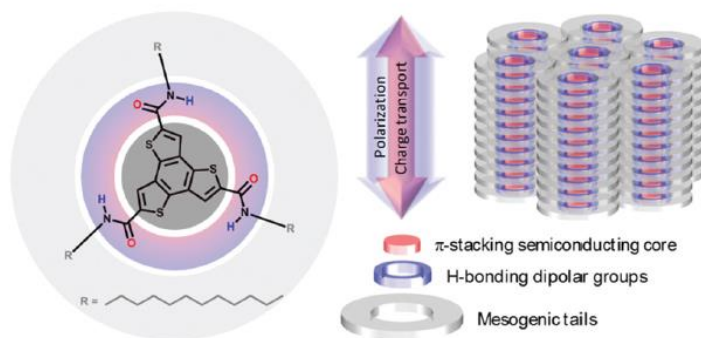


## Resistive Switching in an Organic Supramolecular Semiconducting Ferroelectric

Casellas, M. N.; Urbanaviciute, I.; Cornelissen, T. D.; Berrocal, J. A.; Torres, T.\*; Kemerink, M.\*; García-Iglesias, M.\* [Chem. Commun. 2019, ASAP](#)

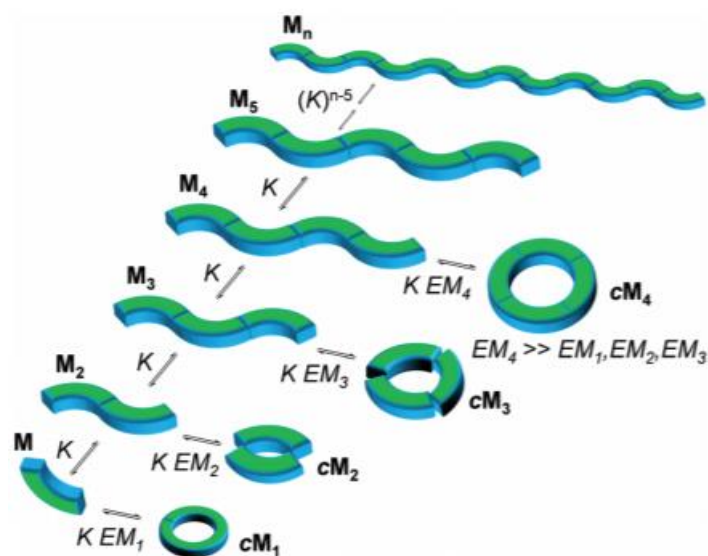


The combination of switchable dipolar side groups and the semiconducting core of the newly synthesized C3-symmetric benzotrithiophene molecule (BTTTA) leads to an ordered columnar material showing continuous tunability from injection- to bulk-limited conductivity modulation.

**Comment:** Like triarylamine, the self-assemblies of BTTTA in this paper also exhibit conducting property. They did comprehensive characterizations from morphology to properties.

## Guidelines for the Assembly of Hydrogen-Bonded Macrocycles

Aparicio, F.; Mayoral, M. J.; Montoro-García, C.; González-Rodríguez, D\* [Chem. Commun. 2019, ASAP](#)



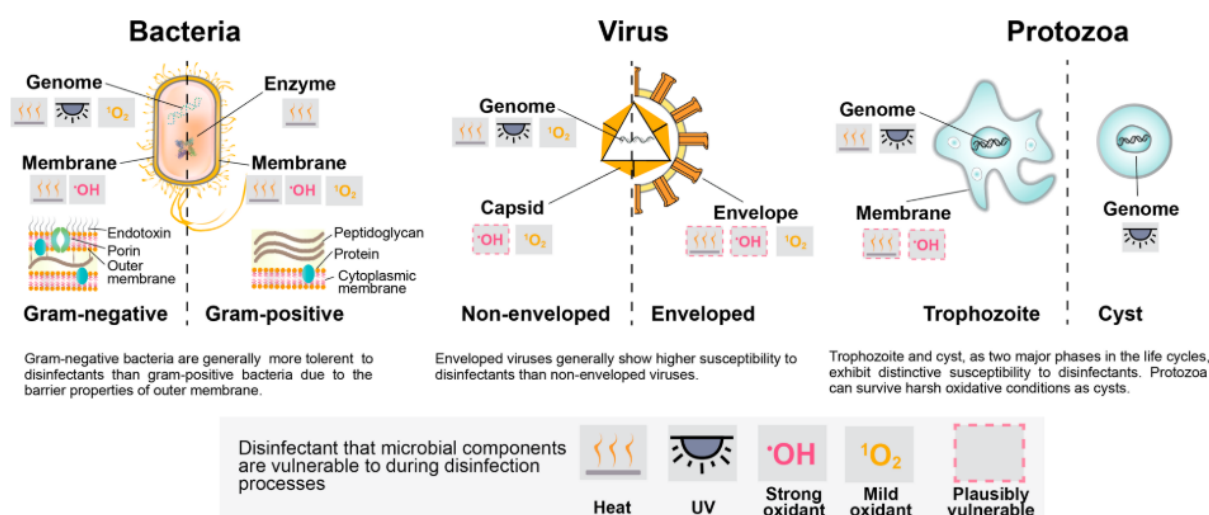
The formation of well-defined, discrete self-assembled architectures relies on the interplay between non-covalent interactions and cooperative phenomena. In particular, chelate or

intramolecular cooperativity is responsible for the assembly of closed, cyclic structures in competition with open, linear oligomers, and it can be enhanced in several ways to increase the stability of a given cycle size. **In this article, we review the work of several researchers on the synthesis of hydrogen-bonded macrocycles from ditopic molecules and analyze the main factors, often interrelated, that influence the equilibrium between ring and chain species.** Emphasis will be set on the diverse features that can increase cyclization fidelity, including monomer geometry, template effects, conformational effects, intramolecular interactions and H-bonding pattern.

**Comment:** I (highly!) recommend this feature article because it includes so many examples about hydrogen bonding macrocycles, which are composed by different components with distinct hydrogen bonds and angles. This article could provide some knowledge for our further molecular design, especially, for self-assembly of small molecules or dynamic polymers.

