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A tool, an app and a field: supramolecular photochemistry discoveries from Sri Lanka and Northern Ireland

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A main principle underlying fluorescent sensors is based on photoinduced electron transfer (PET), according to which a switching 'on' of fluorescence in response to chemical[1] and biochemical[2] analytes can be designed. Since its generalization in Colombo, Sri Lanka, it involves >1200 laboratories worldwide. Some of these sensors are serving in critical care units in hospitals and in ambulances, performing blood diagnostics (Figure 1). These form the basis of a billion-dollar industry.[3] Other sensors visualize intracellular players. Yet others map species distributions in nanometric spaces.[4] These spaces are too small for the tiniest silicon-based wireless devices to enter. Our introduction of molecular logic gates[5,6] from Belfast, Northern Ireland, allows us to build more complex sensors and micro-object identification systems. More complex logic operations[7-9] and even human-scale computations, e.g. edge detection of objects[10] and outline drawing (Figure 2), are achieved. >1965 laboratories have contributed to this field. For a short video, see: www.youtube.com/watch?v=sLGnZDP5Ecg.

Keywords: Fluorescent sensors; fluorescent PET sensors; Photoinduced electron transfer; Molecular logic-based computation; OPTI blood electrolyte and gas analyzer

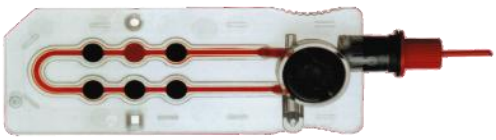

Figure 1. Blood electrolyte analyser

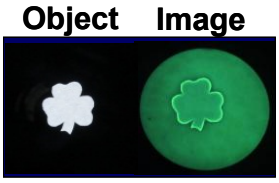

Figure 2. Molecular outline drawing

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Organic reaction development using Digital Chemistry

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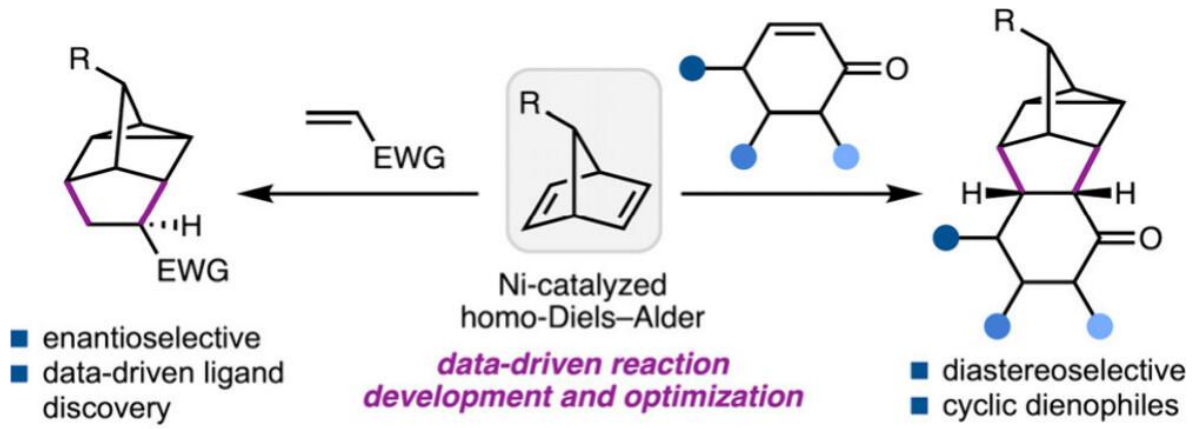
Digital Chemistry is a transformative field combining high-throughput experimentation/automation, computation, and machine learning which can accelerate reaction discovery and optimization in organic synthesis. By systematically interrogating reaction space, it enables rapid identification of reactivity trends and mechanistic insight. Here, this approach is demonstrated through the development of a Ni-catalyzed homo-Diels–Alder (hDA) reaction (*J. Am. Chem. Soc.* **2025**, *147*, 31175).

