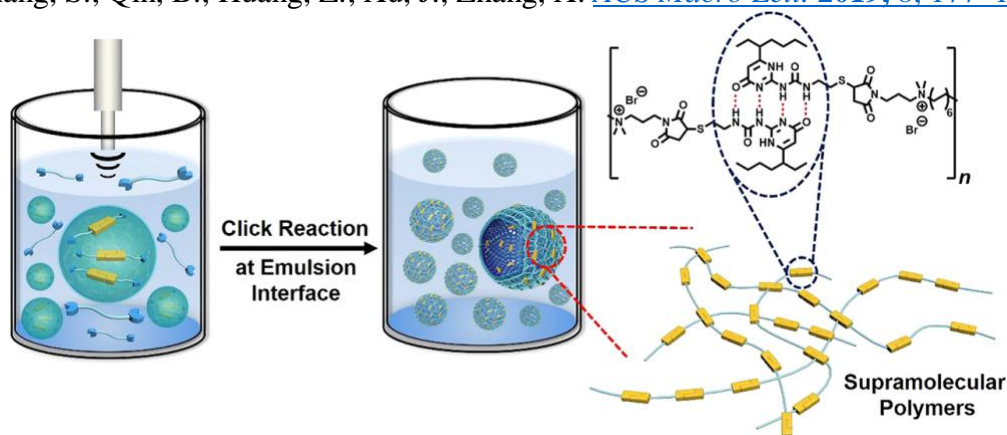


## Supramolecular Emulsion Interfacial Polymerization

Zhang, S.; Qin, B.; Huang, Z.; Xu, J.; Zhang, X. *ACS Macro Lett.* **2019**, *8*, 177–182.

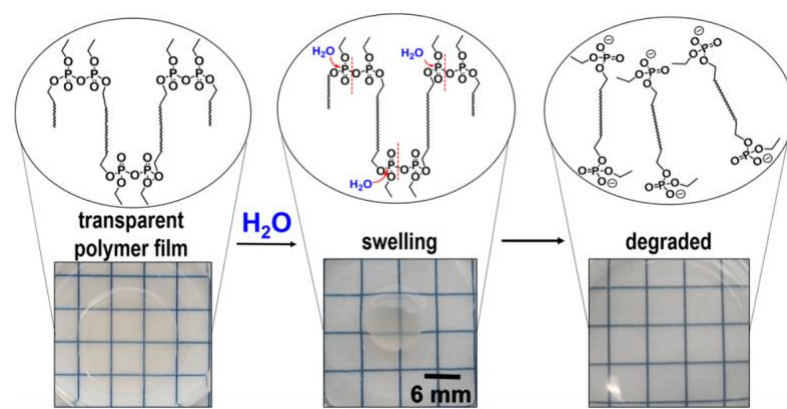


A new method of supramolecular emulsion interfacial polymerization is developed, which can be used to fabricate supramolecular polymeric nanospheres. We designed a water-soluble monomer containing two maleimide end groups, acting as both building block and surfactant, and an oil-soluble supramonomer bearing two thiol groups connected by quadruple hydrogen bonds. With the assist of ultrasonication, hollow nanospheres can be controllably prepared by thiol-maleimide reaction of two monomers at the emulsion interface, which exhibit good stability and dynamic property. In addition, the encapsulated guest molecules could be controllably released from the supramolecular polymeric nanospheres, owing to their stimuli-responsiveness. It is anticipated that this approach will enrich the methodology of supramolecular polymerization and can be applied to constructing supramolecular materials with controllable structures and functions.

**Comment:** In this paper the authors demonstrated that supramolecular emulsion interfacial polymerization is a simple and controllable method to prepare supramolecular polymeric nanospheres, which are component-defined, highly stable and degradable.

## Aliphatic Long-Chain Polypyrophosphates as Biodegradable Polyethylene Mimics

Tee, H. T.; Lieberwirth, I.; Wurm, F. R.\* *Macromolecules* **2019**, *52*, 1166–1172.



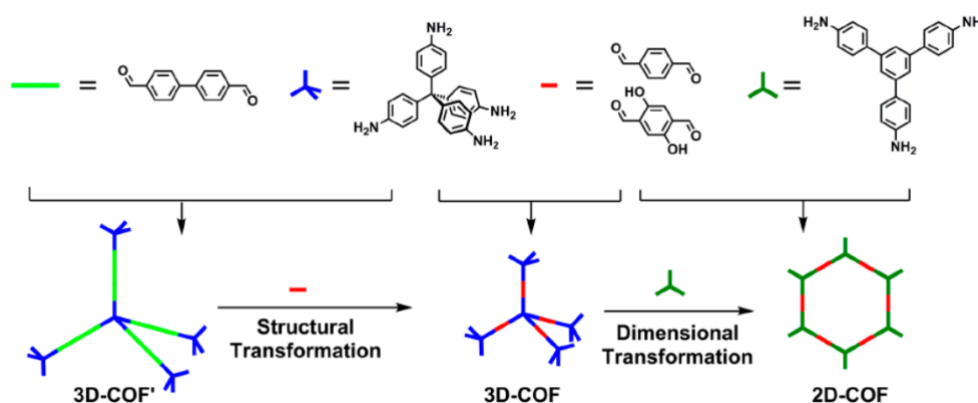
Biodegradable polyethylene mimics have been synthesized by the introduction of **pyrophosphate groups** into the polymer backbone, allowing not only hydrolysis of the backbone but also further degradation by microorganisms. Because of cost, low weight, and good mechanical

properties, the use of polyolefins has increased significantly in the past decades and has created many challenges in terms of disposal and their environmental impact. The durability and resistance to degradation make polyethylene difficult or impossible for nature to assimilate, thus making the degradability of polyolefins an essential topic of research. The biodegradable polypyrophosphate was prepared via acyclic diene metathesis polymerization of a diene monomer. The monomer is accessible via a three-step synthesis, in which the pyrophosphate was formed in the last step by DCC coupling of two phosphoric acid derivatives. **This is the first report of a pyrophosphate group localized in an organic polymer backbone.** The polypyrophosphate was characterized in detail by NMR spectroscopy, size exclusion chromatography, FTIR spectroscopy, differential scanning calorimetry, and thermogravimetry. X-ray diffraction was used to compare the crystallization structure in comparison to analogous polyphosphates showing poly(ethylene)-like structures. In spite of their hydrophobicity and water insolubility, **the pyrophosphate groups exhibited fast hydrolysis, resulting in polymer degradation when films were immersed in water.** Additionally, the hydrolyzed fragments were further biodegraded by microorganisms, rendering these PE mimics potential candidates for fast release of hydrophobic cargo, for example, in drug delivery applications.

**Comment:** I guess that we are all aware of the rising need for biodegradable commodity polymers. In this paper, as you can see in the abstract, the authors use pyrophosphate groups that can easily hydrolyse. Something bothers me, however: the term “polyethylene mimics”. They compare their polymer to PE because of some crystalline features and good mechanical properties. PE is widely used for packaging applications, but, in my opinion, they cannot replace it with their easily-hydrolysable polymer; comparing the two sounds like greenwashing to me.

### Structural and Dimensional Transformations between Covalent Organic Frameworks via Linker Exchange

Li, Z.; Ding, X.\*; Feng, Y.\*; Feng, W.\*; Han, B.-H.\* [Macromolecules 2019, 52, 1257–1265.](#)



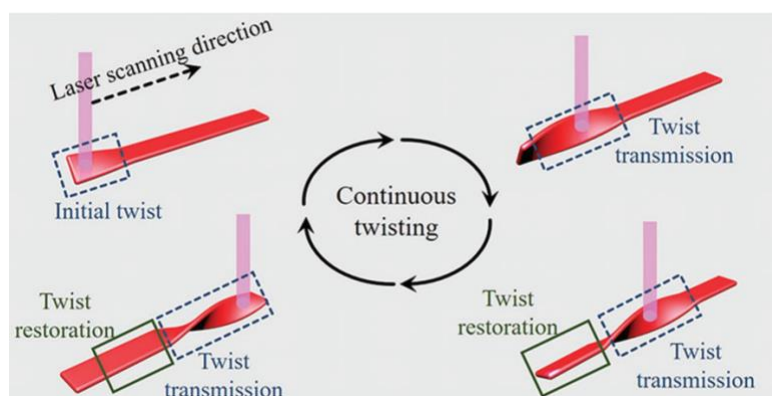
**Covalent organic frameworks (COFs)** are porous crystalline materials with well-controlled structures and extensive potential applications. However, construction of new COFs has tremendous challenges including design and synthesis of building block monomers, selection of reaction solvent systems, and investigation of reaction time and temperature. Here, we performed the **structural transformation** in the three-dimensional (3D) COFs (COF-300 and COF-320) and **dimensional transformation** from 3D (COF-301) to 2D (IPB-DHTP-COF) via the **dynamic covalent chemistry** principle, and the successful realization of these exchanges was confirmed by FT-IR,  $^1\text{H}$  NMR, PXRD, and nitrogen sorption isotherm measurements. Based on the results of PXRD patterns and  $^1\text{H}$  NMR spectra, the COF-320-to-COF-300 transformation was successfully achieved, and an efficiency as high as 89% was attained. A new network structure rather than COF-320 was obtained after transformation in the opposite direction (from COF-300 to COF-320),

which contains the two aldehyde building units biphenyl-4,4'-dicarbaldehyde (BPDA) and terephthalaldehyde (TA) in one framework. Meanwhile, the dimensional transformation of COFs with 78% exchange efficiency for COF-301-to-TPB-DHTP-COF is also achieved. These results not only reveal the COF-to-COF transformation in 3D-to-3D and 3D-to-2D but also provide a new way for constructing new COFs that is difficult to synthesize in one step.

**Comment:** Metal Organic Frameworks (MOFs) sound more familiar to me than COFs. It seems these materials have the same properties as their non-covalent counterpart, because they also have a highly porous structure. The authors showed that the framework can be tuned by using different building blocks linked with an imine bond. One major advantage of the COFs may be the better mechanical properties of the material compared to MOFs.

### Light-Driven Continuous Twist Movements of Microribbons

Zhang, Y.; Gong, Y.; Li, B.; Ma, R.-M.\*; Che, Y.\*; Zhao, J. *Small* **2019**, *15*, 1804102.

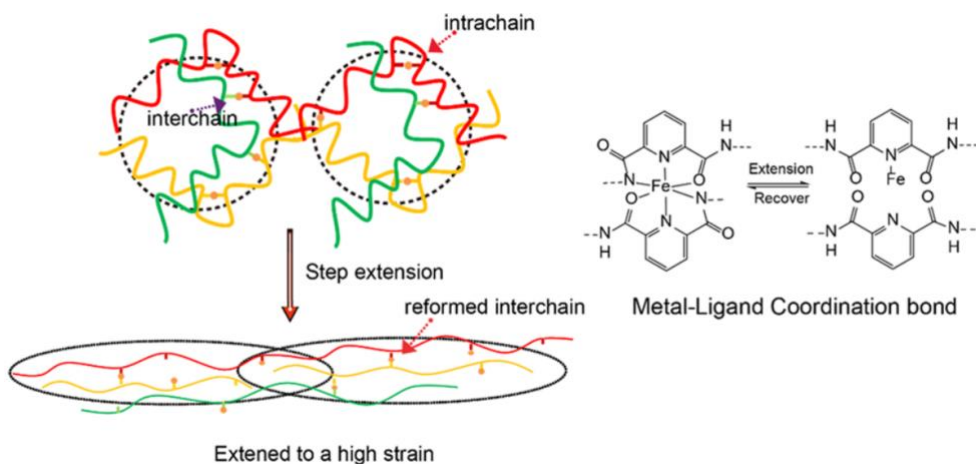


Despite many advances in the development of artificial systems with helical twist motions or deformations, obtaining materials that can undergo continuous twist movements upon an energy input remains a great challenge. In this work, a **continuous twist movement** of microribbons driven by **scanning laser irradiation**, a process that a twist generates initially at one end of the microribbon and is continuously transmitted to the other end and then kept twisting, is reported. Key factors to the achievement of this movement are the fabrication of elastic microribbons that possess relatively low elastic modulus and diagonal photoinduced  $\pi$ -stacking distortion relative to the microribbon long axis. Furthermore, the scanning laser irradiation is required to drive the  $\pi$ -stacking distortion with the spatiotemporal coordination for the continuous twist movement of microribbons. These findings may be extended to the achievement of other sophisticated continuous movements of microscale systems.

