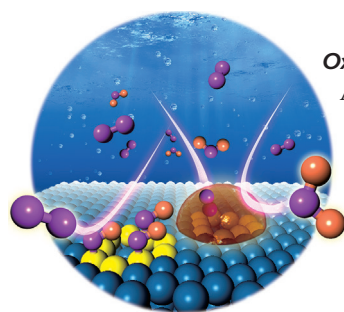
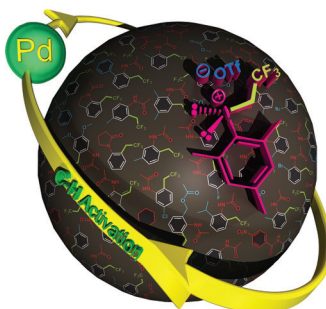




... to enter an enchanted world where corannulenes fall from the sky and form porous crystalline scaffolds. In their Communication on page 2195 ff., N. B. Shustova et al. merge the inherent properties of hybrid crystalline frameworks with the intrinsic properties of redox-active π -bowls for the first time. The dancing corannulene ballerinas are the opening act to the innovative corannulene science developed herein (cover design: Ekaterina A. Dolgoplova).

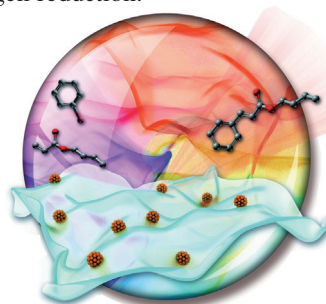
Trifluoroethylation

In their Communication on page 1988 ff., Z. Novák and co-workers describe the use of a trifluoroethyl-(mesityl)iodonium salt for the simple and efficient palladium-catalyzed trifluoroethylation of aromatic compounds by C–H activation.



Oxygen Reduction Reaction

A hydrophobic ionic liquid protects catalytically active Pt sites from being poisoned. B. J. M. Etzold and co-workers show in their Communication on page 2257 ff. that this approach leads to dramatically boosted oxygen reduction.



Nanosheets

In their Communication on page 2167 ff., Y. Yin, T. Zhang, et al. show how transition-metal hydroxide nanosheets less than 5 nm thick can be modified to form highly active Heck reaction catalysts.

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Service

Spotlight on Angewandte's Sister Journals

1952 – 1955

Author Profile



"In a spare hour, I enjoy a good glass of red wine and Bill Evans.

My biggest inspiration is my family. ..."

This and more about Haoshen Zhou can be found on page 1956.

Haoshen Zhou _____ 1956

News

MacArthur Fellowships:
W. R. Dichtel and P. Yang _____ 1957

Deutscher Zukunftspreis:
J.-P. Stasch _____ 1957

AAAS Marion Milligan Mason Award:
A. R. Fout, K. R. M. Mackey,
K. N. Parent, and
L. Whittaker-Brooks _____ 1957

Elected to the Royal Swedish
Academy of Engineering Sciences:
M. Antonietti _____ 1958



W. R. Dichtel



P. Yang



J.-P. Stasch



A. R. Fout



K. R. M. Mackey



K. N. Parent



L. Whittaker-Brooks



M. Antonietti

Books

Introduction to Molecular Magnetism

Cristiano Benelli, Dante Gatteschi

reviewed by E. J. L. McInnes* 1959

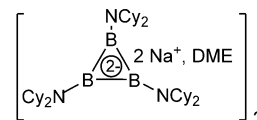
Highlights

Aromaticity

B. Wrackmeyer* 1962–1964

A Cyclotriborane Dianion and the Triboron Cation: “Light Ends” of the Hückel Rule

The lightest synthetically accessible Hückel π -aromatic system, a triboracyclopropenyl dianion bearing three dicyclohexylamino groups, has been obtained by reduction of dicyclohexylaminoboron dichloride and structurally characterized as the dimeric disodium salt (see scheme; Cy = cyclohexyl, DME = 1,2-dimethoxyethane).

