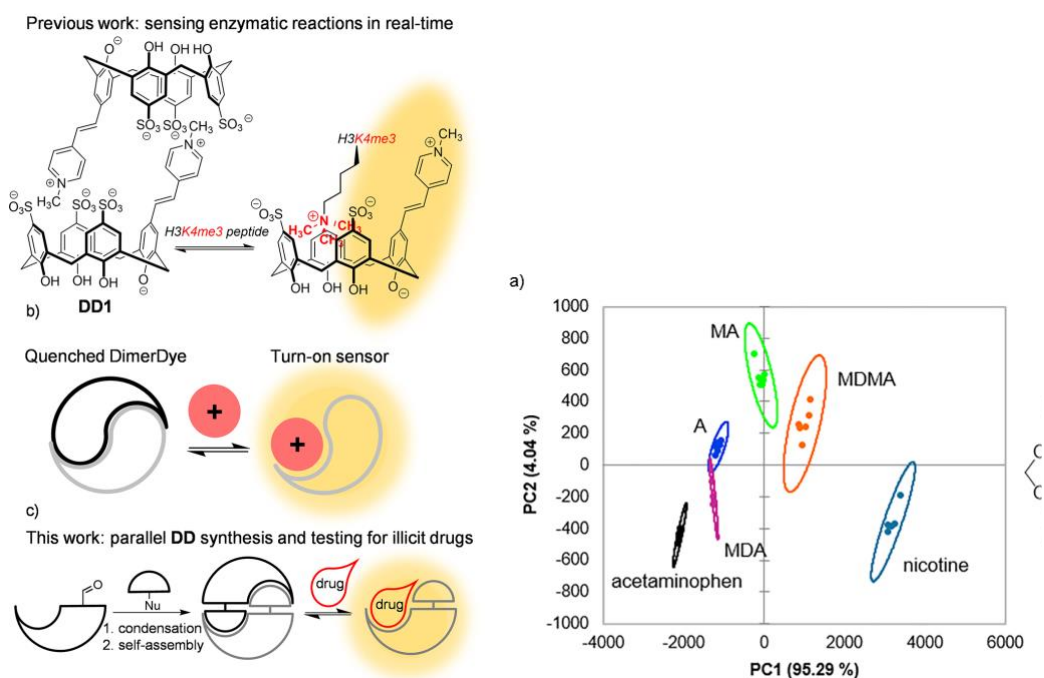


Parallel Synthesis and Screening of Supramolecular Chemosensors That Achieve Fluorescent Turn-on Detection of Drugs in Saliva

Beatty, M. A.; Selinger, A. J.; Li, Y.; Hof, F.* *J. Am. Chem. Soc.* **2019**, *141*, 16763–16771.

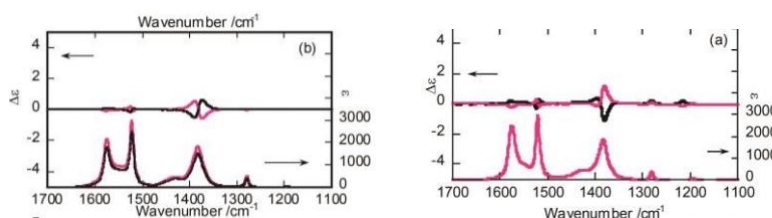


Programming and controlling molecular recognition in aqueous solutions is increasingly common, but creating supramolecular sensors that detect analytes in biologically relevant solutions remains a nontrivial task. We report here a parallel synthesis-driven approach to create a family of self-assembling dimeric sensors that we call DimerDyes and its use for the rapid identification of salt-tolerant sensors for illicit drugs. We developed an efficient method that involves parallel synthesis and screening in crude form without the need to purify each potential sensor. Structurally diverse “hit” DimerDyes were resynthesized and purified and were each shown to assemble into homodimers in water in the programmed way. DimerDyes provided a “turn-on” fluorescence detection of multiple illicit drugs at low micromolar concentrations in water and in saliva. The combination of multiple agents into a sensor array was successfully able to detect and discriminate between closely related drugs and metabolites in multiple important drug families.

Comment: Pattern recognition, basically the way your nose works to identify virtually any smell – and distinguish among them – has an intrinsic beauty that fascinates me. It is weird that if we understand this relatively well we refuse to see it beyond this narrow field.

A New Horizon for Vibrational Circular Dichroism Spectroscopy: A Challenge for Supramolecular Chirality

Sato, H. *Phys. Chem. Chem. Phys.* **2020**, ASAP.

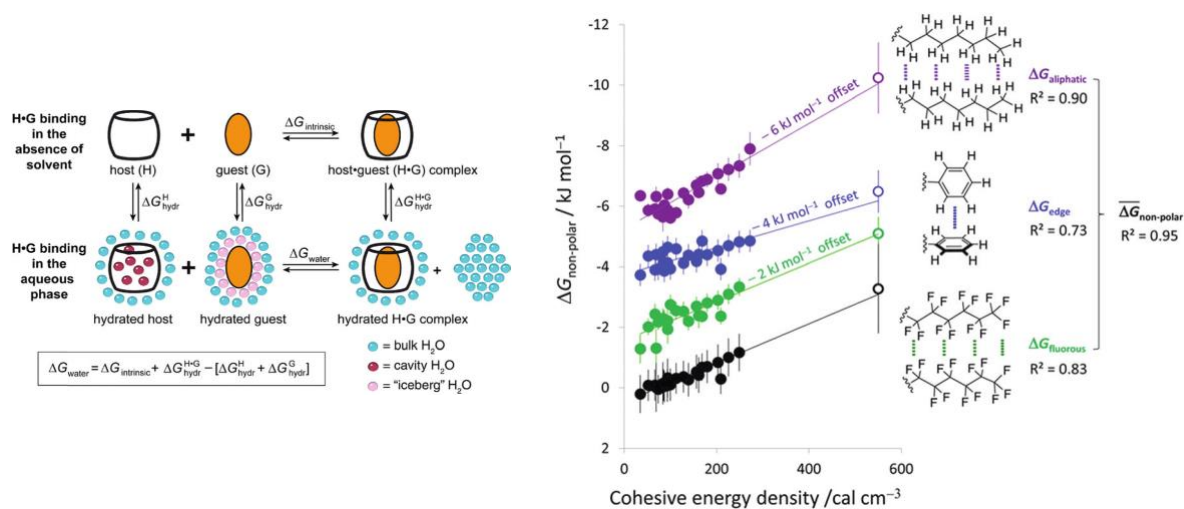


Vibrational circular dichroism (VCD) spectroscopy is an extension of circular dichroism spectroscopy into the infrared and near-infrared regions where vibrational transitions occur in the ground electronic state of a molecule. The method offers the advantage of studying the chiroptical properties of a wide range of molecules in non-crystalline states. However, because of the smallness of the signals, one measurement requires several hours to yield reliable results. Accordingly, its targets were limited to a stable molecule in a solution. To overcome this difficulty, our group applied the VCD method to supramolecular systems. The work was launched based on the finding that the VCD signal remarkably enhances when low-molecular mass gelators form gels. By analysing a number of well-resolved VCD peaks, the detailed conformation of a component molecule was deduced. This provided a clue to elucidating the molecular organization in supramolecular architectures. Our final goal was to clarify the route from microscopic molecular chirality to supramolecular chirality. For this purpose, a **time-step VCD measurement method was developed for the *in situ* monitoring of the progress of chirality amplification.**

Comment: VCD the less known version of circular dichroism that only few people use and even less understand. Worth a read nonetheless, is always nice to know something others are likely to not know!