**Comment:** The examples of motion of objects containing perylene diimide (PDI) are not rare but achieving a twist movement is not trivial. In this paper, the authors scan the microribbon with light. It causes a local change in the  $\pi$ - $\pi$  stacking of the PDI and generates a twist of the material that can be propagated along with the scanning spot.

### Disassociation and Reformation Under Strain in Polymer with Dynamic Metal-Ligand Coordination Cross-Linking

Zhang, Q.; Zhu, X.; Li, C.-H.; Cai, Y.; Jia, X.; Bao, Z\*. *Macromolecules* **2019**, *52*, 660–668.

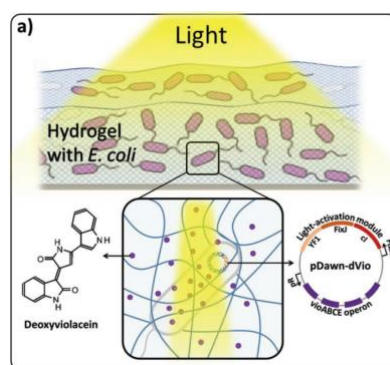


Dynamic bonding is a key factor for self-healing and stimuli-response polymeric materials. However, a detailed molecular-level understanding on effects from dynamic bond break and reform from interchain and intrachain cross-linking remains unclear. Here, we apply nonlinear rheological characterization on a polymer cross-linked with **dynamic coordination cross-linking**. Startup shear measurements and small amplitude oscillatory shear (SAOS) were used to confirm the existence of metal–ligand bond disassociation and reformation; Step extensional measurement was used to further characterize the dynamics of the interchain and intrachain cross-linkings’ disassociation and reformation. We found that dynamic bonds, which are shear rate sensitive, **could be disassociated by continuous shear**. When the shear or extension rate was much higher than the relaxation rate, both interchain and intrachain coordinated bondings may disassociate, and polymer chains became highly aligned; thus, more interchain reformation took place after extension cessation at higher strain since chain configurations have changed.

**Comment:** The authors investigated the effect of shear in networks crosslinked by dynamic bonds, both on the intra- and intermolecular reticulation points. This article could be interesting for those, for instance, who are trying to apply a force to such networks with photoactive molecules.

### Optoregulated Drug Release from an Engineered Living Material: Self-Replenishing Drug Depots for Long-Term, Light-Regulated Delivery

Sankaran, S.\*; Becker, J.; Wittmann, C.; del Campo, A.\* [Small 2019, 15, 1804717](https://doi.org/10.1002/smll.201904717).



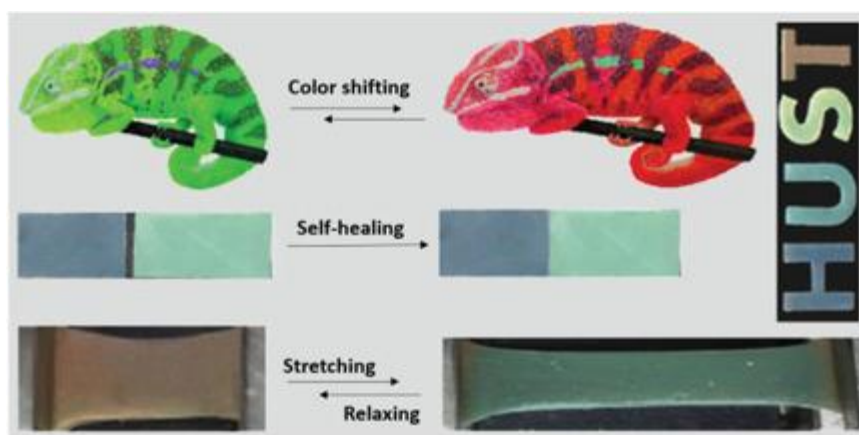
**On-demand and long-term delivery of drugs** are common requirements in many therapeutic applications, not easy to be solved with available smart polymers for drug encapsulation. This work presents a fundamentally different concept to address such scenarios using a self-replenishing and optogenetically controlled living material. **It consists of a hydrogel containing an active endotoxin-free *Escherichia coli* strain.** The bacteria are metabolically and optogenetically

engineered to secrete the antimicrobial and antitumoral drug deoxyviolacein in a light-regulated manner. The permeable hydrogel matrix sustains a viable and functional bacterial population and permits diffusion and delivery of the synthesized drug to the surrounding medium at quantities regulated by light dose. Using a focused light beam, the site for synthesis and delivery of the drug can be freely defined. The living material is shown to maintain considerable levels of drug production and release for at least 42 days. These results prove the potential and flexibility that living materials containing engineered bacteria can offer for advanced therapeutic applications.

**Comment:** Can you imagine, in the near future, wearing or having a material inside you, full of bacteria that will work for your health? I find this perspective equally fascinating as scaring: this is basically “controlled symbiosis”. This article is not a breakthrough in this field but I chose it to share the topic with you.

### Metallosupramolecular Photonic Elastomers with Self-Healing Capability and Angle-Independent Color

Tan, H.; Lyu, Q.; Xie, Z.; Li, M.; Wang, K.; Wang, K.; Xiong, B.; Zhang\*, L.; and Zhu,\* J. [Adv. Mater. 2019, 31, 1805496](#)

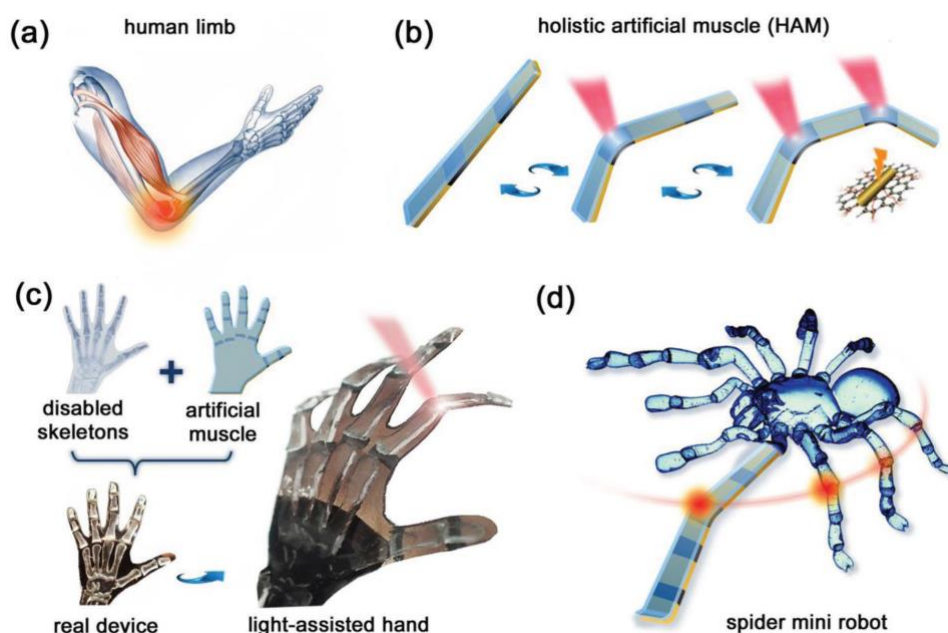


Photonic elastomers that can change colors like a chameleon have shown great promise in various applications. However, it remains a challenge to produce artificial photonic elastomers with desired optical and mechanical properties. Here, the generation of metallosupramolecular polymer-based photonic elastomers with tunable mechanical strength, angle-independent structural color, and self-healing capability is reported. The photonic elastomers are prepared by incorporating isotropically arranged monodispersed SiO<sub>2</sub> nanoparticles within a supramolecular elastomeric matrix based on metal coordination interaction between amino-terminated poly(dimethylsiloxane) and cerium trichloride. The photonic elastomers exhibit angle-independent structural colors, while Young's modulus and elongation at break of the as-formed photonic elastomers reach 0.24 MPa and 150%, respectively. The superior elasticity of photonic elastomers enables their chameleon-skin-like mechanochromic capability. Moreover, the photonic elastomers are capable of healing scratches or cuts to ensure sustainable optical and mechanical properties, which is crucial to their applications in wearable devices, optical coating, and visualized force sensing.

**Comment:** An example of self-healing color-based stress sensor. The angle independency of the seen color is to be noticed, even if it limits the applications. The coloration is due to the size of the SiO<sub>2</sub> NPs, and their average distance.

## Plasmonic-Assisted Graphene Oxide Artificial Muscles

Han, B.; Zhang\*, Y. L.; Zhu, L.; Li, Y.; Ma, Z. C.; Liu, Y. Q.; Zhang, X. L.; Cao, X. W.; Chen, Q. D.; Qiu,\* C. W.; and Sun\*, H. B. [Adv. Mater. 2019, 31, 5](#)

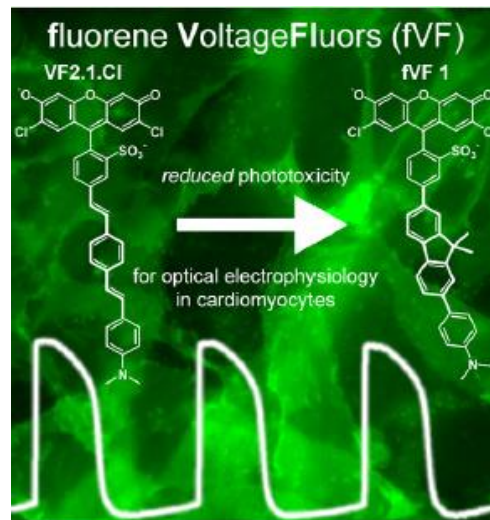


Muscles and joints make highly coordinated motion, which can be partly mimicked to drive robots or facilitate activities. However, most cases primarily employ actuators enabling simple deformations. Therefore, a mature artificial motor system requires many actuators assembled with jointed structures to accomplish complex motions, posing limitations and challenges to the fabrication, integration, and applicability of the system. Here, a holistic artificial muscle with integrated light-addressable nodes, using one-step laser printing from a bilayer structure of poly(methyl methacrylate) and graphene oxide compounded with gold nanorods (AuNRs), is reported. Utilizing the synergistic effect of the AuNRs with high plasmonic property and wavelength selectivity as well as graphene with good flexibility and thermal conductivity, the artificial muscle can implement full-function motility without further integration, which is reconfigurable through wavelength-sensitive light activation. A biomimetic robot and artificial hand are demonstrated, showcasing functionalized control, which is desirable for various applications, from soft robotics to human assists.

