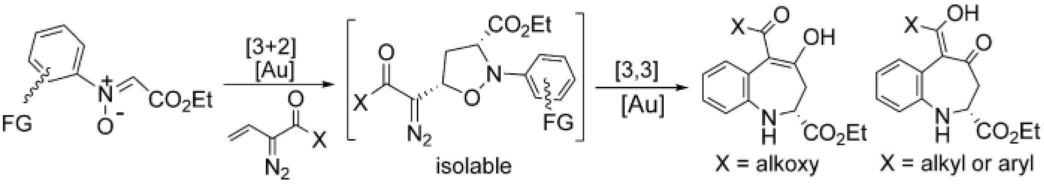
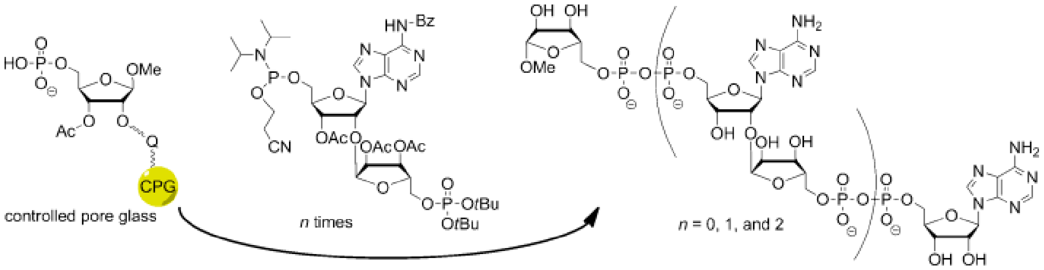
- Gold-Catalyzed Cycloaddition Reactions of Ethyl Diazoacetate, Nitrosoarenes, and Vinyldiazo Carbonyl Compounds: Synthesis of Isoxazolidine and Benzo[b]azepine Derivatives
  Pagar, V. V.; Liu, R.-S. *Angew. Chem. Int. Ed.* **2015**, *54*, 4923 –4926.
  Abstract:



Gold-catalyzed cycloadditions of ethyl diazoacetate, nitrosoarenes, and vinyldiazo carbonyl species to yield isoxazolidine derivatives stereoselectively are described. Treatment of these isoxazolidine products with the same catalyst results in a novel 1,2-H shift/[3,3] rearrangement to give benzo[b]azepine compounds. The mechanism of this skeletal rearrangement is elucidated with deuterium-labeling experiments.

- Synthesis of Well-Defined Adenosine Diphosphate Ribose Oligomers
  Kistemaker, H. A. V.; Lameijer, L. N.; Meeuwenoord, N. J.; Overkleeft, H. S.; van der Marel, G. A.; Filippov, D. V. *Angew. Chem. Int. Ed*. **2015**, *54*, 4915 –4918.
  Abstract:



The post-translational modification of proteins that is known as adenosine diphosphate ribosylation (ADPr) regulates a wide variety of important biological processes, such as DNA-damage repair and cellular metabolism. This modification is also involved in carcinogenesis and the process of aging. Therefore, a better understanding of the function of ADP-ribosylation is crucial for the development of novel therapeutics. To facilitate the elucidation of the biology of ADPr, the availability of well-defined fragments of poly(ADP-ribose) is essential. Herein we report a solid-phase synthetic approach for the preparation of ADP-ribose oligomers of exactly defined length. The methodology is exemplified by the first reported synthesis of an ADP-ribose dimer and trimer.

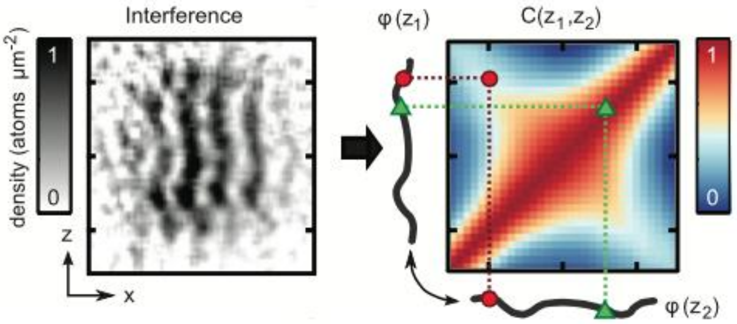
- Self-templated chemically stable hollow spherical covalent organic framework
  Kandambeth, S.; Venkatesh, V.; Shinde, D. B.; Kumari, S.; Halder, A.; Verma, S.; Banerjee, R. *Nature Comm.* **2015**, *6*, 6786.
  Abstract:

Covalent organic frameworks are a family of crystalline porous materials with promising applications. Although active research on the design and synthesis of covalent organic frameworks has been ongoing for almost a decade, the mechanisms of formation of covalent organic frameworks crystallites remain poorly understood. Here we report the synthesis of a hollow spherical covalent organic framework with mesoporous walls in a single-step template-free method. A detailed time-dependent study of hollow sphere formation reveals that an inside-out Ostwald ripening process is responsible for the hollow sphere formation. The synthesized covalent organic framework hollow spheres are highly porous (surface area ~1,500 $m^2g^{-1}$), crystalline and chemically stable, due to the presence of strong intramolecular hydrogen bonding. These mesoporous hollow sphere covalent organic frameworks are used for a trypsin immobilization study, which shows an uptake of 15.5 $\mu mol.g^{-1}$ of trypsin.

- Iterative design of a helically folded aromatic oligoamide sequence for the selective encapsulation of fructose
  Chandramouli, N.; Ferrand, Y.; Lautrette, G.; Kauffmann, B.; Mackereth, C. D.; Laguerre, M.; Dubreuil, D.; Huc, I. *Nature Chem.* **2015**, *7*, 334-341.
  Abstract:

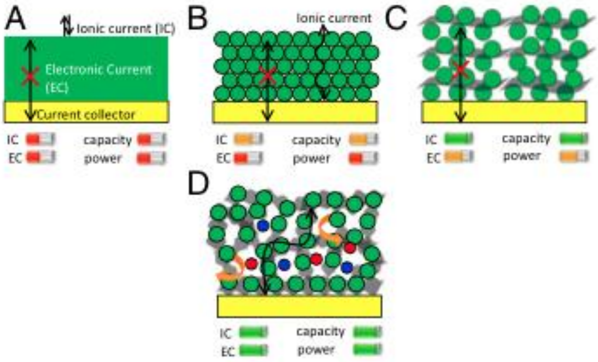
The *ab initio* design of synthetic molecular receptors for a specific biomolecular guest remains an elusive objective, particularly for targets such as monosaccharides, which have very close structural analogues. Here we report a powerful approach to produce receptors with very high selectivity for specific monosaccharides and, as a demonstration, we develop a foldamer that selectively encapsulates fructose. The approach uses an iterative design process that exploits the modular structure of folded synthetic oligomer sequences in conjunction with molecular modelling and structural characterization to inform subsequent refinements. Starting from a first-principles design taking size, shape and hydrogen-bonding ability into account and using the high predictability of aromatic oligoamide foldamer conformations and their propensity to crystallize, a sequence that binds to β-D-fructopyranose in organic solvents with atomic-scale complementarity was obtained in just a few iterative modifications. This scheme, which mimics the adaptable construction of biopolymers from a limited number of monomer units, provides a general protocol for the development of selective receptors.

