Stretchable Materials: Enabling Soft, Smart, and Self-Healing Polymers for Advanced Applications

Periodic Self-Healing Dynamic Polymers

Nature's self-healing abilities, as observed in biological organisms, have served as a source of inspiration for the development of self-healing materials.

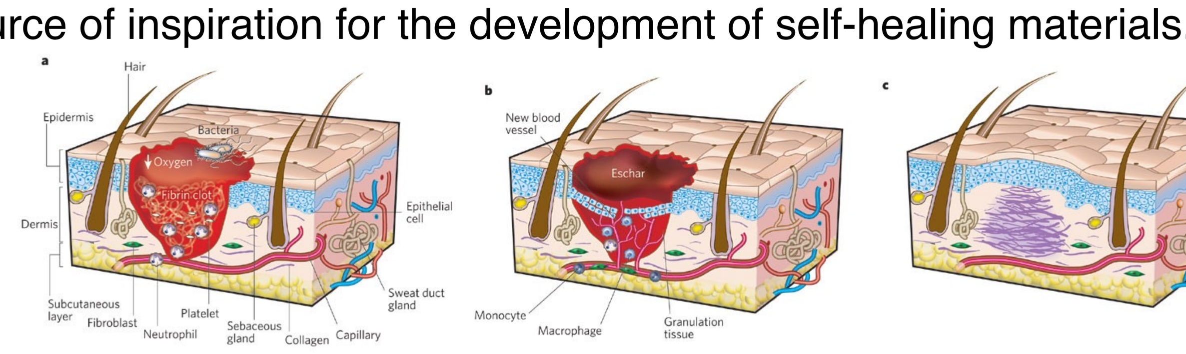
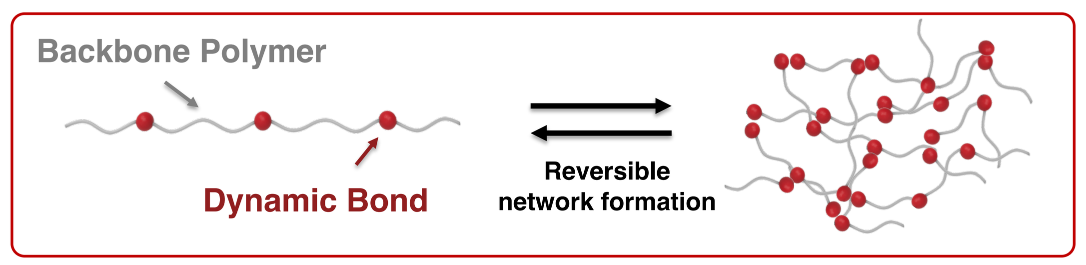


Fig 1. *Stages of wound repair.* [1]

Self-healing in networks occurs by diffusion of polymer chains across fractured interface, reformation of dynamic bonds and re-entanglement of polymer chains.

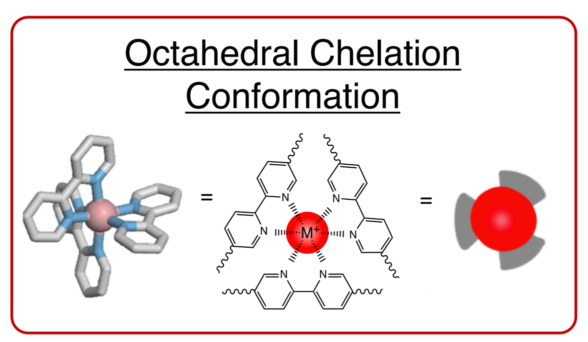


Network contains physical bonds that can reconfigure or heal over time using external stimuli

Acc. Mater. Res. 2022, 3, 9, 948 – 959

Metal-Ligand Coordination Chemistry

Metal–ligand coordination interactions are attractive due to their broad range of molecular parameters: (1) metal ions, (2) counter anions, and (3) ligands, which can be varied to tune the bond strengths, endowing desirable self-healing material properties.