**Comment:** A very nice proof of concept. The authors describe here an easy way to produce artificial muscles: it only requires to burn a DVD with an appropriate coating. These muscles are therefore only 2D, paper thin, muscles.

## New Molecular Scaffolds for Fluorescent Voltage Indicators

Bogess, S. C.; Gandhi, S. S.; Siemons, B. A.; Huebsch, N.; Healy, K. E.; Miller, E. W.\* [ACS Chem. Bio. 2019, ASAP](#)

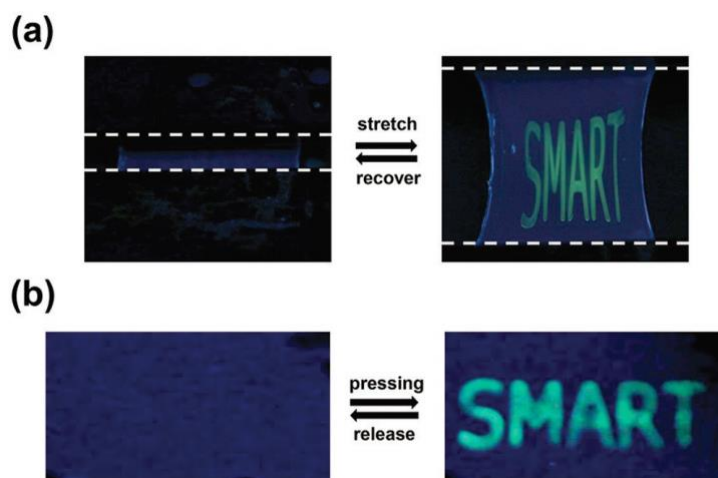


The ability to non-invasively monitor membrane potential dynamics in excitable cells like neurons and cardiomyocytes promises to revolutionize our understanding of the physiology and pathology of the brain and heart. Here, we report the design, synthesis, and application of a new class of fluorescent voltage indicators that make use of a fluorene-based molecular wire as a voltage-sensing domain to provide fast and sensitive measurements of membrane potential in both mammalian neurons and human-derived cardiomyocytes. We show that the best of the new probes, fluorene VoltageFluor 2 (fVF 2), readily reports on action potentials in mammalian neurons, detects perturbations to the cardiac action potential waveform in human induced pluripotent stem cell-derived cardiomyocytes, shows a substantial decrease in phototoxicity compared to existing molecular wire-based indicators, and can monitor cardiac action potentials for extended periods of time. Together, our results demonstrate the generalizability of a molecular wire approach to voltage sensing and highlight the utility of fVF 2 for interrogating membrane potential dynamics.

**Comment:** Voltage-sensing seems interesting. I do not understand well the principle behind it.

### A Double-Layer Mechanochromic Hydrogel with Multidirectional Force Sensing and Encryption Capability

Zhu, Q.; Vliet, K.\*; Holten-Andersen, N.\*; Miserez, A.\* [Adv. Func. Mater 2019, 1808191](#)

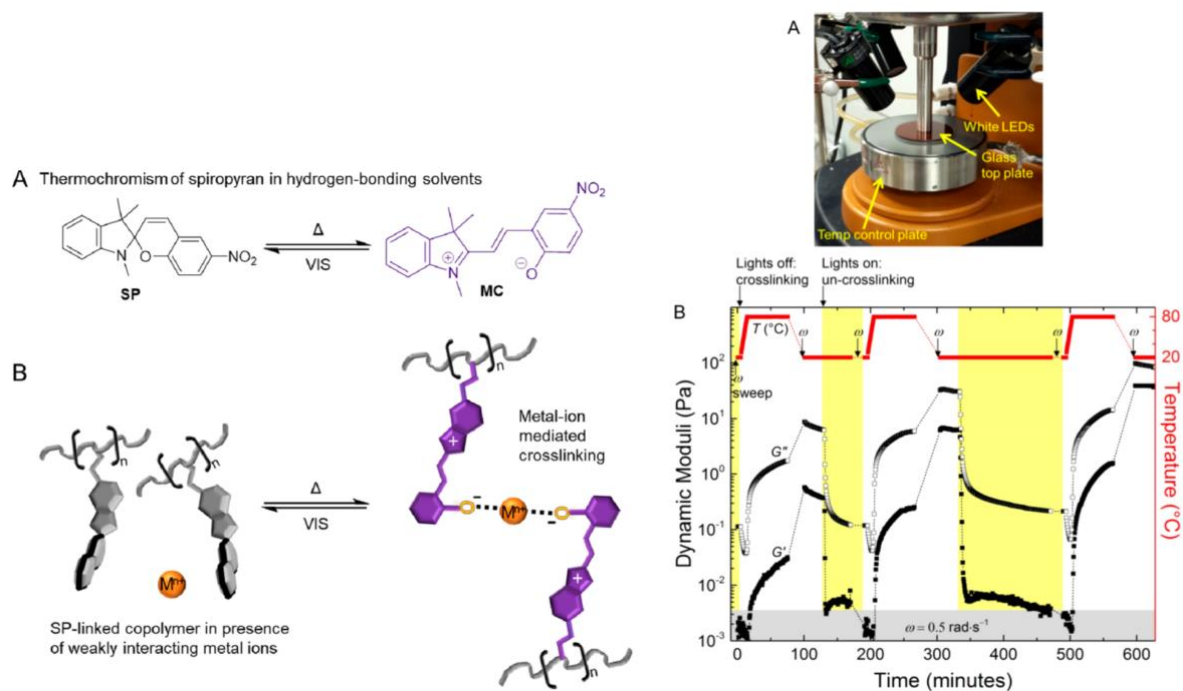


Hydrogel-based soft mechanochromic materials that display colorimetric changes upon mechanical stimuli have attracted wide interest in sensors and display device applications. A common strategy to produce mechanochromic hydrogels is through photonic structures, in which mechanochromism is obtained by strain-dependent diffraction of light. Here, a distinct concept and simple fabrication strategy is presented to produce luminescent mechanochromic hydrogels based on a double-layer design. The two layers contain different luminescent species—carbon dots and lanthanide ions—with overlapped excitation spectra and distinct emission spectra. The mechanochromism is rendered by strain-dependent transmittance of the top-layer, which regulates light emission from the bottom-layer to control the overall hydrogel luminescence. An analytical model is developed to predict the initial luminescence color and color changes as a function of uniaxial strain. Finally, this study demonstrates proof-of-concept applications of the mechanochromic hydrogel for pressure and contact force sensors as well as for encryption devices. A luminescent mechanochromic hydrogel is fabricated using a novel double-layer design and a simple fabrication process. The mechanochromism can be tuned by altering the top layer UV-absorber concentration and can be predicted based on strain-transmittance relationships. This new type of mechanochromic hydrogel could be used for pressure and force sensing as well as for encryption devices containing hidden information that can only be revealed by applying strain under UV light.

**Comment:** Very interesting work. The mechanochromism is achieved by changing the transmittance of light through the stretching or pressing of the gel. The ideal is simple and it really worked well.

### Modulating Noncovalent Cross-links with Molecular Switches

Epstein, E. S.; Martinetti, L.; Kollarigowda, R. H.; Carey-De La Torre, O.; Moore, J. S.; Ewoldt, R. H.; Braun, P. V.\* *J. Am. Chem. Soc.* **2019**, *141*, 3597–3604.



Spiropyran molecular switches, in conjunction with transition metal ions, are shown to operate as reversible polymer cross-linkers. Solutions containing a spiropyran-functionalized

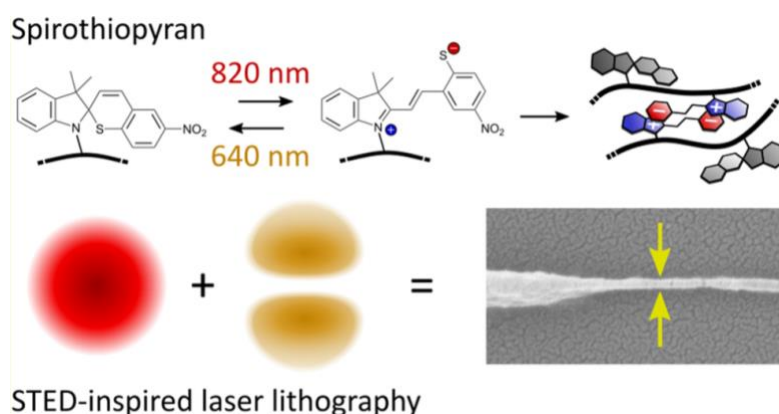


polymer and transition metal ions underwent reversible thermally triggered (light-triggered) transient network formation (disruption) driven by the association (dissociation) of metal-ligand cross-links. **Heat triggers metal-ion-mediated cross-linking via thermal isomerization of spiropyran** to its open, merocyanine form, and exposure to visible light triggers dissociation of polymer cross-links. Cross-linking is found to depend on both the valence of the ion as well as the molar ratio of spiropyran to metal salt. We envision this to be a starting point for the design of many types of reversible, stimuli-responsive polymers, utilizing the fact that spiropyrans have been shown to respond to a variety of stimuli including heat, light, pH, and mechanical force.

**Comment:** To be fair, I was tempted by the use of spiropyrans to do “something” and stayed because of the rheology under light irradiation. A very simple set-up and work like a charm.

### STED-Inspired Laser Lithography Based on Photoswitchable Spirothiopyran Moieties

Müller, P.; Müller, R.; Hammer, L.; Barner-Kowollik, C.\*; Wegener, M.; Blasco, E.\* [Chem. Mater. 2019, ASAP.](#)

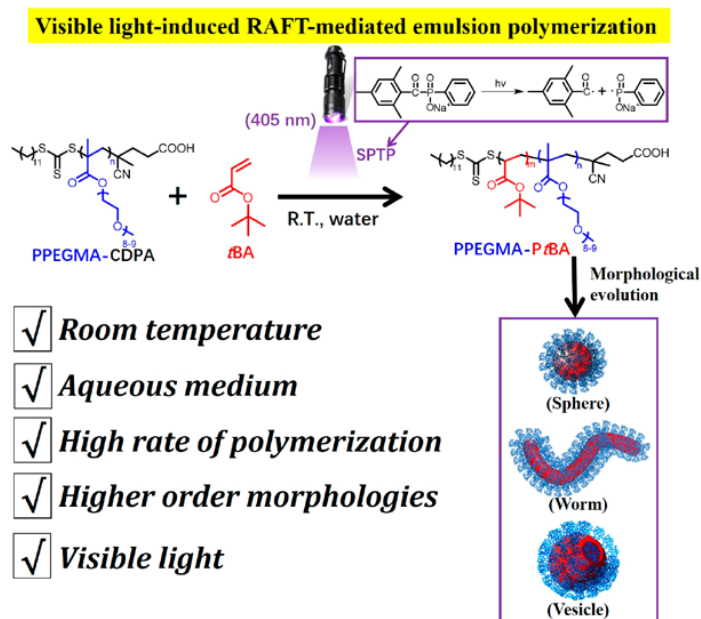


We introduce a photoresist based on methacrylate copolymers bearing photochromic spirothiopyran moieties as side groups that can crosslink via supramolecular interaction between the chromophores. **Upon two-photon excitation, the resist is capable of generating freestanding three-dimensional structures and offers an inhibition channel, which allows for stimulated-emission depletion-inspired laser lithography.** Reversible inhibition, linewidth narrowing, and resolution enhancement are demonstrated.