Quantification of Noncovalent Interactions – Promises and Problems

Schneider, H.-J. *New J. Chem.* **2019**, *43*, 15498–15512.

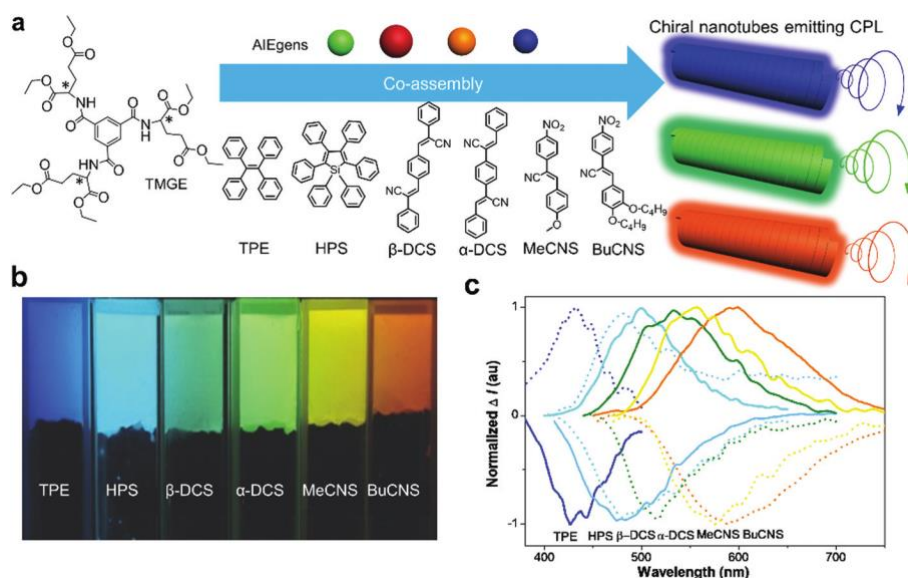


The quantification of noncovalent interactions is of paramount importance for the understanding of binding mechanisms, including biological systems, for the design of drugs and their delivery as well as for the design of host compounds for separations, sensors, actuators, smart materials etc. Although thousands of publications address noncovalent interactions it is often not clear how much these really contribute to complex formation. One complication is that the interaction between molecules is often determined by several different mechanisms. The present review intends to illustrate the presently available methods for the quantification of intermolecular interactions, mostly between organic compounds, and especially based on measurements with supramolecular complexes. Major emphasis will be given to limitations of the present methodology, which present a challenge for future work. The discussion will be grouped into several methods, from small molecule interactions to crystallographic and computational approaches, with highlights of some typical results and problems.

Comment: They often say that we should study history so that we do not repeat the errors of the past. Well, I guess it might also be true for listening to what the past generation of researchers might have to say. If someone that has spent his lifetime studying non-covalent interactions writes a review/perspective on a 'small' journal, it is certainly worth the detour.

Circularly Polarized Luminescence from AIEgens

Song, F.; Zhao, Z.; Liu, Z.; Lam, J. W. Y.; Tang, B. Z.* *J. Mater. Chem. C* 2020, 8, 3284–3301.



Circularly polarized luminescence (CPL) based on aggregation-induced emission luminogens (AIEgens) has received increasing attention owing to the increase of applications to generate CPL devices and biologically active probes. CPL-active materials fabricated from AIEgens exhibit higher photoluminescence quantum yields and larger dissymmetry factors in the solid state in comparison with single molecules in solution. We called the phenomenon of enhanced CPL in an aggregated state **aggregation-induced circularly polarized luminescence (AICPL)**. This review highlights recent research in the CPL generation of AIEgens from (macro)molecular luminogens and supramolecular assemblies based on the AIE mechanism of restriction of intramolecular motion. In addition, some cases in which CPL-active AIEgens have enabled advancement in stimulus response materials and application in CPL devices, especially circularly polarized electroluminescence. Thus, CPL-active AIEgens are highly promising candidates for a wide range of technologically oriented multidisciplinary fields.

Comment: A review by Tang and co-workers regarding AIE of circularly polarized luminescence. He is obviously very well informed on the field so I can but guess the review is comprehensive.

The future of molecular machines

Aprahamian, I.* [ACS Cent. Sci. 2020, 6, 347-358](#)

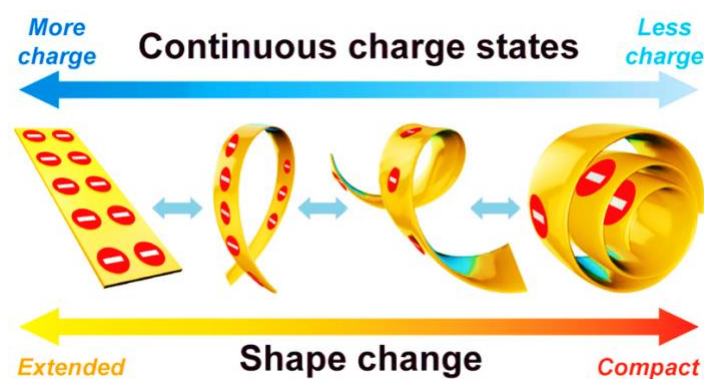


Artificial molecular machines have captured the imagination of scientists and nonscientists alike for decades now, given their clear potential to transform and enhance all aspects of human life. In this Outlook, I use a bicycle as an analogy to explain what a molecular machine is, in my opinion, and work through a representative selection of case studies to specify the significant accomplishments made to date, and the obstacles that currently stand between these and the field's fulfillment of its great potential. The hope of this intentionally sober account is to sketch a path toward a rich and exciting research trajectory that might challenge current practitioners and attract junior scientists into its fold. Considering the progress we have witnessed in the past decade, I am positive that the future of the field is a rosy one.