I. Dual-Strength Metal–Ligand Coordination Bonds within PDMS Backbone

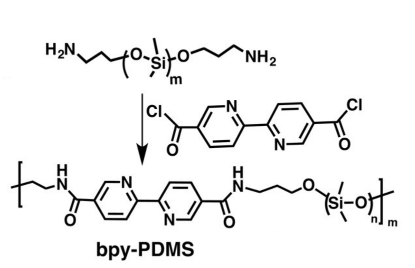
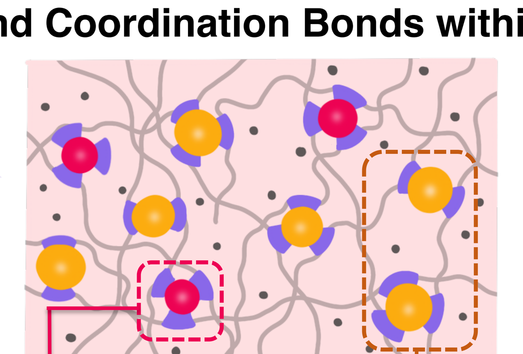
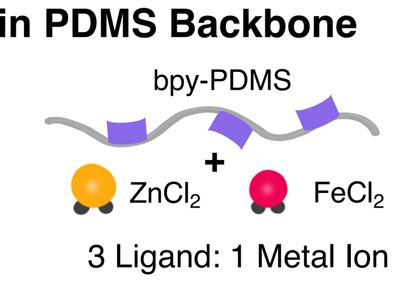


Fig 2. *Synthesis route of bpy-PDMS*





Strong Interaction
- Length: 1.97 Å
- ↓ Kinetically Labile
- Orthogonal Conformation

Weak Interaction
- Length: 2.15 Å
- ↑ Kinetically Labile
- Vary Conformations

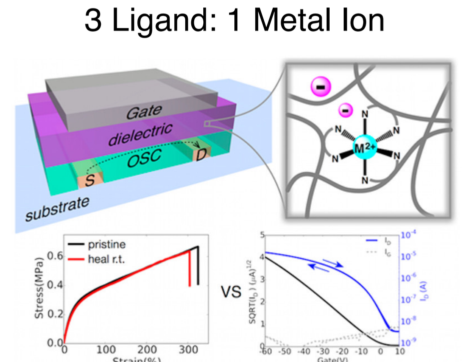
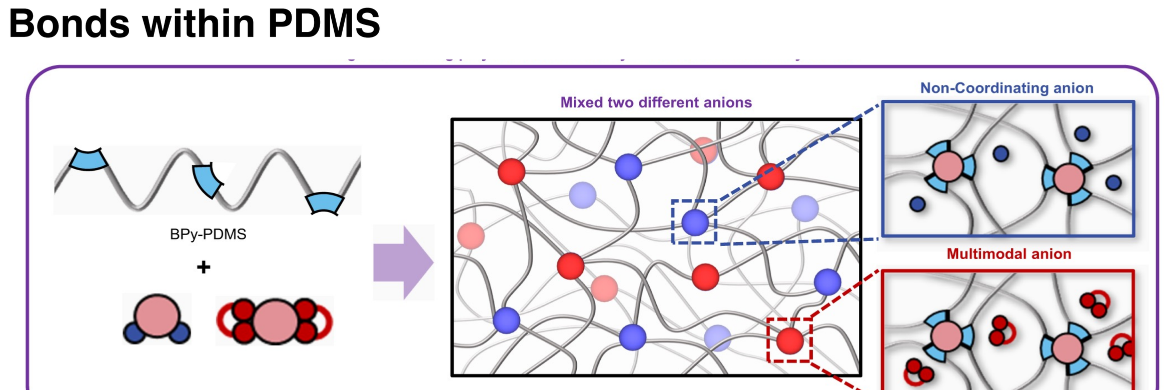


Fig 3. *OFET device structure* [2]