**Comment:** Neat. The merocyanine units can aggregate by ion pairing which entails the formation of solid materials. The other form of the switch, the spiropyran, cannot. Since you have two states, both promoted by independent light frequencies, you can apply tricks learned from super-resolution microscopy.

### Photoinitiated Polymerization-Induced Self-Assembly via Visible Light-Induced RAFT-Mediated Emulsion Polymerization

Tan, J.; Dai, X.; Zhang, Y.; Yu, L.; Sun, H.; Zhang, L. [ACS Macro Lett. 2019, 8, 205–212.](#)

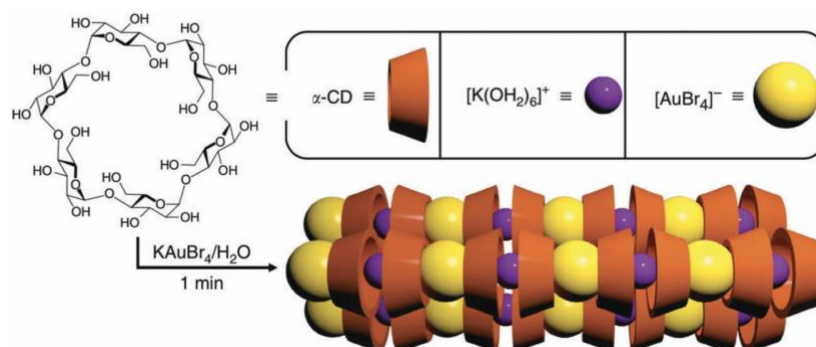


Aqueous emulsion polymerization is one of the most commonly used techniques in industry for the production of polymer latexes. In this contribution, we present photoinitiated polymerization-induced self-assembly (photo-PISA) based on aqueous visible light-induced reversible addition–fragmentation chain transfer (RAFT)-mediated emulsion polymerization at room temperature. A wide range of morphologies including spheres, worms, and vesicles have been achieved at room temperature by modulating reaction parameters. Additionally, this method enables access to inorganic nanoparticles-loaded vesicles by adding inorganic nanoparticles at the beginning of the polymerization. Finally, an oxygen-tolerant RAFT-mediated emulsion polymerization has been developed, allowing the synthesis of polymer nano-objects at low volumes (e.g., in a 96-well plate). This study is expected to expand the scope of photo-PISA for the preparation of various block copolymer nano-objects in water at room temperature.

**Comment:** The authors developed a novel photo-PISA method via visible light-induced RAFT-mediated emulsion polymerization for the synthesis diblock copolymer nano-objects.

### Practical Applications of Supramolecular Chemistry

Kolesnichenko, I. V.; Anslin, E. V.\* [Chem. Soc. Rev. 2017, 46, 2385–2390](#).

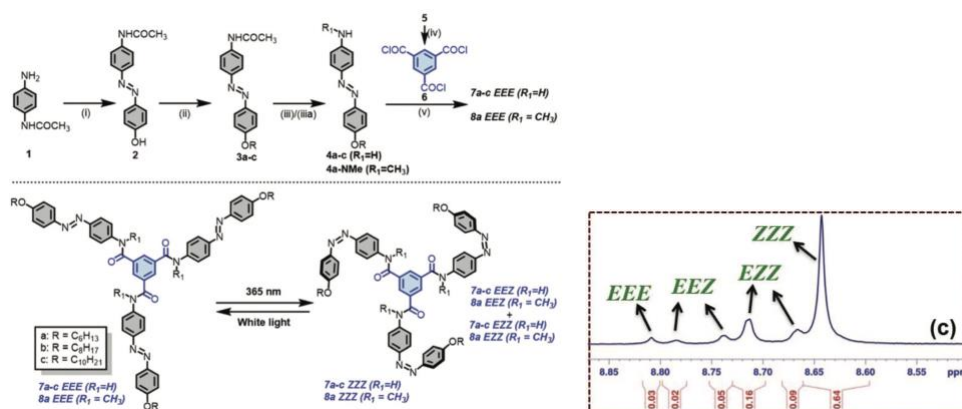


This year marks the 50th anniversary of Charles Pedersen's discovery of crown ethers, what is widely considered the birth of supramolecular chemistry. Since then, the field has progressed greatly, winning two Nobel Prizes and **seeing the implementation of many practical applications**. In commemoration, we are exploring the more recent advances of the field, which have made it past the realm of chemistry, into the real world. Though not a comprehensive review, the topics that we discuss here are supramolecular sensors, imaging for medical applications, metal extraction from ores and nuclear waste, as well as drug delivery.

**Comment:** More a joke on my part that something else: on the one hand, Fraser would hit Eric with a chair or something because of his claim that the 2016 Nobel Prize was given to supramolecular chemistry (it was given to molecular chemistry he'd say). And, second, the authors do mention actual applications of supramolecular systems, which is good to have around.

### Reversibly Photoswitchable Alkoxy Azobenzenes Connected Benzenetricarboxamide Discotic Liquid Crystals with Perpetual Long Range Columnar Assembly

Devi, S.; Bala, I.; Gupta, S. P.; Kumar, P.; Pal, S. K.; Venkataramani, S. \* [Org. Biomol. Chem.](#) **2019**, *17*, 1947–1954.



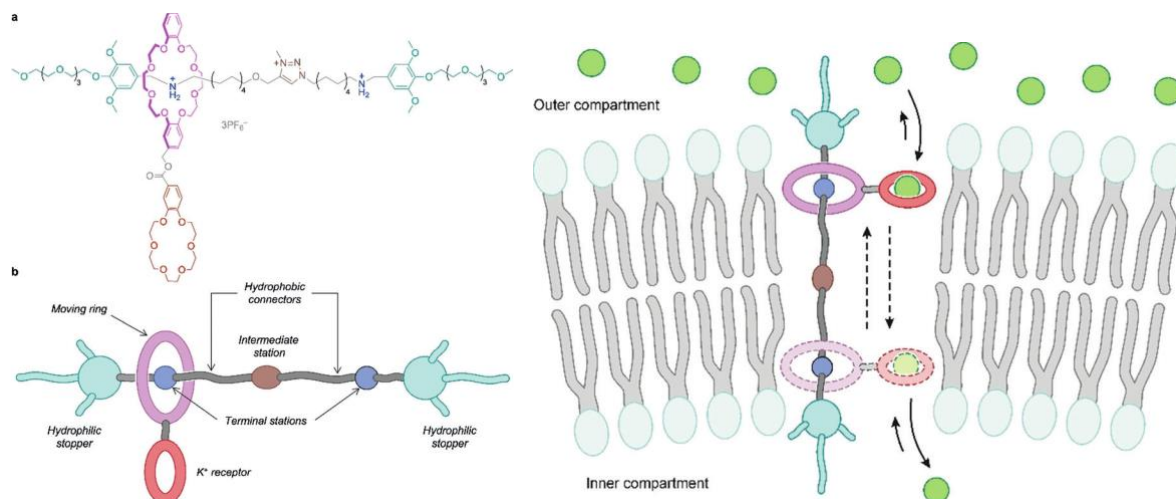
Liquid crystals (LCs) with photoswitchable groups are very interesting owing to their dual applications. In this regard, we report the synthesis of long chain alkoxy azobenzene incorporated benzenetricarbox- amides **7a–c** based room temperature columnar LCs. Apart from the light induced isomerization in the solution phase, the salient feature of these systems is the reversible photoisomerization even in the bulk state with perpetual columnar self-assembly at room temperature. Based on the observation of meso- morphic textures under polarised optical microscopy (POM) and grazing incidence small/wide angle X-ray scattering (GISAXS/GIWAXS) studies, the columnar assembly was found to be stable upon photo- isomerization. However, subtle changes in height profile have been observed in AFM measurements after photoswitching. Interestingly, a temperature dependent change between rectangular and hexagonal mesophases in **7a** has been observed. Upon extending the alkoxy chain length, only the hexagonal meso- phase was observed. For comparison, the corresponding N-methylated derivative of **7a** has also been syn- thesized. Despite the better photoswitching behaviour, due to the lack of planarity and H-bonding, **8a** did not show any columnar mesophase.

**Comment:** So much is wrong here! Although wrong is a strong word, if you ever dare to put a compound number in an abstract it has gone too far. The molecules, though, are nice; simple but nice. But I disagree with their conclusions regarding the reason behind the lack of switching in concentrated solutions (i.e. notice the amide proton rather on the high ppm region). All in all, I

think the exact same system, studied more carefully, would end up in a better journal (provided the good names are among the authors).

### Molecular Cable Car for Transmembrane Ion Transport

Credi, A.\* [Angew. Chem. Int. Ed. 2019, ASAP.](#)

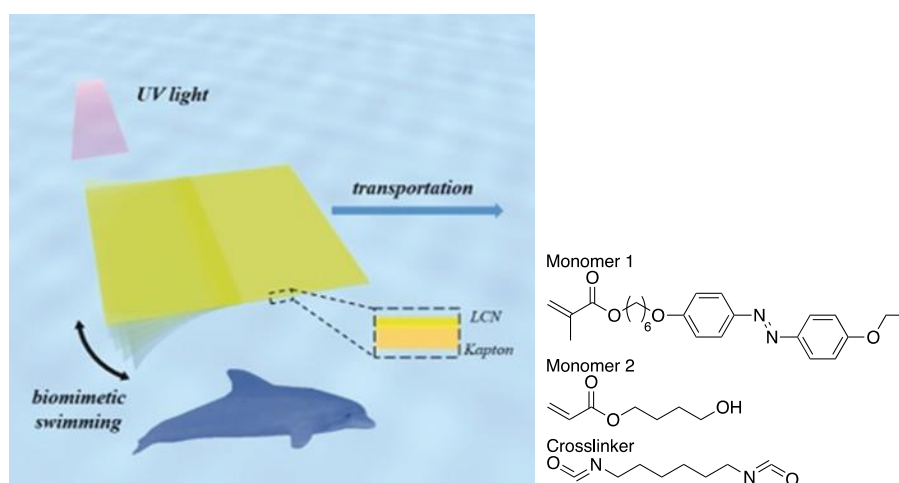


The controlled transport of molecular and ionic substrates across bilayer membranes is a fundamental task for the operation of living organisms. It is also a highly fascinating and demanding challenge for artificial molecular machines. The recent report of a **synthetic transmembrane molecular shuttle** that can **transport potassium ions selectively down a gradient in a liposomal system** makes a small but significant step towards this goal.

**Comment:** This brief paper allows the reader to understand better the different challenges that remain to be addressed in the field of controlled molecular transport. Moreover, I find this system cleverly designed, even if it is not extremely useful at the moment.

