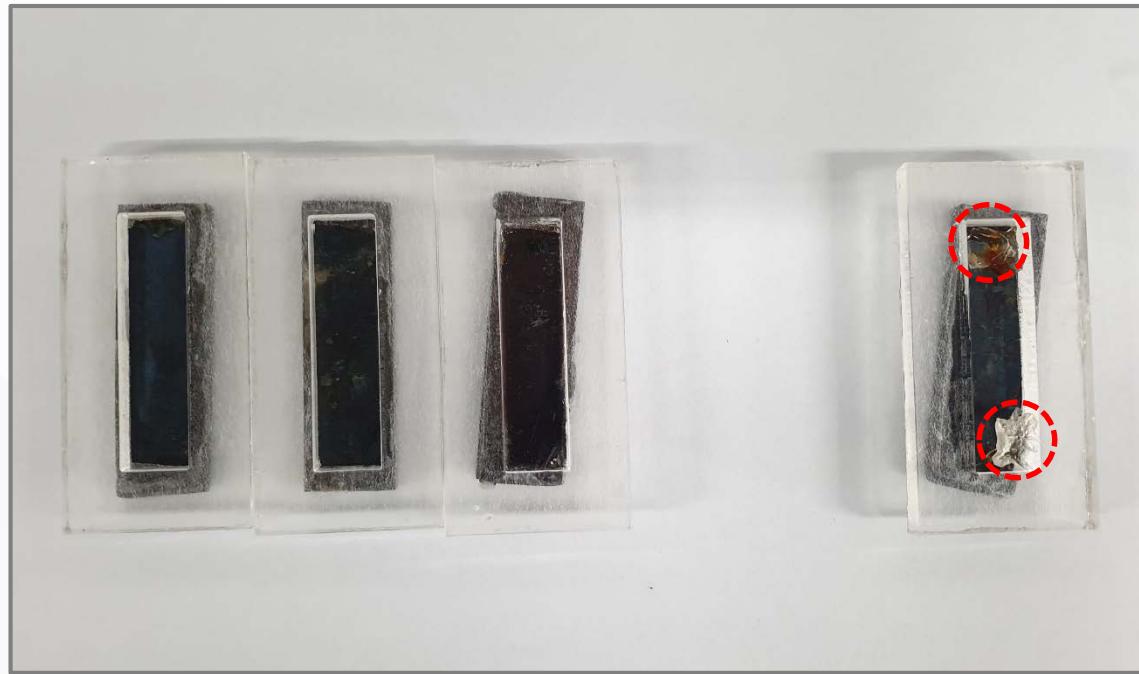
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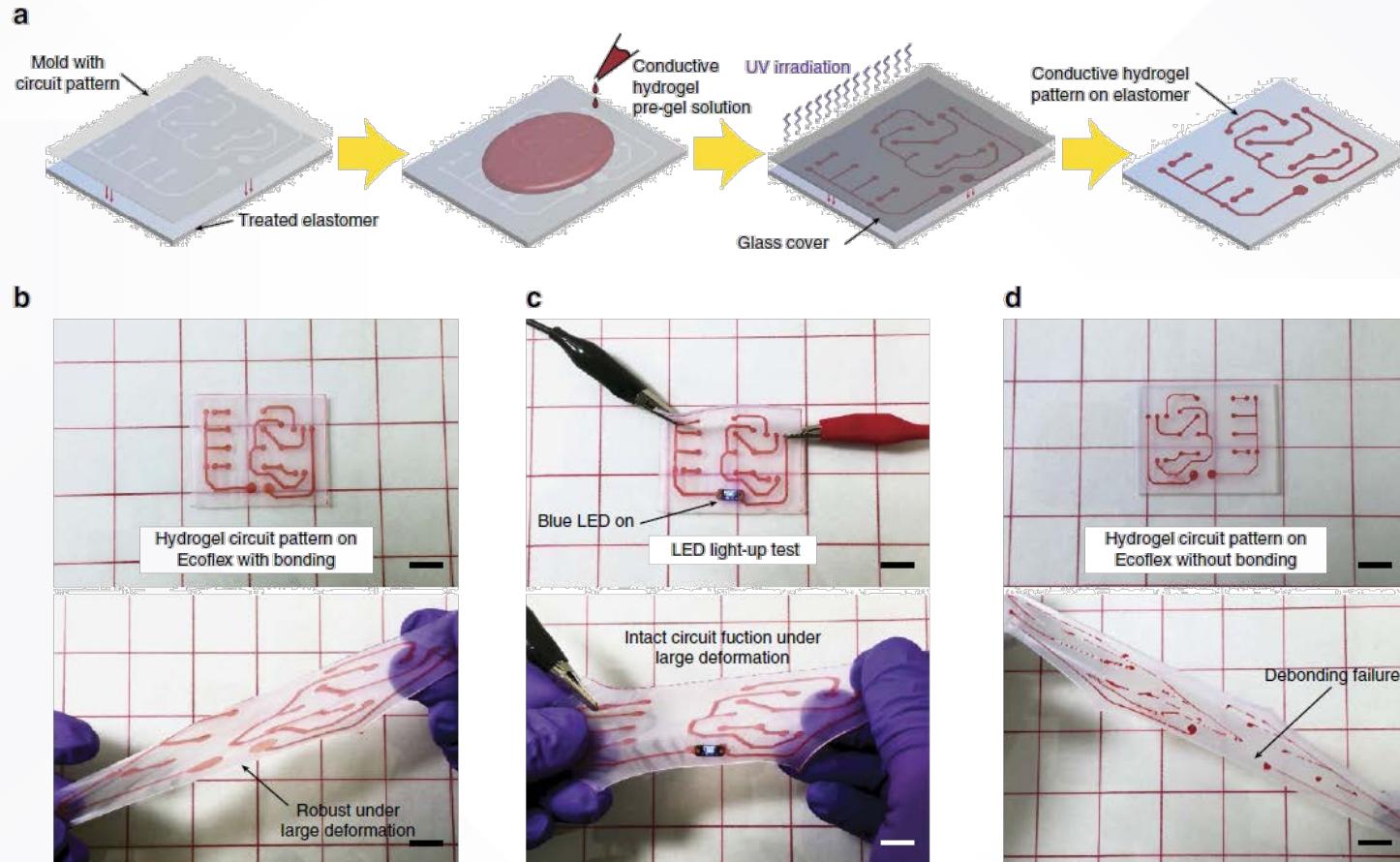
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