II. Mixing Non-Coordinating and Multimodal Anions in Metal–Ligand Coordination Bonds within PDMS



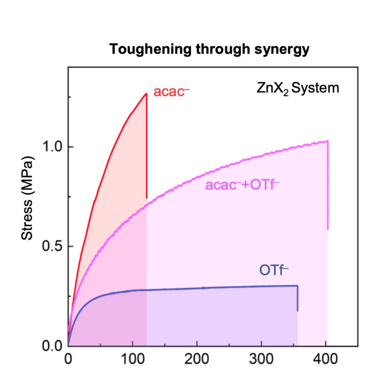


Fig 4. *Stress-Strain Curves of individual and mixed anion.* [3]

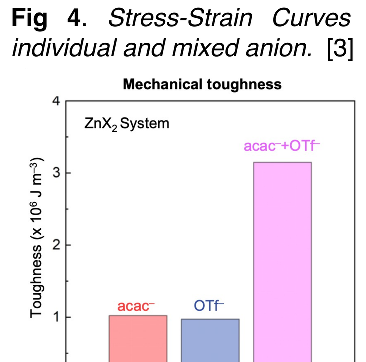


Fig 5. *Toughness values of individual and mixed anion.* [3]

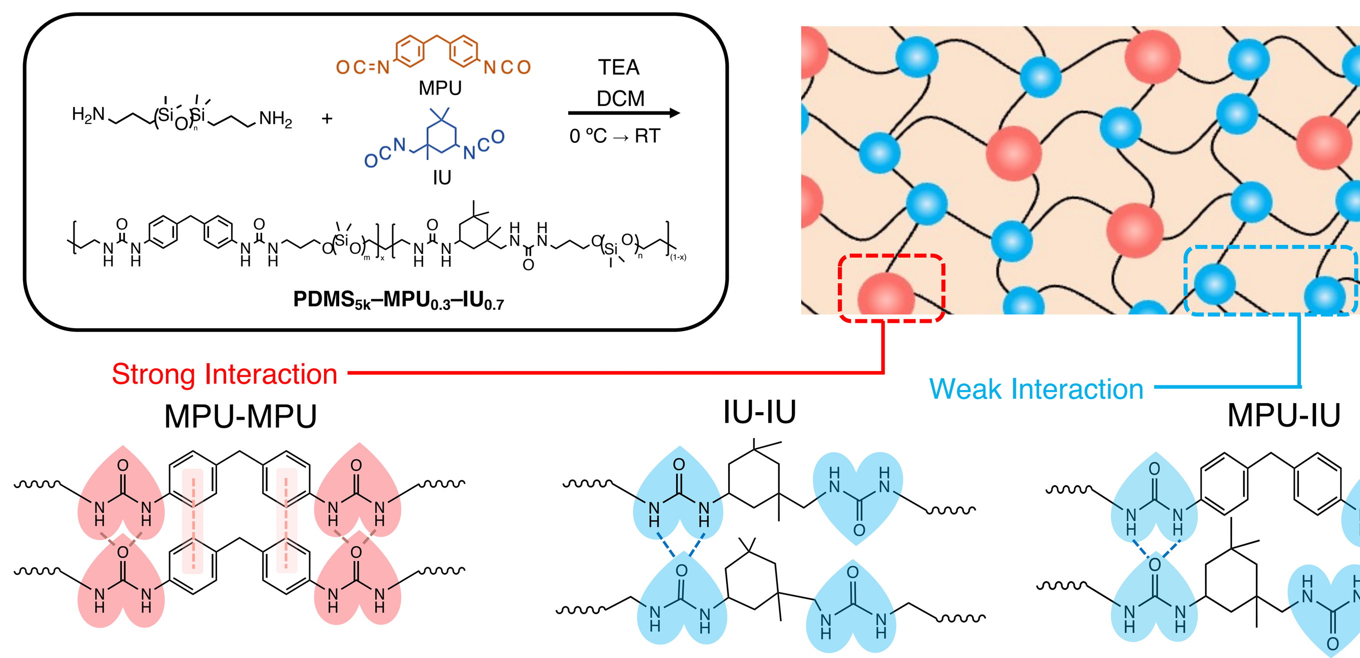
- Non-coordinating anions help dissipate energy quickly, but slow down the self-healing process
- Multimodal anions create additional energy dissipation pathways
- Mix of anions enhances mechanical toughness and self-healing, regardless of the metal used.