The Ni-catalyzed hDA reaction is a convergent yet underexplored strategy for constructing bridged bicyclic systems. Using the *kraken* monophosphine descriptor library, reactions of acyclic and cyclic electron-deficient olefins were studied, and key ligand features governing reactivity were identified. This enabled the discovery of (*S*)-AntPhos as a chiral ligand for the enantioselective had of acyclic dienophiles, although these conditions were not compatible with cyclic substrates.

Mechanistic and computational studies suggested the involvement of Ni(I) species and revealed divergence between cyclic and acyclic enone dienophiles. Guided by these insights, reaction space and Bayesian optimization identified conditions that enable reactivity with cyclic substrates, expanding the scope. The resulting cycloadducts were further transformed into bicycloheptane frameworks *via* cyclopropane cleavage, providing rapid access to structurally complex molecules.

Overall, this work demonstrates how Digital Chemistry can uncover reactivity, resolve mechanistic differences, and accelerate catalytic reaction development.

Keywords: Digital chemistry; Cycloaddition; Ni catalysis; Mechanism



Reprogramming alkylamine reactivity for direct C–C bond formation

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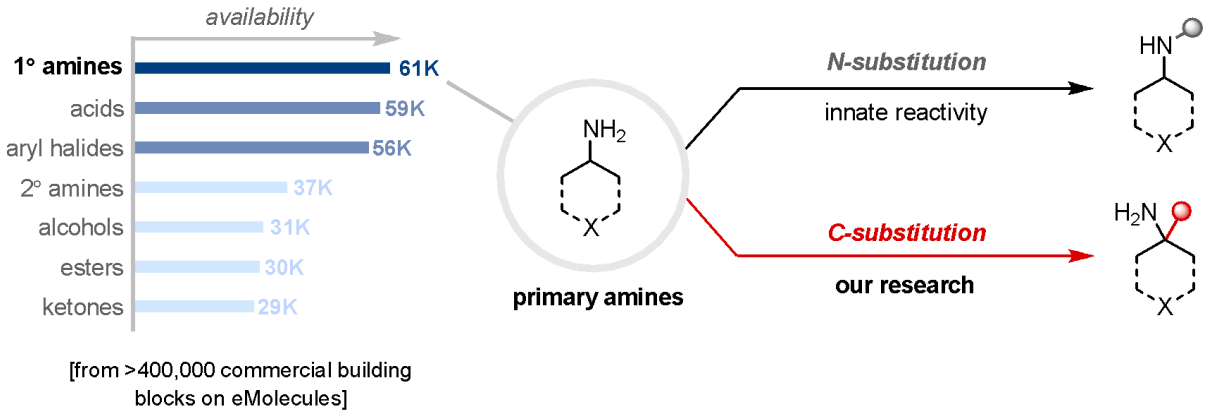
We have uncovered a new mode of reactivity for unprotected alkylamines, enabling them to participate in direct C–C bond formation as carbon nucleophiles, rather than conventional nitrogen nucleophiles. This conceptually distinct strategy for amine synthesis, enabled by the merger of photoredox and hydrogen atom transfer (HAT) catalysis, provides rapid access to α-tertiary and spirocyclic amine architectures through selective α-C–H functionalisation. The process is readily translated to automated and continuous-flow platforms, offering a scalable route to structurally complex amines with broad potential for application in medicinal chemistry and drug discovery.