Comment: Oh my god! If one day I am invited to write a perspective article, I would like to do it in the way Professor Aprahamian did it in this article. Such a nice explanation of different concepts in the area of molecular machines, with good examples and analogies, showing the direction in which the field is moving towards. **I highly recommend this outlook.**

The Relationship between Static Charge and Shape

Pandey, R. K.; Ao, C. K.; Lim, W.; Sun, Y.; Di, X.; Nakanishi, H.*; Soh, S.* [ACS Cent. Sci., 2020, ASAP](#)



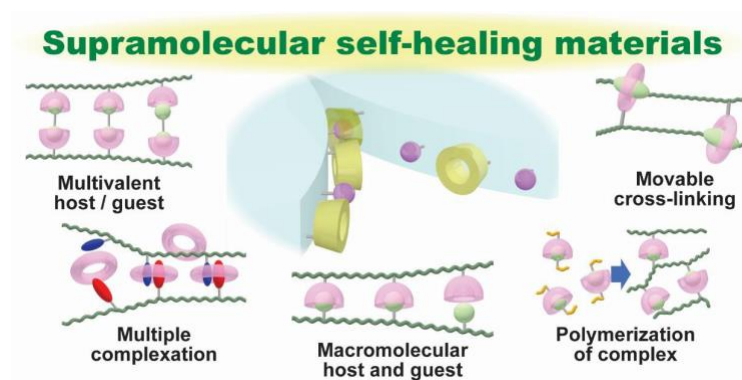
The amount of charge of a material has always been regarded as a property (or state) of materials and can be measured precisely and specifically. This study describes for the first time a fundamental physical–chemical phenomenon in which the amount of charge of a material is actually a variable—

it depends on the shape of the material. Materials are shown to have continuously variable and reversible ranges of charge states by changing their shapes. The phenomenon was general for different shapes, transformations, materials, atmospheric conditions, and methods of charging. **The change in charge was probably due to a dynamic exchange of charge from the material to the surrounding atmosphere as the shape changed via the reversible ionization and deposition of air molecules.** Similar changes in charge were observed for self-actuating materials that changed their shapes autonomously. This fundamental relationship between geometry and electrostatics via chemistry is important for the broad range of applications related to the charge of flexible materials.

Comment: Interesting article about the relationship between the shape of the materials (geometry) and how their charges change (electrostatics). So, when a material is extended or becomes more compact due to mechanical manipulation, or in an autonomous way, the amount of charge decreases. This study can be of relevance in the fields of stimuli-responsive materials that are flexible.

Supramolecular self-healing materials from non-covalent crosslinking host-guest interactions

Sinawang, G.; Osaki, M., Takashima, Y. *; Yamaguchi, H.*; Harada, A.* [Chem. Commun. 2020, ASAP](#)

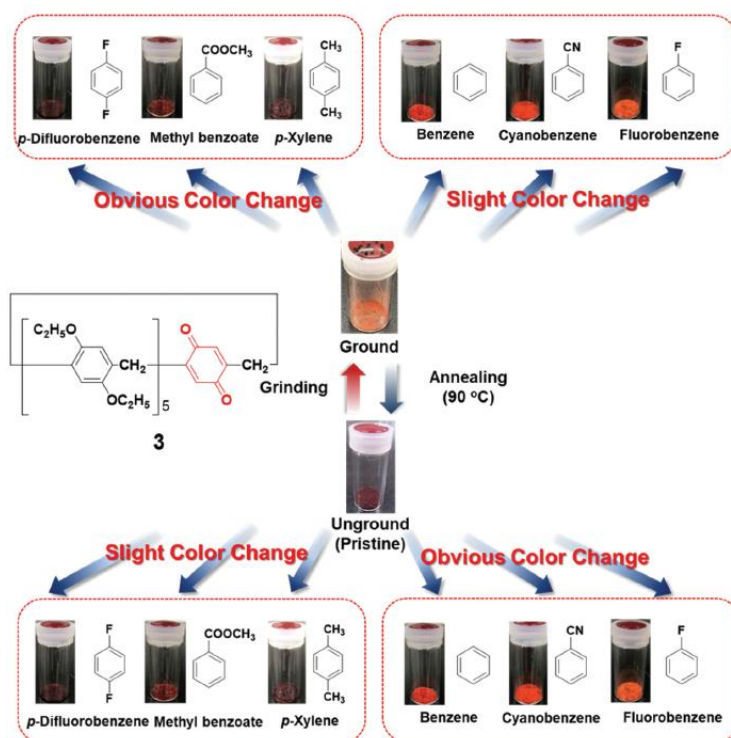


The introduction of non-covalent bonds is effective for achieving self-healing properties because they can be controlled reversibly. One approach to introduce these bonds into supramolecular materials is use of host-guest interactions. This feature article summarizes the development of supramolecular materials constructed by non-covalent cross-linking through several approaches, such as host-guest interactions between host polymers and guest polymers, 1:2-type host-guest interactions, and host-guest interactions from the polymerization of host-guest inclusion complexes. Host-guest interactions show self-healing functions while also enabling stimuli-responsiveness (redox, pH, and temperature). The self-healing function of supramolecular materials is achieved by stress dispersion arising from host-guest interactions when stress is applied. Reversible bonds based on host-guest interactions have tremendous potential to expand the variety of functional materials.

Comment: Compared to conventional polymers, some supramolecular polymers display versatile novel properties, for example, self-healing, high elasticity and so on. This feature article comprehensively describes host-guest interaction based supramolecular polymers, all which show excellent self-healing behaviour.

Obvious vapochromic color changes of a pillar[6]arene containing one benzoquinone unit with a mechanochromic change before vapor exposure

Wada, K.; Kakuta, T.; Yamagishi, T.; Ogoshi, T.* [Chem. Commun. 2020, ASAP](#)

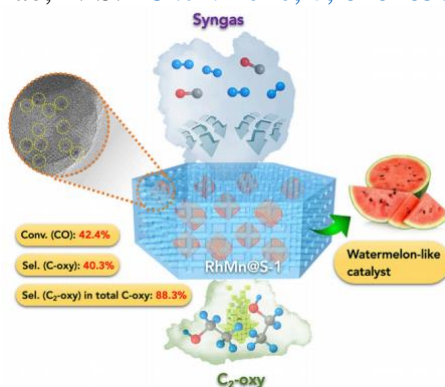


We developed a color changeable aromatic vapor detection system by combining the mechanochromism and vapochromism of pillar[6]arene containing one benzoquinone unit. The color of pillar[6]arene solid was changed by mechanochromism before vapor exposure. Different aromatic vapors then induced an obvious vapochromic color change from dark red to light orange or *vice versa*.

Comment: It is an interesting paper, the introduction of benzoquinone results in the possibility of visualization after uptake of aromatic guest vapor. It is a smart design and also provides a novel host towards non-porous adaptive crystals.

Direct Conversion of Syngas to Ethanol within Zeolite Crystals

Wang, C.; Zhang, J.; Qin, G.; Wang, L.*; Zuidema, E.; Yang, Q.; Dang, S.; Yang, C.; Xiao, J.*; Meng, X.; Mesters, C.; Xiao, F.-S.* [Chem 2020, 6, 646–657](#).