J. Am. Chem. Soc. 2016, 138, 18, 6020 – 6027; Nat. Comm. 2023, 14, 1, 5026

Hydrogen Bonding

Hydrogen bonding interactions are a useful approach due to their synthetic simplicity, cooperative binding mechanism, and fast bond breaking and reformation dynamics, which enhances a material's robustness.

I. Multi-Strength Hydrogen Bonds within PDMS Backbone



While individual HB are comparatively weak (with an energy range of ~5–30 kJ/mol), one tenth of a covalent bond (~345 kJ/mol C–C bonds), they still affect the viscoelasticity, crystallinity and phase separation structure of polymer.

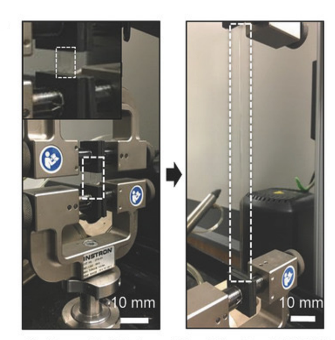


Fig 6. *Mechanical Test of PDMS film* [5]

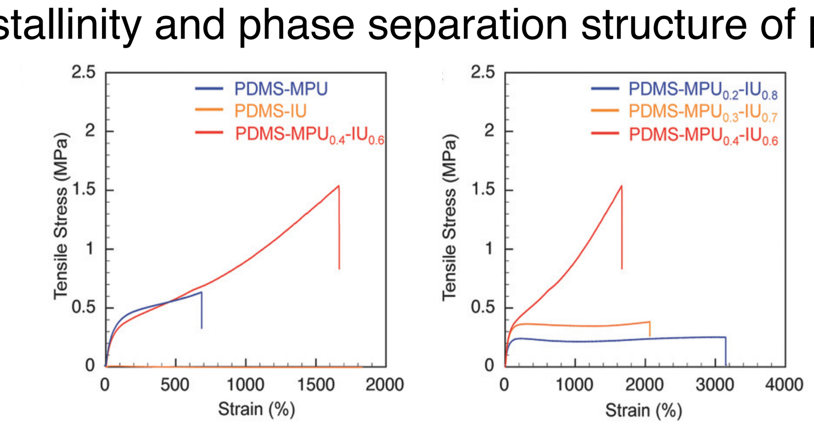


Fig 7. *Stress-Strain Curves for PDMS-MPU-IU variations* [5]

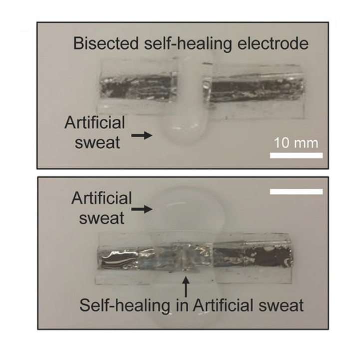


Fig 8. *Self-Healing Electrode* [5]

New polymeric material approach with multi-strength hydrogen bonding achieves high extensibility, water and notch insensitivity, and toughness to withstand wear and tear, while simultaneously self-healing properties.

Adv. Mat. 2018, 30, 18, 1706846; ACS Appl. Polym. Mater. 2020, 2, 9, 4127 – 4127

4. Immiscibility of Periodic Dynamic Polymers

Challenges

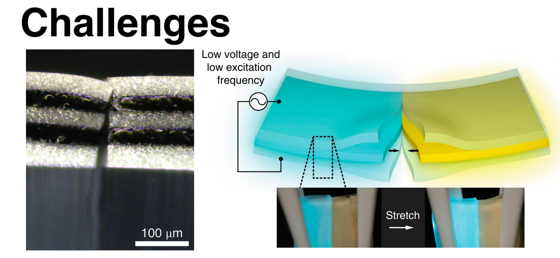


Fig 9. *Damaged Multilayer Polymer Devices* [7]