Keywords: Photoredox catalysis; C–H functionalisation; Radicals; Amines; Heterocycles



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Synergistic hybrid nanogenerators for respiration-driven smart face masks

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Polyvinylidene difluoride (PVDF) polymers are key materials for enhancing the performance of triboelectric nanogenerators (TENGs) due to their exceptional dielectric and piezoelectric properties. In this study, we systematically investigate a series of PVDF homopolymers, copolymers and terpolymers to explore the impact of dielectric and piezoelectric properties on TENG output. A detailed analysis of structural evolution is conducted to understand the microscopic features of dielectric and piezoelectric polarization kinetics during contact electrification (CE). Our findings reveal that optimizing both dielectric and piezoelectric properties of PVDF materials significantly improves TENG performance. Terpolymers with high dielectric constants suppress electron backflow after CE, while the piezoelectric polarization of β-phase PVDF alters electron energy levels before CE. These complementary mechanisms were characterized using electric displacement–electric field (*D–E*) hysteresis loops and dielectric measurements under DC bias. A phase change from relaxor ferroelectric to regular ferroelectric was observed in PVDF terpolymers as the temperature was cooled from room temperature to -40 ℃, leading to enhanced piezoelectric polarization and TENG outputs. It is found that the dielectric and piezoelectric contributions to TENG performance do not act synergistically. Experimental phase transitions confirm a trade-off between the two, being reported for the first time through electric-field-induced measurements. The relaxor phase is dominated by dielectric properties due to weak dipole interactions, while the regular ferroelectric phase is piezoelectric-dominant because of coherent cooperative dipole interaction. In a practical demonstration, we integrate PVDF fiber-based respiration-driven TENGs (R-TENGs) into face masks to harvest mechanical energy through breathing. R-TENGs achieve self-powered electroporation by harvesting mechanical energy from the bi-directional motion of breathing. Leveraging a lightning-rod effect to amplify the local electric field, the device enables rapid disinfection through nanowire-assisted electroporation. By integrating energy harvesting with real-time disinfection in the R-TENGS, this work paves the way for self-sustained smart protective gear that sets a new benchmark in personal safety.

Keywords: Polyvinylidene difluoride; triboelectric nanogenerators; face mask

Fluorescent chemosensors and imaging agents

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Sensors and imaging agents can be used to monitor analytes within physiological, environmental, and industrial scenarios. The interactions between the "chemosensor" and an analyte of choice occurs on a molecular level and as such gathering and processing the information is challenging. Therefore, I will outline the trials and challenges encountered in the development of several robust chemical molecular sensors "chemosensors" able to detect such analytes selectively and signal or map their concentration in a biological or environmental scenario. During the talk you will be introduced to a variety of fluorescent probes designed for diols (D-glucose), and redox imbalance. With the goal being the development of chemosensors capable of determining the concentration (and location) of a target species in any medium. Particular attention will be paid to the underlying chemistry associated with the construction of practical chemosensors for both sensing and imaging applications.

Keywords: Fluorescence; Chemosensors; Imaging agents

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Design of peptides for functional cell-interfacing assembly

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Designing peptides that self-assemble at cellular interfaces holds great promise for sensing and modulating cellular activities. A key challenge lies in the control of inter-peptide packing and alignment via residue-residue interactions within folded peptide scaffolds. We reported a strategy that integrates multiple cohesive faces or sites into individual α-helical coiled-coil peptides, enabling modular assembly with high degree of compositional and morphological control. On the basis of this platform, we recently fabricated dynamic one-dimensional assemblies that elongate differently responding to the quantities of proteins on encountering cell surfaces. This sets the foundation of generating a cell recognition system targeting the feature (i.e. quantity and identity) of cell-surface molecular groups, rather than solely to individual molecular identities. The system was further translated into bifunctional cell decorations, which confer cell survival under environmental stressors via segregation and support cell-cell engagements to maintain intercellular communication and interactions. Applied to immune cells (natural killer cells), this system elicited enhanced and specific antitumor effects both in vitro and in vivo.

