Mechanically triggered adaptive materials

Yixiao Dong, Pin-Ruei Huang, Elina Ghimire, Charlie A. Lindberg, Farsa Ram,
Stuart J. Rowan,* and Aaron Esser-Kahn*

Adaptive materials, characterized by their dynamic response to external stimuli, undergo changes in their physical or chemical properties to suit varying environmental conditions. Mechanical forces, ubiquitous in our surroundings, are widely utilized by living systems to reinforce tissues and bones. This natural process has spurred numerous studies in recent decades. This article first focuses on recent advancements in mechanically triggered adaptive materials, examining the underlying principles, fabrication methods, and potential applications across diverse disciplines. Challenges in the field are identified along with future insight and opportunities, such as leveraging the chemistry toolbox and emerging engineering technologies to develop adaptations with higher order and longer time-scale response. Future adaptive materials are envisioned to possess unique properties and functionalities, enabling applications such as self-repair, biomimetic smart systems, and information storage across diverse scenarios.

Introduction

Adaptation behavior is a crucial surviving trait found in all biological species. Living systems develop a variety of strategies, such as tropism (movement in response to an external stimulus), phenotypic plasticity (environment affected gene expression), and adaptive immunization (immunization that is tailored to recognize and target specific pathogens), to adapt to the specific environmental conditions in which they find themselves. Inspired by the trainability of biological systems such as force-induced bone stiffening and muscle growing, the concept of adaptation has been applied to materials with the goal of improving performance and/or imparting new functionalities.[1,2] In general, adaptive materials refer to a class of substances engineered with the capability to respond dynamically to external stimuli, such as mechanical,[3,4] thermal,[5] (bio) chemical,[6] or electromagnetic cues,[7] by altering their properties or behavior. Mechanical adaptation of materials depends on their structure and associated activation mechanisms, which allows them to alter ideally desired mechanical properties without the need for remaking or reprocessing.[8,9] Mechanical force-induced mechanical adaptation is particularly interesting as all systems commonly experience a range of mechanical forces and as such materials that can adapt to such stresses can allow access to new application areas.

Mechanical vibrations and cyclic loading by stretching, compression, or shear are the most common ways of inducing mechanical adaptation in materials such as dense suspensions,[10] gels,[11] and soft polymeric materials.[3] A primary requisite of mechanical adaptation is the material's ability to change its structure in response to mechanical loads. Mechanophores (mechanically sensitive molecules) and piezoelectric particles can respond to mechanical forces and drive chemical, physical, configurational, topological, or structural changes in the materials that result in mechanical adaptation.[12,13] For example, mechanophores that generate radicals in the presence of mechanical forces can result in secondary network formation via polymerization of monomers and results in self-strengthening.[3] In addition to mechanophore-induced adaptation, mechanical adaption can be realized through other mechanisms that include (1) strain-induced alignment

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Yixiao Dong, Pritzker School of Molecular Engineering, The University of Chicago, Chicago, USA; ydong2@uchicago.edu
Pin-Ruei Huang, Pritzker School of Molecular Engineering, The University of Chicago, Chicago, USA; huangpr@uchicago.edu
Elina Ghimire, Pritzker School of Molecular Engineering, The University of Chicago, Chicago, USA; elinaghimire@uchicago.edu
Charlie A. Lindberg, Pritzker School of Molecular Engineering, The University of Chicago, Chicago, USA; lindbeca@uchicago.edu
Farsa Ram, Pritzker School of Molecular Engineering, The University of Chicago, Chicago, USA; Interdisciplinary Centre for Energy Research, Indian Institute of Science, Bangalore, India; ramf@iisc.ac.in
Stuart J. Rowan, Pritzker School of Molecular Engineering, The University of Chicago, Chicago, USA; stuartrowan@uchicago.edu
Aaron Esser-Kahn, Pritzker School of Molecular Engineering, The University of Chicago, Chicago, USA; aesserkahn@uchicago.edu
*Corresponding author

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of high aspect ratio fillers;[14] (2) strain-induced mesogenic realignment in liquid-crystal elastomers (LCEs);[15,16] (3) mineralization deposition on charged surfaces induced by mechanical activation;[9] and (4) strain-induced rearrangement of dynamic covalent, or noncovalent bonds.[17–19] These fundamental studies have spurred the development of new adaptive materials with properties, such as anti-fatigue,[13] self-healing,[16] and self-strengthening,[3,20] which enable enhanced functionality and performance of smart materials that have impacted a range of fields and applications, such as soft robotics,[21] biomedical devices,[22] energy-harvesting systems,[23] and smart textiles.[24]

There are existing reviews that have highlighted the use of mechanochemistry[25] or the design of mechanical adaptation in polymer science.[26] While various mechanistic approaches have been developed to impart mechanical adaptation, this article will focus on examining mechanically triggered adaptative polymers by using the development and design of three different classes of mechanical-induced adaptivity in materials as examples: (1) covalent bond formation through piezo-initiated polymerization; (2) mechano-activated mineralization process in polymer matrix as the adaptivity is contributed from inorganic crystallization; (3) noncovalent aggregation such as mesogenic realignment in LCEs (**Figure 1**). In the end, we discuss the challenges and opportunities of mechanically triggered adaptive materials and give an insight into their future applications.

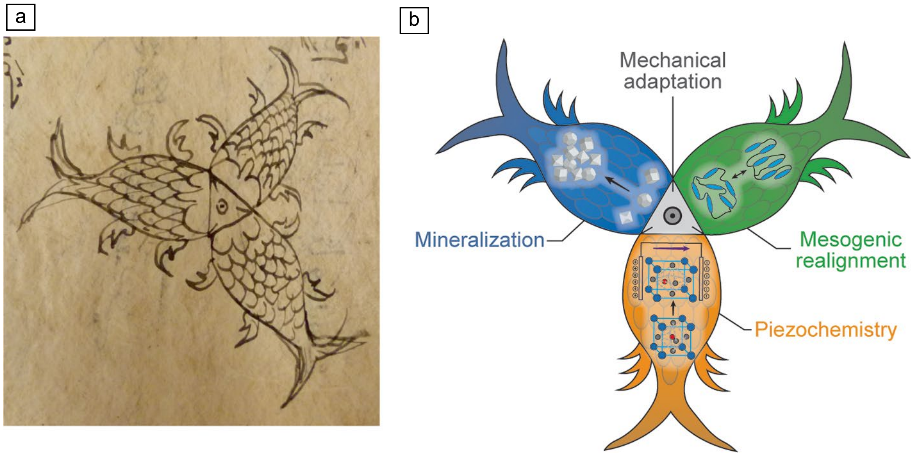

Figure 1. Three mechanistic approaches to access mechanically triggered adaptive behavior. (a) A Javanese manuscript (AD 1790/1) that depicts three fish with one head (British Library, MSS Jav 77, f. 16v) and (b) the corresponding concept of this review that will focus on three different mechanisms (fish bodies), piezochemistry, mineralization, and mesogenic realignment, to access the same goal (fish head) of mechanical adaptation.