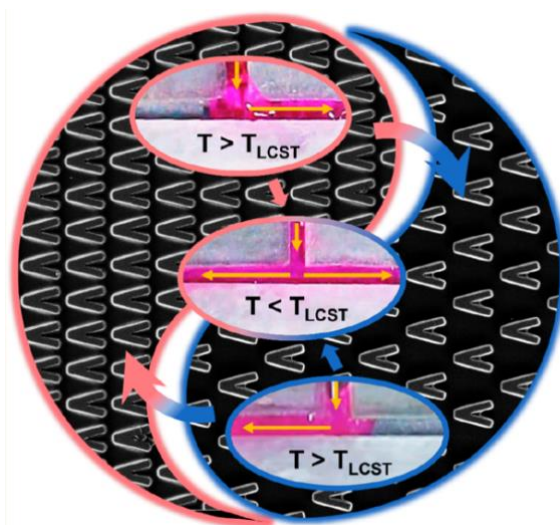


The direct transformation of syngas into C₂-oxygenates (e.g., ethanol) is in great demand, but it is still challenging because of the low selectivity and poor catalyst stability. Here, we overcome the issues and report a zeolite-fixation methodology to strengthen the transformation. By employing the extensively investigated RhMn catalyst, the **silicate-1 zeolite-fixed RhMn nanoparticles** with core-shell structure (RhMn@S-1) **exhibit high efficacy for the C₂ oxygenates production** from syngas, giving oxygenate selectivity at 40.3% under CO conversion of 42.4% (C₂-oxygenate selectivity at 88.3% in the total oxygenates), which decidedly outperforms the previous catalysts in direct syngas-to-ethanol transformation. Multiple studies demonstrate that the rigid zeolite framework efficiently stabilizes the Mn-O-Rh^{δ+} structure, known as the crucial site for C₂-oxygenate formation. In the long-term test, the RhMn@S-1 catalyst exhibits constant C₂-oxygenate productivities with superior durability. This investigation demonstrates the powerful siliceous zeolite support in the C₂-oxygenate production, which has not been explored previously.

Comment: The immobilization of catalytic nanoparticles on porous rigid frameworks (like zeolites) to improve their efficacy or stability is not new. The recycling of syngas is, however, an important topic for sustainable development and it is worthwhile to synthesize ethanol from this mixture. I did not know the journal *Chem*, it focuses on sustainable development in the chemistry field so have a look if you are interested!

Switchable Direction of Liquid Transport via an Anisotropic Microarray Surface and Thermal Stimuli

Zhang, Q.; He, L.; Zhang, X.; Tian, D.*; Jiang, L. *ACS Nano* 2020, 14, 1436–1444.



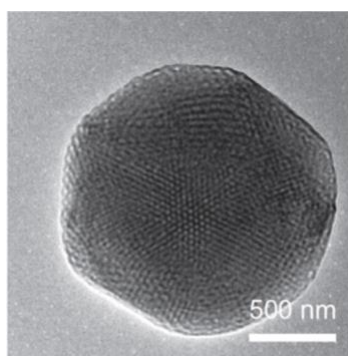
Design and construction of special surface microstructures has made many amazing breakthroughs in directional liquid transport. Despite much progress in this field, challenges still remain in on-demand switchable direction transport of liquid *in situ* and real-time *via* transforming the arrangement of the surface microstructure and external stimuli. Herein, we demonstrate a strategy to achieve switchable direction transport of liquid *via* a tunable anisotropic microarray surface, that is, assembling a V-shaped prism microarray (VPM) surface, which can also be intelligently manipulated by thermal stimuli. By transforming the parallel and staggered prism microstructure arrangement of the VPM, switchable direction transport of a liquid can be successfully achieved on the VPM surface. Flow direction switching among unidirectional transport, bidirectional transport, and reverse unidirectional transport is also achieved on the temperature-adaptive VPM surface by thermal stimuli, which can be used for ondemand liquid transport according to the paths of the microfluidic channels. The work provides a way for precise

liquid manipulation in desired liquid transport, which may be utilized in nonpower conveying systems, autolubrication, life fluid medical instruments, and other microfluidic devices.

Comment: After reading the abstract and before reading the paper, I thought the paper presented a way to switch from a parallel to staggered topology of the surface microstructures. In fact, the authors prepared these two different topologies separately. I do not know what you think about it, maybe my English is not good enough to understand the subtlety of the language... Anyway, this paper is still a proof that not only the chemical nature and nanotopology play a role in hydrophilicity but also the microstructure of the surface.

Polymer Cubosomes: Infinite Cubic Mazes and Possibilities

Ha, S.; La, Y.; Kim, K. T.* [*Acc. Chem. Res.* **2020**, *53*, 620–631.](#)