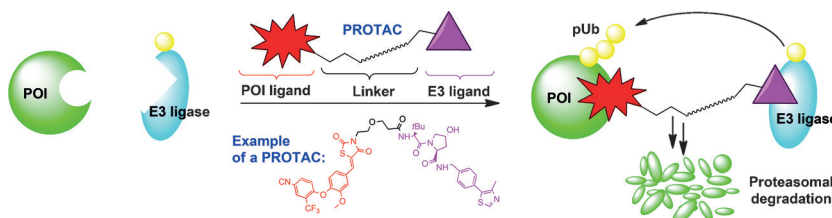


Minireviews

Medicinal Chemistry

M. Toure, C. M. Crews* 1966–1973

Small-Molecule PROTACS: New Approaches to Protein Degradation



Now we're just falling apart: The use of small-molecule proteolysis-targeting chimeras (PROTACs) to induce protein degradation offers an alternative therapeutic strategy to the traditional inhibitor-based approach. Recent progress in developing

small-molecule PROTACs to mediate the proteasomal degradation of targeted proteins is presented, and the opportunities and challenges of this technology for drug discovery are discussed.

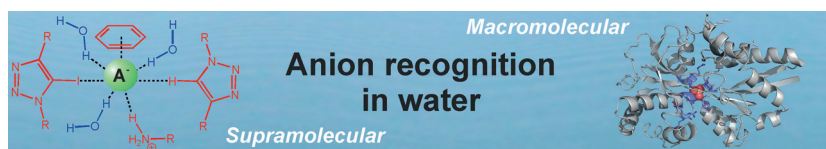
Reviews

Molecular Recognition



M. J. Langton, C. J. Serpell,*
P. D. Beer* 1974–1987

Anion Recognition in Water: Recent Advances from a Supramolecular and Macromolecular Perspective



The quest for recognition: The recognition of anions in water is a great challenge for supramolecular chemistry. Recently, great strides have been made in this area by exploiting exotic interactions such as C–H hydrogen bonding and halogen bonding,

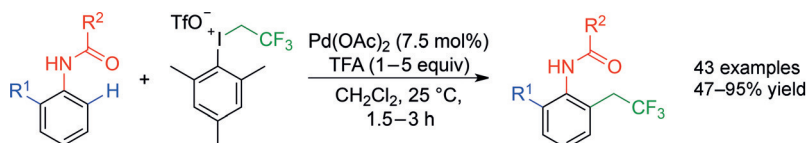
as well as exercising control over the hydrophobic effect. Recognition strategies based on biomolecules, polymers, and nanoparticles also offer a complementary approach to anion binding in the aqueous phase.

For the USA and Canada:

ANGEWANDTE CHEMIE International Edition (ISSN 1433-7851) is published weekly by Wiley-VCH, PO Box 101161, 69451 Weinheim, Germany. US mailing agent: SPP, PO Box 437, Emigsville, PA 17318. Periodicals postage

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electronic delivery); for individuals who are personal members of a national chemical society prices are available on request. Postage and handling charges included. All prices are subject to local VAT/sales tax.



Simple: Anilides can be trifluoroethylated with a 2,2,2-trifluoroethyl-substituted iodonium salt in the presence of a palladium catalyst under mild conditions (see

scheme). The reaction proceeds by C–H activation at the *ortho* position and features a broad substrate scope.

Communications

Trifluoroethylation

B. L. Tóth, Sz. Kovács, G. Sályi,
Z. Novák* — 1988 – 1992

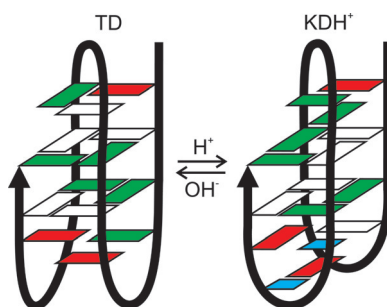
Mild and Efficient Palladium-Catalyzed Direct Trifluoroethylation of Aromatic Systems by C–H Activation



Frontispiece



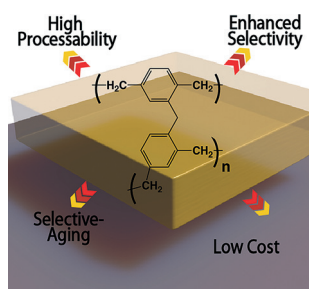
Reversible G-quadruplex pH switch: A human telomere DNA sequence has been found to adopt two distinct two G-quartet antiparallel basket-type G-quadruplexes, a TD and KDH⁺ form, which can reversibly transform to each other by variation of pH. The protonation and deprotonation of A20 is the key for the transformation. Reversibility offers possibilities for its utilization as conformational switch in a highly polymorphic system.