Piezo-initiated polymerization: Mechanically driven covalent bond formation

As a subcategory of piezochemistry, piezo-initiated polymerization (PIP) opens the door to new approaches to mechanically active adaptive materials. Since the first example of piezo-initiated controlled atom transfer radical polymerization (ATRP) was demonstrated in 2016 (**Figure 2**a),[27] the toolbox of such chemistry has been greatly expanded from design

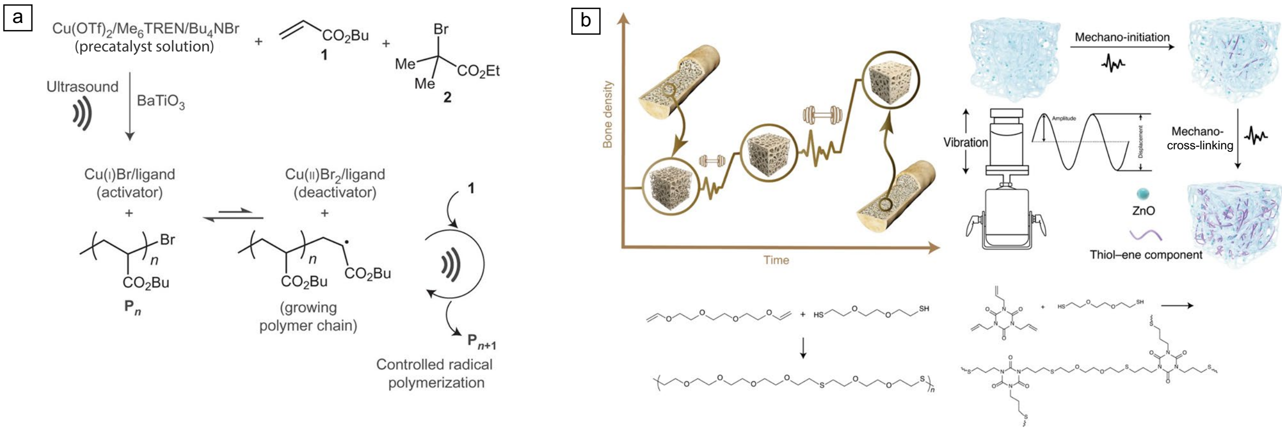

Figure 2. Adaptation that is driven by piezo-initiated covalent bond formation. (a) Ultrasound triggered living polymerization through $BaTiO_3$ piezonanoparticles. (b) Adaptive ZnO composite that can be self-stiffened in response to external vibration signals, akin to a bone stiffening process. Reprinted with permission from References 3 and 27. © 2021, 2017 Springer Nature.

principles to material types.[3,28–30] Other traditional controlled/polymerization methods, such as reversible addition fragmentation transfer (RAFT), have been initiated by mechanical activation of piezoelectric particles.[29] Commonly proposed mechanisms[31,32] attribute radical generation via two primary processes: (1) Mechanical deformation induces polarization on the surface of piezoelectric particles, creating electron (e^-) and hole (h^+) domains. (2) Redox reactions occur between the polarized domain and chemical initiators, ultimately generating a sufficient number of radicals to sustain the radical polymerization process. More specific mechanisms highly depend on an applied piezo system. For example, a $BaTiO_3$-initiated RAFT polymerization system may rely on the formation of hydroxy radicals from water oxidization,[33] whereas the ZnO-initiated RAFT polymerization system could rely on alkyl radicals generated by the reductive cleavage of carbon-halogen bonds.[29] Building off the development of PIP has led access to new adaptive materials that exploit this mechanically activated chemistry. In general, the external forces applied to synthetic materials result in a destructive, irreversible bond cleavage process leading to common effects such as fatigue, deformation, and eventual failure. However, this is not always the case for living systems. In nature, living systems often develop mechanical adaptations to withstand destroying forces. For example, human skin could develop calluses, and bones could increase in density in response to intense physical labor. Inspired from such natural examples, Wang et al. developed a dual network system that could adapt to external vibration or sonication.[3] A methyl cellulose gel was embedded with thiol-ene monomers and ZnO piezoparticles (Figure 2b). When the piezoparticles sense the mechanical input, they will generate radicals that initiate the thiolene polymerization to form a covalent thiol-ene network. Based on this mechanism, these composite materials can detect and adapt to the vibrational input energy, resulting in a modulus increase of up to 66 times relative to its original value. This demonstration established a potential approach for creating adaptive materials that stiffen themselves to withstand external forces, akin to human tissue or bone (Figure 2b).

PIP: Challenges and perspectives

While these initial studies in PIP-based adaptive materials start to show the promise of this approach, there are still numerous challenges and opportunities that need to be addressed if their full potential is to be realized. For example, in many of the finely tuned pathways found in living systems, multiple enzymatic cascade reactions may underlie a single response. Each enzymatic process plays a role in determining and adjusting the nature of the adaptation. In contrast to such sophisticate biological approaches, existing PIP-based adaptive materials operate in a simplistic, one-way reaction manner. Therefore, in addition to the principles and mechanisms established for PIP, developing cascade pathways that facilitate fine-tuning and control could enable materials to adapt in a more precise manner. As preliminary research has indicated the feasibility of establishing an enzymatic cascade *in vitro*,[34] one future effort could concentrate on integrating piezo-based cascade reactions into designed adaptive systems. However, the challenge is that the living systems developed a hierarchical and rather sophisticated structure to accomplish the task of adaptation. The structures were genetically coded and expressed by the cellular systems, which can be hard to mimic using existing technology. In future research, a promising approach could be to focus on developing substitution methods that mimic these structural designs using different advanced technologies, which involves integrating various materials and technologies to replicate the adaptation behavior seen in biological systems.

Beyond the cascade reactions commonly found in living systems, the incorporation of suitable feedback loops is also crucial for living organisms to adapt to their environment. A feedback loop serves not only to determine when adaptation is required, but also to regulate when the adaptive response should cease. Essentially, implementing a feedback loop requires an efficient communication system, wherein a sender element perceives a stimulus, encodes a message for a receiver element, which interprets the message and provides feedback to the initial element. A fundamental illustration of a communication feedback loop is exemplified in the study by Llopis-Lorente et al.,[35] wherein researchers examined interactive communication models at the nanoscale employing two distinct types of Janus nanoparticles. Each variant of Janus nanoparticle functions either as a sender or a receiver. In this case, a hierarchical design of functional nanoparticles is essential for fulfilling the roles of both sender and receiver (**Figure 3**). This concept could be further expanded to facilitate the creation of intelligent PIP adaptive materials. Here, mechano-chemical conversion signals could be communicated and controlled via the interactions between particles or between particles and polymers. For example, future studies could target a multilevel particle communicating system, where a composite system consisted of force transducer particles, energy gating particles, feedback particles, and potentially information storage particles. Eventually, the force adaptivity behavior will no longer render from one or two functional components, but the collaborative or interactive system akin to many well-known biological processes such as adaptive immunization.