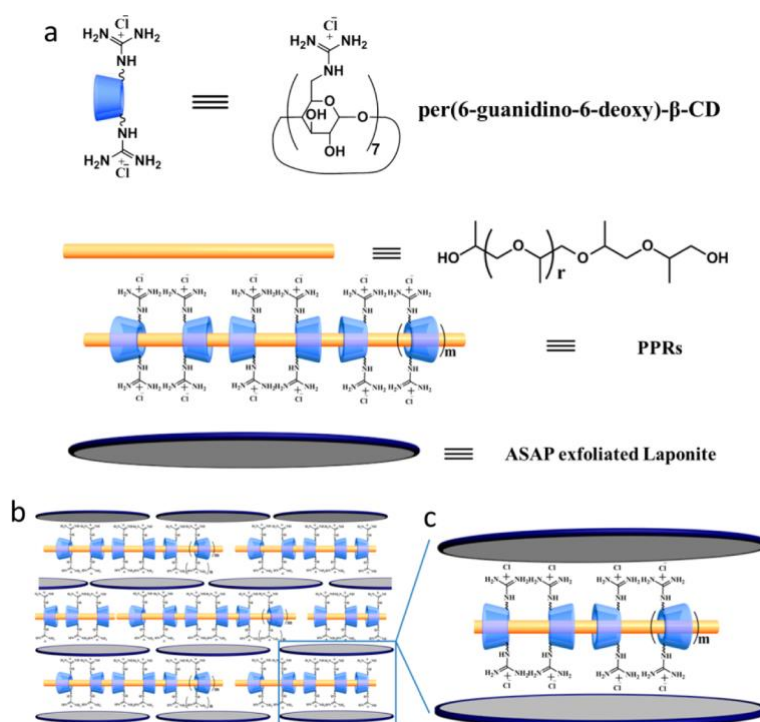
Block copolymers (BCPs) have been indispensable building blocks to create a range of soft nanostructures including discrete particulates (micelles and vesicles) and periodic structures via spontaneous assembly in bulk and in solution. The size, shape, and phase of these structures can be controlled by the rational design of the molecular structure of building blocks based on the structural analogy of BCPs to lipids and small molecule surfactants. Inverse bicontinuous cubic mesophases of polymers, or polymer cubosomes when in colloidal forms, are emerging nanostructures composed of triply periodic minimal surfaces (TPMSs) of block copolymer bilayers. Created by spontaneous assembly of BCPs in solution, polymer cubosomes internalize two nonintersecting nanochannel networks arranged in a cubic crystalline order. As well-defined porous particles with highly ordered internal structures and high surface-area-to-volume ratios, polymer cubosomes can be used for chemical reactors or bioreactors, carriers capable of cargo loading and release, and scaffolds for nanotemplating. However, despite their structural similarity to lipid cubosomes and applicability, polymer cubosomes have been only sporadically observed as an outcome of serendipity until recent studies demonstrated that BCPs could form well-defined polymer cubosomes in solution. In this Account, we describe our recent progress in creating polymer cubic mesophases and their colloidal particles (polymer cubosomes) in dilute solution. BCPs with nonlinear architectures (dendritic–linear, branched–linear, and branched–branched BCPs) preferentially self-assembled to inverse mesophases in solution when the block ratio (f), defined as a molecular weight ratio of the hydrophilic block to that of the hydrophobic block, was small ($<10\%$). The resulting lyotropic structures transformed from flat bilayers to cubic phases of primitive cubic and double diamond lattices and finally to inverted hexagonal phases as f decreased. We proposed that the architecture of a BCP plays an important role in the preferential formation of polymer cubosomes in solution. The presence of the bulky hydrophilic block limited chain stretching of the hydrophobic polymer block, which would increase the packing parameter of the BCP to greater than unity, a prerequisite for inverse mesophase formation. The structural characteristics of polymer cubosomes, such as lattice symmetries, pore sizes, and lattice parameters, could also be controlled by fine-tuning the structural parameters of BCPs. We also suggested nonsynthetic methods to precisely control the phase and internal lattice of inverse mesophases of

BCPs by the coassembly of two BCPs with different block ratios (mix-and-match approach) and the modulation of the affinity of the common solvent toward the hydrophobic block of the BCP. To investigate the potential applications of polymer cubosomes, we prepared inorganic photonic crystals using a cubosome-templated synthesis. We also discussed the utilization of cubosomes as chemical reactors by functionalization of the surface and the covalent stabilization of transient self-assembled structures via cross-linking of the hydrophobic domain. This Account reflects the efforts of synthetic chemists to understand the selfassembly behavior of BCPs to form complex morphologies in solution. We hope that our Account inspires efforts from chemists and other scientists to further understand these structures with infinite mazes of complexity and possibility.

Comment: In this Account, the authors mainly discuss the design of polymeric backbones to achieve a specific morphology of polymeric cubosomes. They also quickly discuss the possible applications of these structures, namely drug delivery, catalysis...

Mechanical Behaviors of Highly Swollen Supramolecular Hydrogels Mediated by Pseudorotaxanes

Li, Z.; Zhang, Y. M.; Wang, H. Y.; Li, H.; Liu, Y. *Macromolecules* **2017**, *50*, 1141–1146.

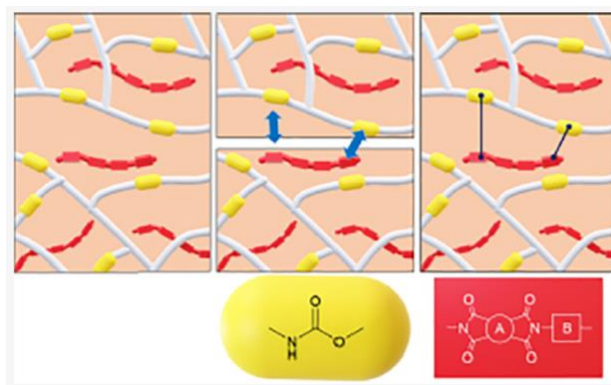


Supramolecular self-assemblies based on multiple noncovalent forces have emerged as an effective way to fabricate multistimuli responsive and self-healable intelligent materials. However, the development of supramolecular nanostructures as advanced functional materials in practical fields is always subjected to their relatively weaker mechanical strength and overuse of organic components. In this work, we proposed a new strategy to construct self-supported supramolecular hydrogels with the features of tunable mechanical strength, high water content, and self-healing properties through the hierarchically organic–inorganic hybridization of Laponite matrix with cyclodextrin-based pseudopolyrotaxanes. Notably, the mechanical properties of the obtained hydrogels can be conveniently modulated by tuning the molecular weight of polymeric chains in the central pseudopolyrotaxanes, which may provide a feasible way to promote the practical application of supramolecular metamaterials in miscellaneous fields.

Comment: Relatively uncommon materials. They combine organic supramolecular assemblies (Cyclodextrines complexing short PPG chains) with inorganic Laponite frameworks. They succeed at tuning the mechanical properties of their materials.

Highly Self-Healable Polymeric Blend Synthesized Using Polymeric Glue with Outstanding Mechanical Properties

Lee, S.; Hong, P. H.; Kim, J.; Choi, K.; Moon, G.; Kang, J.; Lee, S.; Ahn, J. B.; Eom, W.; Ko, M. J.; et al. *Macromolecules* **2020**, ASAP



A highly self-healable polymeric system with enhanced mechanical properties is prepared by blending conventional polyurethane (PU) with functional polyimide (PI). PU and PI synthesized in this study are miscible with each other and the thermal stability of the self-healable blend (PUPI) is improved by incorporation of PI into PU. Interestingly, on adding only a small amount of PI to PU, PUPI exhibits higher self-healing efficiency and faster self-healing kinetics. Furthermore, unlike conventional self-healing materials, PUPI also has superior surface and bulk mechanical properties. A model for the mechanism for the improvement of self-healing and mechanical properties is derived by analyzing FT-IR spectra. The outstanding self-healing and mechanical properties are attributed to the unique intermolecular networks resulting from the strong supramolecular interactions between urethane groups in PU and imide groups in PI. As a result, the PI chain acts as a polymeric glue inside the PU matrix of PUPI, which results in significant enhancements in both properties mentioned previously.

Comment: This article describes a good way to enhance mechanical properties: additives. Here, the additives are polyimide of low molecular mass. The strengthening of H bonds of the urethanes moieties fasten the relaxation time and thus the self-healing properties, at a relatively high temperature ($T_g + 10^\circ\text{C} = \sim 80^\circ\text{C}$ here)

Highly Selective Fluoride Recognition by a Small Tris-Urea Covalent Cage

Delecluse, M.; Colombar, C.*; Chatelet, B.; Chevallier-Michaud, S.; Moraleda, D.; Dutasta, J.-P.; Martinez, A.* *J. Org. Chem.* **2020**, ASAP.