- Experimental observation of a generalized Gibbs ensemble
  Langen, T.; Erne, S.; Geiger, R.; Rauer, B.; Schweigler, T.; Kuhnert, M.; Rohringer, W.; Mazets, I. E.; Gasenzer, T.; Schmiedmayer, J. *Science* **2015**, *348*, 207-211.
  Abstract:



The description of the non-equilibrium dynamics of isolated quantum many-body systems within the framework of statistical mechanics is a fundamental open question. Conventional thermodynamical ensembles fail to describe the large class of systems that exhibit nontrivial conserved quantities, and generalized ensembles have been predicted to maximize entropy in these systems. We show experimentally that a degenerate one-dimensional Bose gas relaxes to a state that can be described by such a generalized ensemble. This is verified through a detailed study of correlation functions up to 10th order. The applicability of the generalized ensemble description for isolated quantum many-body systems points to a natural emergence of classical statistical properties from the microscopic unitary quantum evolution.

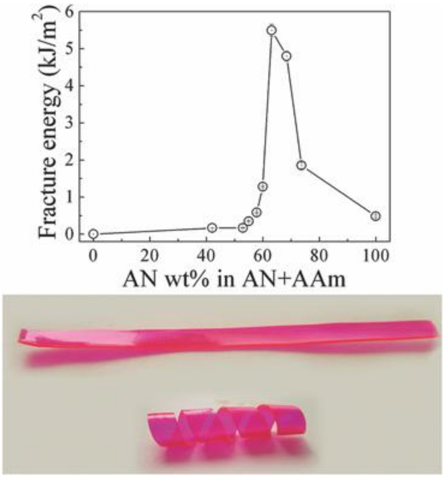
- Engineering three-dimensional hybrid supercapacitors and microsupercapacitors for high-performance integrated energy storage
  El-Kady, M. F.; Ihns, M.; Li, M.; Hwang, J. Y.; Mousavi, M. F.; Chaney, L.; Lech, A. T.; Kaner, R. B. *Proc. Nat. Acad. Sci. USA* **2015,** *112*, 4233-4238.
  Abstract:



Supercapacitors now play an important role in the progress of hybrid and electric vehicles, consumer electronics, and military and space applications. There is a growing demand in developing hybrid supercapacitor systems to overcome the energy density limitations of the current generation of carbon-based supercapacitors. Here, we demonstrate 3D high-performance hybrid supercapacitors and microsupercapacitors based on graphene and $MnO_2$ by rationally designing the electrode microstructure and combining active materials with electrolytes that operate at high voltages. This results in hybrid electrodes with ultrahigh volumetric capacitance of over 1,100 F/cm$^3$. This corresponds to a specific capacitance of the constituent $MnO_2$ of 1,145 F/g, which is close to the theoretical value of 1,380 F/g. The energy density of the full device varies between 22 and 42 Wh/l depending on the device configuration, which is superior to those of commercially available double-layer supercapacitors, pseudocapacitors, lithium-ion capacitors, and hybrid supercapacitors tested under the same conditions and is comparable to that of lead acid batteries. These hybrid

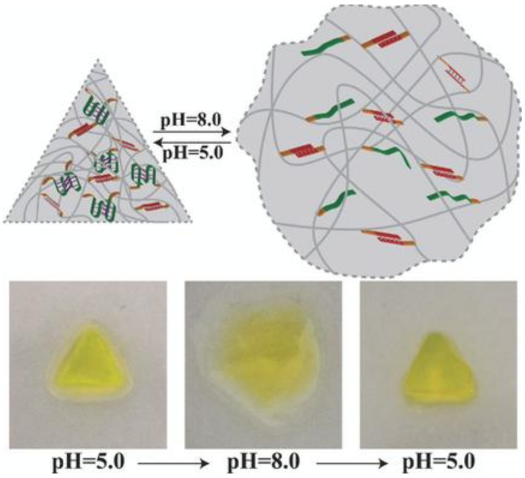
supercapacitors use aqueous electrolytes and are assembled in air without the need for expensive "dry rooms" required for building today's supercapacitors. Furthermore, we demonstrate a simple technique for the fabrication of supercapacitor arrays for high-voltage applications. These arrays can be integrated with solar cells for efficient energy harvesting and storage systems.

- Dipole–Dipole and H-Bonding Interactions Significantly Enhance the Multifaceted Mechanical Properties of Thermoresponsive Shape Memory Hydrogels
  Zhang, Y.; Li, Y.; Liu, W. *Adv. Funct. Mater.* **2015**, *25*, 471–480.
  Abstract:



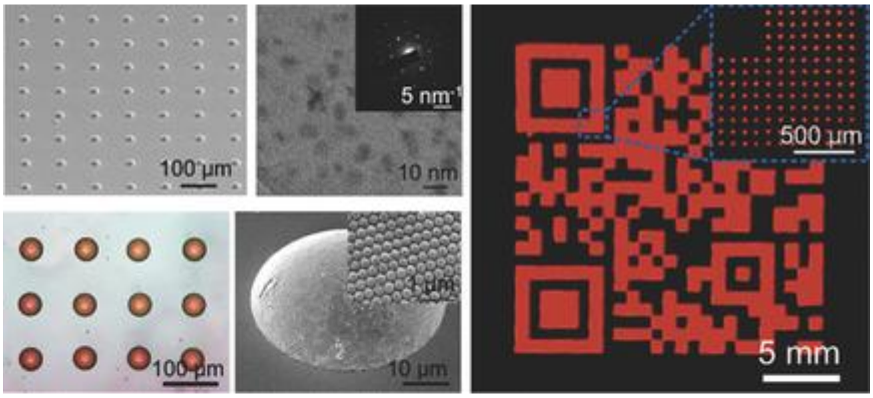
High strength hydrogels were previously constructed based on dipole–dipole and hydrogen bonding reinforcement. In spite of the high tensile and compressive strengths achieved, the fracture energy of the hydrogels strengthened with sole noncovalent bondings was rather low due to the lack in energy dissipating mechanism. In this study, combined dipole–dipole and hydrogen bonding interactions reinforced (DHIR) hydrogels are synthesized by one-step copolymerization of three feature monomers, namely acrylonitrile (AN, dipole monomer), acrylamide (AAm, H-bonding monomer), and 2-acrylamido-2-methyl-1-propanesulfonic acid (AMPS, anionic monomer) in the presence of PEGDA575, a hydrophilic crosslinker. The electrostatic repulsion from PAMPS allows the gel network to absorb water readily, and meanwhile the synergistic effect of dipole–dipole and H-bonding interactions enable the DHIR hydrogel to withstand up to 8.3 MPa tensile stress, 4.8 MPa compressive stress and 140–716% elongation at break with the fracture energy reaching as high as 5500 J/m$^2$. In addition, this DHIR hydrogel exhibits reversible mechanical properties after undergoing cyclic loading and unloading. Interestingly, the DHIR hydrogels with appropriate compositions demonstrate temperature-tunable mechanical properties as well as accompanied shape memory effect. The dual noncovalent bonding strengthening mechanism reported here offers a universal strategy for significantly enhancing the comprehensive mechanical properties of hydrogels.