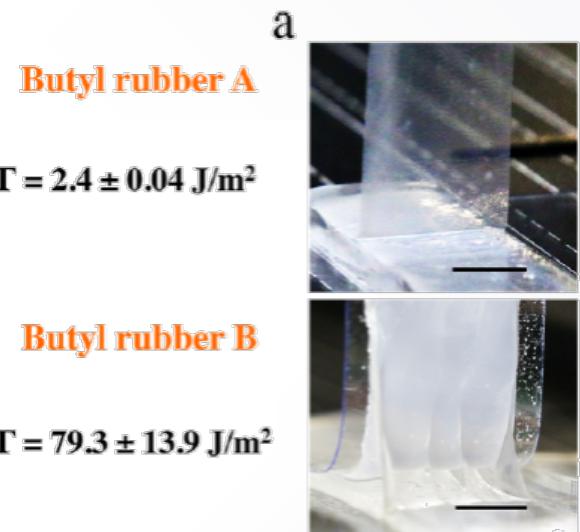
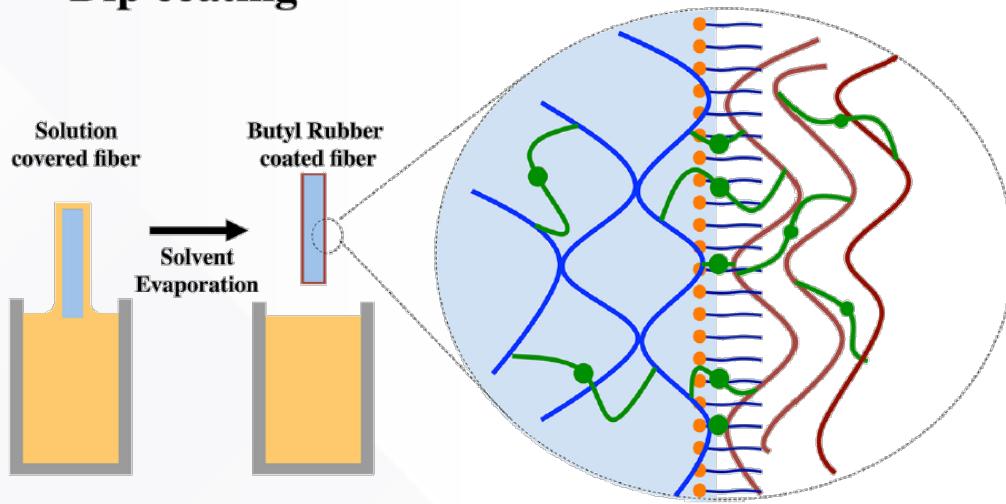


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