## Water Disinfection in Rural Areas Demands Unconventional Solar Technologies

Chu, C.; Ryberg, E. C.; Loeb, S. K.; Suh, M.-J.; Kim, J.-H.\* [Acc. Chem. Res. 2019, 52, 5, 1187-1195.](#)



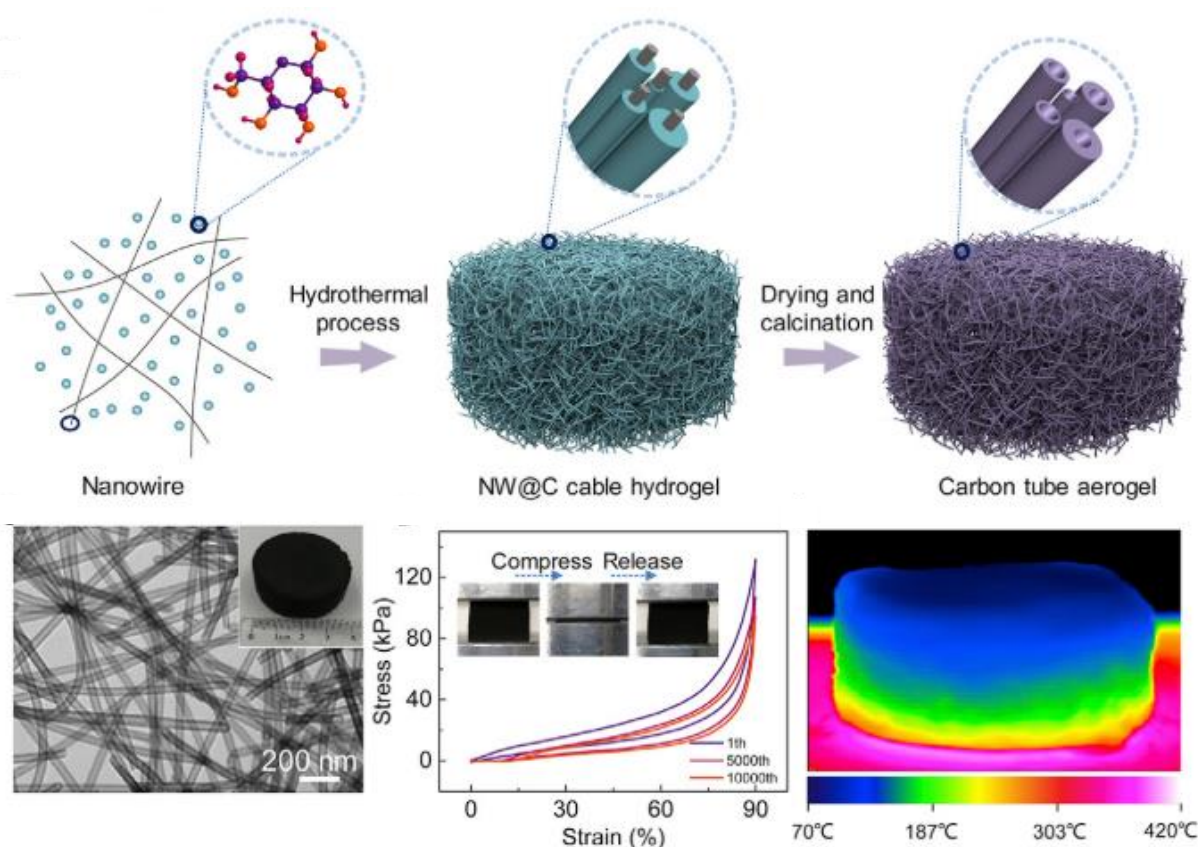
Providing access to safe drinking water is a prerequisite for protecting public health. Vast improvements in drinking water quality have been witnessed during the last century, particularly in urban areas, thanks to the successful implementation of large, centralized water treatment plants and the distribution of treated water via underground networks of pipes. Nevertheless, infection by waterborne pathogens through the consumption of biologically unsafe drinking water remains one of the most significant causes of morbidity and mortality in developing rural areas. In these areas, the construction of centralized water treatment and distribution systems is impractical due to high capital costs and lack of existing infrastructure. **Improving drinking water quality in developing rural areas** demands a paradigm shift to **unconventional, innovative water disinfection strategies** that are low cost and simple to implement and maintain, while also requiring minimal infrastructure.

**Comment:** This paper is, obviously, absolutely not related to the different subjects we are working on in our team. This article, however, allows to have an overview of a simple problem which is the access to drinkable water. It is taken for granted in our society but remains an important challenge in other countries. Here, the authors briefly sum up the different advances in

this field during the past century and how it impacted humankind. Then they present different approaches based on solar energy in order to purify water in rural areas. The chemistry behind these processes is quite simple but I think it is always useful to keep in mind such practical implementations given that our team is mainly focused on fundamental chemistry. At the end, I would recommend reading this paper just for the general knowledge it can offer.

### Biomimetic Carbon Tube Aerogel Enables Super-Elasticity and Thermal Insulation

Zhan, H.-J.; Wu, K.-J.; Hu, Y.-L.; Liu, J.-W.; Li, H.; Guo, X.; Xu, J.; Yang, Y.; Yu, Z.-L.; Gao, H.-L.; et al. *Chem* 2019, ASAP.



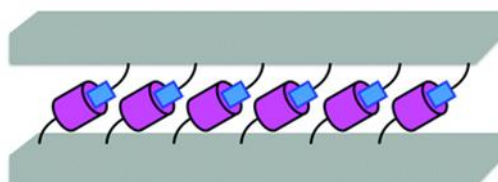
Inspired by microstructures of polar bear hair, herein, we describe a simple solution-based strategy to fabricate a **macroscopic-scale** and **lightweight carbon tube aerogel with super-elasticity** and **excellent thermal insulation**. The microstructure-derived thermal conductivity and super-elasticity are strongly dependent on the shell thickness of the interconnected tubes, as well as the aperture of the aerogel. Remarkably, the optimized aerogel can maintain structural integrity after more than **one million compress-release cycles at 30% strain and 10,000 cycles at 90% strain**. Moreover, this biomimetic aerogel offers a fast and accurate dynamic piezoresistive response to broad bandwidth frequency forces. Particularly, the super-elasticity is further confirmed by its fastest **rebounding speed of 1,434 mm s<sup>-1</sup>** among the traditional elastic materials measured by a standard falling steel ball. Furthermore, the optimized minimum **thermal conductivity is as low as 23 mW m<sup>-1</sup> K<sup>-1</sup>** which performs better than the thermal conductivity of dry air.

**Comment:** This paper describes an amazing system which possesses promising mechanical properties. From the synthesis to the characterization, the authors performed a complete study of

this material. In this aspect, I definitely recommend reading this article, as it is short but easily understandable. Thus, it can be really useful to all the rookies in material chemistry.