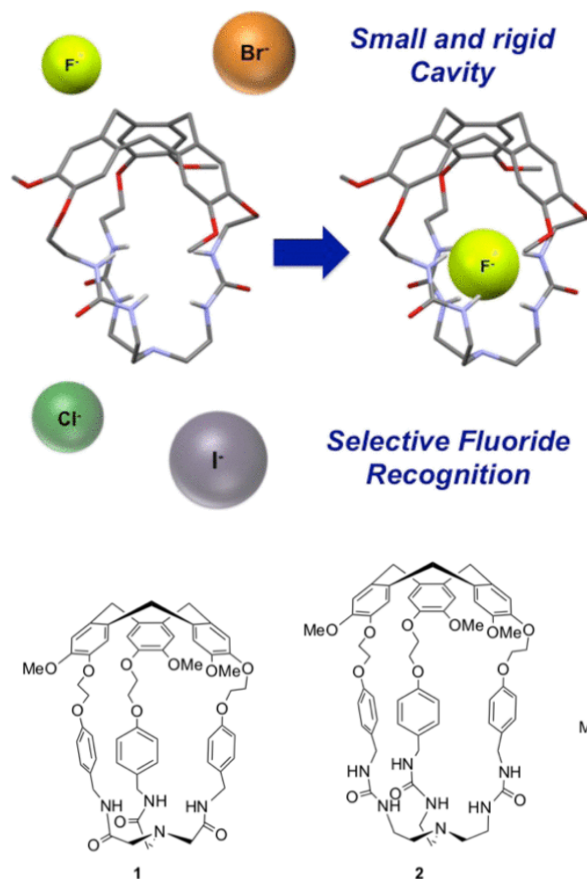


Table 1. Comparison of the association constants $K_a(\text{M}^{-1})$ for the 1:1 complexes formed between anion guests and hemicryptophanes hosts **2** and **3**.

Guest ^[a]	$K_a(\text{M}^{-1})$, Host 2 ^[b]	$K_a(\text{M}^{-1})$, Host 3 ^[c]
F ⁻	3900	1200
Cl ⁻	4100	<1
Br ⁻	1050	<1
I ⁻	80	<1
AcO ⁻	1400	<1
H ₂ PO ₄ ⁻	184200	216

[a] TBA⁺ salt. [b] From reference 10. [c] K_a was extracted from the ¹H NMR titration curves (CDCl₃, 500 MHz, 298K) for the proton H_b (δ = 4.71 ppm for the free cage) using Bindfit software.¹⁵ Estimated error 10%.

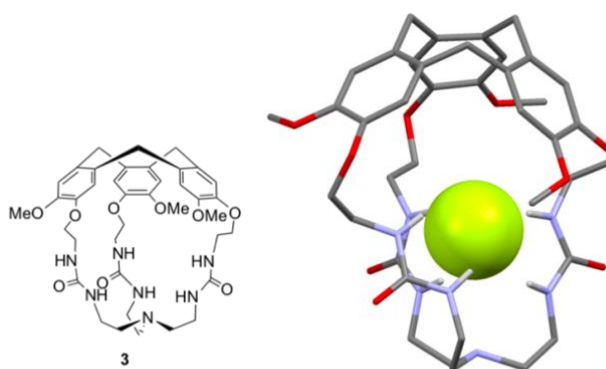


Figure 5. X-ray structure of [3·F⁻]. TBA⁺ has been omitted for clarity.

Figure 1. Structure of hemicryptophanes **1**, **2** and **3**.

A highly selective recognition of fluoride was achieved through the design of a small hemicryptophane cage (**3**) presenting a southern tris-urea hosting moiety. The resulting host–guest complex has been characterized by electrospray ionization-high-resolution mass spectrometry, ¹H and ¹⁹F NMR, and X-ray diffraction techniques. In particular, X-ray diffraction analysis of [3·F⁻] reveals that the encapsulation of one fluoride, within **3**, occurs through NH⋯F⁻ H-bonding with the six NH residues of the tris-urea ligand. An association constant of 1200 M⁻¹ was extracted from ¹H NMR titration experiments, indicating that efficient fluoride binding also occurs in solution. Finally, in sharp contrast with previously reported urea-based hemicryptophane hosts, the small preorganized cavity found in **3** allows for an exclusive selectivity for fluoride over other competing halides.

Comment: This work can selectively control an encapsulation of “F⁻” in the hemicryptophane cage. Owing to not only host-guest interaction of the small cavity but also hydrogen-bonding between “F⁻” and the tris-urea groups. The association constant of K_a is over 1000 which is extremely higher than the case of the other anions (Cl⁻, Br⁻, I⁻). Moreover, this work revealed the encapsulation of “F⁻” confirmed by ¹H and ¹⁹F NMR and single crystal X-ray analysis.

Ionic Dopant-Encapsulating Single-Walled Carbon Nanotube Films with Metal-Like Electrical Conductivity

Y. Iihara, T. Kawai* and Y. Nonoguchi* *Chem.Asian.J.* **2020**, *15*, 590.

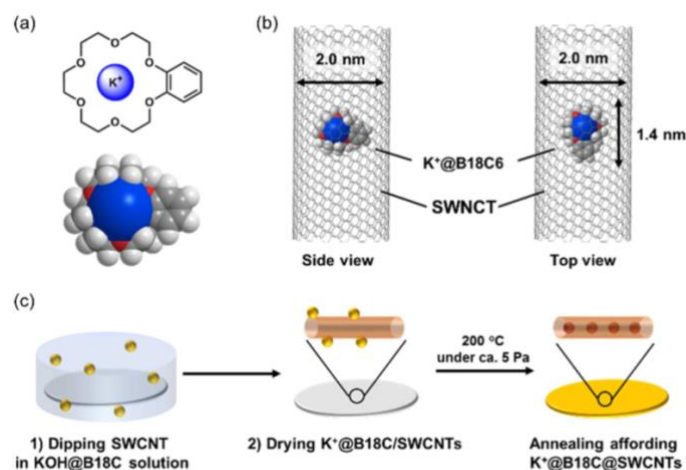
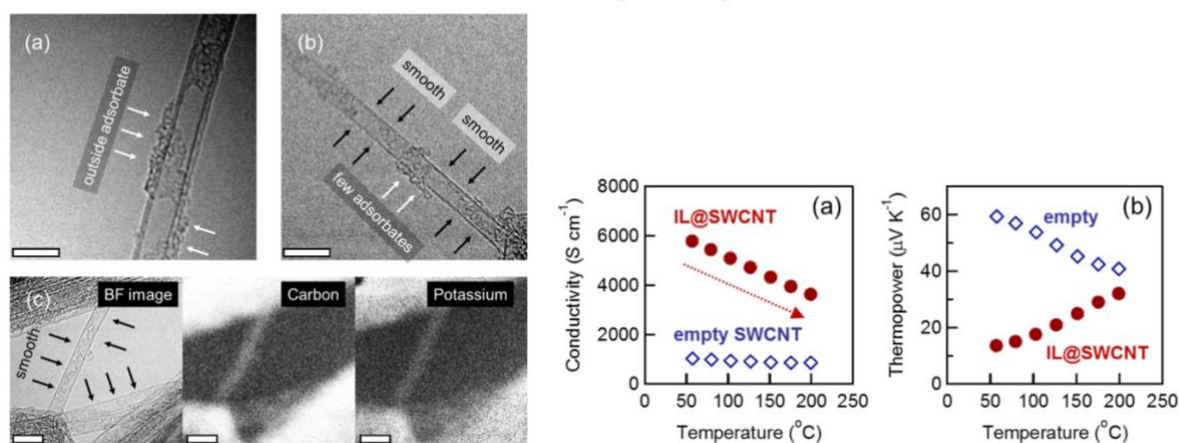