In situ mineralization: Cultivating "bone-like" structures within polymeric materials

Mineralization is a prevalent biological tactic, which serves as a route to fortify tissue with enhanced mechanical resilience and structural reinforcement.[36] Although there are numerous studies of growing inorganic crystals in solutions, few studies have shown the mineralization chemistry in a soft polymer matrix with the goal of accessing adaptive materials. Kang et al. have recently demonstrated the mineralization process initiated via piezoelectric materials (**Figure 4**a).[9] The polarization charges produced from mechanical bending of a bulk PVDF can initiate the growth of hydroxyapatite (HAP) in a solution that mimics biological fluids. This discovery could

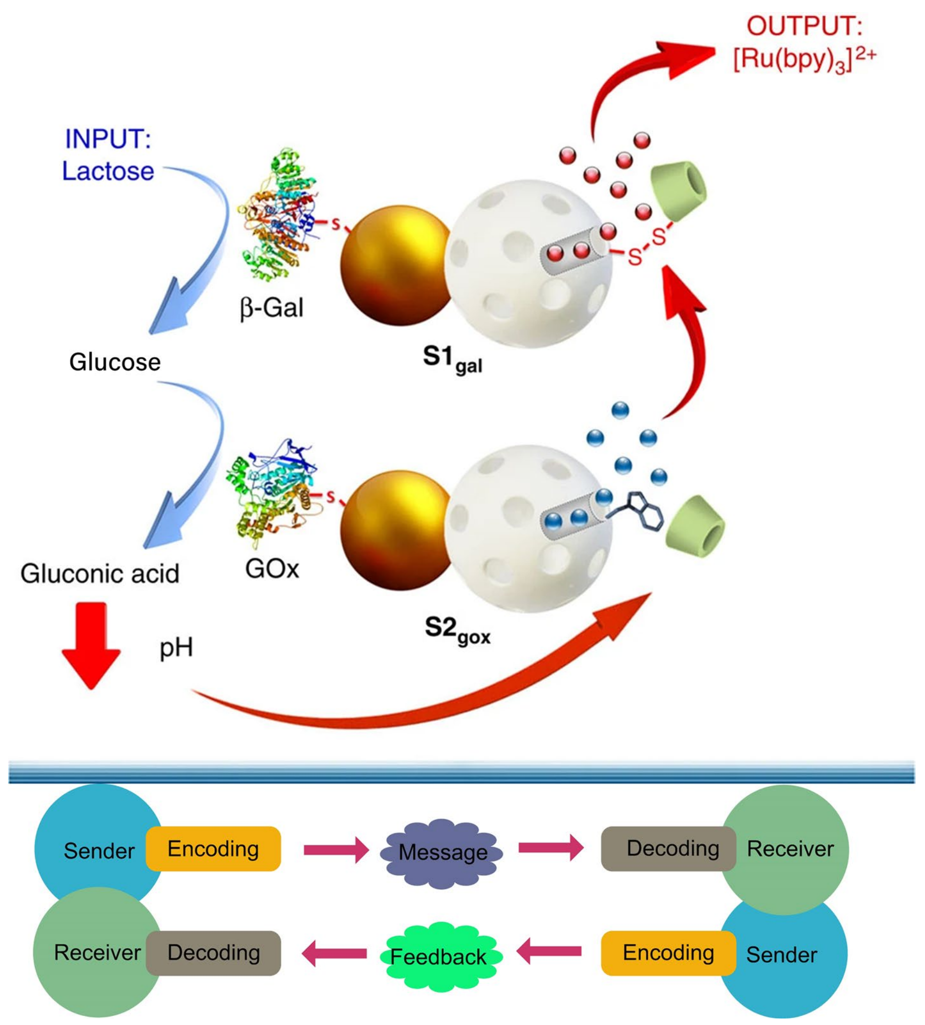

Figure 3. Concept illustration of the interactive model of particle communication: a sender first receives a stimulus, encodes a message for the receiver. The receiver then interprets it and provides feedback to the sender. Reprinted with permission from Reference 35. © 2017 Springer Nature.

open an interesting avenue of smart coating technology for self-stiffening adaptive materials. The study also leads to a new question: instead of using bulk piezomaterial, could a polymer composite that contains mechanoresponsive nanoparticles initiate a similar mineralization process? In fact, a recent study found that ZnO piezoparticles can promote benzyl disulfide cross-linking in synthetic organic gels.[37] In this system, the precise control over the bulk material mineralization is achievable through adjustments in the duration and intensity of mechanical inputs. This manipulation involves tuning parameters such as frequency and force, leveraging molecular-scale bond activations, giving direct correlation between mechanical stimuli and polymer network growth. Building off this work, recent studies have shown that mechanical stimulation can induce mineralization in a system that contains thiadiazole thiol (McMT) and ZnO piezoparticles.[38] The force-triggered mineralization can transform the spherical nanoscale ZnO into micro-level $Zn(McMT)_n$ crystalline rods (Figure 4b). The morphology of these microrods (length and size) is significantly influenced by the intensity and duration of mechanical stimulation (frequency and duration) and the surrounding reaction environment such as solvent or synthetic matrix. When the mixture of ZnO and McMT is reacted in a gel, the rod can be mineralized *in situ* allowing the viscoelasticity of the overall polymer network to be tuned according to the mechanical input. In other words, the adaptivity of the polymer composite was directed by the mineralization and growth of $Zn(McMT)_n$. These force-triggered adaptable features of composites were further confirmed by dynamic mechanical analysis, showing the potential of mimicking the biomineralization process, such as the growth of bone, in biological tissues.

Challenges of *in situ* mineralization

While the previous examples show the potential of translating mechanical input information toward mechanical adaptability, several challenges remain. One major challenge is to grow the mineral component in a more spatially and directionally controlled manner. As many bone/skeleton structures formed in a genetic coded anisotropic and hierarchical way, mechanically induced *in situ* mineralization currently lacks an effective mechanism for directing formation and growth within specific spatial regions. To tackle this challenge, a mechanically targeted approach would be advantageous. For instance, high-intensity focused ultrasound (HIFU)[39,40] could offer valuable insights into addressing this issue. Another major challenge is to develop a reversible mineralization process, akin to some biomineralization process that allows the replacement or growth of mineral structures. As the current material studies only enable a one-way adaptation process, it is not easy to adapt with a more dynamically changing environment. To