- pH-Stimulated DNA Hydrogels Exhibiting Shape-Memory Properties
  Guo, W.; Lu, C.-H.; Orbach, R.; Wang, F.; Qi, X.-J.; Cecconello, A.; Seliktar, D.; Willner, I. *Adv. Mater.* **2015**, *27*, 73–78.
  Abstract:



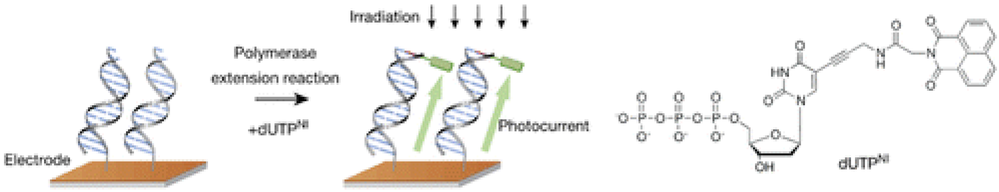
**Nucleic acid-functionalized polyacrylamide chains** that are cooperatively cross-linked by i-motif and nucleic acid duplex units yield, at pH 5.0, DNA hydrogels exhibiting shape-memory properties. Separation of the i-motif units at pH 8.0 dissolves the hydrogel into a quasi-liquid phase. The residual duplex units provide, however, a memory code in the quasi-liquid allowing the regeneration of the hydrogel shape at pH 5.0.

- Patterning Fluorescent Quantum Dot Nanocomposites by Reactive Inkjet Printing
  Bao, B.; Li, M.; Li, Y.; Jiang, J.; Gu, Z.; Zhang, X.; Jiang, L.; Song, Y. *Small* **2015**, *11*, 1649–1654.
  Abstract:



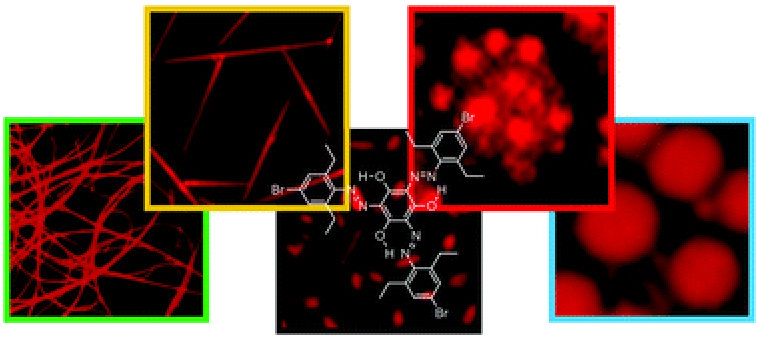
Fluorescent quantum dot nanocomposites, including polymer and photonic crystal quantum dots, have been fabricated by reactive inkjet printing. This reactive inkjet printing method has the potential to be broadened to fabrication of other functional nanomaterials, which will find promising applications in optoelectronic devices.

- Photoresponsive DNA Monolayer Prepared by Primer Extension Reaction on the Electrode
  Takada, T.; Takemura, M.; Kawano, Y.; Nakamura, M.; Yamana, K. *Langmuir* **2015**, *31*, 3993–3998.
  Abstract:



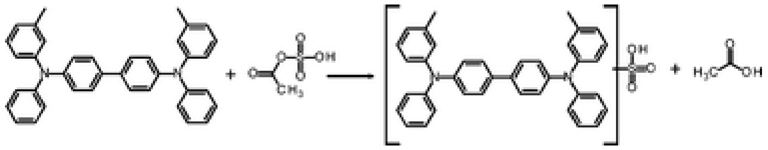
We describe a simple and convenient method for the preparation of photoresponsive DNA-modified electrodes using primer extension (PEX) reactions. A naphthalimide derivative was used as the photosensitizer that was attached to the C5-position of 2′-deoxyuridine-5′-triphosphate (dUTP$^{\mathrm{NI}}$). It has been found that dUTP$^{\mathrm{NI}}$ is a good substrate for the PEX reactions using KOD Dash and Vent (exo-) enzymes in solutions to incorporate naphthalimide (NI) moieties into the DNA sequences. On the electrode surface immobilized with the primer/template DNA, the PEX reactions to incorporate dUTP$^{\mathrm{NI}}$ molecules into the DNA sequence were found to efficiently proceed. With this solid-phase method, the DNA monolayers capable of generating photocurrent due to the photoresponsive NI molecule can be constructed. It was shown that the photocurrent generation was significantly suppressed by a single-nucleotide mismatch included in the primer/template DNA, which is applicable for the design of photoelectrochemical sensors to discriminate single-nucleotide sequences.

- Facile morphological control of fluorescent nano/microstructures via self-assembly and phase separation of trigonal azobenzenes showing aggregation-induced emission enhancement in polymer matrices
  Han, M.; Takeoka, Y.; Seki, T. *J. Mater. Chem. C* **2015**, *3*, 4093-4098.
  Abstract:



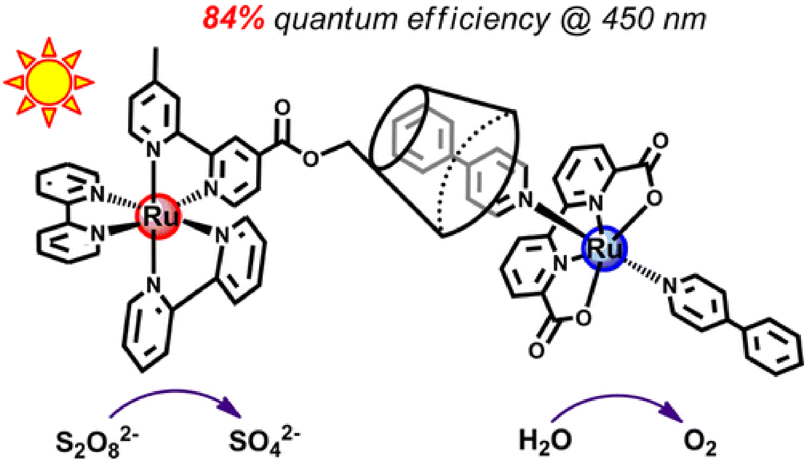
We report a facile and mild strategy for constructing diverse fluorescent nano/microstructures *via* self-assembly and phase separation of trigonal azobenzene chromophores (3N1s) showing aggregation-induced emission enhancement (AIEE) in polymer matrices [poly(methyl methacrylate) (PMMA) and/or poly(4-chlorostyrene) (PSCl)]. Thermal treatment above the glass transition temperature enhances the large-scale molecular motions of the polymer chains, which causes AIEE-active 3N1 molecules to assemble into fluorescent nanorods and long nanosticks in the confined homopolymer (PMMA and PSCl, respectively) matrices. Strikingly, as-prepared 3N1–PMMA–PSCl ternary mixtures exhibit a splendid raspberry-like morphology. In other words, the uneven island-like surfaces with micrometer-scale round protuberances are decorated with red fluorescent nanospheres. This result can be interpreted as surface-directed phase separation of immiscible PMMA and PSCl during quick solvent evaporation, which would help the 3N1 components instantaneously assemble into nanospheres on uneven surfaces. By annealing above the glass transition temperature, a distinct morphological transformation from a raspberry-like to a bead-like structure could readily be visualized *via* (i) the inherent assembly of 3N1 molecules into red fluorescent spherical or 1D aggregates and (ii) the selective fluorescence marking due to the difference in compatibility between 3N1 and PMMA or PSCl.