Current Approaches

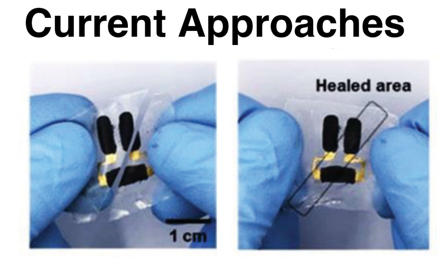


Fig 10. *Manual Alignment*

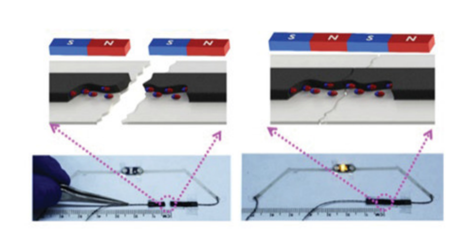
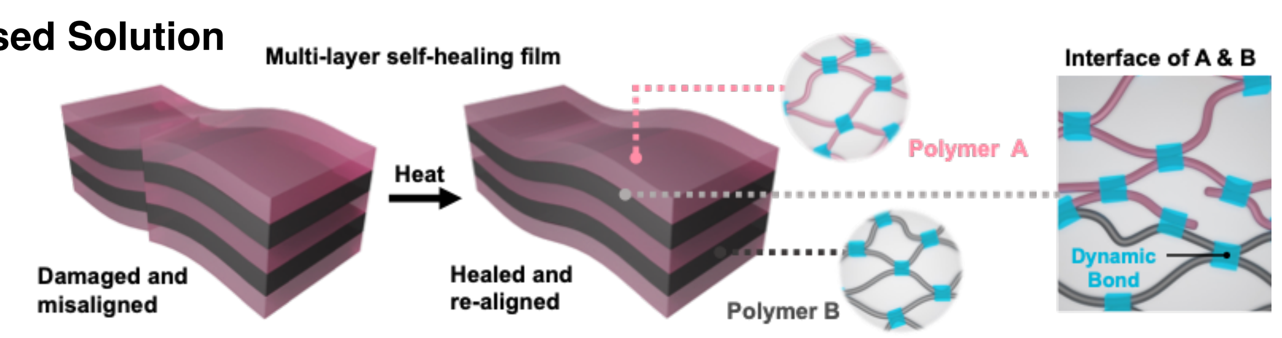


Fig 11. *Magnetic Manipulation* [6]

Proposed Solution



Broad Interfacial Width = Adhesion

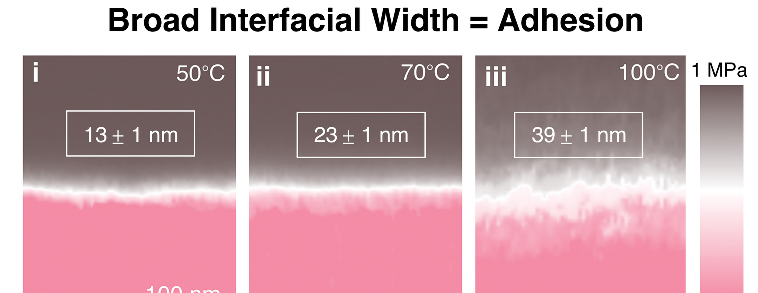


Fig 12. *AFM characterization of modulus gradient across the interfaces of bilayer films at 50 C, 70 C, and 100 C.* [7]