### A Light-Activated Polymer Composite Enables On-Demand Photocontrolled Motion: Transportation at the Liquid/Air Interface

Ma, S.; Li, X.; Huang, S.; Hu, J.; Yu, H.\* [Angew. Chem. Int. Ed. 2019, ASAP.](#)

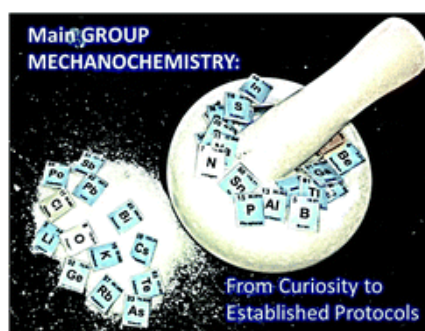


Swimmers at liquid/air interface have drawn enormous attention for their potential applications while conventional devices face problems such as dependence on fuel, difficulty in motion control and accompanying high temperature. Herein, we show one novel **light-driven swimmer** based on bimorph composite structure of **photoresponsive liquid-crystalline polymer** network and commercially available **polyimide** (Kapton) whose motion is highly controllable with photoirradiation. The bilayer-structured film shows **quickly photoinduced bending** towards the Kapton side **on exposure to UV light** and **recovers immediately after removal of light**. When placed on liquid surface, the swimmer propels itself continually though rhythmic beating the liquid like a dolphin moving forward with its tail fin. Besides, light-powered rotation of the swimmer is successfully achieved by simply changing the length-width ratio and the irradiation site, mimicking the function of a dolphin's pectoral fin. Combining the forward movement and rotation motion together, on-demand directional control of the photo-driven swimmer can be readily obtained at room temperature, showing promising for miniaturized transportation

**Comment:** This paper should definitely be part of our review related to collective motion, unfortunately it has been published too recently to be included. In this system, a Liquid-Crystal Polymer (LCP) integrates an azobenzene unit. The latter induces a photo-controlled bending of the material. This article is interesting in order to discover the world of LCP moving materials. However, the different schemes and pictures are so far just unreadable because of their bad quality. I hope an improved version will be published as soon as possible.

### Main Group Mechanochemistry: From Curiosity to Established Protocols

Tan, D.; García, F.\* [Chem. Soc. Rev. 2019, ASAP.](#)

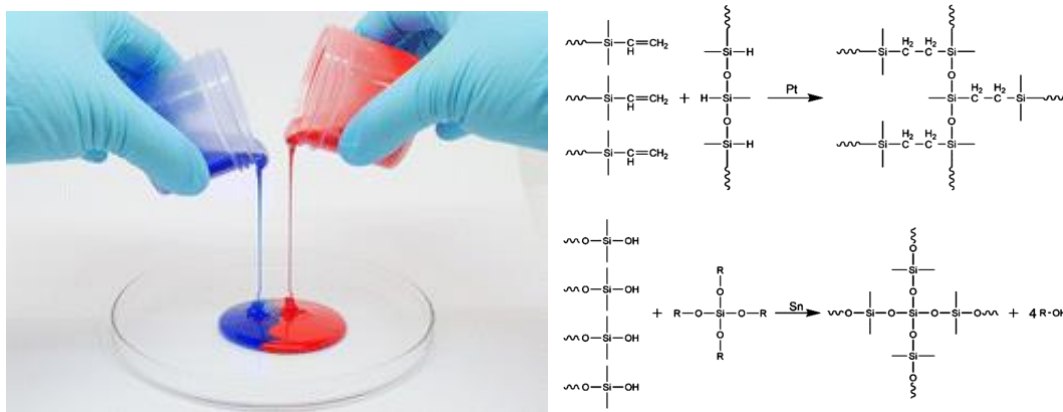


In the last few decades, **mechanochemistry** has become rapidly established as a powerful tool enabling environmentally-benign and sustainable chemical syntheses. Not only have these techniques been demonstrated as viable alternatives to traditional solution-based syntheses, but they have also received attention for their ability to enable new reactivity and “unlocking” novel compounds inaccessible by conventional methods. Reflecting the rising popularity of mechanochemistry, many excellent reviews highlighting its benefits have recently been published. Whilst the scope of most of these focuses on organic chemistry, transition-metal catalysis, porous framework materials, coordination compounds and supramolecular synthesis, few have addressed the use of mechanochemical ball milling for the synthesis of compounds containing s- and p-block elements. This tutorial review turns the spotlight towards **mechanochemical research in the field of inorganic main group chemistry**, highlighting significant advantages that solid-state inorganic reactions often possess, and the potential for these to drive the development of greener methodologies within the modern main group arena.

**Comment:** It is the second time I propose a paper related to mechanochemistry. I am convinced that one day it could be useful to someone working in organic synthesis. Although this article is more related to inorganic chemistry, it remains essential to get a better comprehension of this relatively new domain.

## How to Tailor Flexible Silicone Elastomers with Mechanical Integrity: A Tutorial Review

Mazurek, P.; Vudayagiri, S.; Skov, A. L.\* [Chem. Soc. Rev. 2019.](#)

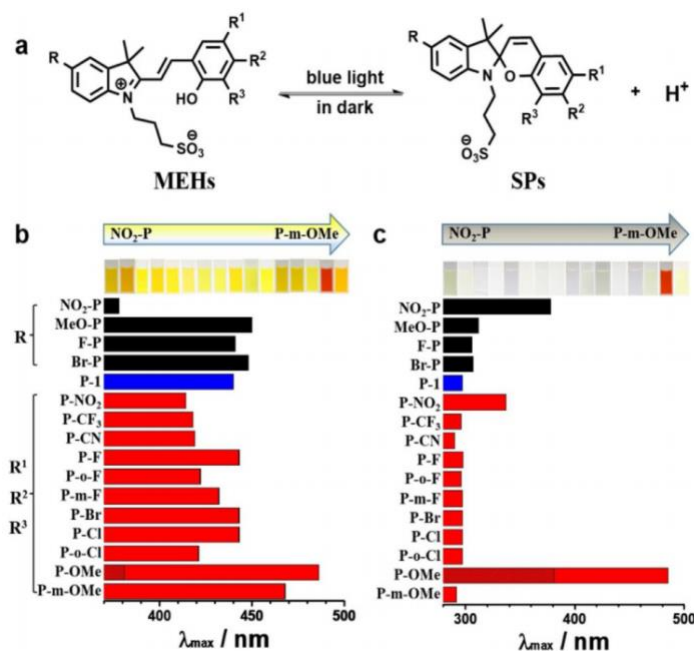


The **tutorial** aims to equip the beginners in **silicone** research with the knowledge to **formulate recipes and process elastomer networks**, targeting specific properties related to soft applications such as stretchable electronics without compromising the mechanical integrity of the elastomer. In doing so, we hope to stimulate further research in the area of tailor-made soft silicone elastomers for novel applications and allow researchers to bypass the limitations imposed by the use of commercially available silicone elastomer formulations. Silicone elastomers are widely used due to the favourable properties, such as flexibility, durable dielectric insulation, barrier properties against environmental contaminants and stress-absorbing properties over a wide range of temperatures  $\sim -100^\circ\text{C}$  to  $250^\circ\text{C}$ . **For research on flexible electronics** and other emerging technologies, the most commonly utilized silicone elastomer formulation is **Sylgard 184** which is **easier to process** than most other commercially available silicone elastomers, due to the fact that the premixes have low viscosity. Furthermore, curing is robust and not as sensitive to poisoning as other silicone elastomer formulations. However, Sylgard 184 is not suitable for all fields of research that require flexible and stretchable silicones. When much softer networks are needed, the Sylgard 184 premixes are either mixed in non-stoichiometric ratios, or they are blended with softer types of commercially available elastomers, which compromise the mechanical integrity of the elastomer. Therefore, it is advantageous for researchers to formulate their own custom-made silicone elastomers and not depend on premade formulations which often harbour a few unknown components.

**Comment:** This paper is an excellent introduction to read before working on silicone-based materials. Although the reader has to keep in mind that it is not a comprehensive review.

## Effects of Substituents on Metastable-State Photoacids: Design, Synthesis, and Evaluation of their Photochemical Properties

Liu, J.; Tang, W.; Sheng, L.; Du, Z.; Zhang, T.; Su, X.; Zhang, X.; [Chem. Asian J. 2019, 14, 438-445.](#)

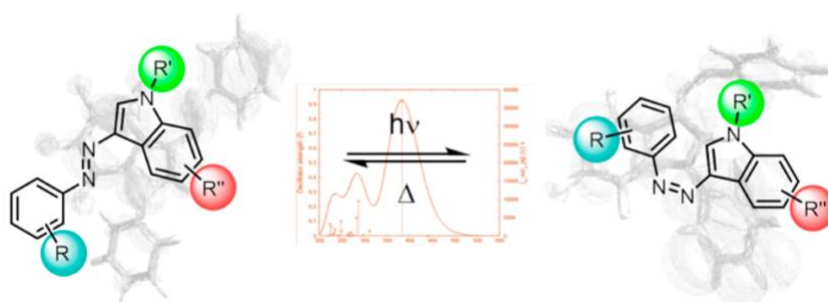


Recently, metastable-state photoacids have been widely used to control proton transfer in numerous chemical and biological processes as well as applications with visible light. Generally, substituents have a great influence on the photochemical properties of molecules, which will further affect their applications. Yet, the effects of substituents on metastable-state photoacids have not been studied systematically. In this work, 16 metastable-state photoacid derivatives were designed and synthesized on the basis of substituents having a large range of s-p electron-donor-acceptor capabilities. The effects of substituents on the color display [or maximum absorption band(s)], solubility, pK<sub>a</sub>, dark/photoacidity, photosensitivity, and relaxation kinetic(s) were investigated in detail. This study will be helpful for the targeted design and synthesis of promising photoacids and the application of their photocontrolled proton-release processes in functional materials/devices.

**Comment:** These results confirmed that the photoreaction characteristics of these photoacids mainly depended on the nature and position of the substituents. Taken together, photoacids modified with conjugated halogens at R and R<sub>1</sub> positions exhibited the best-balanced properties in their photoreactions/processes.

### Substituent Effects on 3-Arylazoindole Photoswitches

Simeth, N.; Bellisario, A.; Crespi, S.\*; Fagnoni, M.; König, B.\* *J. Org. Chem.* **2019**, ASAP.

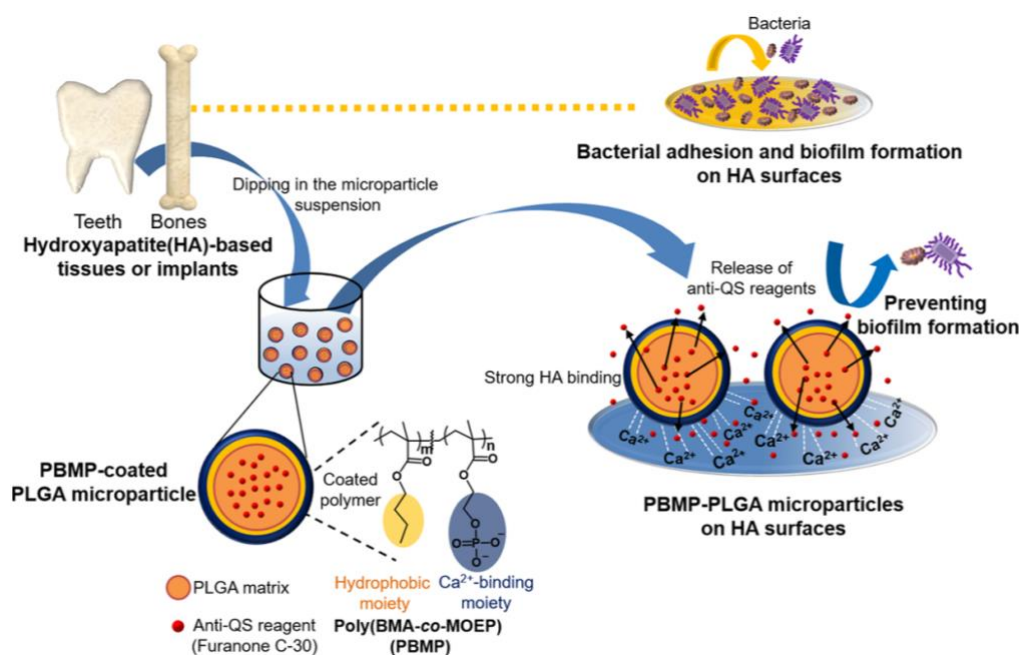


The development and investigation of heteroazo switches has flourished in recent years. Because of their specific photophysical and photochemical properties, they find versatile applications from material science to medicine. However, a deep mechanistic understanding is needed to be able to predict the properties of such azoswitches. In particular, the effect of different substituents on the azo chromophore is of great interest as they are often crucial for embedding the molecular switch into a system of interest. Herein, we provide a detailed spectroscopic and computational study on the influence of substituents on 3-phenylazoindoles chosen as models. We will point out changes in absorption properties and analyze the photostationary state of the thermally labile Z isomers through computational means to provide a general structure–property relationship guideline for further use of these compounds.