- Functionalization of CdSe semiconductor nanocrystals with organic charge-transporting ligands
  Liang, Y.; Moon, J.-S.; Mu, R.; Winiarz, J. G. *J. Mater. Chem. C* **2015**, *3*, 4134-4140.
  Abstract:



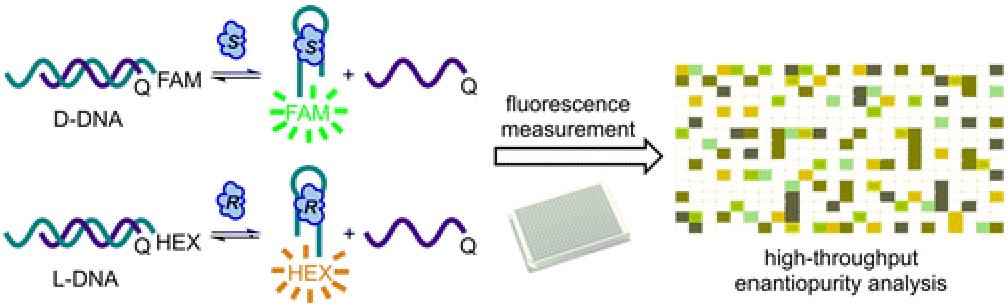
[1,1′-Biphenyl]-4,4′-diamine-*N*,*N*′-bis(3-methylphenyl)-*N*,*N*′-diphenyl (TPD), a well-studied hole transporting material, has been sulfonated using acetyl sulfate and subsequently used as a passivating ligand in the synthesis of CdSe quantum dots (QDs). It is further demonstrated that QDs synthesized through this approach are able to serve as efficient photosensitizers in photoconductive (PC) inorganic–organic hybrid composites. Fourier-transform infrared spectroscopy confirms that the sulfonate group has been bonded to the TPD molecule. UV/visible absorption and photoluminescence (PL) spectroscopy of the sulfonated TPD (STPD) indicate that sulfonation does not significantly alter the electronic properties of TPD. Furthermore, mass spectrometry shows STPD is primarily mono-sulfonated. The STPD capped CdSe QDs (STPD-QCdSe) clearly exhibit the effect of quantum confinement in their UV/visible absorption spectra. The PL spectra of STPD-QCdSe suggest the STPD is attached to the surface of QCdSe. The morphology of STPD-QCdSe has been studied using transmission electron microscopy. The results indicate the STPD-QCdSe are approximately spherical with diameters of [3.1, 3.9] nm and highly crystalline. To demonstrate the enhancement in charge-transfer efficiency associated with STPD-QCdSe, two types of PC composites were fabricated. The first was photosensitized through the inclusion of STPD-QCdSe, and the other included CdSe QDs capped with trioctylphosphine (TOPO-QCdSe), with molecular TPD serving as the charge transport matrix in both cases. The PC was measured as a function of the external electric field, *E*, with the PC of the composite sensitized with STPD-QCdSe exceeding that of the composite sensitized with TOPO-QCdSe by a factor of ~15 with $E = 10\ \mathrm{V\ \mu m^{-1}}$.

- Visible Light-Driven Water Oxidation Promoted by Host–Guest Interaction between Photosensitizer and Catalyst with A High Quantum Efficiency
  Li, H.; Li, F.; Zhang, B. B.; Zhou, X.; Yu, F. S.; Sun L. C. *J. Am. Chem. Soc.* **2015**, *137*, 4332-4335.
  Abstract:



A highly active supramolecular system for visible light-driven water oxidation was developed with cyclodextrin-modified ruthenium complex as the photosensitizer, phenyl-modified ruthenium complexes as the catalysts, and sodium persulfate as the sacrificial electron acceptor. The catalysts were found to form 1:1 host–guest adducts with the photosensitizer. Stopped-flow measurement revealed the host–guest interaction is essential to facilitate the electron transfer from catalyst to sensitizer. As a result, a remarkable quantum efficiency of 84% was determined under visible light irradiation in neutral aqueous phosphate buffer. This value is nearly 1 order of magnitude higher than that of noninteraction system, indicating that the noncovalent incorporation of sensitizer and catalyst is an appealing approach for efficient conversion of solar energy into fuels.

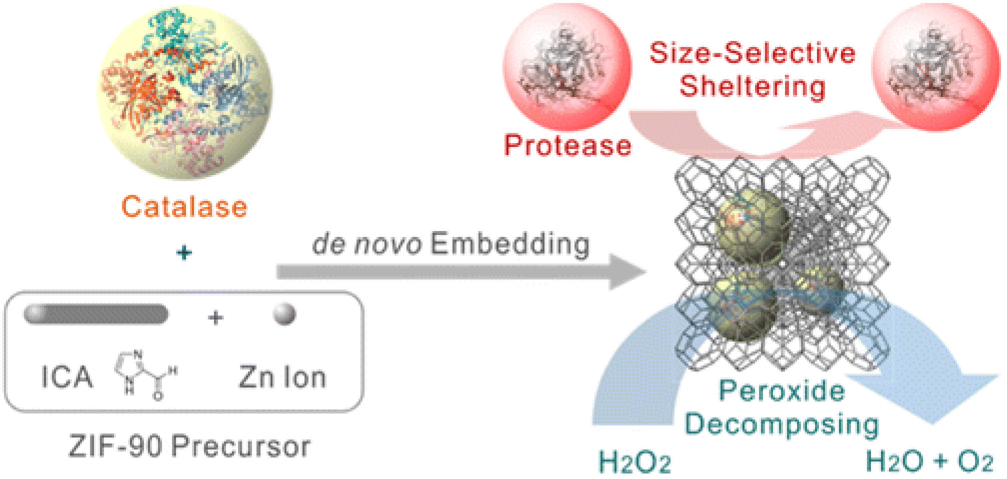
- High-Throughput Enantiopurity Analysis Using Enantiomeric DNA-Based Sensors
  Trevor, A. F.; David, P. V. O.; Zachary, C. H.; Jennifer, M. H. *J. Am. Chem. Soc.* **2015**, *137*, 4198-4206.
  Abstract:



Distinguishing between the two enantiomers of a molecule is a challenging task due to their nearly identical physical properties. Time-consuming chromatography methods are typically required for this task, which greatly limits the throughput of analysis. Here we describe a fluorescence-based method for the rapid and high-throughput analysis of both small-molecule enantiopurity and concentration. Our approach relies on selective molecular recognition of one enantiomer of the target molecule using a DNA aptamer, and the ability of aptamer-based biosensors to transduce the presence of a target molecule into a dose-dependent fluorescence signal. The key novel aspect of our approach is the implementation of enantiomeric DNA biosensors, which are synthesized from d- and l-DNA, but labeled with orthogonal fluorophores. According to the principle of reciprocal chiral substrate specificity, these biosensors will bind to opposite enantiomers of the target with equal affinity and selectivity, enabling simultaneous quantification of both enantiomers of the target. Using the previously reported DNA biosensor for l-tyrosinamide (l-Tym), we demonstrate the ability to rapidly and accurately measure both enantiopurity and concentration for mixtures of l- and d-Tym. We also apply our enantiomeric biosensors to the optimization of reaction conditions for the synthesis of d-Tym and provide mathematical modeling to suggest that DNA biosensors having only modest binding selectivity can also be used for fluorescence-based enantiopurity measurement. This research provides a generalizable method for high-throughput analysis of reaction mixtures, which is anticipated to significantly accelerate reaction optimization for the synthesis of high-value chiral small molecules.