### Adhesive supramolecular polymeric materials constructed from macrocycle-based host-guest interactions

Ji, X.; Ahmed, M.; Long, L.;\* Khashab, N. M.;\* Huang, F.;\* Sessler, J. L.\* [\*Chem. Soc. Rev.\* \*\*2019\*\*, \*48\*, 2682-2697.](#)

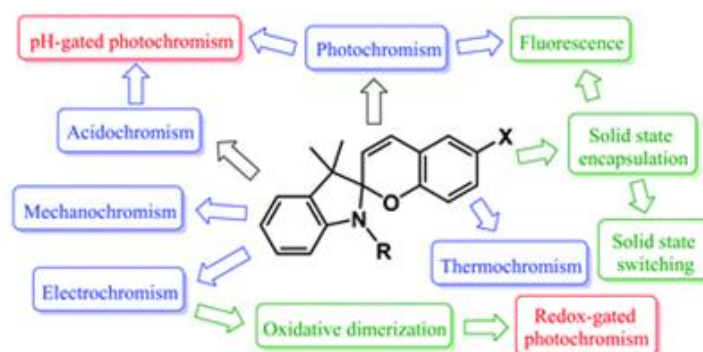


Nature uses the power of non-covalent interactions as the basis for many kinds of adhesion phenomena. Inspired by nature, scientists have prepared various synthetic adhesive materials that rely on a number of non-covalent interactions at the interfaces. Commonly used non-covalent interactions include hydrogen bonding,  $\pi$ - $\pi$  stacking, charge transfer interactions, electrostatic interactions, hydrophobic interactions, macrocycle-based host-guest interactions, among others. Within this context, macrocycle-based host-guest interactions are of particular interest. Often they give rise to distinct properties, such as multiple combined noncovalent interactions and a diversity of stimuli-based responsiveness. In this tutorial review, we will summarise recent advances in adhesive supramolecular polymeric materials that rely primarily on macrocycle-based host-guest interactions. An overview of future challenges and a perspective of this sub-field are also provided.

**Comment:** This is a summary for those who are interested in adhesive supramolecular polymeric materials based on host-guest interactions. The host-guest interactions are described based on macrocycle used: cyclodextrin, crown ether, cucurbituril, tetracationic imidazolium and calix[4]pyrrole.

### The evolution of spiropyran: fundamentals and progress of an extraordinarily versatile photochrome

Kortekaas, L.; Browne, W. R.\* [\*Chem. Soc. Rev.\*, \*\*2019\*\*, \*Advance Article\*.](#)

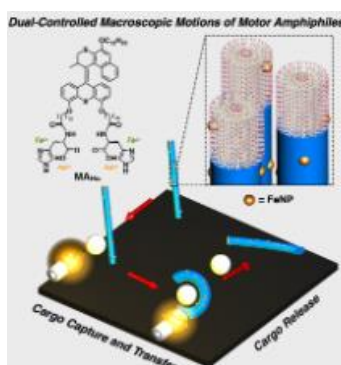


Spiropyrans have played a pivotal role in the emergence of the field of chromism following their discovery in the early 20th century, with almost ubiquitous use in materials applications especially since their photochromism was discovered in 1952. Their versatility continues to lend them to application in increasingly diverse fields not least due to recent discoveries of properties that have expanded their utility extensively. This review provides an overview of their rich history and highlights the contemporary relevance of the spiropyran.

**Comment:** A new review about the photochrome spiropyran describing their history and their multifunctionality.

### Dual-Controlled Macroscopic Motions in A Supramolecular Hierarchical Assembly of Motor Amphiphiles

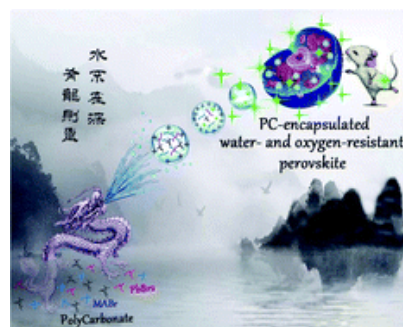
Leung, F. K.-C.; Kajitani, T.; Stuart, M. C. A.; Fukushima.; Feringa, B. L.\* [\*Angew. Chem. Int. Ed.\* \*\*2019\*\*, ASAP](#)



Three-dimensional unidirectionally aligned and responsive supramolecular hierarchical assemblies have much potential in adaptive materials for biomedical and soft actuator applications. However, to achieve systematical control of the motion of stimuli-responsive materials by orthogonal external stimuli and to complete a series of complicated tasks remains a grand challenge. Herein, we demonstrate a novel designed hybrid supramolecular assembly of molecular motor amphiphiles that also serves as a template for iron nanoparticles growth, and as a consequence this soft hybrid material is orthogonally controlled by dual light/magnetic stimuli. Macroscopic motor amphiphile strings, decorated with iron nanoparticles, provide fast response photoactuations and magnet induced movements that allows a precisely controlled cargo transport process.

### Accelerated Crystallization and Encapsulation for the Synthesis of Water- and Oxygen-Resistant Perovskite Nanoparticles in Micro-Droplets

Long, Z. Wang, Y., Fu, Q., Ouyang, J., Lixin, H., Na, N. [\*Nanoscale\*, \*\*2019\*\*, \*11\*, 11093–11098](#)