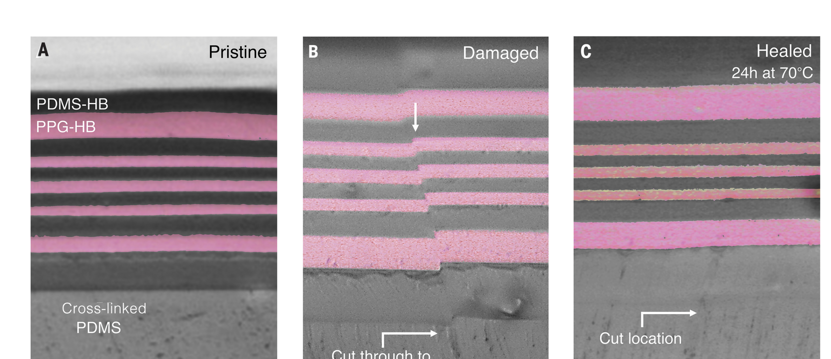
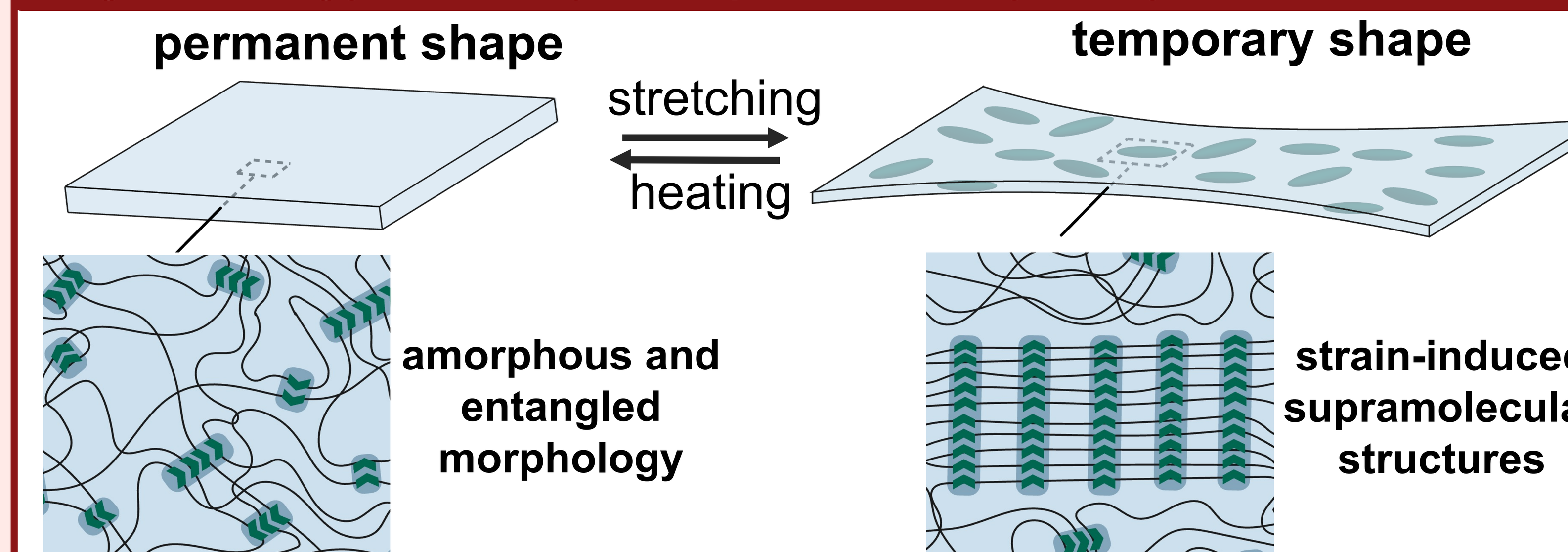


Fig 12. *Cross-sectional optical microscope images of pristine, damaged, and healed multilayer after 24 hr annealing at 70 C.* [7]

First demonstration of a thin polymer multi-layer film with autonomous realignment during self-healing using two immiscible dynamic polymers with different backbones.