Keywords: peptide assembly; peptide sequence; inter-peptide interactions; cell recognition; cell-surface molecules

Chirality of peptidomacrocycle

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We recently reported a β-templated macrocyclization protocol that allows a feasible access to the peptidomacrocycles containing 4 amino acid residues (F. Gou, et al., *J. Am. Chem. Soc.*, 2023, *145*, 9530). 2,6-Pyridinedicarbonylaminoacidhydrazide was allowed to react with 1,4-benzenedithioisocyanate to form "2+2" 46-membered macrocycles. Depending on the chirality of the amino acid residues in the dihydrazide reactant, the macrocycles could have the chirality combinations of LLLL, DDDD, LLDD, LDDL and LDLD (clockwise). It is interesting to observe when the racemate LL-/DD-dihydrazide or the meso LD-dihydrazide was used to react, the exclusive or preferential products are LLDD- and LDDL-macrocycles, respectively. We found that this preferential is due to the strong and ordered intermolecular packing in the solid (microcrystalline) products of them, that the heterochiral products in the solid state is more stable. We also found that the binding of these stereoisomers with achiral sulfate anion is not the same, which means that a conformation change upon anion binding could have happened. We thank the contributions of students of our group and the supports from the NSF of China, the MOE of China, and the Xiamen City.

Asymmetric site-induced oxidation pathways in acidic oxygen evolution reactions

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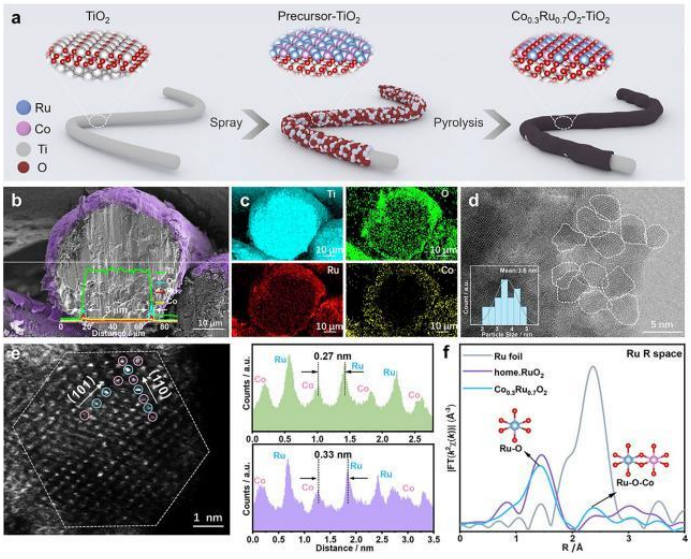
Hydrogen, as a vital energy carrier, serves as an effective solution to the intermittency and volatility of wind and solar power generation. Proton exchange membrane hydrogen production technology offers low resistance, high energy efficiency, compact systems, high pressure tolerance, and rapid start-stop capabilities, making it particularly suitable for fluctuating inputs from renewable energy sources. However, the efficiency of this technology is currently limited by the need for high-performance, low-cost oxygen evolution reaction (OER) electrocatalysts that can operate under strong acidic and oxidative conditions.In this study, we present a cobalt-doped ruthenium dioxide on TiO_2 ($Co_{0.3}Ru_{0.7}O_2$-TiO_2) as electrocatalysts toward an acidic oxygen evolution reaction (AOER). The $Co_{0.3}Ru_{0.7}O_2$-TiO_2 achieves an impressive overpotential of 322 mV at 1 A cm^{-2} and demonstrates stable operation for over 1000 hours at 500 mA cm^{-2} in a PEMWE device, with hydrogen production costs well below \$2/kg. Comprehensive experimental and theoretical calculation results demonstrate that the doped Co atoms into the rutile RuO_2 lattice optimize geometric configuration and enable Ru-Co dual-site parallel oxidation pathway for efficient *O-O* bond formation. The matched lattices of $Co_{0.3}Ru_{0.7}O_2$ and TiO_2 promote electron rearrangement leading to the parallel ad/desorption of intermediates across the dual sites. This work provides valuable insights into the design of high-performance and stable electrocatalysts for AOER, driving the path toward scalable PEMWE technology.