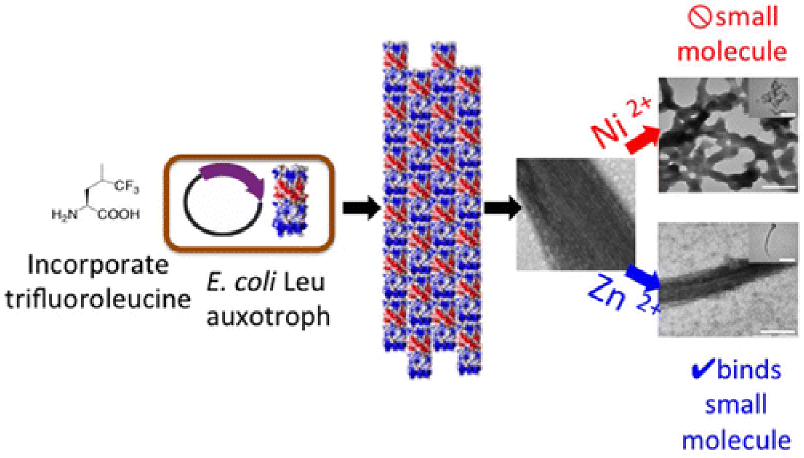
- Imparting Functionality to Biocatalysts via Embedding Enzymes into Nanoporous Materials by a de Novo Approach: Size-Selective Sheltering of Catalase in Metal–Organic Framework Microcrystals
  Shieh, F.-K.; Wang, S.-C.; Yen, C.-I; Wu, C.-C.; Dutta, S.; Chou, L.-Y.; Morabito, J. V.; Hu, P.; Hsu, M.-H.; Wu, K. C.-W.; Tsung, C.-K. *J. Am. Chem. Soc.* **2015**, *137*, 4276-4279.

Abstract:



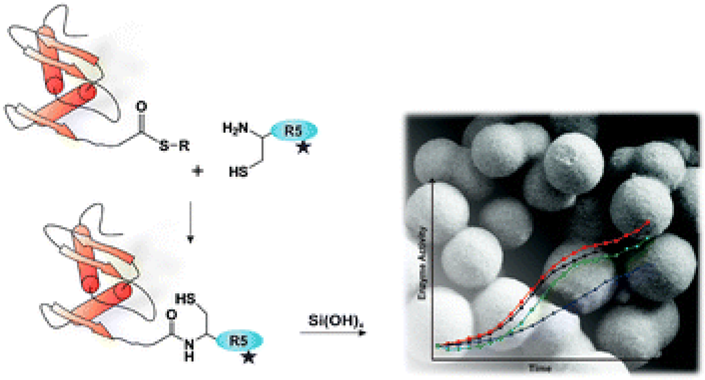
We develop a new concept to impart new functions to biocatalysts by combining enzymes and metal–organic frameworks (MOFs). The proof-of-concept design is demonstrated by embedding catalase molecules into uniformly sized ZIF-90 crystals via a de novo approach. We have carried out electron microscopy, X-ray diffraction, nitrogen sorption, electrophoresis, thermogravimetric analysis, and confocal microscopy to confirm that the ~10 nm catalase molecules are embedded in 2 µm single-crystalline ZIF-90 crystals with ~5 wt % loading. Because catalase is immobilized and sheltered by the ZIF-90 crystals, the composites show activity in hydrogen peroxide degradation even in the presence of protease proteinase K.

- Influence of Fluorination on Protein-Engineered Coiled-Coil Fibers
  More, H. T.; Zhang, K. S.; Srivastava, N.; Frezzo, A.J.; Montclare, K. J. *Biomacromolecules* **2015**, *16*, 1210–1217.
  Abstract:



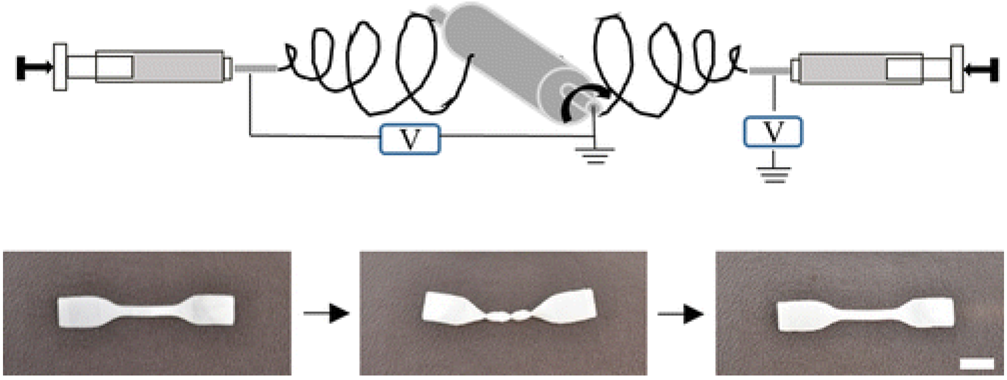
We describe the design and characterization of fluorinated coiled-coil proteins able to assemble into robust nano- and microfibers. Fluorination is achieved biosynthetically by residue-specific incorporation of 5,5,5-trifluoroleucine (TFL). The fluorinated proteins C+TFL and Q+TFL are highly α-helical as confirmed via circular dichroism (CD) and more resistant to thermal denaturation compared to their nonfluorinated counterparts, C and Q. The fluorinated proteins demonstrate enhanced fiber assembly at pH 8.0 with higher order structure in contrast to nonfluorinated proteins, which are unable to form fibers under the same conditions. Ionic strength dependent fiber assembly is observed for fluorinated as well as wild-type proteins in which the fluorinated proteins exhibited more stable, thicker fibers. The fluorinated and nonfluorinated proteins reveal metal ion-dependent small molecule recognition and supramolecular assemblies. In the presence of Zn (II), enhanced thermal stability and fiber assembly is observed for the fluorinated proteins and their nonfluorinated counterparts. Whereas Ni (II) promotes aggregation with no fiber assembly, the stabilization of α-helix by Zn (II) results in enhanced binding to curcumin by the fluorinated proteins. Surprisingly, the nonfluorinated proteins exhibit multiple-fold increase in curcumin binding in the presence of Zn (II). In the context of the growing number of protein-based fiber assemblies, these fluorinated coiled-coil proteins introduce a new paradigm in the development of highly stable, robust self-assembling fibers under more physiologically relevant pH conditions that promotes the binding and release of small molecules in response to external cues.