Figure 1. (a) The molecular structure of $K^+@B18C6$. (b) The Schematic image of $K^+@B18C6@SWCNT$. (c) A schematic experimental procedure.

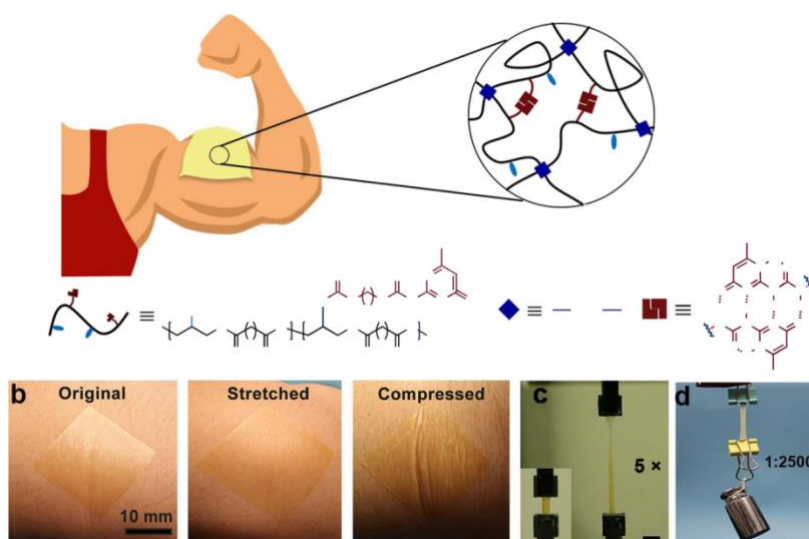


Heavy doping is inevitable for utilizing single-walled carbon nanotubes for wiring. However, the electrical conductivity of their films is currently as low as one tenth of the films made from typical metal pastes. Herein we report on metal-comparable electrical conductivity from single-walled carbon nanotube network films. We use ionic liquids and crown ether complexes for p-type and n-type doping, respectively. The encapsulation of counterions into carbon nanotubes promotes the conductivities in the range of $7000\ S\ cm^{-1}$, approximately ten times larger than those of undoped films.

Comment: This work could enhance the conductivity of Single-walled carbon nanotubes (SWCNTs) by chemical doping of potassium crown ether molecule ($[K^+@B_{18}C_6][OH^-]$). It looks very simple and interesting to enhance the conductivity. But the author did not address the possibility of ion conductivity derived from $[K^+@B_{18}C_6][OH^-]$ itself not only into the SWCNTs but also outside of the SWCNTs. Because they did not measure the conductivity the single fiber of doped-SWCNTs and the behavior of conductivity changed from the 1st and 2nd scan. Therefore, it will look much more interesting when the author carefully investigates the mechanism of this type of doping.

Mechanically and biologically skin-like elastomers for bio-integrated electronics

Chen, S.; Sun, L.; Zhou, X.; Guo, Y.; Song, J.; Qian, S.; He, C.; You, Z.;* [Nat. Commun.](#) **2020**, *11*, 1107

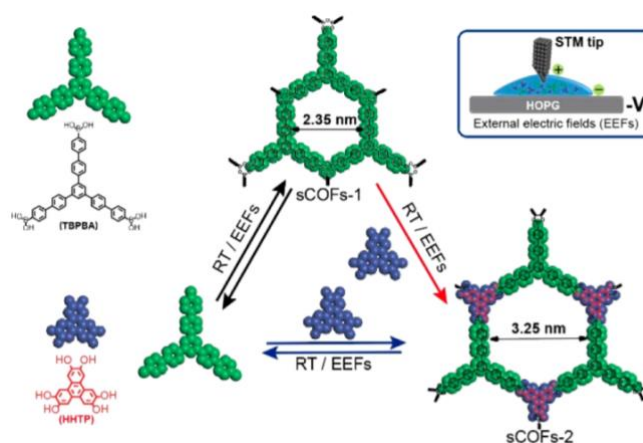


The bio-integrated electronics industry is booming and becoming more integrated with biological tissues. To successfully integrate with the soft tissues of the body (eg. skin), the material must possess many of the same properties including compliance, toughness, elasticity, and tear resistance. In this work, we prepare mechanically and biologically skin-like materials (PSeD-U elastomers) by designing a unique physical and covalent hybrid crosslinking structure. The introduction of an optimal amount of hydrogen bonds significantly strengthens the resultant elastomers with 11 times the toughness and 3 times the strength of covalent crosslinked PSeD elastomers, while maintaining a low modulus. Besides, the PSeD-U elastomers show nonlinear mechanical behavior similar to skins. Furthermore, PSeD-U elastomers demonstrate the cytocompatibility and biodegradability to achieve better integration with tissues. Finally, piezocapacitive pressure sensors are fabricated with high pressure sensitivity and rapid response to demonstrate the potential use of PSeD-U elastomers in bio-integrated electronics.

Comment: In this work, skin-like mechanical properties were readily achieved by a combination of hydrogen bonds and covalently crosslinked networks to mimic the roles of collagen and elastin fibers in natural skin. The **ratio between hydrogen bond and covalent crosslinks** was found to play a critical role for the mechanical properties of the “skin”

Real-Time Molecular-Scale Imaging of Dynamic Network Switching between Covalent Organic Frameworks

Zhan, G.; Cai, Z. F.; Martínez-Abadia, M.; Mateo-Alonso, A.; De Feyter, S. * [J. Am. Chem. Soc.](#) **2020**.