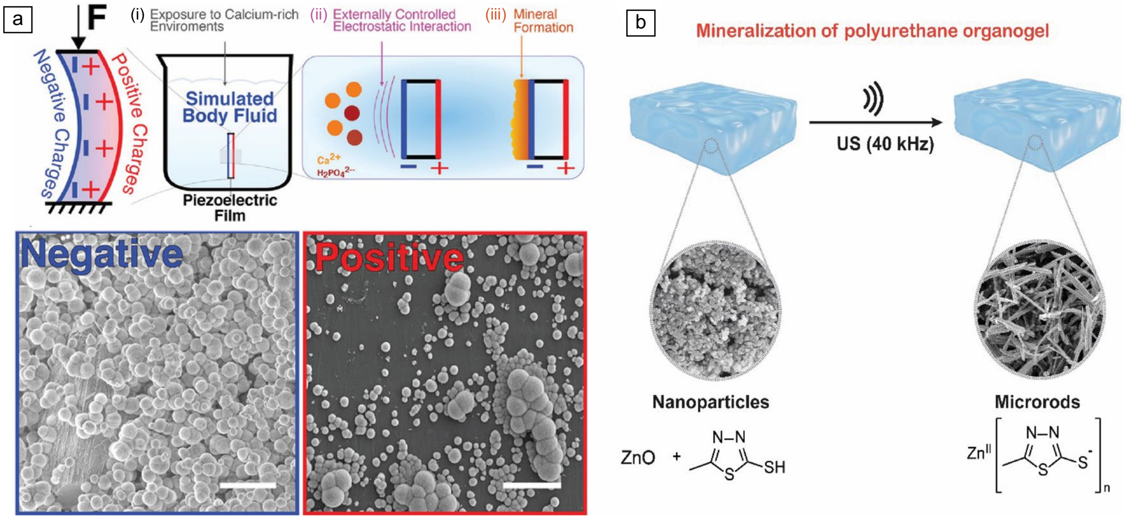

Figure 4. Mechanical force-induced mineralization. (a) Mineralization process induced from the surface of a bent piezoelectric (PVDF) film. Hydroxyapatite mineralization is preferentially grown on the negatively charged side of the PVDF. Scale bars are 10 μm. Reprinted with permission from Reference 9. © 2020 Wiley. (b) *In situ* mineralization of Zn(McMT)$_n$ compound that is grown within a polyurethane matrix. Upon continuous ultrasound stimulation, the morphology of nanoparticles switched from nanospheres to long micron-sized rods. Reprinted with permission from Reference 38. © 2023 Springer Nature.

achieve fully reversible features, the development of new chemistry as well as mineralization strategies is necessary. This development of creating reversible processes will allow continuous mechanical adaptation over a larger time scale.

To advance the state of adaptive materials and further enable a higher level of mechanical response such as information encoding and storage in future works, an adaptive material system would require a hierarchical design and multiresponsive mechanism(s), integrating insights from chemistry, materials science, and mechanical engineering. For example, by harnessing the dynamically reactive nature of thiol and disulfide chemistry, it becomes possible to develop a novel category of mechanically adaptive materials incorporating a mechanical transducer. Consequently, these materials could establish a direct connection between mechanical response at molecular level, chemical interactions, and electrochemical reactions, while possessing adaptable multifunctionality. This innovation would broaden the scope of metamaterials, inspiring applications across various fields, including soft robotics and biomedical devices, among others.

Dynamic LCE networks: Adaptivity from mesogenic realignment

Liquid-crystal elastomers are lightly cross-linked polymer networks that contain mesogenic units and as such they exhibit the entropic elasticity of an elastomer combined with the anisotropy of small molecule liquid crystals (**Figure 5**a). One key property of LCEs is their "soft elasticity," namely, a large stress plateau where the application of strain results in little-to-no increase in stress (Figure 5b) as the additional mechanical load leads to reorientation of the mesogens. As such, polydomain LCEs can dissipate substantial amounts of energy by undergoing a transition to an anisotropic monodomain state under uniaxial mechanical load.[40–43] The mechanism of energy dissipation in LCEs is dependent on temperature as well as loading conditions, such as uniaxial versus high and low strain cyclic loading. The tunability of energy dissipation in LCEs makes them highly suitable to adapt to environmental conditions where the intensity of mechanical stress as well as other stimuli is generally nonmonotonic. The inherent anisotropy of the mesogens provides LCEs with a host of thermally triggerable mechano-optical properties that stem from the alignment and stability of the domains within the network.[44] In addition, LCEs also exhibit reversible actuation (Figure 5c–d) (upon transition from the LC to isotropic state) resulting in large dimensional deformation, which makes them highly suitable in self-adaptive applications such as soft robotics and artificial muscle systems.

The characteristic features of LCEs such as soft elasticity and reversible actuation are highly desirable for mechanical adaptation; however, a network comprising irreversible covalent cross-links can weaken and damage over multiple use cycles. Therefore, dynamic bonds, which can controllably break and reform, have been incorporated into LCEs to further improve their mechanical adaptability (Figure 5a).[15,45,46] Dynamic liquid-crystal elastomers (DLCEs) are stimuli-responsive networks characterized by the inclusion of both domain-forming mesogenic units as well as dynamic covalent bonds,[44] which are well suited as

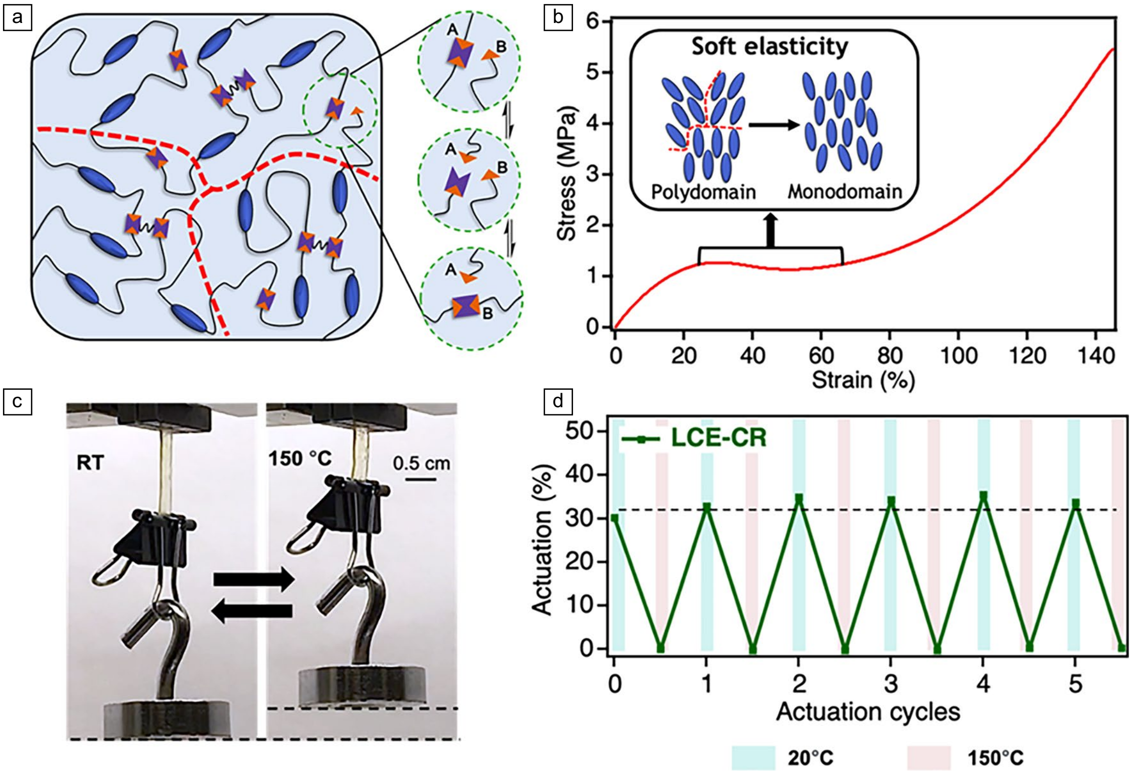

Figure 5. Characteristic features of dynamic liquid-crystal elastomer (DLCE) networks. (a) Schematic of polydomain dynamic LCE, (b) tensile curve of DLCE showing soft elasticity during polydomain to monodomain transition, (c) images of an LCE during actuation between room temperature (RT) and 150°C, (d) multiple thermal actuation cycles of an LCE with the catalyst removed (CR). Reprinted with permission from Reference 45. © 2024 American Chemical Society.