Keywords: Proton exchange membrane water electrolysis (PEMWE); Acidic oxygen evolution reaction (AOER); Ru-based catalysts; Oxidation pathway



Supramolecular sensor devices based on organic field-effect transistors

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Supramolecular materials have the potential to be applied to analytical tools owing to their fascinating functions. However, these materials have not been satisfactorily used for practical sensor devices. In this presentation, we propose an approach for the development of supramolecular sensor devices based on organic field-effect transistors (OFETs).[1] OFETs are electronic devices whose switching characteristics are controlled by applying a gate voltage. The device characteristics are governed by the assembly of organic semiconducting molecules; therefore, OFETs can be referred to as supramolecular devices. Owing to their beneficial device properties, OFETs functionalized with appropriate molecular recognition materials can offer advantages in sensitive detection compared with conventional electrochemical sensing methods. In receptor design, biological materials such as enzymes and antibodies have been employed because of their favorable specificity toward analytes based on the lock-and-key recognition principle. However, the range of detectable analytes is limited by the available library of biological recognition materials. Thus, the use of synthetic receptors based on molecular recognition chemistry is a promising approach for designing recognition sites.

In this study, molecularly imprinted polymers (MIPs) were applied as recognition materials for selective detection. MIPs provide three-dimensional recognition sites for specific analytes, as pre-organized structures consisting of a template and functional monomers can be optimized using quantum chemical calculations. Such optimized MIP structures contribute to selective detection even in the presence of interferents.[2] In contrast, the inherent cross-reactivity of supramolecular receptors can be applied to simultaneous detection using pattern recognition methods.[3] This presentation will highlight the potential of this approach for the realization of supramolecular sensor devices based on the integration of organic electronics, molecular recognition chemistry, and polymer chemistry.

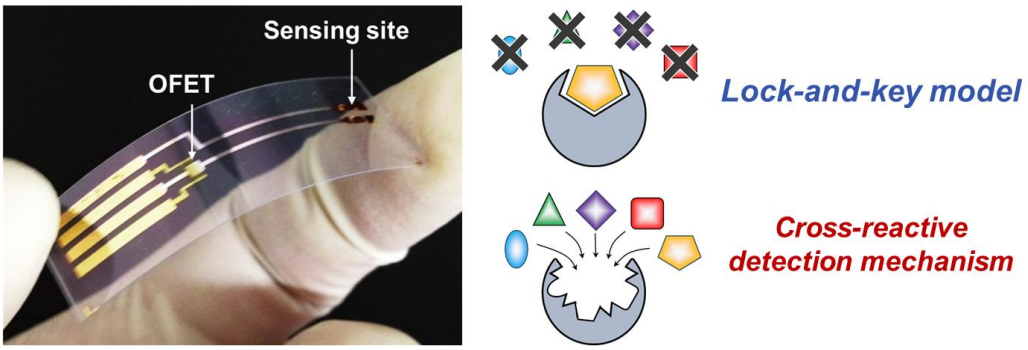

Figure 1. OFET-based chemical sensor device functionalized with receptors designed based on molecular recognition chemistry.

Keywords: Organic transistor; Chemical sensor; Molecularly imprinted polymer; Supramolecular receptor; Real-sample analysis

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The benzannulated xanthenoid NIR/SWIR dyes for in vivo bioimaging and biosensing

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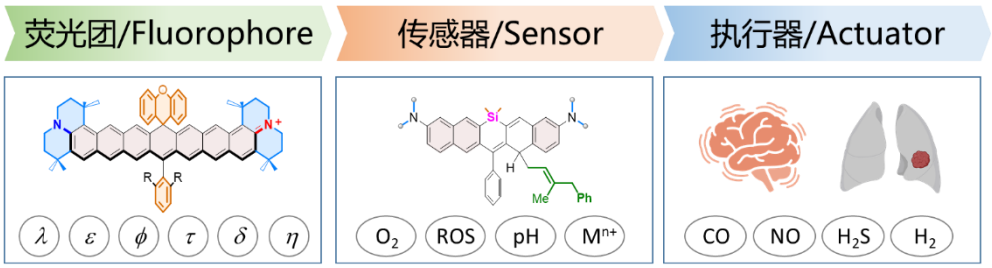
The NIR/SWIR light, i.e., 8000-2000 nm, exhibits high tissue-penetration and biocompatibility, and is a premium spectral window for in vivo bioimaging and biosensing. Yet, exploiting the NIR/SWIR window remains challenging primarily due to the scarcity of high-performance dyes absorbing and emitting in the NIR/SWIR.