- Immobilising proteins on silica with site-specifically attached modified silaffin peptides
  Lechner, C. C.; Becker, W. F. C. *Biomater. Sci.* **2015**, *3*, 288-297.
  Abstract:



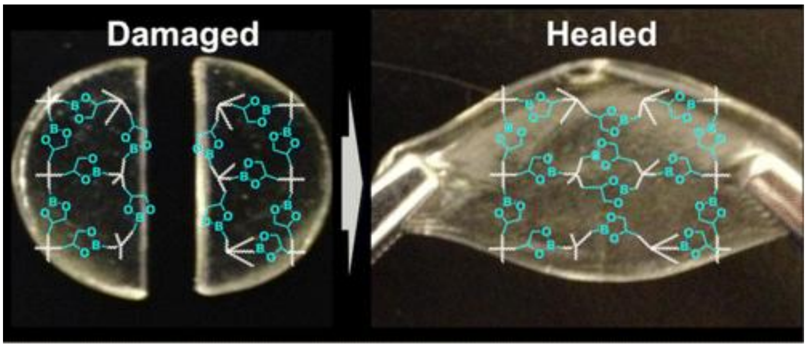
Immobilisation of proteins on solid supports such as silica is commonly applied to improve performance of enzymes under detrimental conditions and to allow enzyme recycling. Silica biomineralisation processes occurring in nature have recently inspired approaches towards mild, biomimetic silica formation. In diatoms, complex posttranslationally modified silaffin peptides are directly involved in formation and patterning of silica cell walls. Here, chemically modified silaffin peptides are used to establish a novel strategy for silica immobilisation of target proteins. Silaffin variants carrying different modifications are covalently linked to eGFP and thioredoxin using expressed protein ligation. Covalent eGFP- and thioredoxin-silaffin conjugates are able to efficiently precipitate silica and control silica properties by choice of different silaffin modifications leading to functional encapsulation of these proteins in silica particles. Covalent protein-silaffin conjugates lead to a distinctly more efficient and homogenous encapsulation of proteins in silica, superior to random protein entrapment resulting from simple co-precipitation. Silica-immobilised proteins are confirmed to be fully active and stabilised against denaturation.

- Dual-Spun Shape Memory Elastomeric Composites
  Robertson, J. M.; Nejad, H. B.; Mather, P. T. *ACS Macro. Lett.* **2015**, *4*, 436—440.
  Abstract:



Thermally responsive shape memory polymers (SMPs) are typically relatively stiff due to the need to vitrify the polymer chains to fix a temporary shape. A need exists for elastomeric SMPs with mechanical properties that more closely match those of human tissue. In this communication, we present a novel approach to fabricate a fully thermoplastic elastomeric SMP. Two polymers are simultaneously electro-spun, or dual-spun, forming a composite fiber mat with a controllable composition. The two polymers were chosen such that one assists in “shape fixing” and the other in “shape recovery”. We envision that the versatility and simplicity of this fabrication approach will allow for large scale production of shape memory elastomeric composites (SMECs) for a wide range of applications.

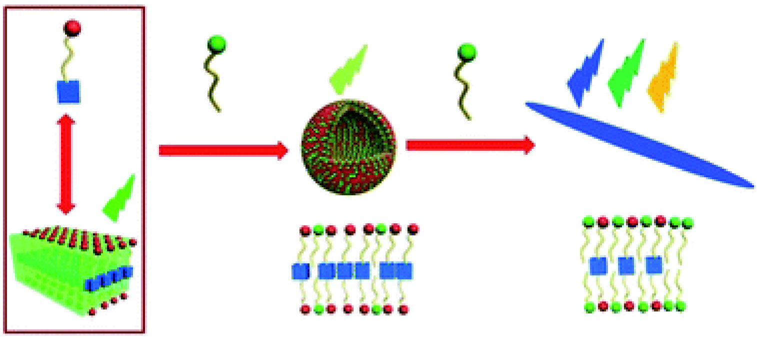
- Room-Temperature Self-Healing Polymers Based on Dynamic-Covalent Boronic Esters
  Cash, J. J.; Kubo, T.; Bapat, A. P.; Sumerlin, B. S. *Macromolecules* **2015**, *48*, 2098–2106.
  Abstract:



Cross-linked polymers constructed with dynamic-covalent boronic esters were synthesized via photo-initiated radical thiol–ene click chemistry. Because the reversibility of the boronic ester cross-links was readily accessible, the resulting materials were capable of undergoing bond exchange to covalently mend after failure. The reversible bonds of the boronic esters were shown to shift their exchange equilibrium at room temperature when exposed to water. Nevertheless, the materials were observed to be stable and hydrophobic and absorbed only minor amounts of water over extended periods of time when submerged in water or exposed to humid environments. The facile reversibility of the networks allowed intrinsic self-healing under ambient conditions. Highly efficient self-healing of these bulk materials was confirmed by mechanical testing, even after subjecting a single site to multiple cut–repair cycles. Several variables were considered for their effect on materials properties and healing, including cross-link density, humidity, and healing time.

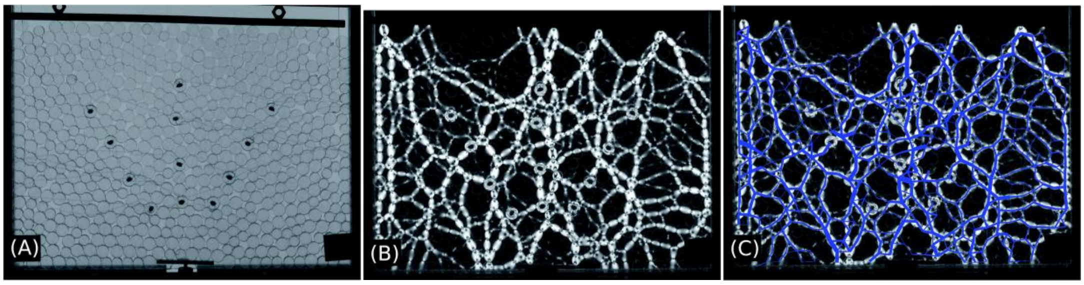
- One platform solid multicolour emission of terthiophene compounds controlled by mixed self-assembly

Liu, S.; Zhao, L.; Yan, Y.; Huang, J. *Soft Matter* **2015**, *11*, 2752-2757.
Abstract:



Via the mixed self-assembling procedure, solid multicolour emission materials based on an amphiphilic terthiophene compound are obtained from a unimolecular platform. Upon controlling the concentration of the cationic surfactant dodecyltriethyl ammonium bromide (DEAB) in the precipitate–monomer equilibrium system of the terthiophene compound TTC4L, mixed self-assembly of TTC4L–DEAB results in diverse structures (including plates, spheres, and needles) with different emission colours. The multicolour emissions are triggered by the different distances between the terthiophene groups in these mixed self-assemblies. Each distance corresponds to a specific molecular state of terthiophene groups, so that emissions corresponding to the monomers, excimers, and aggregates are obtained. Upon variation of the ratio of DEAB and TTC4L, the relative fraction of emissions corresponding to the monomers, excimers, and aggregates of TTC4L changes. This approach may act as a simple method to control the stacking mode of the oligothiophene group which is anticipated to realize unimolecular-platform multicolour emissions.