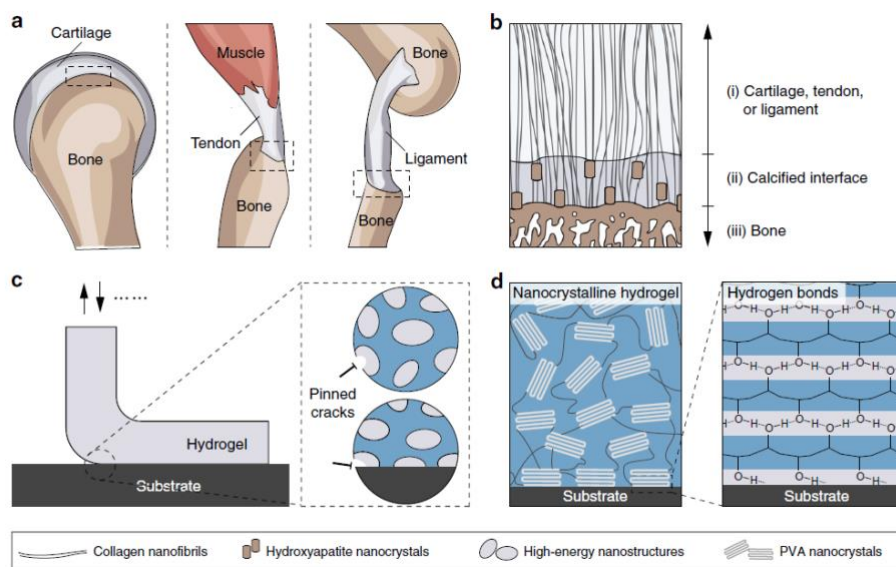


The in situ on-surface conversion process from boroxine-linked covalent organic frameworks (COFs) to boronate ester-linked COFs is triggered and catalyzed at room temperature by an electric field and monitored with scanning tunneling microscopy (STM). The adaptive behavior within the generated dynamic covalent libraries (DCLs) was revealed, providing in-depth understanding of the dynamic network switching process.

Comment: Dynamic boronic-ester linked COF monitored by STM (beautiful images!), effector: external electric fields.

Fatigue-resistant adhesion of hydrogels

Liu, J.; Lin, S.; Liu, X.; Qin, Z.; Yang, Y.; Zang, J.;* Zhao, X.;* *Nat. Commun.* **2020**, *11*, 1071



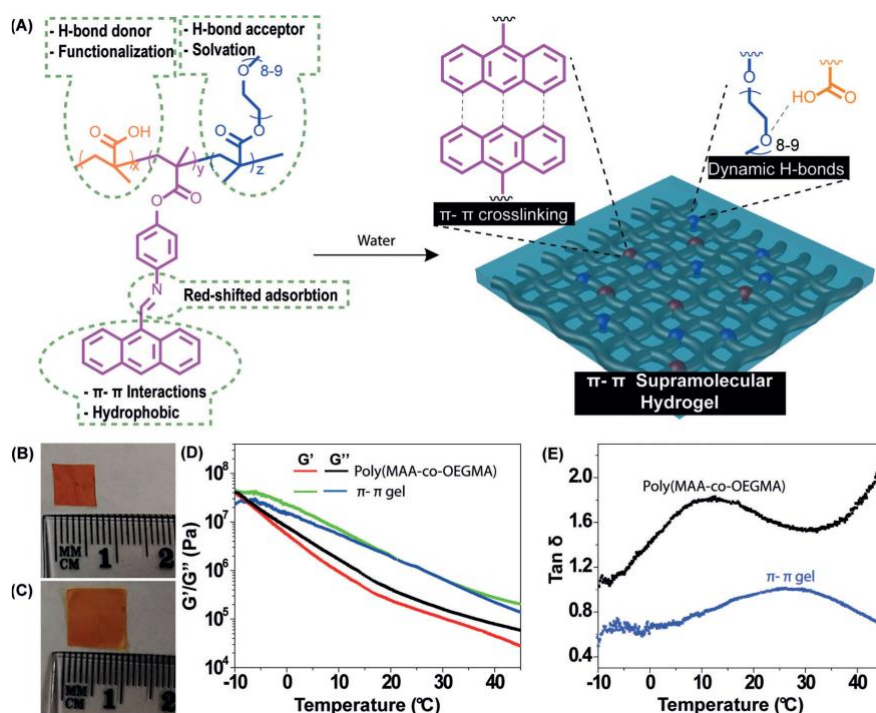
The adhesion of soft connective tissues (tendons, ligaments, and cartilages) on bones in many animals can maintain high toughness ($\sim 800 \text{ J m}^{-2}$) over millions of cycles of mechanical loads. Such fatigue-resistant adhesion has not been achieved between synthetic hydrogels and engineering materials, but is highly desirable for diverse applications such as artificial cartilages and tendons, robust antifouling coatings, and hydrogel robots. Inspired by the nanostructured interfaces between tendons/ligaments/cartilages and bones, we report that bonding ordered nanocrystalline domains of synthetic hydrogels on engineering materials can give a fatigue-resistant adhesion with

an interfacial fatigue threshold of 800 Jm^{-2} , because the fatigue-crack propagation at the interface requires a higher energy to fracture the ordered nanostructures than amorphous polymer chains. Our method enables fatigueresistant hydrogel coatings on diverse engineering materials with complex geometries. We further demonstrate that the fatigue-resistant hydrogel coatings exhibit low friction and low wear against natural cartilages.

Comment: A facile yet general strategy for the fatigue-resistant adhesion of hydrogels on diverse engineering materials through bonding ordered nanostructures in the hydrogels.

Strong, Self-healable, and Recycle Visible-Light Responsive Hydrogel Actuators

Jiang, Z.; Tan, M. L.; Taheri, M.; Yan, Q.; Tsuzuki, T.; Gardiner, M. G.; Connal, L. A. C.*
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The most pressing challenges for light-driven hydrogel actuators include reliance on UV light, slow response, poor mechanical properties, and limited functionalities. Now, a supramolecular design strategy is used to address these issues. Key is the use of a benzylimine-functionalized anthracene group, which red-shifts the absorption into the visible region and also stabilizes the supramolecular network through π - π interactions. Acid-ether hydrogen bonds are incorporated for energy dissipation under mechanical deformation and maintaining hydrophilicity of the network. This double-crosslinked supramolecular hydrogel developed via a simple synthesis exhibits a unique combination of high strength, rapid self-healing, and fast visible-light-driven shape morphing both in the wet and dry state. As all of the interactions are dynamic, the design enables the structures to be recycled and reprogrammed into different 3D objects.

Comment: This work used a benzylimine-functionalized anthracene group, which could redshift the absorption into the visible region and also stabilizes the supramolecular network through π - π interaction. Combining the acid-ether hydrogen bonds for energy dissipation under mechanical deformation and maintaining the hydrophilicity of the network. All of these dynamic interactions enable the structure to be recycled and reprogrammed into different 3D objects.