G-Quadruplexes

P. Galer, B. Wang, P. Šket,*
J. Plavec* — 1993 – 1997

Reversible pH Switch of Two-Quartet G-Quadruplexes Formed by Human Telomere

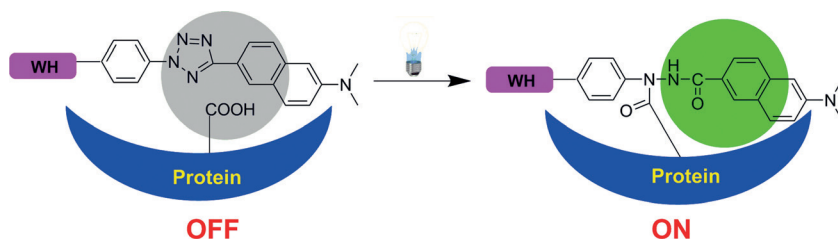


Giving membranes eternal youth: Preventing the collapse of pores in gas-separation membranes vastly improves their lifetime. In fact, the high performance membranes improve with age. The hypercrosslinked polymer additive used for this is cheap to manufacture and has very high processability. This brings the reality of high-performance membranes in the market much closer.

Gas-Separation Membranes

C. H. Lau,* X. Mulet, K. Konstas,
C. M. Doherty, M.-A. Sani, F. Separovic,
M. R. Hill,* C. D. Wood* — 1998 – 2001

Hypercrosslinked Additives for Ageless Gas-Separation Membranes



“Tie” and “see”: By reassessing the bio-orthogonality of tetrazole photoclick chemistry, an alternative use of tetrazoles in chemical biology is proposed, as general photo-crosslinkers with potential

fluorescence Turn-ON properties. This method could be used to develop affinity-based probes for live-cell imaging of endogenous kinase activities. WH = warhead.

Photoclick Chemistry

Z. Li, L. Qian, L. Li, J. C. Bernhammer,
H. V. Huynh, J.-S. Lee,
S. Q. Yao* — 2002 – 2006

Tetrazole Photoclick Chemistry: Reinvestigating Its Suitability as a Bioorthogonal Reaction and Potential Applications



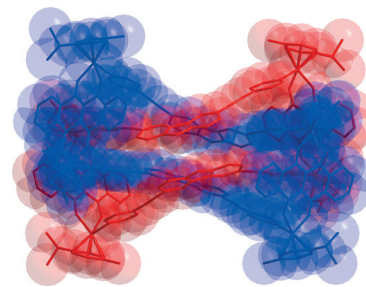
Supramolecular Chemistry

Y. H. Song, N. Singh, J. Jung,* H. Kim,
E.-H. Kim, H.-K. Cheong, Y. Kim,
K.-W. Chi* ————— 2007 – 2011



Template-Free Synthesis of a Molecular
Solomon Link by Two-Component Self-
Assembly

Link up: A molecular Solomon link has been prepared by using the title reaction. This template-free approach favors the doubly interlocked [2]catenane because of multiple π - π and $\text{CH}\cdots\pi$ interactions, as evidenced by X-ray crystal structure and computational analysis.



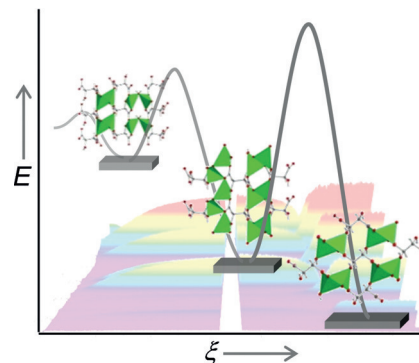
Crystal Growth

H. H.-M. Yeung,* Y. Wu, S. Henke,
A. K. Cheetham, D. O'Hare,
R. I. Walton* ————— 2012 – 2016



In Situ Observation of Successive
Crystallizations and Metastable
Intermediates in the Formation of
Metal–Organic Frameworks

Changing phases: The relative stabilities and activation energies for the solvothermal formation of dense lithium *meso*-tartrate metal–organic frameworks (MOFs) have been mapped out using high-energy in situ X-ray powder diffraction. The thermodynamic product containing a non-equilibrium ligand conformation is accessed through the successive crystallization and dissolution of two metastable intermediate phases. ξ = reaction progress.