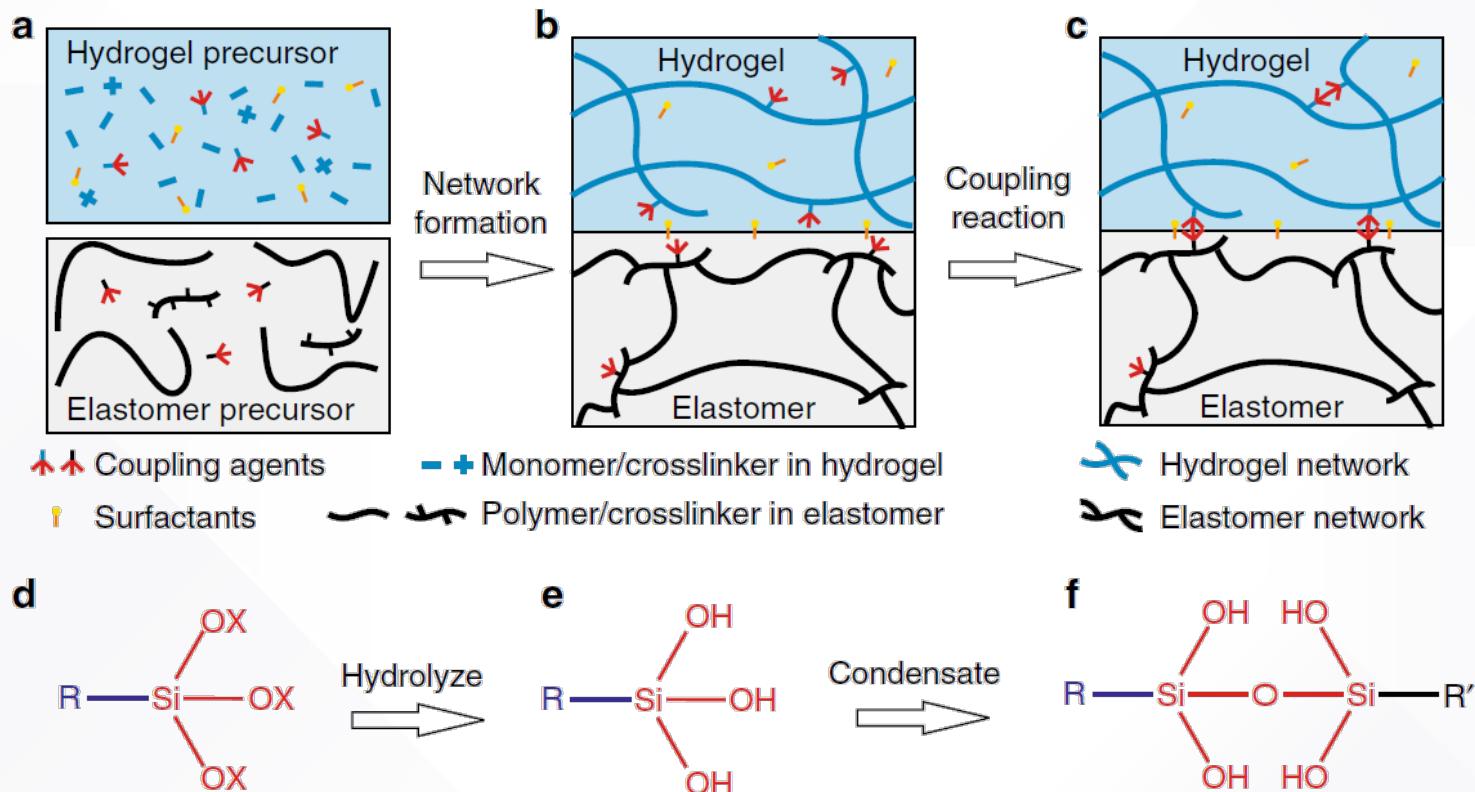


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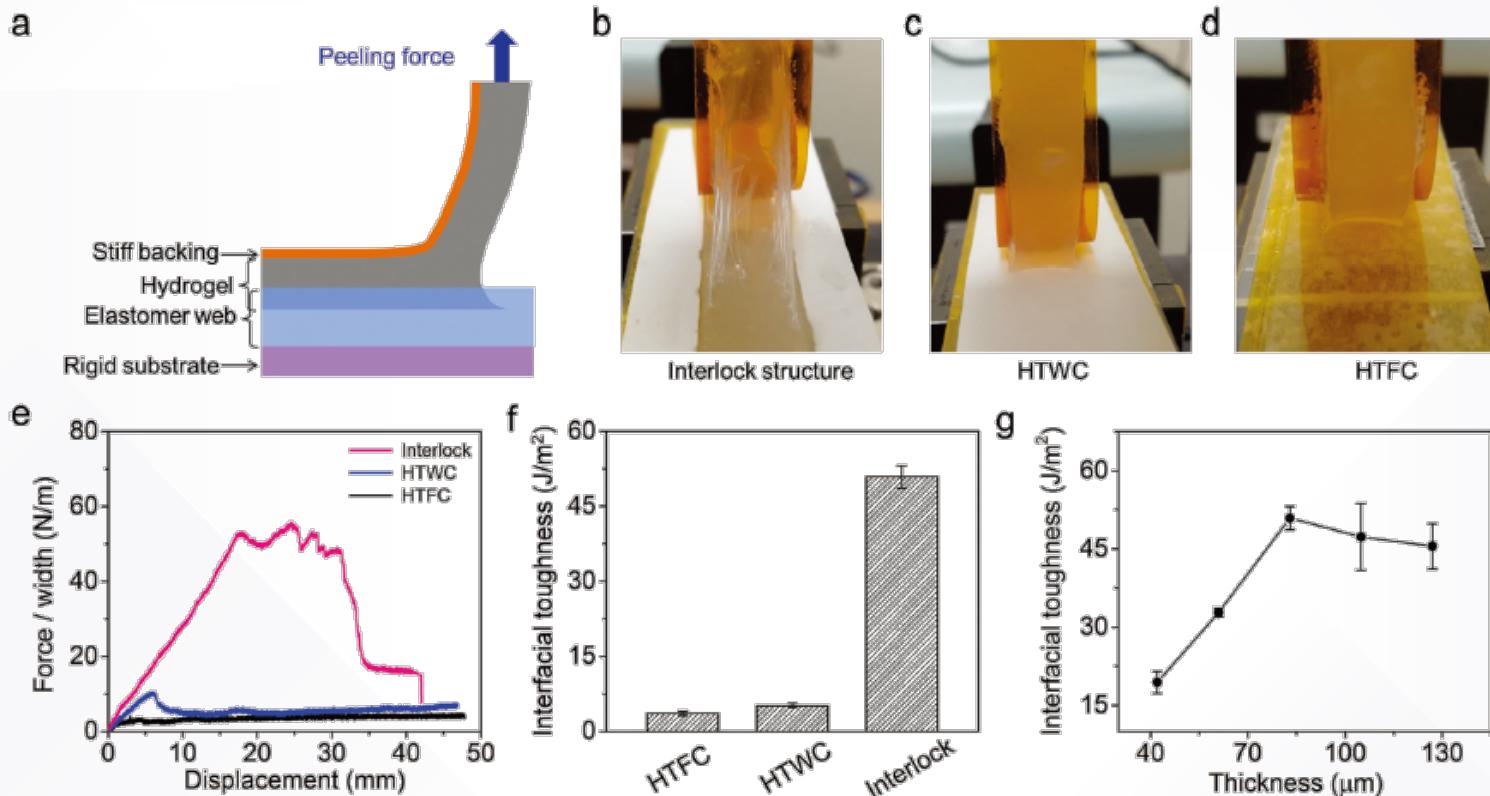
## Dip coating



# Tough interfaces between metals and hydrogels



# Tough interfaces between metals and hydrogels



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**HANK YOU**  
SNU MSE MFSM 송원준