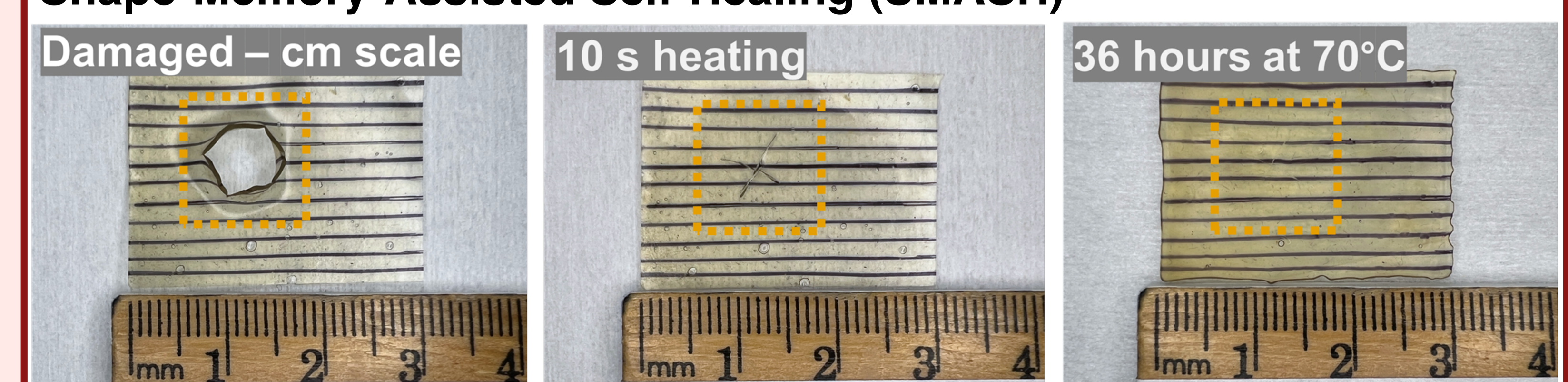
Science. 2023, 380, 6648, 935 – 941

High Energy Density Shape Memory Polymers



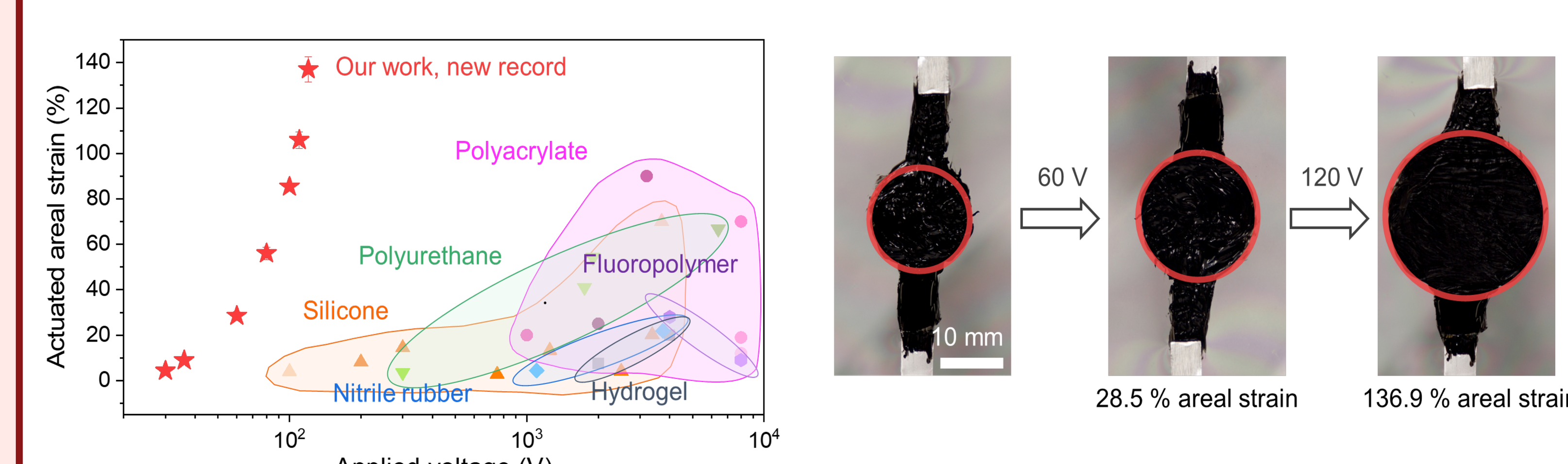
The alignment of the polymer chains, from stretching, induces the **formation of supramolecular structures** due to H-bonds.