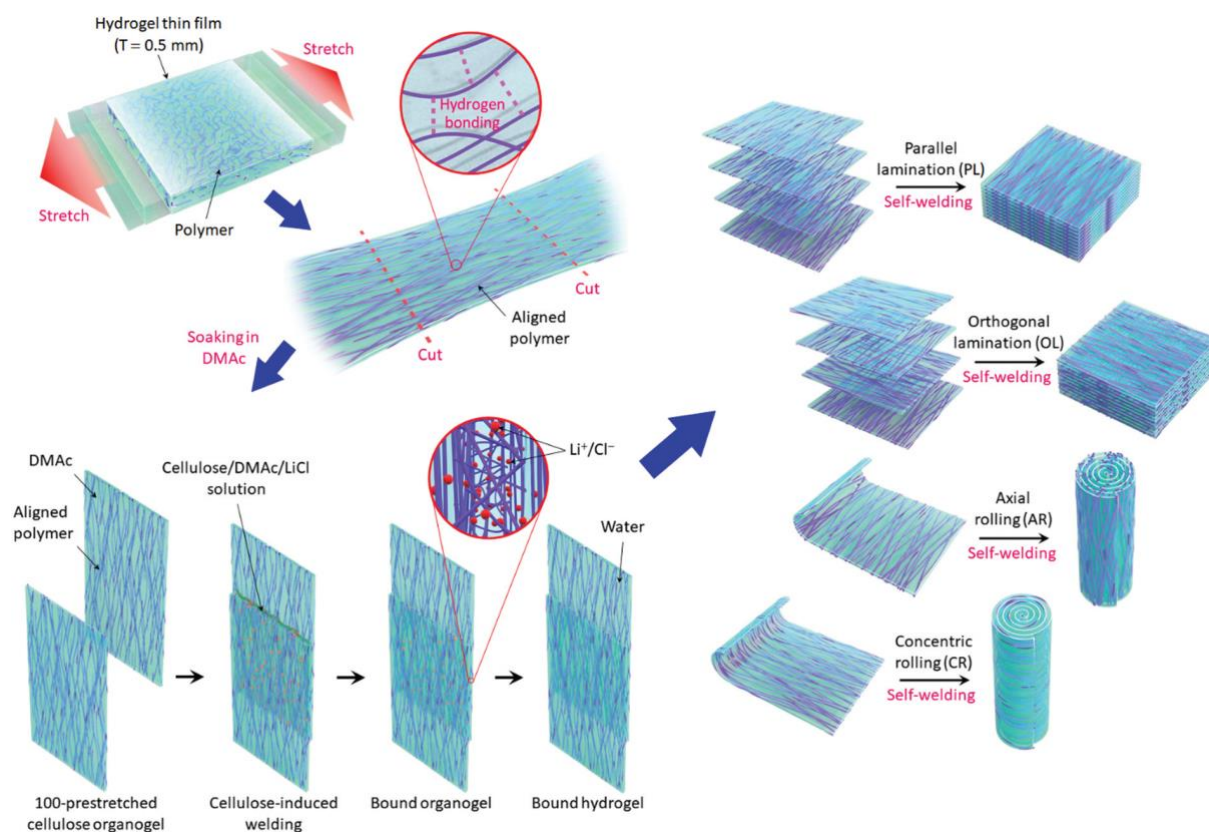


Water- and oxygen-resistant perovskite nanoparticles (PC-PNPs) were synthesized in milliseconds via a single-step microdroplet reaction. It was found through kinetics studies that the synthesis was accomplished within 100 ms in microdroplet reactors, with ejection at  $\sim 1.5 \text{ m s}^{-1}$ . Being small-sized, stable, less toxic and biocompatible, PC-PNPs were feasible for cell or in vivo imaging, and selective sensing for  $\text{Fe}^{3+}$  and Cys, which will inspire further expanded studies of perovskite materials.

**Comment:** I definitely do not understand why there is a purple dragon spitting polycarbonate to a microdroplet hold by a dancing mouse, with Chinese annotations and green glitters in supplement...

## Anisotropic Tough Multilayer Hydrogels with Programmable Orientation

M. T. I. Mredha; H. H. Le; V. T. Tran; P. Trtik; J. Cui\*; I. Jeon\* *Mater. Horiz.* 2019, ASAP



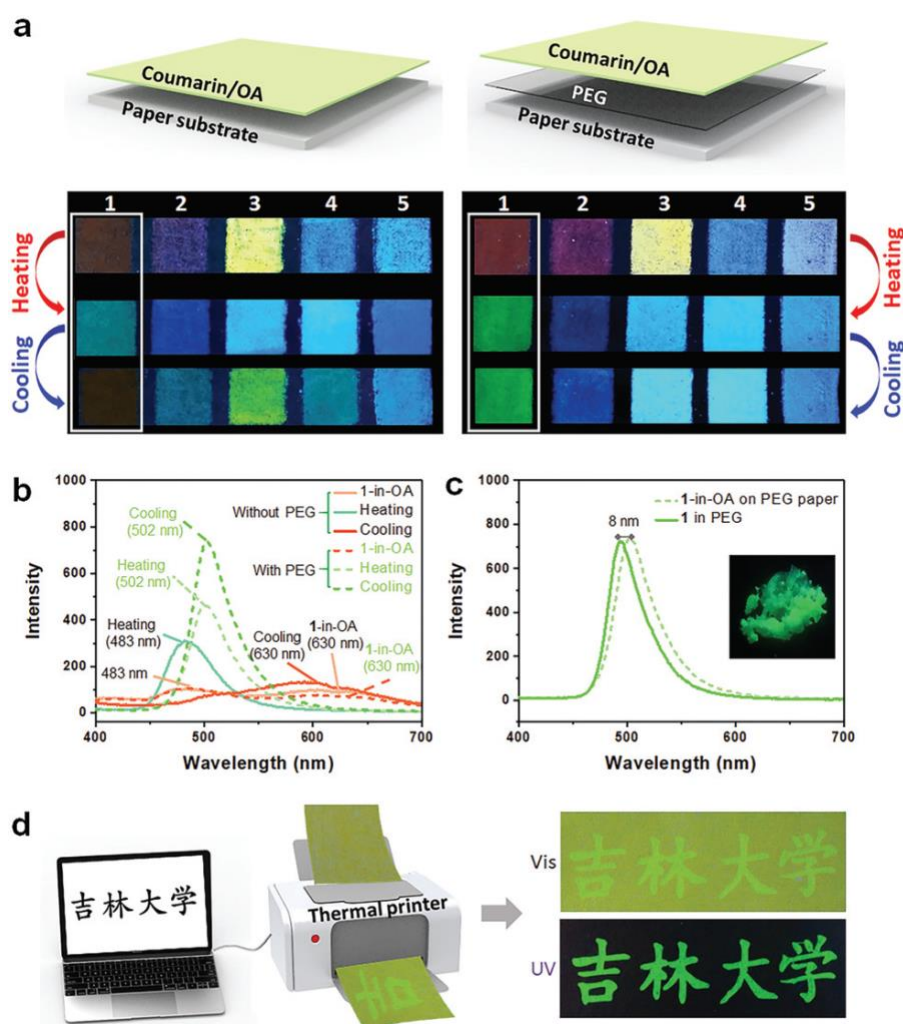
Hydrogels are normally isotropic and weak, in contrast to their natural analogs that possess hierarchically oriented structure and excellent mechanical properties. Inspired by the unique structure of natural materials, we develop an approach for preparing highly anisotropic hydrogels with programmable oriented polymer structure and extraordinary mechanical properties. Our method is based on a novel welding technique for stretched cellulose hydrogel films, in which interfacial reconfiguration occurs, allowing full integration without compromising the highly aligned polymer orientation. We demonstrate the versatility of this method by fabricating four types of anisotropic tough multilayer hydrogels with differently oriented hierarchies: parallel lamination ( $\parallel$ ), orthogonal lamination ( $\perp$ ), axial rolling, and concentric rolling. These high-water-content hydrogels ( $\sim 68 \text{ wt}\%$ ) exhibit extremely high anisotropy (birefringence  $\geq 0.006$ , the highest reported

for hydrogels) and superior mechanical properties (Young's modulus of  $\sim 140$  MPa, tensile strength of  $\sim 47$  MPa, and work of extension of  $\sim 20$  MJ m $^{-3}$ ). Moreover, these hydrogels also show interesting anisotropic electrical conductivity and asymmetric shape deformation. Because this method is easy to apply and offers flexibility in designing complex hierarchical hydrogel structures, our work opens a new window to designing novel hydrogel materials for engineering and biomedical applications.

**Comment:** Orienting polymer by stretching to obtain programmable hydrogen orientation is quite interesting. Different oriented hydrogel layers can be connected to each other by welding methods without changing its orientation, which is an attractive concept. A better understanding of welding is realized in this paper.

