DMR-1921835, 1921858, 1921923/2049518

Collaborative research: Strain-adaptive materials

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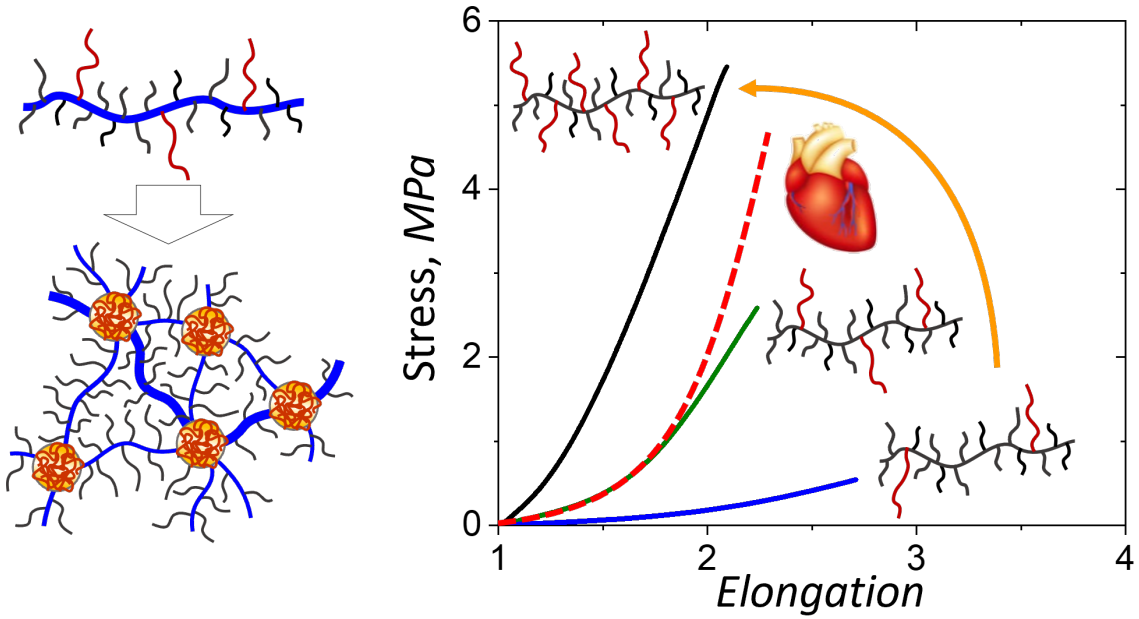
Soft-yet-firm and elastic-yet-damping are opposing traits that synergistically define the unique mechanics of biological tissues. Until now, no single class of synthetic materials including elastomers and gel could provide independent control of these distinct mechanical characteristics. This challenge was addressed by molecular engineering of brush-like building blocks that self-assemble into thermoplastic elastomers with architecturally tunable softness and firmness. These materials can reach strength of ~10MPa on par with stress-supporting tissues such as blood vessel, muscle, and skin. Furthermore, brush-based materials exhibit viscoelastic relaxation in a broad frequency range, which is vital for shock absorbance and vibration damping.

The distinct feature of the design-by-architecture approach is the ability to encode a broad range of mechanical characteristics in network architecture without altering its chemical composition. This opens opportunities for artificial intelligence in the design of soft biomimetic materials with programmable properties.

E. Dashtimoghadam, M. Maw, A.N. Keith, F. Vashahi, Y. Tian, A.V. Dobrynin, M. Vatankhah, S.S. Sheiko , *Super-soft, firm, and strong elastomers toward replication of tissue viscoelastic response*, Materials Horizons **9**, 3022 (2022). DOI: 10.1039/D2MH00844K

One molecule – multiple strands: Soft, strong, and damping materials with programmable properties



Self assembly of brush-like graft copolymers yields thermoplastic elastomers that uniquely combine softness and firmness of biological tissues. The strength of tissue like blood vessels and muscles was archived by tuning the brush architecture.