**Comment:** The energy of the transition band depends on the solvent and the substituents. Indeed, functional groups which extend the  $\pi$ -system, or generate an azo-push-pull system, lead to more pronounced bathochromic shifts in the absorption spectra.

### Calcium-Binding Polymer-Coated Poly(lactide-co-glycolide) Microparticles for Sustained Release of Quorum Sensing Inhibitors to Prevent Biofilm Formation on Hydroxyapatite Surfaces

Kang, M.; Kim, S.; Kim, H.; Song, Y.; Jung, D.; Kang, S.; Seo, J.-H.; Nam, S.\*; Lee, Y.\* [ACS Appl. Mater. Interfaces](#) **2019**, *11*, 7686–7694.



**Quorum sensing (QS) inhibitor-based therapy** is an attractive strategy to inhibit bacterial biofilm formation without excessive induction of antibiotic resistance. Thus, we designed Ca<sup>2+</sup>-binding poly(lactide-co-glycolide) (PLGA) microparticles that can maintain a sufficient concentration of QS inhibitors around hydroxyapatite (HA) surfaces in order to prevent biofilm formation on **HA-based dental or bone tissues or implants** and, therefore, subsequent pathogenesis. Poly(butyl methacrylate-co-methacryloyloxyethyl phosphate) (PBMP) contains both Ca<sup>2+</sup>-binding phosphomonoester groups and PLGA-interacting butyl groups. The PBMP-coated PLGA (PLGA/PBMP) microparticles exhibited superior adhesion to HA surfaces without altering the sustained release properties of uncoated PLGA microparticles. PLGA/PBMP microparticle-

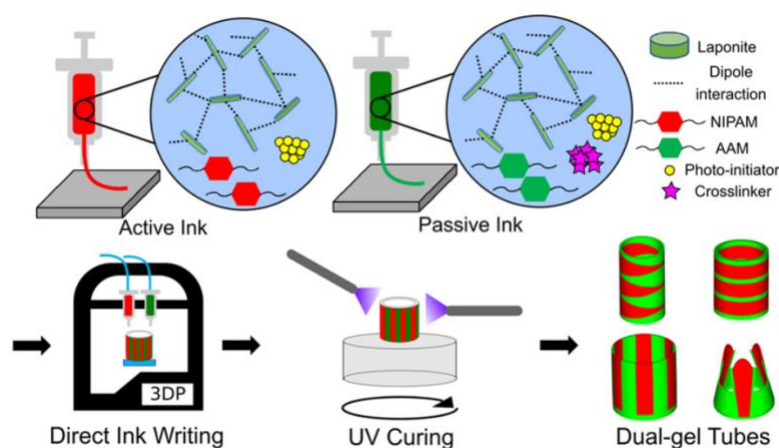


encapsulating furanone C-30, a representative QS inhibitor, effectively inhibited the growth of *Streptococcus mutans* and its ability to form biofilms on HA surface for prolonged periods of up to 100 h, which was much longer than either furanone C-30 in its free form or when encapsulated in noncoated PLGA microparticles.

**Comment:** Hydroxyapatite-based implants are used to replace bones and teeth, but they are suffering from the formation of biofilms at their surface like many other materials. A solution to inhibit the formation of such films is to suppress the communication between bacteria (quorum sensing [QS]). In this study, furanone C-30, a QS inhibitor, is embedded in a polymeric microparticle coated with a copolymer. These microparticles are further coated on a biomaterial, effectively preventing the biofilm formation for a long period of time due to the sustained release of the inhibitor.

### Dual-Gel 4D Printing of Bioinspired Tubes

Liu, J.; Erol, O.; Pantula, A.; Liu, W.; Jiang, Z.; Kobayashi, K.; Chatterjee, D.; Hibino, N.; Romer, L. H.; Kang, S. H.; Nguyen, T. D.; Gracias, D. H.\* [ACS Appl. Mater. Interfaces 2019, 11, 8492–8498.](#)

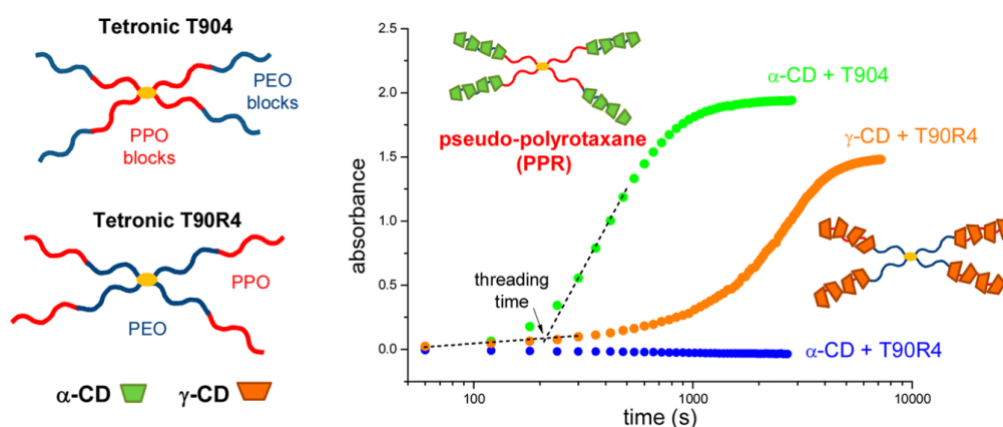


The distribution of periodic patterns of materials with radial or bilateral symmetry is a universal natural design principle. Among the many biological forms, tubular shapes are a common motif in many organisms, and they are also important for bioimplants and soft robots. However, the simple design principle of strategic placement of 3D printed segments of swelling and nonswelling materials to achieve widely different functionalities is yet to be demonstrated. Here, we report the design, fabrication, and characterization of **segmented 3D printed gel tubes** composed of an active **thermally responsive** swelling gel (poly-*N*-isopropylacrylamide) and a passive **thermally nonresponsive** gel (polyacrylamide). Using finite element simulations and experiments, we report a variety of shape changes including uniaxial elongation, radial expansion, bending, and gripping based on two gels. Actualization and characterization of thermally induced shape changes are of key importance to robotics and biomedical engineering. Our studies present rational approaches to engineer complex parameters with a high level of customization and tunability for additive manufacturing of dynamic gel structures.

**Comment:** Various shape changes of responsive materials are commonly attained by the introduction of bulk anisotropy. In this report, the authors engineered a 3D-printing method to create a composite material, containing a responsive gel and its non-responsive counterpart.

## Pseudo-Polyrotaxanes of Cyclodextrins with Direct and Reverse X-Shaped Block Copolymers: A Kinetic and Structural Study

Puig-Rigall, J.; Serra-Gómez, R.; Stead, I.; Grillo, I.; Dreiss, C. A.\*; González-Gaitano, G.\* *Macromolecules* **2019**, *52*, 1458–1468.

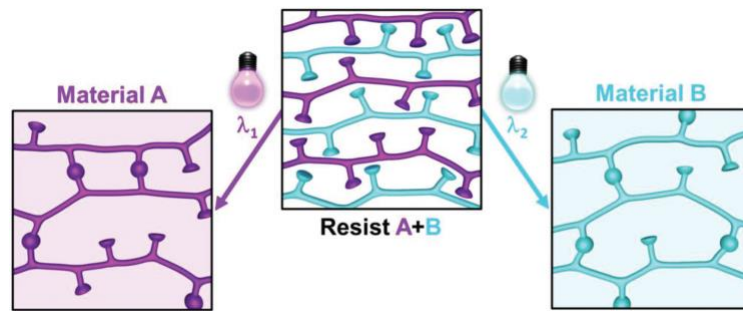


Pseudo-polyrotaxanes (PPRs) are supramolecular host–guest complexes constituted by the reversible threading of a macrocycle along a polymer chain. The resulting dynamic “molecular necklaces” offer potential applications in nanotechnology, drug delivery, and biomaterials. We report the formation of PPRs by **threading of cyclodextrins (CDs), cyclic oligosaccharides, onto X-shaped PEO–PPO block copolymers** with two opposite presentation of their hydrophobic and hydrophilic blocks: Tetronic 904 (T904) and its reverse counterpart Tetronic 90R4 (T90R4). We assess the effects that relative block position on the polymeric surfactants and cavity size of CD have on the composition, morphology, thermodynamics, and kinetics of PPRs by using a combination of X-ray diffraction, scanning electron microscopy, NMR, UV–vis spectroscopy, and time-resolved small-angle neutron scattering (TR-SANS). Solid PPRs with lamellar microstructure and crystalline channel-like structures are obtained with native CDs and both Tetronics above a threshold concentration of the macrocycle, which varies with the type of CD and surfactant. While  $\gamma$ -CD can form PPRs with both Tetronics,  $\alpha$ -CD only forms a PPR with T90R4 at high concentrations. The results can be explained in terms of the preferential complexation of  $\alpha$ -CD with EO and  $\gamma$ -CD with PO monomers, which also has a direct impact on the kinetics of PPR formation. Thermodynamic parameters of the reaction were obtained from the analysis of the stoichiometries and threading times as a function of temperature by using a model based on the Eyring equation. Negative enthalpies and positive entropies are obtained in all cases, and reactions are thermodynamically most favorable in the case of  $\alpha$ -CD with T904 and  $\gamma$ -CD with T90R4. TR-SANS experiments reveal an increase in the radius of gyration of the unimers over time, consistent with CD threading and further expansion of the PPR. Above the CMT,  $\alpha$ -CD threads the unimers to form the PPR, with no effect on the structure of T904 micelles, whose volume fraction decreases due to the shift of micellization equilibrium.

**Comment:** Pseudo-polyrotaxanes of cyclodextrin derivatives threaded onto PEO and PPO chains can self-assemble in various structures. This paper studies these supramolecular complexes for star-shaped block copolymers, and the results are interesting from the fundamental point of view of rational design of the resulting assemblies.