The Qian/Yang group has focused on developing NIR/SWIR dyes with a xanthenoid scaffold, e.g., **EC5** and **ESi5** for 800-900 nm, and **EC7** and **ESi7** for 1150-1300 nm. The highlight of these dyes is a rigid push-pull scaffold, which imparts high fluorescence quantum yield. Additionally, bulky groups were installed to shield the central methine carbon from nucleophilic attack. Besides, tetramethylated julolidine moiety was a premium electron-donating headgroup for super-high molar absorptivity, *i.e.*, 3.95 × 10^5 $cm^{-1}M^{-1}$ for **EC7** in CH_2Cl_2. The use of a silicon-based bridging group enhances fluorescence quantum yield by suppressing low-frequency vibration normal modes. One-color and two-channel high-contrast in vivo images of mouse vasculature, liver, gallbladder, intestine, and kidney will be presented. With the growth in the use of small animals in biological and medical studies, NIR/SWIR fluorescent probes for all biorelevant substrates are sought after. The difficulty is the lower chemical stability of NIR/SWIR dyes compared to the visible dyes.

EC5/**ESi5** are stable and bright, and hence superior templates for developing NIR fluorescent probes, compared to the polymethine-type dyes. We report that asymmetric addition of a sensing moiety to the push-pull conjugative backbone is a feasible probe-design strategy with benzannulated xanthenoid dyes. We recently designed a NIR-probe for the early detection of ferroptosis. Highly oxidative radicals implicated in lipid peroxidation abstract the benzylic hydrogen of the probe (LHA930), and release a **ESi5** fluorophore via a radical-initiated rearrangement/β-fragmentation cascade, with a concomitant fluorescence turn-on of 200 fold. The probe was used in real-time monitoring of the drug-induced liver injury.



Xanthenoid Dyes	Ferroptosis Probe	Gas-Walz Therapy
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Real-time Sensing of Microalgal Photosynthesis Using a Droplet-Based Triboelectric Nanogenerator

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Our climate is changing. And the climate change is primarily due to the "greenhouse effect" associated with human activities, which is an unavoidable consequence of an ever-advancing civilization. Studies show that the average global temperature over its fluctuation during the past 10 centuries followed natural events (such as volcanic eruptions and variations in solar flux) very well until the 20th century. Then there was rapid rise, associated with the increase in atmospheric carbon dioxide, from its preindustrial level (from 1850 to 1900) of 280 parts per million (ppm) to the present level of 420+ ppm, which is still accelerating. Today, carbon capture and storage has emerged as a promising technology to combat greenhouse gas emissions, drawing growing attention as the world seeks solutions to flatten the climate curve. A promising mitigation strategy is the development of artificial carbon sinks to enhance carbon dioxide removal through biological fixation. Microalgae, among the most efficient natural carbon sinks, offer superior carbon fixation rates and faster growth compared to terrestrial plants, making them ideal candidates for engineered carbon capture systems. Therefore, understanding microalgal photosynthesis dynamics is critical for enhancing cultivation efficiency and maximising carbon fixation potential. However, conventional photosynthesis rate monitoring techniques often suffer from low temporal resolution or limited accuracy, hindering real-time optimisation of photometric parameters and leading to suboptimal system performance. Here, a new technique is developed to monitor the photosynthesis dynamics of *Chlorella vulgaris* using a droplet triboelectric nanogenerator (D-TENG), enabling real-time insights into its response to various photometric excitations through varying degrees of charge interactions at the liquid-solid interface. For the first time, this work demonstrates the potential of D-TENG as a promising real-time, non-invasive, and accurate tool for monitoring microalgal photosynthesis, opening new avenues for probing the carbon fixation capabilities of various microalgae species under diverse lighting conditions.