Combining SMP & accessible terminal flow temperatures:
Shape-Memory-Assisted Self-Healing (SMASH)



ACS Cent. Sci. 2021, 7, 10, 1657–1667

Low Voltage Dielectric Elastomer Actuators

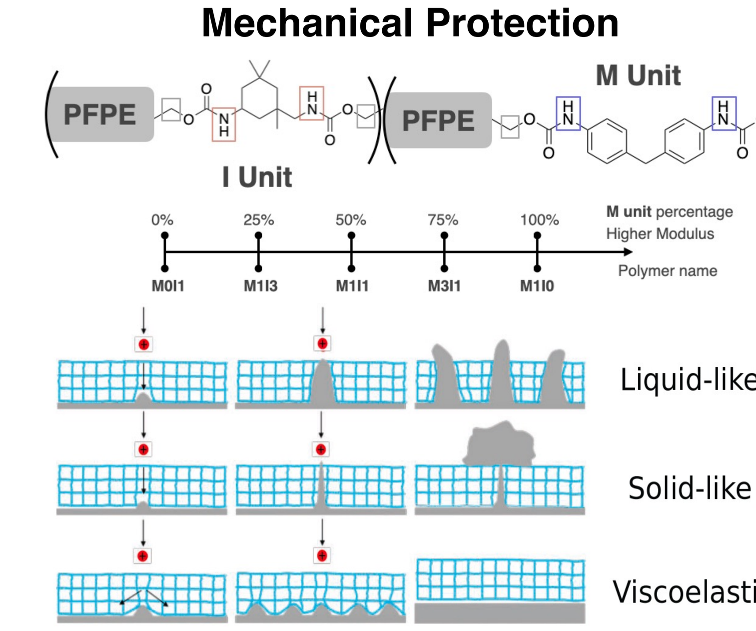


Dielectric elastomers with **high dielectric constant** and **low elastic modulus** results in low voltage actuations for haptic applications

Battery Coatings

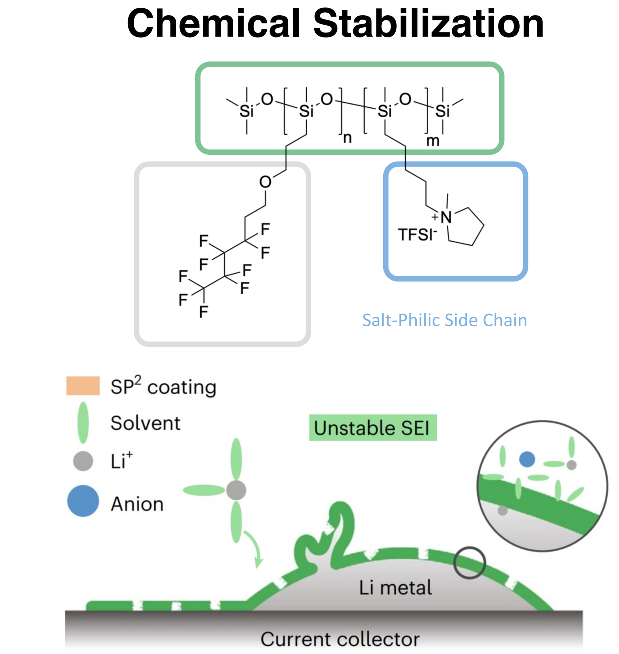
Soft, stable, and dynamic polymers can serve as protective anode coatings for high-performance, safe Li metal batteries as next generation energy storage solutions.

Mechanical Protection



- a fine-tuned coating modulates lithium plating morphology and maintains uniform ion transport through its viscoelasticity and dynamic, self-healable properties.

Chemical Stabilization



- by tuning salt and solvent affinity, the coatings can modulate SEI formation through selectively transport ion and solvent, thus increasing the anode stability.

Nat. Energy. 2023, 8, 577-585; Adv. Energy Mater. 2022, 32, 5, 21013187