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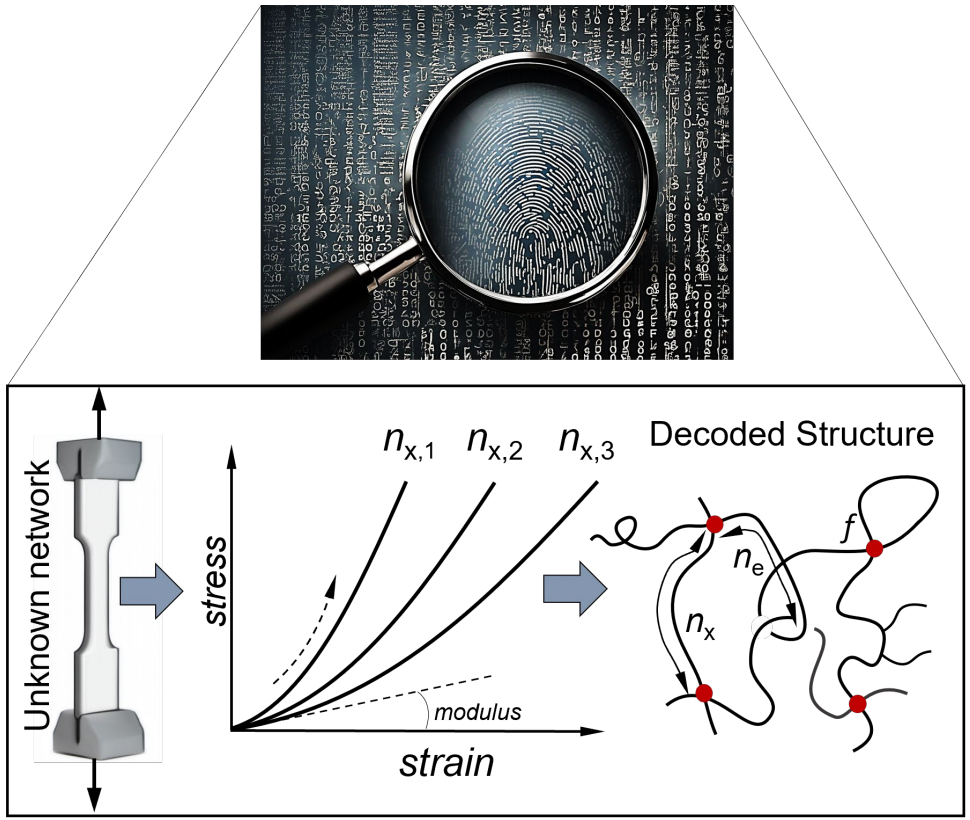
This project has culminated with the development of a forensics methodology for non-invasive evaluation of polymer network topology, which ultimately defines all properties of polymer networks from elasticity to swelling. Any reasonable efforts to project network architecture by specifying formulation and pathway of chemical reactions are instantly scrambled by erratic node formation processes generating a stochastic distribution of structural elements. This problem was resolved by developing a forensic-like methodology for deciphering the network structure from its deformation response.

These capabilities will have a manyfold impact on accelerated design of soft material. First, it allows verification of the agreement between targeted and actual network structure, which is especially crucial for confirming architectural codes generated by artificial intelligence. Second, it provides an architectural blueprint for a desired set of mechanical properties to meet the needs of specific applications. Third, it enables quality control of synthesized polymer networks to detect and minimize human errors.

A.V. Dobrynin, Y. Tian, M. Jacobs, E.A. Nikitina, D.A. Ivanov, M. Maw, F. Vashahi, S.S. Sheiko, *Forensics of Polymer Networks*, Nature Materials **22**, 1394 (2023). DOI: 10.1038/s41563-023-01663-5

M. Maw,… K. Matyjaszewski, A.V. Dobrynin, S.S. Sheiko "Brush Architecture and Network Elasticity: Path to the Design of Mechanically Diverse Elastomers" Macromolecules **55**, 2940 (2022). DOI: 10.1021/acs.macromol.2c00006

Forensics of polymer networks



Schematics of network forensic approach based on the analysis of non-linear response of elastomers to deformation.

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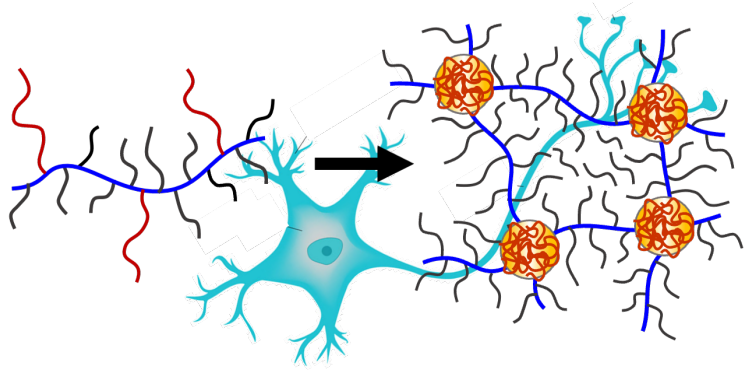
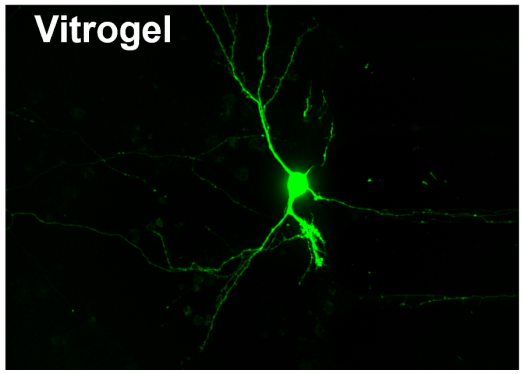
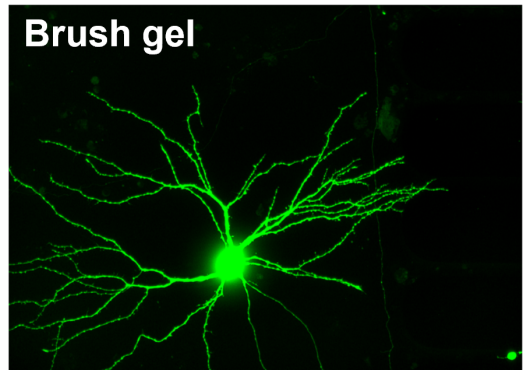
We have partnered with Xona Microfluidics, Inc. on joint development of microchip devices that enable high throughput screening of novel drugs for Alzheimer's disease. Current materials such as Matrigel and Vitrogel that are used for cell encapsulation are inadequate due to batch-to-batch variability of their composition and mismatch between their mechanical properties and extracellular mechanics. We employed our design-by-architecture platform to synthesize tissue-mimetic hydrogels for encapsulation of neuron cells and subsequent monitoring of the axon growth as a function of network architectural codes.