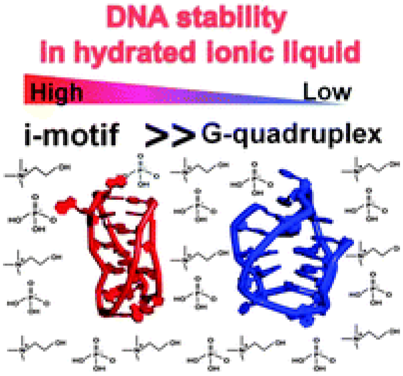
- Extraction of force-chain network architecture in granular materials using community detection
  Bassett, D. S.; Owen, E. T.; Porter, M. A.; Manning, M. L.; Daniels, K. E. *Soft Matter* **2015**, *11*, 2731-2744.
  Abstract:



Force chains form heterogeneous physical structures that can constrain the mechanical stability and acoustic transmission of granular media. However, despite their relevance for predicting bulk properties of materials, there is no agreement on a quantitative description of force chains. Consequently, it is difficult to compare the force-chain structures in different materials or experimental conditions. To address this challenge, we treat granular materials as spatially-embedded networks in which the nodes (particles) are connected by weighted edges that represent contact forces. We use techniques from community detection, which is a type of clustering, to find sets of closely connected particles. By using a geographical null model that is constrained by the particles' contact network, we extract chain-like structures that are reminiscent of force chains. We propose three diagnostics to measure these chain-like structures, and we demonstrate the utility of

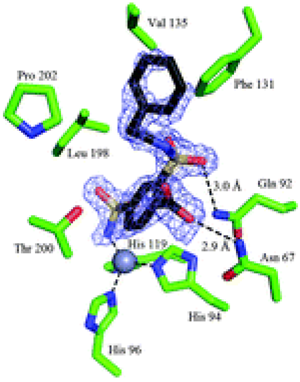
these diagnostics for identifying and characterizing classes of force-chain network architectures in various materials. To illustrate our methods, we describe how force-chain architecture depends on pressure for two very different types of packings: (1) ones derived from laboratory experiments and (2) ones derived from idealized, numerically-generated frictionless packings. By resolving individual force chains, we quantify statistical properties of force-chain shape and strength, which are potentially crucial diagnostics of bulk properties (including material stability). These methods facilitate quantitative comparisons between different particulate systems, regardless of whether they are measured experimentally or numerically.

- i-Motifs are more stable than G-quadruplexes in a hydrated ionic liquid
  Tateishi-Karimata, H.; Nakano, M.; Pramanik, S.; Tanakab, S.; Sugimoto, N. *Chem. Commun.* **2015**, *51*, 6909-6912.
  Abstract:



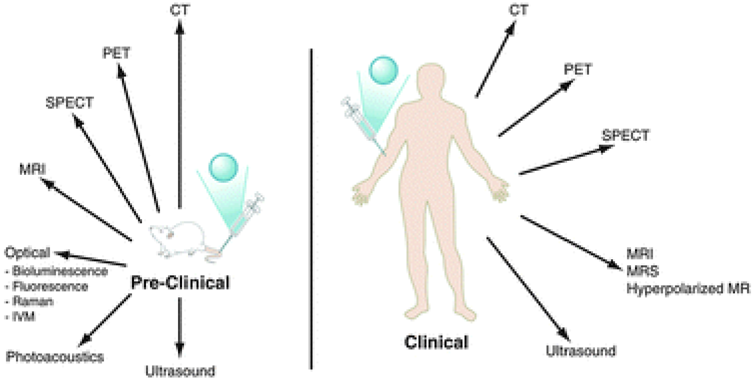
Thermodynamic analyses and molecular dynamics calculations demonstrated that i-motifs in a hydrated ionic liquid of choline dihydrogen phosphate (choline dhp) were more stable than G-quadruplexes due to choline ion binding to loop regions in the i-motifs. Interestingly, the i-motifs formed even at physiological pH in the choline dhp-containing solution.

- X-ray crystallography-promoted drug design of carbonic anhydrase inhibitors
  Ivanova, J.; Leitans, J.; Tanc, M. ; Kazaks, A. ; Zalubovskis, R. ; Supuran, C. T.; Tars, K. *Chem. Commun.* **2015**, *51*, 7108-7111.
  Abstract:



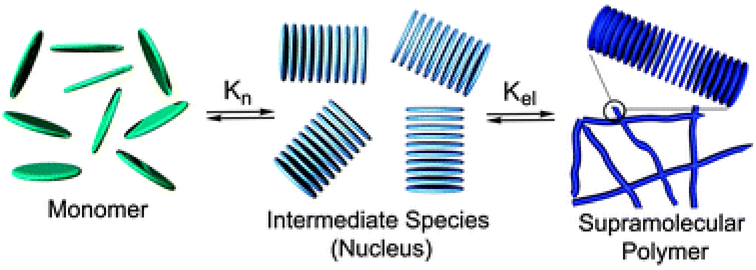
1-N-Alkylated-6-sulfamoyl saccharin derivatives were prepared and assayed as carbonic anhydrase inhibitors (CAIs). During X-ray crystallographic experiments an unexpected hydrolysis of the isothiazole ring was evidenced which allowed us to prepare highly potent enzyme inhibitors with selectivity for some isoforms with medical applications.

- Critical design issues in the targeted molecular imaging of cell surface receptors
  Parker, D.; Sim, N. *Chem. Soc. Rev.* **2015**, *44,* 2122-2134.
  Abstract:



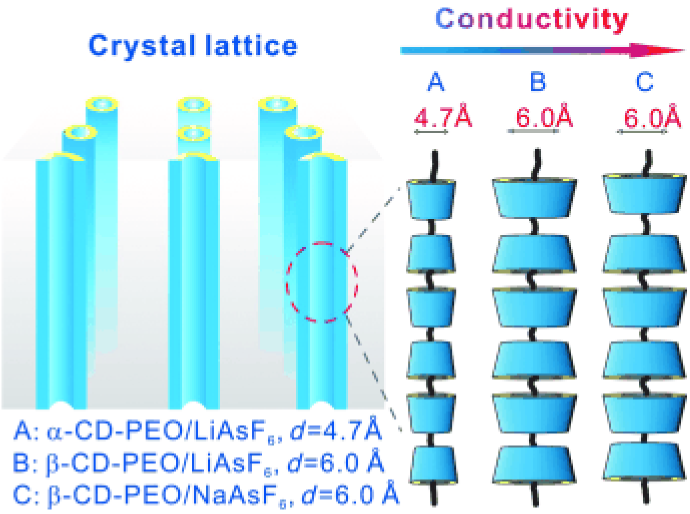
**A cool paper that will explain you what still needs to be done in this very important field.** The imaging of cell-surface receptors can be achieved using several methods, including single photon emission tomography (SPECT) positron emission tomography (PET), optical imaging and magnetic resonance imaging (MRI). The application of targeted MRI contrast agents is particularly well-suited to this task, provided that the agents reach the desired site efficiently and selectively. In addition, they should bind reversibly to the cell-surface receptor and give rise to a large change in cellular relaxation rate, in competition with binding to the natural substrate. Such approaches offer promise in the molecular imaging of neurotransmission in the brain, using conjugates that selectively target dopamine or glutamate receptor sub-types. Strategies based on the use of competitive antagonist vectors offer particular scope, as such conjugates are generally not taken into the target cell following cell surface receptor binding, in contrast to the use of MRI contrast agents based on agonists that tend to be internalised quickly or are designed to target intracellular sites.