### Simple and General Platform for Highly Adjustable Thermochromic Fluorescent Materials and Multi-feasible Applications

Du, J.; Sheng, L.\*; Chen, Q.; Xu, Y.; Li, W.; Wang, X.; Li, M.; Zhang, S. X.-A.\*; *Mater. Horiz.* **2019**, *ASAP*



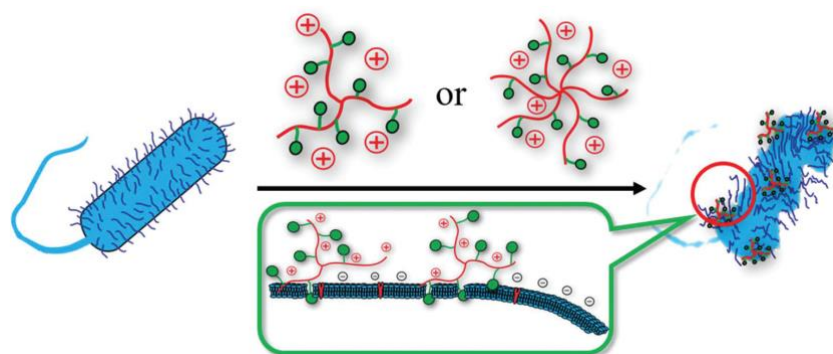
Even though numerous thermochromic fluorescent materials (TFMs) have been reported, effective and universal strategies for the development of ideal TFMs with simple preparation, low cost, excellent performance and practicability are still urgently required. Herein, we report a simple

and general platform for such TFMs via taking biomimetic lipid like fatty acids (FAs) as an effective matrix which is applicable to abundant simple fluorophores. Change of crystallinity of FAs induced by phase transition regulates the reversible interconversion between non-dispersion and dispersion states of fluorescent dyes (e.g., coumarins), thus leading to obvious fluorescence (FL) change of the TFMs based on both. The reaction mechanism was proved in detail by FL spectra, XRD and SEM techniques. The substituent effect of coumarins and the nature of FAs play important roles in adjusting the properties of the TFMs, such as FL colour and responsive temperature. Additionally, their multiple feasible potential applications (i.e., in situ thermometers, thermal response FL papers, FL colour change crayon, anti-counterfeiting and encryption, erasable or long-preservative thermal printing) with selective multi-mode (i.e., groove-fill, writable pen, loading on paper substrate) were developed. These TFMs with the merit of easy preparation, low cost, obvious colour change (ca. 100 nm shifts), fast response speed, good reversibility and high adjustability are very close to industrial application requirements. The design and preparation principles from this work should facilitate the development of other novel stimuli-responsive fluorescent materials.

**Comment:** This work shows a new way to control the fluorescence color changes of thermochromic fluorescent materials (TFMs) by using the biomimetic lipid-like fatty acids (FAs) as an effective matrix. The idea is based on the phase transition of TFMs in those matrixes from dispersion to non-dispersion upon heating and cooling. Such simple ideas can have big applications in materials and that is great.

### Star-Shaped Polypeptides Exhibit Potent Antibacterial Activities

Chen, Y-F., Lai, Y-D., Chang, C-H., Tsai, Y-C., Tanga, C-C., Jan., J-S.\* [Nanoscale, 2019, ASAP](#)



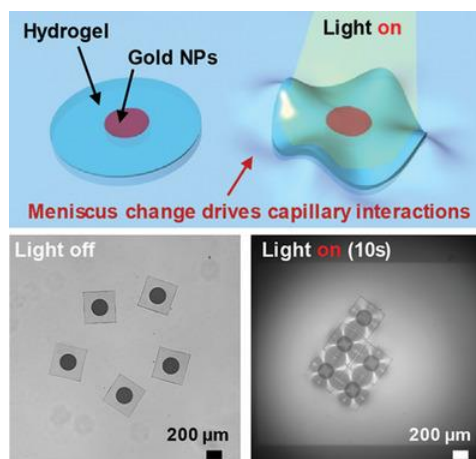
Peptide-based biomaterials are a promising class of **antimicrobial agents** that work by physically damaging bacterial cell membranes rather than targeting intracellular factors, resulting in less susceptibility to drug resistance. Herein we report the **synthesis of cationic, star-shaped polypeptides with 3 to 8 arms** and their evaluation as antimicrobial agents against different types of bacteria. The effects of the arm number and side chain group on their antimicrobial activities were systematically investigated. Compared to their linear counterparts, these star-shaped polypeptides exhibited potent antibacterial activity (which may involve adhesion and disruption processes). The increase of the arm number can efficiently increase the antibacterial activities up until 8 arms, which did not exhibit further improvement of antibacterial activities. Poly(Llysine) (PLL) modified with an indole group (PLL-g-indo) exhibited the best antibacterial activity among all grafted copolypeptides and improved cytotoxic selectivity towards pathogens over mammalian cells without compromising their hemolytic activities. In vivo studies showed that the star-shaped PLL-g-indo can effectively suppress Enterohaemorrhagic E. coli (EHEC) infection and attenuate the clinical symptoms in mice, suggesting that they are promising antimicrobial agents.



**Comment:** This article is very complete, describing as well synthesis and characterization of sequence-defined star shape polypeptides (NMR, GPS-LS), as biological properties with in vitro and in vivo tests. The development of such structures is a promising way to fight bacteria without developing antimicrobial resistance, as they allow the physical damage of bacterial cell membranes.

### Light-Driven Shape Morphing, Assembly, and Motion of Nanocomposite Gel Surfers

Kim, H.; Kang, H.-J.; Zhou, Y.; Kuenstler, A. S.; Kim, Y.; Chen, C.; Emrick, T.;\* Hayward, R. C.\* [\*Adv. Mater.\* 2019, ASAP](#)

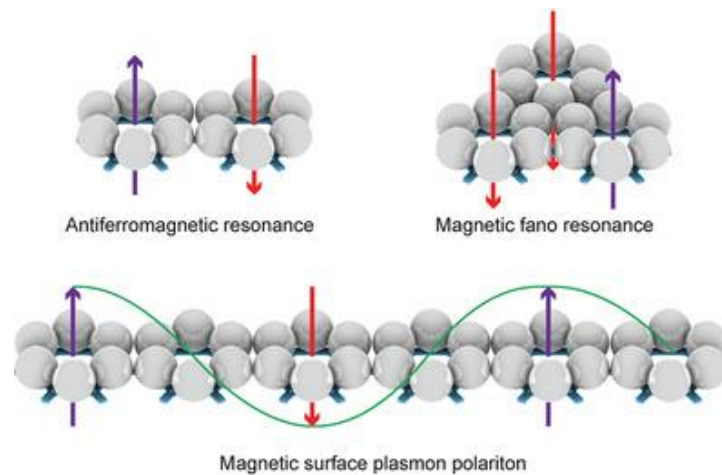


Patterning of nanoparticles (NPs) via photochemical reduction within thermally responsive hydrogel films is demonstrated as a versatile platform for **programming light-driven shape morphing and materials assembly**. Responsive hydrogel disks, containing patterned metal NPs, form characteristic wrinkled structures when illuminated at an air/water interface. The resulting distortion of the three-phase (air/water/hydrogel) contact lines induces **capillary interactions** between two or more disks, which are either attractive or repulsive depending on the selected pattern of light. By programming the shapes of the NP-rich regions, as well as of the hydrogel objects themselves, the number and location of attractive interactions are specified, and the assembly geometry is controlled. Remarkably, appropriately patterned illumination enables sustained rotation and motion of the hydrogel disks. Overall, these results offer insight into a wide variety of shape-programmable materials and capillary assemblies, simply by controlling the NP patterns and illumination of these soft materials.