mechanically adaptive materials. In addition to the properties exhibited by covalently cross-linked LCEs, the addition of dynamic bonds provides for bulk reprocessability as well as the ability to lock in selected alignment configurations through stimuli-induced network reorganization. A variety of factors influence the impact a dynamic bond will have on the bulk network's properties that include the nature of the chemistry, the concentration of the bond, and the placement of the bond within the network.[15] Many DLCEs have been studied with a host of different chemistries and mesogen types being explored producing materials with a vast array of thermomechanical properties that are compatible with a variety of stimuli.[47,48] Given the highly tunable nature of both of these moieties, DLCEs have been used in a wide variety of applications, including soft robotics,[49,50] smart adhesives,[51,52] and tunable optics.[53,54]

Dynamic LCE networks: Challenges and perspectives

Many of the dynamic chemistries employed in DLCEs undergo exchange under elevated temperatures and/or use an associative exchange process both of which can provide improved mechanical robustness at elevated operating temperatures. However, this can result in a tradeoff where more extreme processing conditions are required to reset the material or reprogram it into a new configuration, which is particularly important when considering designing materials capable of mechanical adaptation.[45] As such, a future direction is the development of DLCEs whose constituent polymers and mesogens can be efficiently post-synthetically reoriented under mild processing conditions. One avenue exploring this concept is the incorporation of more labile dynamic dissociative linkages into DLCEs, such as hydrogen bonds. For example, work by Lugger et al. explored the synthesis and processing of linear LC polymers cross-linked with hydrogen bonds formed by thiourethane moieties incorporated into the polymer backbone.[55] With these more labile linkages forming a supramolecular network assembly, the supramolecular LCEs are much more processable than their more robust covalent counterparts and possess the ability to be 3D printed in an aligned state via a direct ink writing process. This material system provides a useful platform as this 3D printing technique is often inaccessible to most covalently cross-linked DLCE materials due to their high viscosities and lack of terminal flow even at high temperatures. These supramolecular LCEs provide a class of materials with the potential to be post-synthetically reprocessed in a manner that allows for the configuration of mesogens to be more readily defined enabling a multilevel material adaptation system.

The prospect of creating mechanically adaptive DLCEs has proven challenging on multiple fronts. One such challenge is the lack of precise spatial control over the rearrangement of mesogens once they have been polymerized into an LCE network. Many groups have studied LCEs as sensors and information storage materials; however, most of these materials are limited to single configurations that are locked in once cross-linking has occurred. For example, work by Ware et al. demonstrated the utility of alignment layers controlled by electric fields for imprinting images into LCEs;[56] however, these materials could only be encoded with information a single time. A more ideal approach for programming in LCE materials would involve the ability to alter information post-synthetically within an existing bulk material. As the high viscosities associated with DLCEs often hinder reprocessing efforts, this would likely necessitate the further development and study of LCEs capable of a post-synthetic reversible viscosity reduction such that mesogens can be precisely aligned via an external field (mechanical, electric, or chemical) as a means of encoding and rewriting the information stored in the LCE *in situ*.

Another aspect of this challenge is the quantity of information that can be stored within a single material. Although many soft information storage materials utilize synthetic backbone sequencing as a means of encoding simple information into a polymer, these methods fall short in that they often rely on a binary encoding structure which limits the complexity of the information stored. One approach that has been explored in the realm of storing images in LCEs is the incorporation of photonic dopants, often in the form of cholesteric mesogens, within the LCE. Work by Zhang et al. explored cholesteric LCE bilayers that were able to store and subsequently display printed images under the addition of a mechanical stress.[57] The ability to store information pictorially whether it be in the form of strings of characters or entire images also allows for the encoding of complex information into a single piece of material without the need for complex decoding procedures. To further expand the utility of these systems, fabrication of layered materials consisting of greater numbers of photonic layers tuned to specific strain values would allow for more complex mechanically adaptive responses to a wide array of stress inputs within a single material.

An additional challenge involves developing DLCEs with higher order complex actuation. In addition to their ability to passively adapt to the external mechanical stress, DLCEs can also be conditioned to utilize the stress to achieve *in situ* alignment. By gradually tuning the alignment of mesogens from a polydomain toward a monodomain state under varying degrees and type (e.g., compression versus tension) of mechanical load and dynamic bond stimuli (heat, light, pH, etc.), a range of reversible anisotropic elastic responses can be achieved toward complex actuation. In tension, a polydomain LCE is soft and stretchable in all directions, whereas a monodomain LCE is only strong and stiff in the direction parallel to the alignment of mesogens. The stress placed into the system during the mesogen alignment can then be potentially utilized to accelerate dynamic bond exchange and could result in two-way shape-memory properties responsible for reversible actuation.[58] Hence, under different conditioning protocols, the alignment of mesogens and therefore the actuation behavior of dynamic LCEs could be spatially as well as temporally controlled. For example, LCEs cross-linked in the presence of light-responsive dynamic bonds could be selectively triggered under different magnitudes of mechanical stress during programming to achieve complex actuation behavior. In addition, by incorporating mechanoresponsive dynamic bonds in LCEs, the alignment of mesogens and dynamic bond exchange could be simultaneously achieved while under stress. The mechanically strained regions with aligned mesogens can undergo stress relaxation via mechanoreduction as the *in situ* alignment progresses. Furthermore, by incorporating cholesteric liquid crystals and/or mechanochromic dynamic bonds, the effect of mechanical stress could also be locally visualized during conditioning[59,60] (**Figure 6**).