Chasing nature's anion binders

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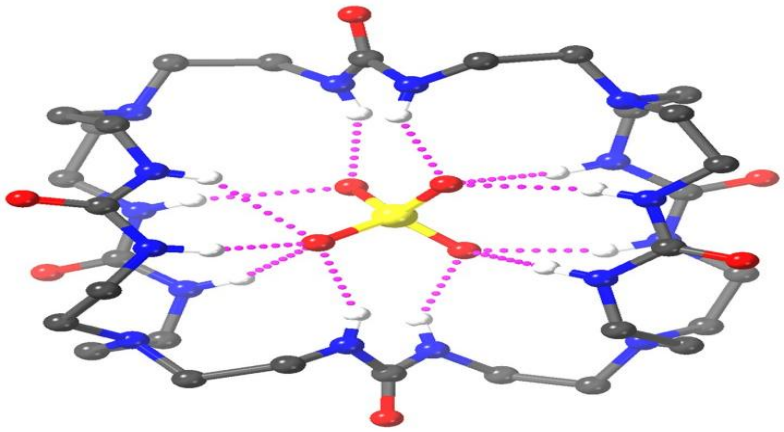
Sulfate binding proteins from bacteria capture strongly-hydrated sulfate anions from the environment as an initial step of nutrient uptake. Surprisingly, their interaction with sulfate was purely hydrogen bonding, without the formation of salt links, yet achieving remarkable sub-micromolar affinities. Supramolecular chemists have faced significant challenges in using hydrogen bonds to attain comparable host-guest binding strengths in water. This presentation will discuss our recent progress in developing uncharged macrocycles and cages that can bind to hydrophilic anions including fluoride, chloride, sulfate and chromate in water, with affinities up to micromolar levels that match nature's counterparts. Contrasting large biological receptors, the high affinity anion binding was achieved with minimalistic molecular receptors with the polar binding sites exposed to water. We further discuss how these macrocycles and cages can assemble to form supramolecular polymers, ionic liquids or solid materials to achieve rapid and selective removal of toxic anions

Keywords: Hydrogen bonds; Anion receptors; Molecular cages; Water purification



Macrocyclic-ring formation enhanced by silver bridging: quantitative fluorescence sensing and specific detection of norepinephrine

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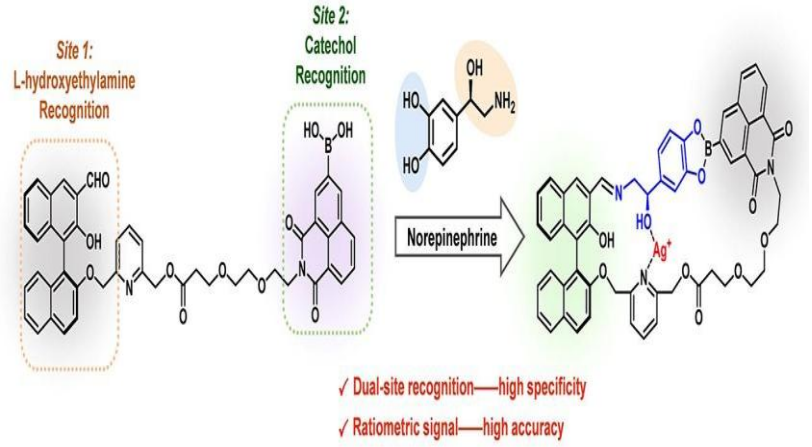
Norepinephrine is an important intermediate in the metabolism of catecholamines, and as such reflects the fluctuation in of catecholamines in biological systems. Abnormal levels of NE are closely related with neuro-degenerative and cardiovascular diseases. Significantly, the accurate monitoring of NE is essential for diagnosing or tracking the progression of diseases. However, its precise quantification in biological fluids by readily available tools is challenging. Herein, focusing on the unique structures of NE including L-hydroxyethylamine and catechol units, we designed and synthesized a dual-site fluorescent probe that can specifically detect NE. The probe can recognize the two characteristic groups of NE to form a macrocyclic-ring, accompanied by a ratiometric fluorescence change. Significantly, the addition of Ag+ significantly reduced the detection time and enhanced the signal due to silver bridging. As such, the probe can be used to quantify NE in serum and urine, and is expected to provide a reliable tool for studying NE-related biology, as well as providing an appropriate sensing component for the development of point of care detection devices.