**Comment:** Although not well known to me, controlled swelling of hydrogels giving rise to capillary attractions to generate motion of nano and micro objects has recently been studied to design micro robots and swimmers. However, up to now we did not control single objects of such an ensemble. Here, by introducing spatial-patterned **Au NPs**, the authors realized reconfigurable capillary assembly and motion through lateral temperature gradients. I think they explain the mechanisms of wrinkling in a clear manner, they provide multiple hypotheses which they test (theory and experimental) and conclude on.

### Magnetic Plasmon Networks Programmed by Molecular Self-Assembly

Wang, P.;\* Huh, J.-H.; Lee, J.; Kim, K.; Park, K. J.; Lee, S.::\* Ke, Y\* [\*Adv. Mater.\* 2019, ASAP](#)

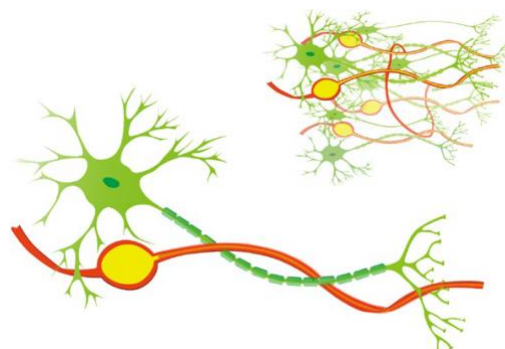


**Nanoscale manipulation of magnetic fields** has been a long-term pursuit in plasmonics and metamaterials, as it can enable a range of appealing optical properties, such as high-sensitivity circular dichroism, directional scattering, and low-refractive-index materials. Inspired by the natural magnetism of aromatic molecules, the cyclic ring cluster of plasmonic nanoparticles (NPs) has been suggested as a promising architecture with induced unnatural magnetism, especially at visible frequencies. However, it remains challenging to assemble plasmonic NPs into complex networks exhibiting strong visible magnetism. Here, a **DNA-origami-based strategy** is introduced to **realize molecular self-assembly of NPs forming complex magnetic architectures**, exhibiting emergent properties including **anti-ferromagnetism**, purely **magnetic-based Fano resonances**, and **magnetic surface plasmon polaritons**. The basic building block, a gold NP (AuNP) ring consisting of six AuNP seeds, is arranged on a DNA origami frame with nanometer precision. The subsequent hierarchical assembly of the AuNP rings leads to the formation of higher-order networks of clusters and polymeric chains. Strong emergent **plasmonic properties are induced by in situ growth of silver upon the AuNP seeds**. This work may facilitate the development of a tunable and scalable DNA-based strategy for the assembly of optical magnetic circuitry, as well as plasmonic metamaterials with high fidelity.

**Comment:** Similar architectures have already been described in literature. However, the structures were relatively large and their organization into complex networks was limited. The authors overcome these limitations by making use of a DNA-origami-based approach. I found, both calculations and experimental results difficult to understand and interpret.

### Bioinspired neuron-like electronics

Yang, X.; Zhou, T.; Zwang, T. J.; Hong, G.; Zhao, Y.; Viveros, R. D.; Fu, T.-M.; Gao, T.; Lieber, C. M.\* [Nat. Mater. 2019, 18, 510-517](#)

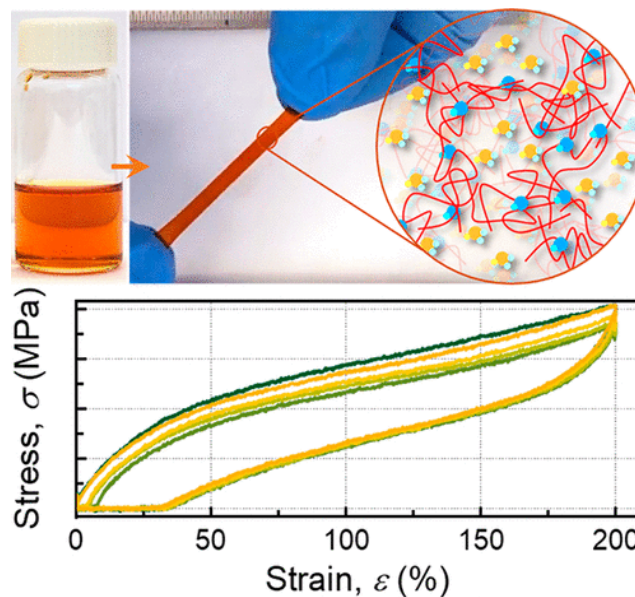


As an important application of functional biomaterials, **neural probes** have contributed substantially to studying the brain. Bioinspired and biomimetic strategies have begun to be applied to the development of neural probes, although these and previous generations of probes have had structural and mechanical dissimilarities from their neuron targets that lead to neuronal loss, neuro inflammatory responses and measurement instabilities. Here, we present a bioinspired design for neural probes - neuron-like electronics (NeuE) - where the **key building blocks mimic the subcellular structural features and mechanical properties of neurons**. Full three-dimensional mapping of implanted NeuE–brain interfaces highlights the structural indistinguishability and intimate interpenetration of NeuE and neurons. Time-dependent histology and electrophysiology studies further reveal a structurally and functionally stable interface with the neuronal and glial networks shortly following implantation, thus opening opportunities for **next-generation brain–machine interfaces**. Finally, the NeuE subcellular structural features are shown to facilitate migration of endogenous neural progenitor cells, thus holding promise as an **electrically active platform for transplantation-free regenerative medicine**.

**Comment:** Although not directly connected to the fields of research in our group, I think this is an intriguing interdisciplinary subject (micro-, nano electronics, material science and biology).