Future perspectives

Three mechanistic approaches, piezo-initiated polymerization, mineralization and mesogenic realignment, have been outlined in this article for mechanically triggered adaptative materials. These approaches correspond to the fundamental pathways of material adaptation: covalent, noncovalent, and inorganic crystallization. However, several challenges remain to be addressed for future works and studies as it is still not possible to recapitulate all of the behavior observed in biological adaptive systems. For example, muscle tissues keep growing and become stronger to adapt with physical labor works. In a more advanced adaptation system, T-cells can transmit piconewton force to antigens with its ligand,[63,64] and it is widely believed that T-cell activation as well as some other immunization process is fine-tuned by cellular mechanotransduction.[64–67] Such mechanoadaptive behaviors have simultaneously included the process of sensing, tuning, communicating, responding, and even memorizing, which is far beyond the capabilities that could be accomplished by current adaptive material systems. To build the “mansion” of mechanically triggered adaptive systems with three fundamental “building blocks” (PIP, mineralization, and mesogenic realignment), it is desirable for future research works to focus on the following aspects: (1) Materials or devices with higher order of adaptation. Given the fact that biosystems have developed thousands of reaction cascades as well as biological pathways for adaptation purposes, mature artificial adaptive systems in future could require a bottom-up design for a more complex material system. (2) Communication and feedback mechanism. The communication between different immune cells, such as B-cell, T-cell, and macrophages, plays an important role

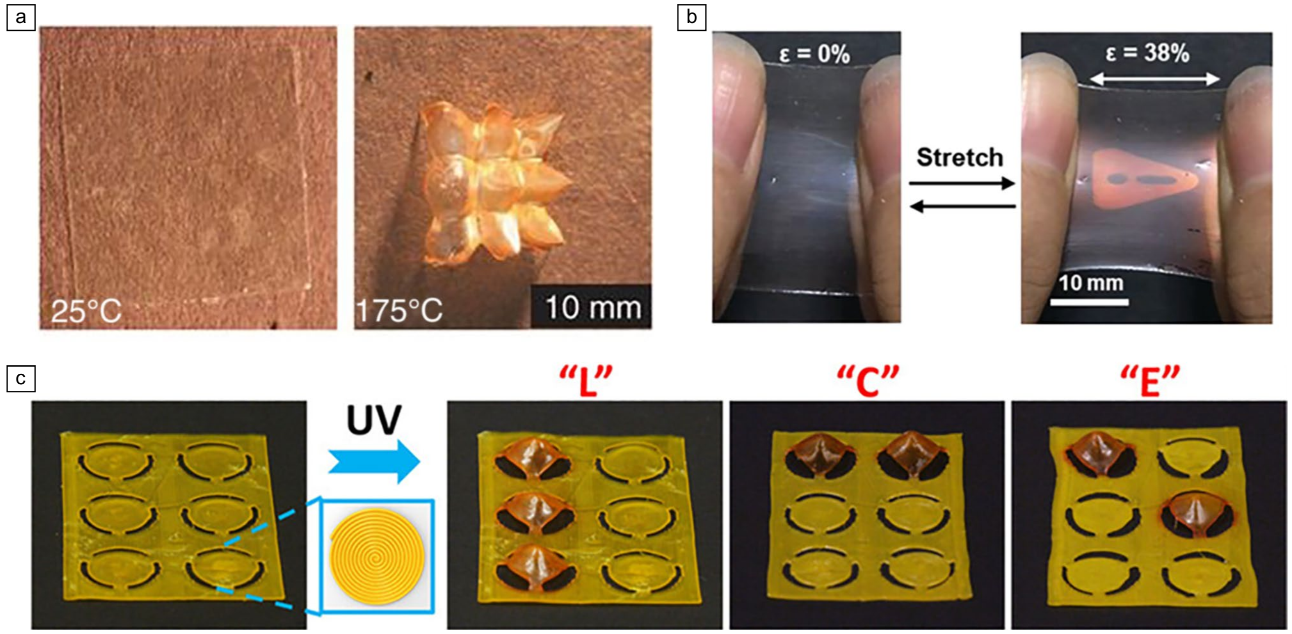

Figure 6. Examples of liquid-crystal elastomers (LCEs) used for complex actuation and information storage. (a) LCE transforming between flat film and complex shape with cones during actuation. Reprinted with permission from Reference 56. © 2015 AAAS. (b) LCE showing imprinted pattern while stretching. Reprinted with permission from Reference 61. © 2020 Wiley. (c) Reconfigurable Braille-like LCE actuators. Reprinted with permission from Reference 62. © 2021 Wiley.

for adaptative immunization. Similarly, an effective communication mechanism could allow the material to fine-tune its adaptation behavior. (3) Long-term memorial adaptation behavior. The unique nature of adaptive immunization as well as genetic coding in the biology world is that it keeps memorial information for future adaptation. Thus, enhancing the adaptive behavior of a material could involve introducing trainable mechanisms that allow the material to memorize specific mechanical inputs. These potential advancements promise to usher a new era of mechanically triggered adaptive materials that are more versatile and highly functional across a wide range of critical applications.

Author contributions

All authors contributed to the conception, discussion, writing, as well as editing of the text, references, and figures.

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Data availability

Not applicable.

Competing interests

The authors declare no competing interests.

References

1. A. Walther, *Adv. Mater.* **32**, 1905111 (2020)
2. R. Merindol, A. Walther, *Chem. Soc. Rev.* **46**, 5588 (2017)
3. Z. Wang, J. Wang, J. Ayarza, T. Steeves, Z. Hu, S. Manna, A.P. Esser-Kahn, *Nat. Mater.* **20**, 869 (2021)
4. L. Hsu, C. Weder, S.J. Rowan, *J. Mater. Chem.* **21**, 2812 (2011)
5. J. Yang, X. Zhang, X. Zhang, L. Wang, W. Feng, Q. Li, *Adv. Mater.* **33**, 2004754 (2021)
6. X. Fan, J.Y. Chung, Y.X. Lim, Z. Li, X.J. Loh, *ACS Appl. Mater. Interfaces* **8**, 33351 (2016)
7. L.R. Arnaut, *IEEE Trans. Antennas Propag.* **51**, 1530 (2003)
8. P. Cai, C. Wang, H. Gao, X. Chen, *Adv. Mater.* **33**, 2007977 (2021)
9. S. Orrego, Z. Chen, U. Krekora, D. Hou, S.Y. Jeon, M. Pittman, C. Montoya, Y. Chen, S.H. Kang, *Adv. Mater.* **32**, 1906970 (2020)
10. E. Brown, N.A. Forman, C.S. Orellana, H. Zhang, B.W. Maynor, D.E. Betts, J.M. DeSimone, H.M. Jaeger, *Nat. Mater.* **9**, 220 (2010)
11. T. Rajasooriya, H. Ogasawara, Y. Dong, J.N. Mancuso, K. Salaita, *Adv. Mater.* **35**, 2305544 (2023)
12. T. Matsuda, R. Kawakami, R. Namba, T. Nakajima, J.P. Gong, *Science* **363**, 504 (2019)
13. S. Lin, J. Liu, X. Liu, X. Zhao, *Proc. Natl. Acad. Sci. U.S.A.* **116**, 10244 (2019)
14. Z. Wang, J. Wu, X. Wei, S. Saleemi, W. Liu, W. Li, I. Marriam, F. Xu, *J. Mater. Chem. C* **8**, 117 (2020)
15. C.A. Lindberg, E. Ghimire, C. Chen, S. Lee, N.D. Dolinski, J.M. Dennis, S. Wang, J.J. de Pablo, S.J. Rowan, *J. Polym. Sci.* **62**, 907 (2024)
16. J. Ma, Y. Yang, C. Valenzuela, X. Zhang, L. Wang, W. Feng, *Angew. Chem. Int. Ed.* **61**, e202116219 (2022)