Keywords: Macrocyclic-ring; Quantitative fluorescence sensing; Specific detection; Silver bridging; Norepinephrine



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Chirality-dependent biomimetic transmembrane transport

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Natural transmembrane channels are intrinsically chiral; however, how chirality governs transport behavior and the structural origin of this phenomenon in natural systems remain largely unexplored. In this report, we address this fundamental question by designing a series of bilateral chiral alkylamide phenylalanine acylhydrazone derivatives that self-assemble into nanopores within biomimetic phospholipid membranes (Figure 1). These artificial nanopores efficiently transport ions and provide a unique platform for systematically investigating chirality-controlled transport.

Our key conceptual advance is the discovery of a distinct V-shaped relationship between transmembrane transport activity and enantiomeric excess (*ee*). Enantiopure transporters exhibit high activity, whereas racemic mixtures lead to a dramatic suppression of ion transport. This finding reveals a nonlinear regulatory mechanism of enantiomeric composition on channel performance, representing a previously unrecognized mode of chirality-controlled function in artificial ion channels. This unique behavior enables the construction of a chirality-gated transport system through the sequential addition of equimolar enantiomers. Mechanistic studies show that heterochiral co-assembly within the membrane induces dense molecular packing, effectively closing the pores and inhibiting ion translocation. This behavior contrasts sharply with the widely studied CD-*ee* relationships in supramolecular chemistry.

We further demonstrate that enantiomers display intrinsically different transport efficiencies in chiral lipid membranes, with L-enantiomers outperforming their D-counterparts due to more favorable membrane insertion. Moreover, in recent work, we efficiently prepared peptide-based acylhydrazone macrocycles and found that they act as efficient anion transporters, exhibiting enantiomer-dependent activity: macrocycles composed of L-amino acids are ten times more active than their D-counterparts. Together, these findings establish a direct structure-activity relationship linking chirality, molecular recognition and assembly, and transmembrane transport function, offering new insight into the functional advantages of homochirality in natural channels.

Keywords: Transmembrane transport; Artificial nanopore; Activity-*ee* dependence; Chirality-gated transport; Peptide-based acylhydrazones

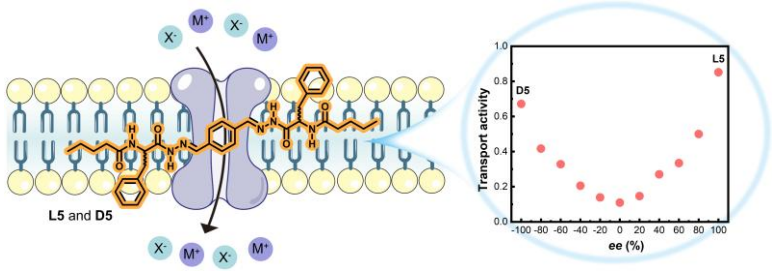

Figure 1. Chirality-dependent biomimetic transmembrane transport. In artificial nanopores assembled from chiral phenylalanine acylhydrazones, ion transport activity exhibits a distinct V-shaped dependence on enantiomeric excess (*ee*).