### A Highly Stretchable Polyacrylonitrile Elastomer with Nanoreservoirs of Lubricant Using Cyano-Silver Complexes

Zhang, S.; Hao, A.; Liu, Z.; Park, J. G.; Liang, R. [Nano Lett. 2019, ASAP.](#)



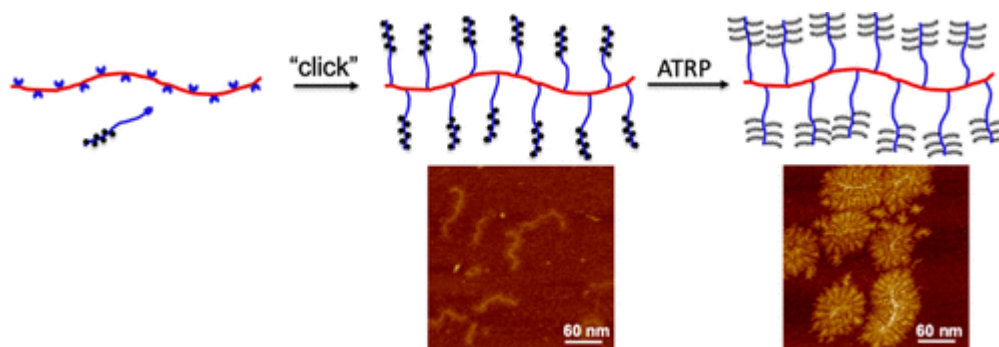
Stretchable materials are indispensable for applications such as deformable devices, wearable electronics, and future robotics. However, designs for new elastomers with high stretchability have undergone only limited research. Here we have fabricated highly stretchable  $\text{Ag}^+$ /polyacrylonitrile elastomer with nanoreservoirs of lubricant using cyano-silver complexes. The prepared products feature nanoconfinement structures of lubricant surrounded by polymer chains with coordination bond through chelates of cyano-silver, resulting in an enhanced stretchability of more than 600% from 2%. The elastomeric properties were investigated, and a mechanical response model was proposed, which explained the structural evolution including the polymer chain fluidity under external deformation. Also, the easy breakage and dynamic reformation of cyano-silver coordination complexes promises a strain recovery under various stretching conditions. This

elastomer itself can directly work as sensors and open paths to alternative substrates for soft electronics development.

**Comment:** This article presents a new strategy to achieve highly stretchable elastomers using only the polymer, the solvent (DMF) and  $\text{AgNO}_3$ . The polyacrylonitrile chains with coordination bonds by cyano-silver complexes create a cellular-like internal structure with nanoreservoirs of DMF endowing the system with high fluidity of the polymer chains. This novel idea could be applied to networks based on molecular motors and also to modulate the physical properties of the double-dynamic polymer system.

### Molecular Bottlebrushes Featuring Brush-on-Brush Architecture

Chen, Y.; Sun, Z.; Li, H.; Dai, Y.; Hu, Z.; Huang, H.; Shi, Y.; Li, Y.; Chen, Y. *ACS Macro Lett.* **2019**, 749–753.

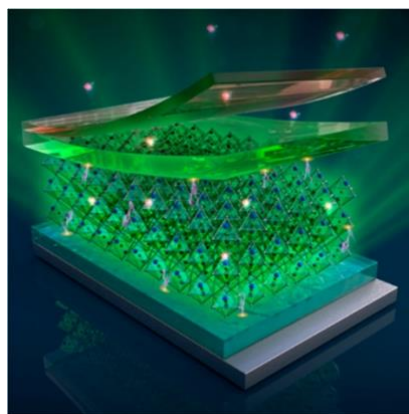


Molecular bottlebrushes featuring brush-on-brush (BoB) architecture were prepared by combining azide–alkyne click chemistry, ring-opening polymerization (ROP), and atom transfer radical polymerization (ATRP). Primary side chains of diblock copolymers with a poly( $\epsilon$ -caprolactone) (PCL) block and a poly( $\alpha$ -bromo- $\epsilon$ -caprolactone) (P(CL-Br)) block were synthesized by ROP and then grafted onto PCL backbone by the click reaction. Then the secondary side chains of poly(oligo(ethylene glycol) acrylate) (POEGA) were grafted from the P(CL-Br) block by ATRP, yielding an amphiphilic core/shell structure. Imaging of individual macromolecules by atomic force microscopy (AFM) demonstrated dramatically thickened wormlike formation with distinct hairy side chains. Interestingly, for the BoB molecular bottlebrushes with enough long primary and secondary side chains, sufficient tension can be generated along the backbone and thus lead to its cleavage.

**Comment:** The authors present the preparation of molecular bottlebrushes with a core–shell structure by combining azide–alkyne click reaction, ROP, and ATRP. As the work of Prof. Manners, the grafting of core-shell structure allows to obtain interesting properties. In this case, bottlebrushes have potential applications as molecular tensile machines and offer more options for a varied mechanochemistries.

### The Rise of Perovskite Light-Emitting Diodes

Wei, Z.; Xing, J.;\* *J. Phys. Chem. Lett.* **2019**, 10, 3035–3042

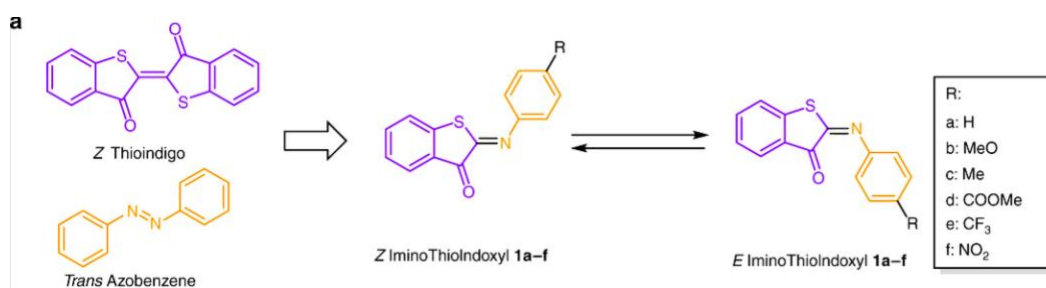


Recently, metal halide perovskite materials have attracted great interest in both photovoltaic and electroluminescent devices. The external quantum efficiency of the perovskite light-emitting diodes (pero-LEDs) has grown to over 20% within four years, and the operational lifetime has been improved to tens of hours. These achievements make pero-LEDs very promising technology in solid lighting and displays. In this Perspective, we first give a general introduction of the pero-LEDs; we then discuss various pero-LEDs with different cell configurations and perovskite emitting structures; to conclude, we propose some efficiency improvement strategies and development opportunities of pero-LEDs. Provided that we can continually improve efficiency and operational lifetime of pero-LEDs, we can make them more and more compatible with other mature technologies, like organic LEDs and inorganic LEDs, and then finally realize their practical application in daily life.

**Comment:** Every time that we take a look at the different journals something related to perovskite solar cells. But in this perspective, the authors present something that I did not know that exists which are the perovskite-LEDs, a promising technology that could be used in the future in our daily life. For those who are interested in photovoltaics and molecular electronics, this perspective could be very interesting.

### Iminothioindoxyl as a Molecular Photoswitch with 100 nm Band Separation in the Visible Range

Hoorens, M.; Medved, M.; Laurent, A.; Di Donato, M.; Fanetti, S.; Slappendel, L.; Hilbers, M.; Feringa, B.\*; Jan Buma, W.\*; Szymanski, W.\* *Nat. Commun.* **2019**, *10*, 2390.