17. S.-M. Lee, E. Pippel, U. Gösele, C. Dresbach, Y. Qin, C.V. Chandran, T. Bräuniger, G. Hause, M. Knez, *Science* **324**, 488 (2009)
18. Y. Lin, M.H. Barbee, C.-C. Chang, S.L. Craig, *J. Am. Chem. Soc.* **140**, 15969 (2018)
19. Y. Peng, Y. Hou, Q. Wu, Q. Ran, G. Huang, J. Wu, *Mater. Horiz.* **8**, 2553 (2021)
20. F. Ling, Z. Liu, M. Chen, H. Wang, Y. Zhu, C. Ma, J. Wu, G. Huang, *J. Mater. Chem. A* **7**, 25324 (2019)
21. C. Wang, K. Sim, J. Chen, H. Kim, Z. Rao, Y. Li, W. Chen, J. Song, R. Verduzco, C. Yu, *Adv. Mater.* **30**, 1706695 (2018)
22. M. Jorfi, M.N. Roberts, E.J. Foster, C. Weder, *ACS Appl. Mater. Interfaces* **5**, 1517 (2013)
23. L.C. Zhao, H.X. Zou, K.X. Wei, S.X. Zhou, G. Meng, W.M. Zhang, *Adv. Energy Mater.* **13**, 2300557 (2023)
24. J. Tabor, K. Chatterjee, T.K. Ghosh, *Adv. Mater. Technol.* **5**, 1901155 (2020)
25. T. Friščić, C. Mottillo, H.M. Titi, *Angew. Chem.* **132**, 1030 (2020)
26. L. Montero de Espinosa, W. Meesorn, D. Moatsou, C. Weder, *Chem. Rev.* **117**, 12851 (2017)
27. H. Mohapatra, M. Kleiman, A.P. Esser-Kahn, *Nat. Chem.* **9**, 135 (2017)
28. Z. Wang, J. Ayarza, A.P. Esser-Kahn, *Angew. Chem.* **131**, 12151 (2019)
29. C. Ding, Y. Yan, Y. Peng, D. Wu, H. Shen, J. Zhang, Z. Wang, Z. Zhang, *Macromolecules* **55**, 4056 (2022)
30. H. Feng, X. Shao, Z. Wang, *ChemPlusChem* e202400287 (2024). https://doi.org/10.1002/cplu.202400287
31. M.D. Nothling, J.E. Daniels, Y. Vo, I. Johan, M.H. Stenzel, *Angew. Chem.* **135**, e202218955 (2023)
32. M. Zhou, Y. Zhang, G. Shi, Y. He, Z. Cui, X. Zhang, P. Fu, M. Liu, X. Qiao, X. Pang, *ACS Macro Lett.* **12**, 26 (2022)
33. Y. Zhang, J. Zhang, S. Xu, G. Shi, Y. He, X. Qiao, X. Pang, *Macromol. Rapid Commun.* **45**(17), 2400235 (2024). https://doi.org/10.1002/marc.202400235
34. C. Hold, S. Billerbeck, S. Panke, *Nat. Commun.* **7**, 12971 (2016)
35. A. Llopis-Lorente, P. Díez, A. Sánchez, M.D. Marcos, F. Sancenón, P. Martínez-Ruiz, R. Villalonga, R. Martínez-Máñez, *Nat. Commun.* **8**, 15511 (2017)
36. P.U. Gilbert, K.D. Bergmann, N. Boekelheide, S. Tambutté, T. Mass, F. Marin, J.F. Adkins, J. Erez, B. Gilbert, V. Knutson, *Sci. Adv.* **8**, eabl9653 (2022)
37. J. Ayarza, Z. Wang, J. Wang, A.P. Esser-Kahn, *ACS Macro Lett.* **10**, 799 (2021)
38. J. Ayarza, J. Wang, H. Kim, P.-R. Huang, B. Cassaidy, G. Yan, C. Liu, H.M. Jaeger, S.J. Rowan, A.P. Esser-Kahn, *Nat. Commun.* **14**, 8319 (2023)
39. S. Honda, M. Oka, K. Fuke, P.T. Khuri-Yakub, C.N. Pai, *Adv. Mater.* **35**, 2304104 (2023)
40. X. Song, Q. Zhang, M. Chang, L. Ding, H. Huang, W. Feng, T. Xu, Y. Chen, *Adv. Mater.* **35**, 2212259 (2023)
41. T.J. White, D.J. Broer, *Nat. Mater.* **14**, 1087 (2015)
42. D.R. Merkel, R.K. Shaha, C.M. Yakacki, C.P. Frick, *Polymer* **166**, 148 (2019)
43. N.A. Traugutt, D. Mistry, C. Luo, K. Yu, Q. Ge, C.M. Yakacki, *Adv. Mater.* **32**, 2000797 (2020)
44. K.M. Herbert, H.E. Fowler, J.M. McCracken, K.R. Schlafmann, J.A. Koch, T.J. White, *Nat. Rev. Mater.* **7**, 23 (2022)
45. E. Ghimire, C.A. Lindberg, T.D. Jorgenson, C. Chen, J.J. de Pablo, N.D. Dolinski, S.J. Rowan, *Macromolecules* **57**(2), 682 (2024)
46. S.J. Rowan, S.J. Cantrill, G.R. Cousins, J.K. Sanders, J.F. Stoddart, *Angew. Chem. Int. Ed.* **41**, 898 (2002)
47. M.O. Saed, A. Gablier, E.M. Terentjev, *Chem. Rev.* **122**, 4927 (2021)
48. C. Valenzuela, Y. Chen, L. Wang, W. Feng, *Chemistry* **28**, e202201957 (2022)
49. X. Zhou, G. Chen, B. Jin, H. Feng, Z. Chen, M. Fang, B. Yang, R. Xiao, T. Xie, N. Zheng, *Adv. Sci.* **11**(23), e2402358-59 (2024)
50. Q. He, Z. Wang, Y. Wang, A. Minori, M.T. Tolley, S. Cai, *Sci. Adv.* **5**, eaax5746 (2019)
51. T. Ohzono, Y. Norikane, M.O. Saed, E.M. Terentjev, *ACS Appl. Mater. Interfaces* **12**, 31992 (2020)
52. Y. Wu, B.D. Clarke, K.M. Liechti, Z.A. Page, *Chem. Mater.* **36**(16), 8066 (2024). https://doi.org/10.1021/acs.chemmater.4c01612
53. M.T. Brannum, A.M. Steele, M.C. Venetos, L.T.J. Korley, G.E. Wnek, T.J. White, *Adv. Opt. Mater.* **7**, 1801683 (2019)
54. E.J. Cooper, M. Reynolds, T. Raistrick, S.R. Berrow, E.I.L. Jull, V. Reshetnyak, D. Mistry, H.F. Gleeson, *Macromolecules* **57**, 2030 (2024)
55. S.J. Lugger, R.M. Verbroekken, D.J. Mulder, A.P. Schenning, *ACS Macro Lett.* **11**, 935 (2022)
56. T.H. Ware, M.E. McConney, J.J. Wie, V.P. Tondiglia, T.J. White, *Science* **347**, 982 (2015)
57. P. Zhang, M.G. Debije, L.T. de Haan, A.P. Schenning, *ACS Appl. Mater. Interfaces* **14**, 20093 (2022)
58. E.C. Davidson, A. Kotikian, S. Li, J. Aizenberg, J.A. Lewis, *Adv. Mater.* **32**, 1905682 (2020)
59. I. Azcune, I. Odriozola, *Eur. Polym. J.* **84**, 147 (2016)
60. Y. Geng, R. Kizhakidathazhath, J.P. Lagerwall, *Nat. Mater.* **21**, 1441 (2022)
61. P. Zhang, X. Shi, A.P. Schenning, G. Zhou, L.T. de Haan, *Adv. Mater. Interfaces* **7**, 1901878 (2020)
62. X. Lu, C.P. Ambulo, S. Wang, L.K. Rivera-Tarazona, H. Kim, K. Searles, T.H. Ware, *Angew. Chem. Int. Ed.* **60**, 5536 (2021)
63. Y. Liu, L. Blanchfield, V.P.-Y. Ma, R. Andargachew, K. Galior, Z. Liu, B. Evavold, K. Salaita, *Proc. Natl. Acad. Sci. U.S.A.* **113**, 5610 (2016)
64. V.P.-Y. Ma, Y. Hu, A.V. Kellner, J.M. Brockman, A. Velusamy, A.T. Blanchard, B.D. Evavold, R. Alon, K. Salaita, *Sci. Adv.* **8**, eabg485 (2022)
65. M.S. Wang, Y. Hu, E.E. Sanchez, X. Xie, N.H. Roy, M. de Jesus, B.Y. Winer, E.A. Zale, W. Jin, C. Sachar, *Nat. Commun.* **13**, 3222 (2022)
66. A.V. Kellner, R. Hunter, P. Do, M. Lee, J. Hamilton, R. Ma, A. Ross, C. Porter, E. Dreaden, C. Henry, *Biophys. J.* **121**, 314a (2022)
67. J. Rogers, R. Ma, A. Foote, Y. Hu, K. Salaita, *J. Am. Chem. Soc.* **146**, 7233 (2024)