- Strategies to create hierarchical self-assembled structures *via* cooperative non-covalent interactions
  Rest, C.; Kanadelli, R.; Fernandez, G. *Chem. Soc. Rev*. **2015**, *44*, 2543-2572.
  Abstract:



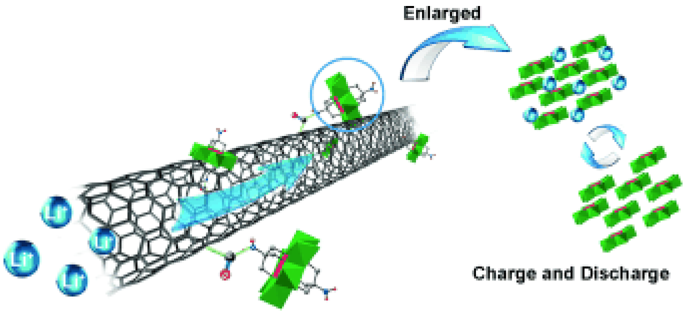
**Good insight into cooperative phenomena. Must read.** Cooperative phenomena are common processes involved in the hierarchical self-assembly of multiple systems in nature, such as the tobacco mosaic virus and a cell's cytoskeleton. Motivated by the high degree of order exhibited by these systems, a great deal of effort has been devoted in the past two decades to design hierarchical supramolecular polymers by combining different classes of cooperative interactions. In this review, we have classified the field of supramolecular polymers depending on the cooperative non-covalent forces driving their formation, with particular emphasis on recent examples from literature. We believe that this overview would help scientists in the field to design novel self-assembled systems with improved complexity and functionalities.

- Ionic Conductivity of β-Cyclodextrin–Polyethylene-Oxide/Alkali-Metal-Salt Complex
  Yang, L.-Y.; Fu, X.-B.; Chen, T.-Q.; Pan, L.-K.; Ji, P.; Yao, Y.-F.; Chen, Q. *Chem. Eur. J.* **2015**, *21*, 6346–6349.
  Abstract:



Highly conductive, crystalline, polymer electrolytes, β-cyclodextrin (β-CD)–polyethylene oxide (PEO)/$LiAsF_6$ and β-CD–PEO/$NaAsF_6$, were prepared through supramolecular self-assembly of PEO, β-CD, and $LiAsF_6$/$NaAsF_6$. The assembled β-CDs form nanochannels in which the PEO/$X^+$ (X=Li, Na) complexes are confined. The nanochannels provide a pathway for directional motion of the alkali metal ions and, at the same time, separate the cations and the anions by size exclusion.

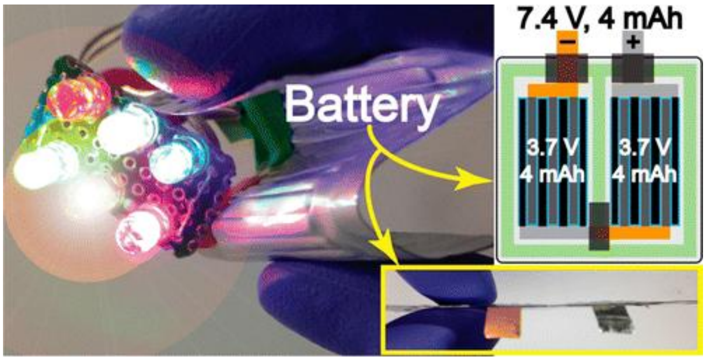
- Covalent Attachment of Anderson-Type Polyoxometalates to Single-Walled Carbon Nanotubes Gives Enhanced Performance Electrodes for Lithium Ion Batteries
  Ji, Y.; Hu, J.; Huang, L.; Chen, W.; Streb, C.; Song, Y.-F. *Chem. Eur. J.* **2015**, *21*, 6469–6474.
  Abstract:



Single-walled carbon nanotubes (SWNTs) covalently functionalized with redox-active organo-modified polyoxometalate (POM) clusters have been synthesized and employed as electrode materials in lithium ion batteries. The Anderson cluster $[MnMo_6O_{24}]^{9-}$ is functionalized with Tris ($NH_2C(CH_2OH)_3$) moieties, giving the new organic–inorganic hybrid $[N(nC_4H_9)_4]_3[MnMo_6O_{18}\{(OCH_2)_3CNH_2\}_2]$. The compound is then covalently attached to carboxylic acid-functionalized SWNTs by amide bond formation and the stability of this nanocomposite is confirmed by various spectroscopic methods. Electrochemical analyses show that the nanocomposite displays improved performance as an anode material in lithium ion batteries

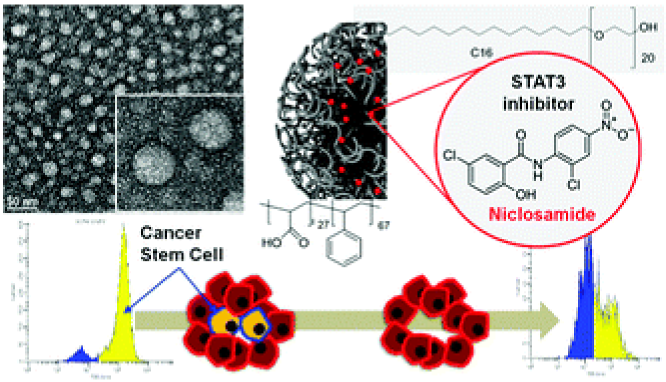
compared with the individual components, that is, SWNTs and/or Anderson clusters. High discharge capacities of up to 932 mAh $g^{-1}$ at a current density of 0.5 mA $cm^{-2}$ can be observed, together with high long-term cycling stability and decreased electrochemical impedance. Chemisorption of the POM cluster on the SWNTs is shown to give better electrode performance than the purely physisorbed analogues.

- A Half Millimeter Thick Coplanar Flexible Battery with Wireless Recharging Capability
  Kim, J.-S.; Ko, D.; Yoo, D.-J.; Jung, D. S.; Yavuz, C. T.; Kim, N.-I.; Choi, I.-S.; Song, J. Y.; Choi J. W. *Nano Lett.* **2015**, *15*, 2350–2357.
  Abstract:



Most of the existing flexible lithium ion batteries (LIBs) adopt the conventional cofacial cell configuration where anode, separator, and cathode are sequentially stacked and so have difficulty in the integration with emerging thin LIB applications, such as smart cards and medical patches. In order to overcome this shortcoming, herein, we report a coplanar cell structure in which anodes and cathodes are interdigitatedly positioned on the same plane. The coplanar electrode design brings advantages of enhanced bending tolerance and capability of increasing the cell voltage by in series-connection of multiple single-cells in addition to its suitability for the thickness reduction. On the basis of these structural benefits, we develop a coplanar flexible LIB that delivers 7.4 V with an entire cell thickness below 0.5 mm while preserving stable electrochemical performance throughout 5000 (un)bending cycles (bending radius = 5 mm). Also, even the pouch case serves as barriers between anodes and cathodes to prevent Li dendrite growth and short-circuit formation while saving the thickness. Furthermore, for convenient practical use wireless charging via inductive electromagnetic energy transfer and solar cell integration is demonstrated.

- Enriched inhibition of cancer and stem-like cancer cells via STAT-3 modulating niclocelles
  Misra, S. K.; Jensena, T. W.; Pan, D. *Nanoscale* **2015**, *13*, 7127–7132.
  Abstract:



We describe for the first time a therapeutic strategy to target stem-like cancer cells *via* STAT-3 modulation using a nanomedicine approach. Niclocelle, a niclosamide loaded rigid core mixed micelle, was synthesized from a self-assembled well-defined amphiphilic diblock copolymer and an FDA-approved signal transducer and activator of transcription factor 3. Followed by a rigorous physico-chemical characterization, niclocelles were evaluated biologically for cytotoxicity and apoptosis in human melanoma (C32) and breast cancer (MDA-MB231 and MCF-7) cells. Niclocelles were found to selectively reduce the CD44+ stem cell population in C32 cells *via* STAT-3 modulation.