Light is an exceptional external stimulus for establishing precise control over the properties and functions of chemical and biological systems, which is enabled through the use of molecular photoswitches. Ideal photoswitches are operated with visible light only, show large separation of absorption bands and are functional in various solvents including water, posing an unmet challenge. Here we show a class of fully-visible-light-operated molecular photo-switches, Iminothioindoxyls

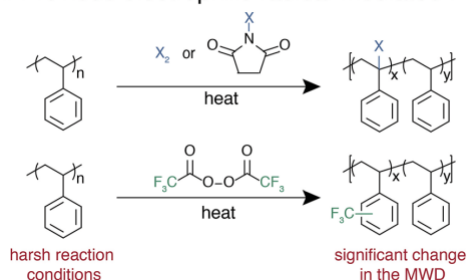
(ITIs) that meet these requirements. ITIs show a band separation of over 100 nm, isomerize on picosecond time scale and thermally relax on millisecond timescale. Using a combination of advanced spectroscopic and computational techniques, we provide the rationale for the switching behavior of ITIs and the influence of structural modifications and environment, including aqueous solution, on their photochemical properties. **This research paves the way for the development of improved photo-controlled systems for a wide variety of applications that require fast responsive functions.**

**Comment:** In this work, the authors describe the **discovery** of iminothioindoxyls which are formed by the combination of photochromic dyes thioindigo and azobenzene. These synthetically accessible visible-light photoswitches with excellent photochemical properties show very fast switching and an absorption band separation of photo-isomers of 100 nm. Also, these photoswitches can be used in aqueous media which opens opportunities for applying these new compounds in biological systems.

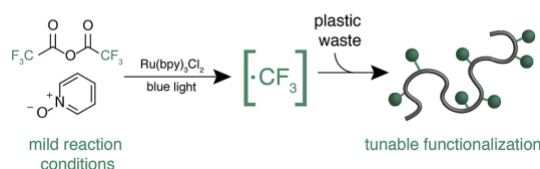
### Upcycling Aromatic Polymers through C–H Fluoroalkylation

Lewis, S. E.; Wilhelmy, B. E.; Leibfarth, F. A. [Chem. Sci. 2019, ASAP.](#)

#### A. Previous electrophilic radical-mediated PPM



#### B. This work: controlled functionalization of commodity plastics



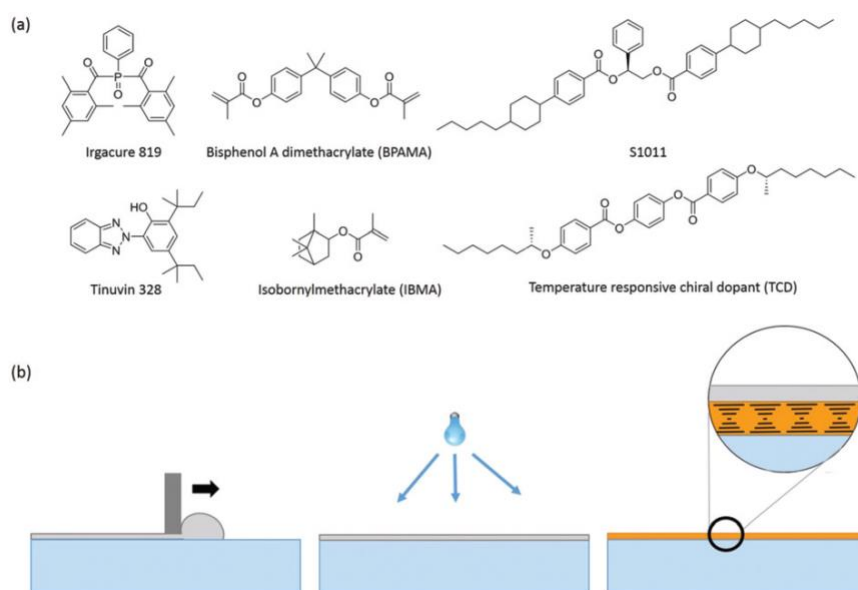
The unique properties imparted by planar, rigid aromatic rings in synthetic polymers make these macromolecules useful in a range of applications, including disposable packaging, aerospace materials, flexible electronics, separation membranes, and engineering thermoplastics. The thermal and chemical stability of aromatic polymers, however, makes it difficult to alter their bulk and/or surface properties and results in challenges during recycling. In response, we report a platform approach for the **C–H functionalization of aromatic polymers** by taking advantage of their innate reactivity with **electrophilic radical intermediates**. The method uses mild reaction conditions to photocatalytically generate electrophilic fluoroalkyl radicals for the functionalization of an array of commercially relevant polyaromatic substrates, including post-industrial and post-consumer plastic waste, without altering their otherwise attractive thermomechanical properties. The density of fluorination, and thus the material properties, is tuned by either increasing the reagent concentration or incorporating longer perfluoroalkyl species. Additionally, the installation of versatile chemical functionality to aromatic polymers is demonstrated through the addition of a bromodifluoromethyl group, which acts as an initiator for atom transfer radical polymerization (ATRP) grafting of vinyl polymers. The method described herein imparts new and versatile chemical functionality to aromatic polymers, enabling an efficient approach to diversify the

properties of these otherwise recalcitrant commodity plastics and demonstrating a **viable pathway to upcycle post-consumer plastic waste**.

**Comment:** Recycling polymers is a very challenging topic. This article describes an **upcycling** but the authors make no mention of the polymer topology: they do not recycle thermosets so, basically, they recycle already recyclable materials. My second concern is the use of Ru-based catalyst in an industrial process to upcycle cheap polymers like polystyrenes.

### Paintable Temperature-Responsive Cholesteric Liquid Crystal Reflectors Encapsulated on a Single Flexible Polymer Substrate

Khandelwal, H.; van Heeswijk, E. P. A.; Schenning, A. P. H. J.; Debije, M. G. *J. Mater. Chem. C* **2019**, *328*.



This work describes the fabrication of temperature-responsive light reflectors deposited on flexible single substrates, a photonic cholesteric liquid crystal system encapsulated by a protective polymer layer generated by photo-enforced stratification. A bendable orange reflector was fabricated by this single application step. Furthermore, a blue shift of 400 nm of an infrared cholesteric reflector was demonstrated upon increasing the temperature from room temperature to 100°C. Such reflectors on flexible substrates could be used for the retrofitting of existing windows to save energy and visible reflectors in clothing or other aesthetically pleasing applications.

**Comment:** The process is simple but very efficient: the phase separation of this polymer is well known but very efficient for encapsulation. This reflecting coating is interesting thanks to its easy process: only a curing step with light is required. The authors propose an application for cloths: this coating changes its color when bended... I can't wait to see it!