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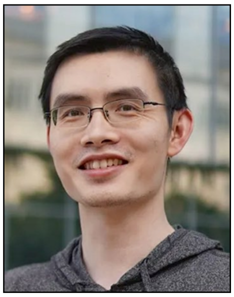
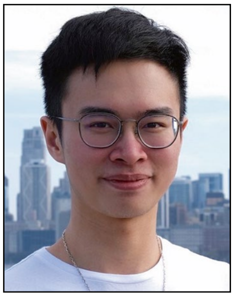


Yixiao Dong has been a postdoctoral researcher in the Pritzker School of Molecular Engineering at The University of Chicago since 2022. He received his BS and MS degrees in polymer materials from Wuhan University of Technology, China. He then received his PhD degree in chemistry from Emory University in 2022. His research spans polymer chemistry, piezochemistry, soft material, and functional nanocomposite. His current work focuses on piezo/inverse piezo effect initiated polymerization. Dong can be reached by email at ydong2@uchicago.edu.



Pin-Ruei Huang has been a doctoral candidate in the Pritzker School of Molecular Engineering at The University of Chicago since 2022. He earned his bachelor's degree in chemistry at the University of Washington—Seattle in 2022, where he studied the synthesis of cyclic polymers and their corresponding organometallic initiators. His current research focuses on piezoelectric composites with dynamic bonds. Huang can be reached by email at huangpr@uchicago.edu.

Elina Ghimire is a doctoral candidate in the Pritzker School of Molecular Engineering at The University of Chicago. She received her bachelor's degree in polymer science from The University of Southern Mississippi (USM) in 2020. At USM, she studied the assembly behavior of block copolymers in the formation of polymer vesicles. Her current research interests include dynamic polymer networks, liquid-crystalline elastomers, and adaptable and trainable materials. Ghimire can be reached by email at elinaghimire@uchicago.edu.

Charlie A. Lindberg is a doctoral candidate at The University of Chicago. She received her BS degree in chemistry with a materials science emphasis from the University of Wisconsin–Eau Claire in 2017. Later that fall, she began her graduate work at what is now the Pritzker School of Molecular Engineering at The University of Chicago. Her current research focuses on the design and development of novel dynamic liquid-crystal elastomer materials. Lindberg can be reached by email at lindbeca@uchicago.edu.

Farsa Ram is an assistant professor in energy research at the Indian Institute of Science (IISc), Bangalore, India. He received his PhD degree in polymer science and engineering (chemical science) from the Academy of Scientific and Innovative Research and CSIR-National Chemical Laboratory, India. After, he completed postdoctoral research at KTH Royal Institute of Technology, Sweden, and The University of Chicago. In 2024, he returned to India to start his research group at IISc. His research has focused on developing functional materials for energy applications while emphasizing sustainability, using renewable and bio-based feedstocks, and energy-efficient processes. Within this umbrella, his research deals with piezo-initiated polymerization, mechanical adaptive materials, polymer composites, polymer electrolyte membranes, radiative cooling coatings, and ambient energy harvesting (mechanical and hydrovoltaic). Ram can be reached by email at ramf@iisc.ac.in.

Stuart J. Rowan is the Barry L. MacLean Professor of Molecular Engineering and professor of chemistry at The University of Chicago. He received his BSc degree in chemistry in 1991 from the University of Glasgow, UK, where he completed graduate school and received his PhD degree in 1995. In 1994, he moved to the Department of Chemistry at The University of Cambridge, UK. He continued his postdoctoral studies at the University of California, Los Angeles, in 1998. In 1999, he was appointed as an assistant professor in the Department of Macromolecular Science and Engineering at Case Western Reserve University, was promoted to associate professor in 2005, and became a full professor in 2008. In 2016, he joined the Institute for Molecular Engineering (now the Pritzker School of Molecular Engineering) and the Department of Chemistry at The University of Chicago. He also has a staff appointment in the Chemical and Engineering Science (CSE) Division at Argonne National Laboratory. The current interests of his group span responsive polymers, sustainable materials, polymers for energy, biomaterials, and new polymer synthesis. Specifically, his group works on supramolecular polymers, dynamic covalent polymers, self-healing materials, responsive adhesives, metal-containing polymers, gels, biomaterials, cellulose nanocrystal/nanocellulose, and interlocked polymeric architectures. Rowan can be reached by email at stuartrowan@uchicago.edu.

Aaron Esser-Kahn is a professor at The University of Chicago. He received his BS degree from the California Institute of Technology in 2004, and his PhD degree from the University of California, Berkeley, in 2009. He later completed his postdoctoral research at the University of Illinois at Urbana-Champaign. He began his independent research career as an assistant professor and then associate professor at the University of California, Irvine. In 2017, he moved the research group to The University of Chicago to build on the areas of immunoengineering and piezo-synthesized materials. Esser-Kahn can be reached by email at aesserkahn@uchicago.edu.