### Access to Disparate Soft Matter Materials by Curing with Two Colors of Light

Bialas, S.; Michalek, L.; Marschner, D. E.; Krappitz, T.; Wegener, M.; Blinco, J.\*; Blasco, E.\*; Frisch, H.\* and Barner-Kowollik, C. *Adv. Mater.* **2019**, *31*, 8

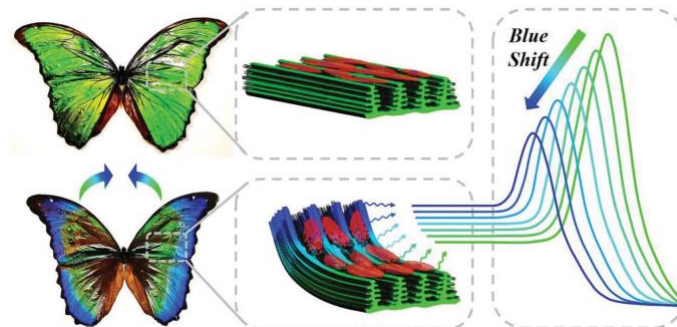


A platform technology for multimaterial photoresists that can be orthogonally cured by disparate colors of light is introduced. The resist's photochemistry is designed such that one wavelength selectively activates the crosslinking of one set of macromolecules, while a different wavelength initiates network formation of a different set of chains. Each wavelength is thus highly selective towards a specific photoligation reaction within the resist. Critically, the shorter wavelength does not induce ligation of the longer wavelength selective species within the same resist mixture, defined as "wavelength orthogonality." Uniquely, this dual-color addressable resist system allows generating spatially resolved soft matter materials by simply selecting the curing wavelength, thus constituting a wavelength-orthogonal multimaterial resist with applications ranging from coatings to 3D additive manufacturing of multimaterial architectures.

**Comment:** The principle is easy to understand but nevertheless interesting: The crosslinking occurs via dimerization of two different species, at two different wavelengths. (o-methyl benzaldehyde and styrylpyrene).

### Cardiomyocytes-Actuated Morpho Butterfly Wings

Chen, Z.; Fu, F.; Yu, Y.; Wang, H.; Shang, Y.; Zhao, Y.\* [Adv. Mater. 2019, 31, 8.](#)



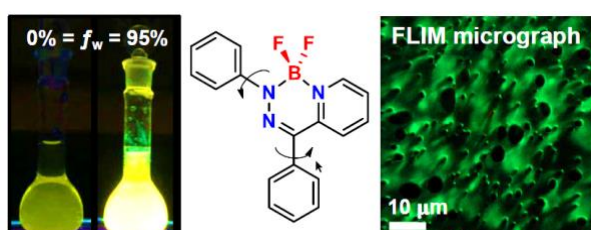
Morpho butterflies are famous for their wings' brilliant structural colors arising from periodic nanostructures, which show great potential value for fundamental research and practical applications. Here, a novel cellular mechanical visualizable biosensor formed by assembling engineered cardiac tissues on the Morpho butterfly wings is presented. The assembled cardiomyocytes benefit from the periodic parallel nanoridges of the wings and can recover their autonomic beating ability with guided cellular orientation and good contraction performance. As the beating processes are accompanied by the cardiomyocytes' elongation and contraction, the elastic butterfly wing substrate undergoes the same cycle of deformations, which causes corresponding synchronous shifts in their structural colors and photonic bandgaps for self-reporting of the cell mechanics. It is demonstrated that this self-reporting performance can be further improved by adding oriented carbon nanotubes in the nanoridges of the wings for the culture. In addition, taking advantage of the similar size of the cardiomyocyte and a single Morpho

wing scale, the investigation of single-cell-level mechanics can be realized by detecting the optical performance of a single scale. These remarkable properties make these butterfly wings ideal platforms for biomedical research.

**Comment:** Using butterfly wings as substrate for culturing cardiomyocytes really is a surprise to me. Due to their amazingly ordered structure, there are a perfect substrate for an almost perfectly ordered conductive carbon nanotubes network, helping the culture of cells.

### Optoelectronic, Aggregation, and Redox Properties of Double-Rotor Boron Difluoride Hydrazone Dyes

Capello, D.; Therien, D. A. B.; Staroverov, V.; Lagugn -Labarthe, F.; Gilroy, J. B.\* [\*Chem. Eur. J.\* \*\*2019\*\*, ASAP.](#)



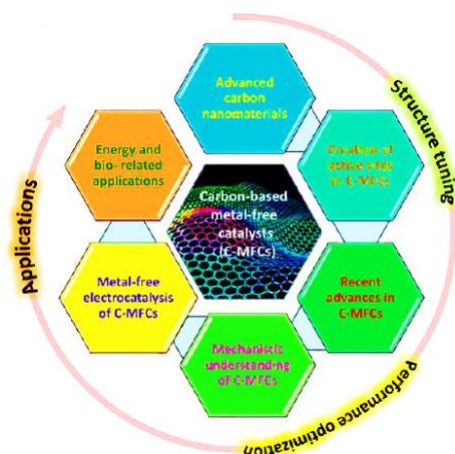
We develop the chemistry of boron difluoride hydrazone dyes (BODIHYs) bearing two aryl substituents and explore their properties. The low-energy absorption bands ( $\lambda_{\text{max}} = 427\text{--}464\text{ nm}$ ) of these dyes depend on the nature of the *N*-aryl groups appended to the BODIHY framework. Electron-donating and extended  $\pi$ -conjugated groups cause a red shift, whereas electronwithdrawing groups result in a blue shift. The title compounds were weakly photoluminescent in solution and strongly photoluminescent as thin films ( $\lambda_{\text{PL}} = 525\text{--}578\text{ nm}$ ) with quantum yields of up to 18% and lifetimes of 1.1–1.7 ns, consistent with the dominant radiative decay through fluorescence. Addition of water to THF solutions of the BODIHYs studied causes molecular aggregation which restricts intramolecular motion and thereby enhances photoluminescence. The observed photoluminescence of BODIHY thin films is likely facilitated by a similar molecular packing effect. Finally, cyclic voltammetry studies confirmed that BODIHY derivatives bearing *para*-substituted *N*-aryl groups could be reversibly oxidized ( $E_{\text{ox1}} = 0.62\text{--}1.02\text{ V vs. Fc/Fc}^+$ ) to their radical cation forms. Chemical oxidation studies confirmed that *para*-substituents at the *N*-aryl groups are required to circumvent radical decomposition pathways. Our findings provide new opportunities and guiding principles for the design of multifunctional boron difluoride complexes that are photoluminescent in the solid state.

**Comment:** This article reports, for the first time, the study of the redox properties of these BODIHY dyes. They also report the synthesis of persistent radical-cation species derived from these dyes, which were able to undergo radical-radical coupling reactions.

It is interesting how the aggregation studies confirmed that the restriction of the molecular motion of the two phenyl groups could be the main factor for the difference in the photoluminescent behaviour in solution and in solid state films.

### Carbon Nanomaterials for Energy and Biorelated Catalysis: Recent Advances and Looking Forward

Hu, Chuangang.; Qu, J.\*; Xiao, Y.; Zhao, S.; Chen, H.; Dai, L.\* [\*ACS Cent. Sci.\* \*\*2019\*\*, ASAP](#)

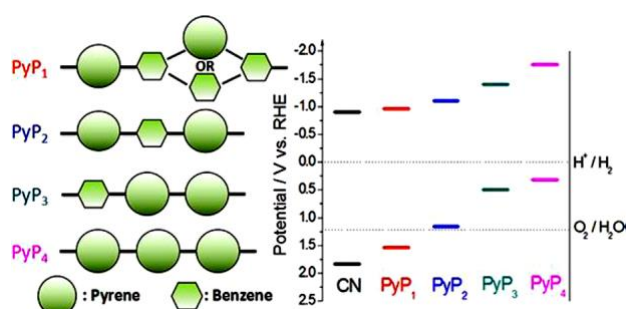


Along with the wide investigation activities in developing carbon-based, metal-free catalysts to replace precious metal (e.g., Pt) catalysts for various green energy devices, carbon nanomaterials have also shown great potential for biorelated applications. This article provides a focused, critical review on the recent advances in these emerging research areas. The structure–property relationship and mechanistic understanding of recently developed carbon-based, metal-free catalysts for chemical/biocatalytic reactions will be discussed along with the challenges and perspectives in this exciting field, providing a look forward for the rational design and fabrication of new carbon-based, metal-free catalysts with high activities, remarkable selectivity, and outstanding durability for various energy-related/biocatalytic processes.

**Comment:** A review for those are interested in the field of carbon based metal free catalysts for different applications. Here, it is highlighted the importance of carbon nanomaterials in energy related technologies like water splitting systems and its integration with fuel cells to generate clean electricity.

### Polymeric Donor-Acceptor Hetero-structures for Enhanced Photocatalytic H<sub>2</sub> Evolution without using Pt Cocatalysts

Zang, S.; Zhang, G.; Yang, P.; Zheng, D.; Wang, X.;\* [Chem. Eur. J. 2019, ASAP.](#)



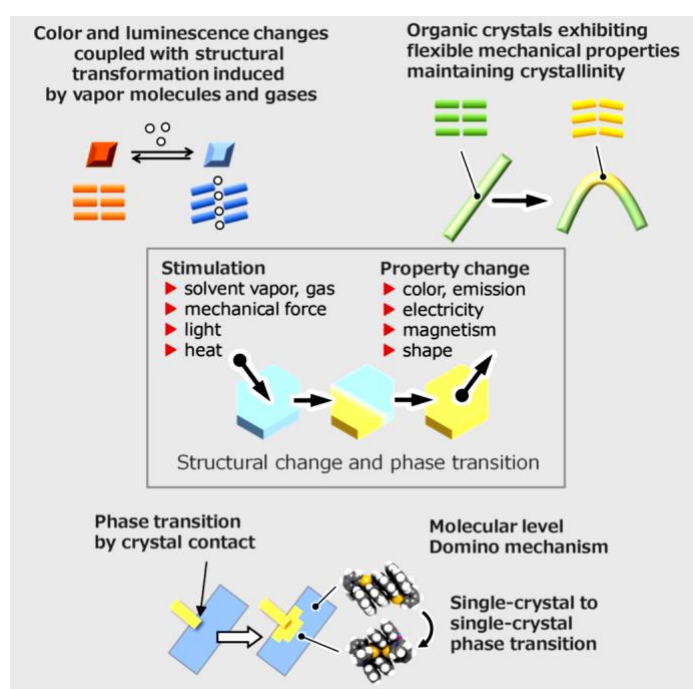
Polymeric carbon nitride (CN) is a promising material for photocatalytic water splitting. However, CN in its pristine form tends to show moderate activity due to fast recombination of the charge carriers. The design of efficient photocatalytic system is therefore highly desired, but it remains a great challenge in chemistry till now. Here we found that pyrene-based polymer, could serve as electron donor to accelerate the interface charge carrier transfer of CN. The construction

of donor-acceptor (D-A) heterojunction is confirmed to significantly restrain the charge recombination and thus improves the proton reduction process. This study provides a promising strategy to achieve solar H<sub>2</sub> production in an efficient and low-cost manner.

**Comment:** This article mentions the lack of good photocatalysts for hydrogen evolution and the potential of conjugated copolymers to improve the charge carrier mobilities when making polymeric junctions with materials such as carbon nitride (CN); the one described in this communication. The authors highlight that “more conjugated polymers can be used as stable, metal-free, semiconductors for the fabrication of efficient D-A junctions with good photocatalytic performance”.