Specifically, we have synthesized a series of linear-brush-linear triblock copolymer with a hydrophilic brush block and two thermosensitive linear blocks with systematically varied architectural parameters. These polymers undergo microphase separation at the body temperature to produce robust physical networks that combine tissue-mimetic softness, firmness, and water content. Our material substantially outperformed the commercial analogs by demonstrating much faster and more vigorous axon growth, indicating the creation of a favorable biomechanical environment by the brush gels.

Collaboration with Xona Microfluidics Inc.: Encapsulation of neuron cells







Neuron cells embedded in different matrixes demonstrate different patterns of axon growth. The brush hydrogel promotes much more vigorous growth as compared to commercial counterparts.

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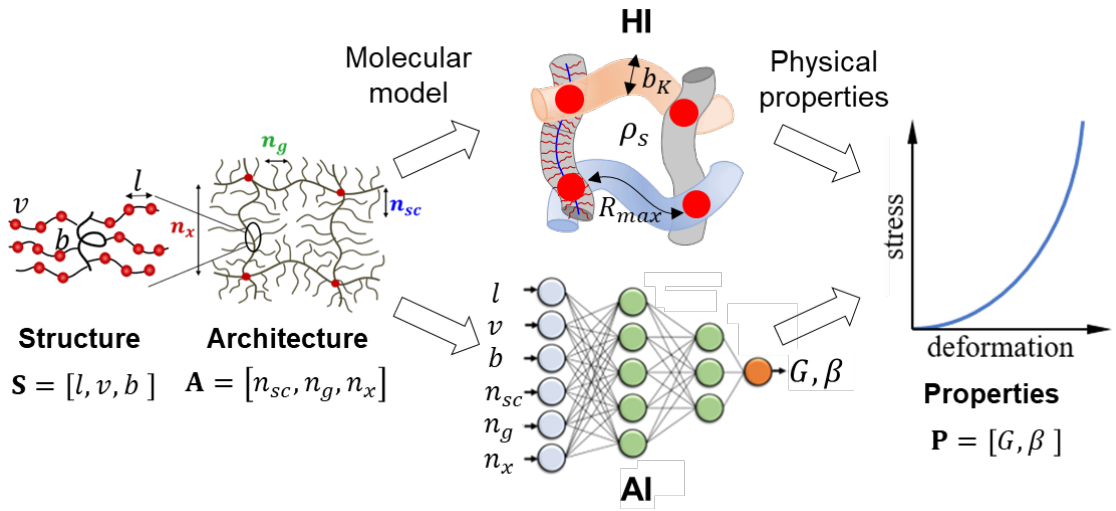
The PIs have established a highly efficient research triangle incorporating soft matter synthesis, characterization, and theory targeting the development of tissue-mimetic materials possessing a strain-adaptive mechanical properties. Theoretical model of brush networks, developed by Dobrynin, guided the network synthesis by Matyjaszewski, followed by verification of predicted structure-property correlations by Sheiko. The closed-loop feedback between the groups allowed refining the theory predictions in the form of materials design rules correlating mechanical and swelling properties with molecular architecture of network strands. Unlike many phenomenological theories currently used in the materials design, our model involves specific architectural parameters that can be synthetically controlled.

The developed design rules have been applied to the synthesis of elastomers, gels, and pastes with predictable mechanical properties. The *design-by-architecture* approach significantly enhances the materials design efficiency and precision in contrast to the conventional trail-and-error approaches based on explorative mixing of reagents. It provides data sets for training of ANN for soft materials design fostering broad application of AI-based techniques.

M. Maw,.. K. Matyjaszewski, A.V. Dobrynin, S.S. Sheiko "Brush Architecture and Network Elasticity: Path to the Design of Mechanically Diverse Elastomers" Macromolecules **55**, 2940 (2022). DOI: 10.1021/acs.macromol.2c00006

Synergizing the Human and Artificial Intelligence in Soft Materials Design



Workflow of HI and AI network design encoded by monomer projection length l, excluded volume v, and Kuhn length b of bare backbone and side chains, and network architecture $\boldsymbol{A} = [n_{sc}, n_g, n_x]$. The network model is defined by the length of fully extended backbone, R_{max}, and effective backbone Kuhn length due to interactions between side chains, b_K.