## Soft Crystals – Flexible Response Systems with High Structural Order

Kato, M.;\* Ito, H.; Hasegawa, M.; Ishii, K. [Chem. Eur. J. 2019, ASAP.](#)

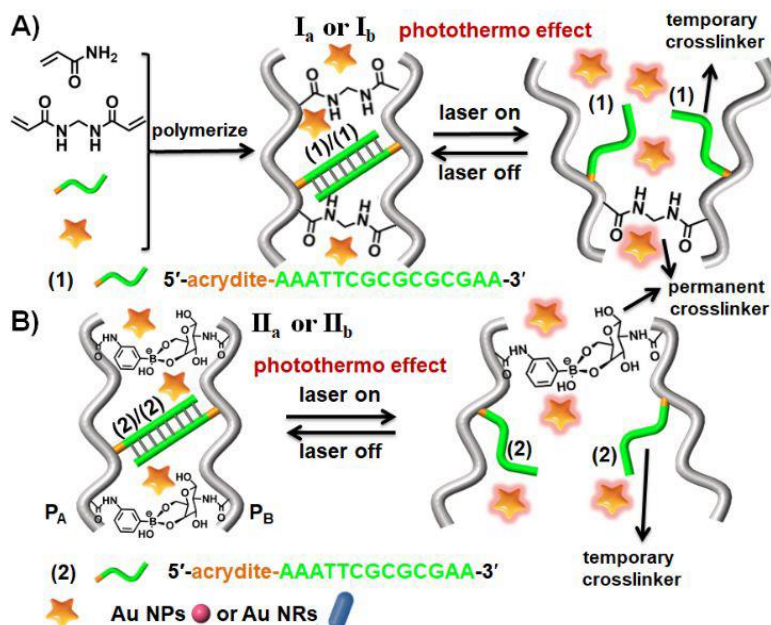


**A new material concept of soft crystals is proposed.** Soft crystals respond to gentle stimuli such as vapor exposure and rubbing but maintain their structural order and exhibit remarkable visual changes in their shape, color, and luminescence. **Various interesting examples of soft crystals are introduced in the article.** By exploring the interesting formation and phase-transition phenomena of soft crystals through interdisciplinary collaboration, new materials having both the characteristics of ordered hard crystals and those of flexible soft matter are expected.

**Comment:** The authors present soft crystals as materials that “possess highly ordered structures, but that can easily change their structure due to the much lower activation energies when compared with those of conventional hard crystals. Therefore, soft crystals respond to gentle stimuli while maintaining their structural order. Furthermore, the changes can be detected visually as changes in color or luminescence.” Also, the paper describes the challenges in this field such as the design of molecules for soft crystals and the understanding of the phase-transition phenomena of these molecular crystals.

## DNA-Based Hydrogels Loaded with Au Nanoparticles or Au Nanorods: Thermoresponsive Plasmonic Matrices for Shape- Memory, Self-Healing, Controlled Release and Mechanical Applications

Wang, C.; Liu, X.; Wulf, V.; Vázquez-González, M.; Fadeev, M.; Itamar, W.\* [ACS Nano 2019, Just accepted.](#)

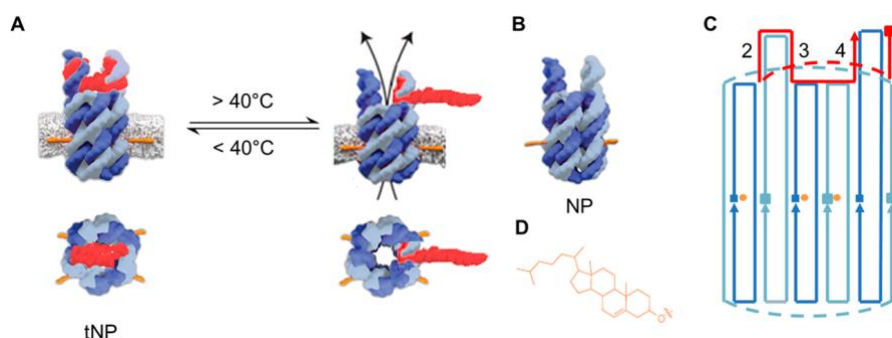


Gold nanoparticles (AuNPs) or gold nanorods (AuNRs) are loaded in polyacrylamide hydrogels cooperatively crosslinked by bis-acrylamide and nucleic acid duplexes or boronate ester-glucosamine and nucleic acid duplexes. The thermoplasmonic properties of AuNPs and AuNRs are used to control the stiffness of the hydrogels. The irradiation of the AuNPs-loaded ( $\lambda = 532$  nm) or the AuNRs-loaded ( $\lambda = 808$  nm) hydrogels leads to the thermoplasmonic heating of the hydrogels, the dehybridization of the DNA duplexes, and the formation of hydrogels with lower stiffness. By ON/OFF irradiation, the hydrogels are switched between low- and high-stiffness states. The reversible control over the stiffness properties of the hydrogels is used to develop shape-memory hydrogels, self-healing soft materials, and to tailor thermoplasmonic switchable drug release. In addition, by designing bilayer composites of AuNPs- and AuNRs-loaded hydrogels, a reversible thermoplasmonic, light-induced bending is demonstrated where the bending direction is controlled by the stress generated in the respective bilayer composite.

**Comment:** Normally, chemist introduce responsive molecular modulators (e.g. molecular machines, molecular switch) to adjust the properties of the materials. In this paper, the stiffness of hydrogels could be modulated by light because the DNA duplexes could be broken due to the high temperature originated from the **photothermal effect** (the wavelength  $\lambda$  of the light is related to the size and the shape of AuNPs) of gold nanoparticles. There is another interesting point in this paper, bilayer hydrogels containing AuNPs and AuNRs loaded composites were constructed, and the bilayer materials could be bended and the bending direction is controlled modulating the light intensity.

### A Temperature-Gated Nanovalve Self-Assembled from DNA to Control Molecular Transport across Membranes

Arnott, P. M.; Howorka, S.\* [ACS Nano 2019, ASAP.](#)

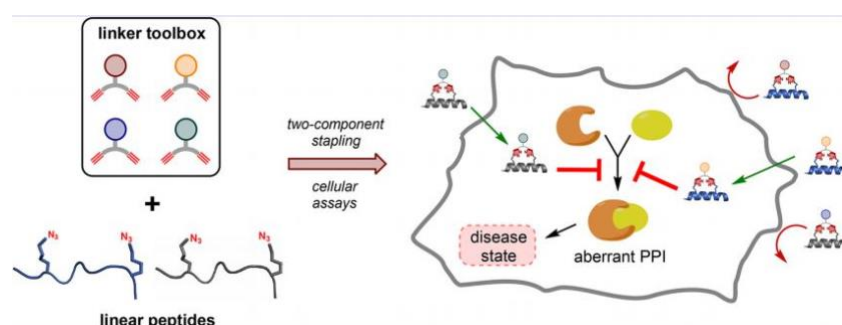


Nanopores are powerful nanodevices that puncture semifluid membranes to enable transport of molecular matter across biological or synthetic thin layers. Advanced nanopores featuring more complex functions such as ambient sensing and reversible channel opening are of considerable scientific and technological interest but challenging to achieve with classical building materials. Here we exploit the predictable assembly properties of DNA to form a multifunctional nanovalve that senses temperature for controlled channel opening and tunable transport. The barrel-shaped valve is formed from solely seven oligonucleotides and is closed at ambient temperatures. At  $>40$  °C a programmable thermosensitive lid opens the barrel to allow transport of small molecules across the membrane. The multifunctional DNA nanodevice may be used to create logic ionic networks or to achieve controlled drug delivery from vesicles.

**Comment:** Nanopores show unpredictable potential in different fields, e.g. biological sensors and molecules sensors. I chose this paper because the structure and construction of the nanopore. They obtained a nanopore with a lid at one side by exploiting the assembly properties of DNA. And the lid as nanovalve is temperature responsive, resulting in the transport switching ON/OFF for small molecules.

### Toolbox of Diverse Linkers for Navigating the Cellular Efficacy Landscape of Stapled Peptides

Wu, Y.; Kaur, A.; Fowler, E.; Wiedmann, M. M.; Young, R.; Galloway, W. R.; Xu, W.\* [ACS Chem. Biol. 2019](#)



Stapled peptides have great potential as modulators of protein–protein interactions (PPIs). However, there is a vast landscape of chemical features that can be varied for any given peptide, and identifying a set of features that maximizes cellular uptake and subsequent target engagement remains a key challenge. Herein, we present a systematic analysis of staple functionality on the peptide bioactivity landscape in cellular assays. Through application of a “toolbox” of diversified dialkynyl linkers to the stapling of MDM2-binding peptides via a double-click approach, we conducted a study of cellular uptake and p53 activation as a function of the linker. Minor changes in the linker motif and the specific pairing of linker with peptide sequence can lead to substantial

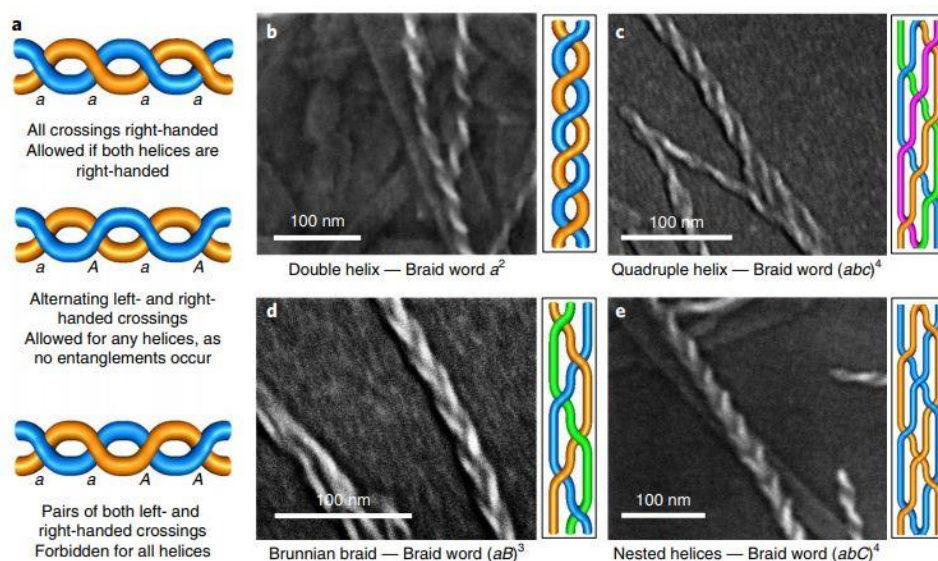


differences in bioactivity, a finding which may have important design implications for peptide-based inhibitors of other PPIs. Given the complexity of the structure–activity relationships involved, the toolbox approach represents a generalizable strategy for optimization when progressing from in vitro binding assays to cellular efficacy studies.

**Comment:** The cellular toolbox which was presented in this paper shows a good potential for the research on protein–protein interactions, which may have implications on the development of medicine.

### Braiding, Branching and Chiral Amplification of Nanofibres in Supramolecular Gels

Jones, C.; Simmons, H.; Horner, K.; Liu, K.; Steed, J.\* [Nat. Chem. 2019, ASAP.](#)



Helical nanofibres play key roles in many biological processes. Entanglements between helices can aid gelation by producing thick, interconnected fibres, but the details of this process are poorly understood. Here, we describe the assembly of an achiral oligo(urea) peptidomimetic compound into supramolecular helices. Aggregation of adjacent helices leads to the formation of fibrils, which further intertwine to produce high-fidelity braids with periodic crossing patterns. A braid theory analysis suggests that braiding is governed by rigid topological constraints, and that branching occurs due to crossing defects in the developing braids. Mixed-chirality helices assemble into relatively complex, odd-stranded braids, but can also form helical bundles by undergoing inversions of chirality. The oligo(urea) assemblies are also highly sensitive to chiral amplification, proposed to occur through a majority-rules mechanism, whereby trace chiral materials can promote the formation of gels containing only homochiral helices.

**Comment:** The beautiful braid image attracted my attention but it's not the only reason for it being shared. After reading it, we could have better understanding of how helix chirality affects the topology of a braid.