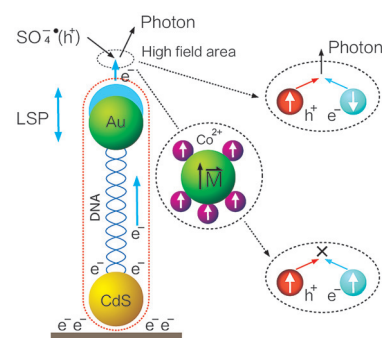
Magnetoplasmonics

Y. Shan, H. Y. Wu, S. J. Xiong, X. L. Wu,*
P. K. Chu ————— 2017 – 2021



Electrochemiluminescent Spin-Polarized
Modulation by Magnetic Ions and Surface
Plasmon Coupling

Electrochemiluminescent modulation: Electrochemiluminescence (ECL) of CdS nanocrystal (NC) film–Au nanoparticle (NP) integrated system is controllably quenched by magnetic Co^{2+} ions without external magnetic field owing to spin-polarized modulation of the electron-hole recombination caused by ferromagnetic alignment of Co^{2+} in close proximity to Au NPs.

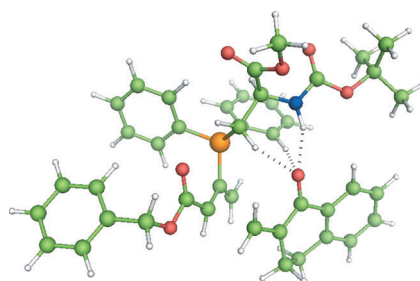


Asymmetric Organocatalysis

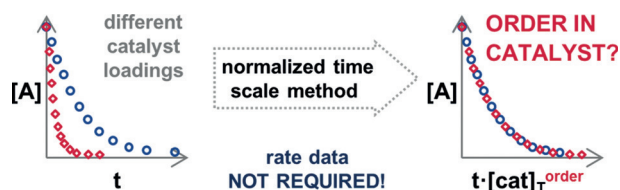
M. C. Holland, R. Gilmour,
K. N. Houk* ————— 2022 – 2027



Importance of Intermolecular Hydrogen
Bonding for the Stereochemical Control of
Allene–Enone (3+2) Annulations
Catalyzed by a Bifunctional, Amino Acid
Derived Phosphine Catalyst



The origin of stereoselectivity in the aminophosphine-catalyzed (3+2) annulation of allenes and enones was investigated on the basis of dispersion-corrected density functional theory. An intermolecular hydrogen bond between the intermediate zwitterion and the enone was found to be the key interaction in the two enantiomeric transition states.



A graphical analysis to elucidate the order in catalyst uses a normalized time scale, $t[\text{cat}]^n$, to adjust entire reaction profiles constructed with concentration data. Compared to methods that use rates, the

normalized time scale analysis requires fewer experiments and minimizes the effects of experimental errors by using information on the entire reaction profile.

Kinetic Analysis

J. Burés* _____ 2028 – 2031

A Simple Graphical Method to Determine the Order in Catalyst

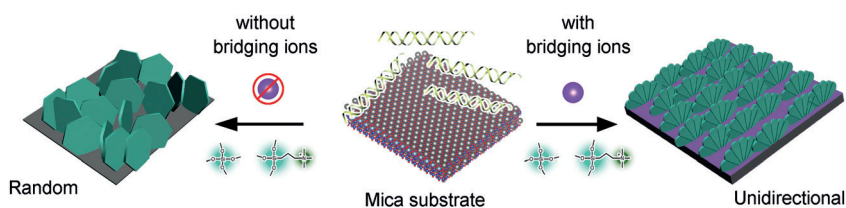


Tricky Teflon: The use of Teflon in making mesostructured carbon materials eliminates toxic HF handling and shortens the synthesis time. Silica template removal and carbonization are combined into a single step. The obtained carbon JNC-1 has a higher surface area (see picture) and pore volume compared to conventionally obtained carbons (such as CMK-3) via the HF etched route.

Mesoporous Materials

D. K. Singh, K. S. Krishna, S. Harish, S. Sampath, M. Eswaramoorthy* _____ 2032 – 2036

No More HF: Teflon-Assisted Ultrafast Removal of Silica to Generate High-Surface-Area Mesostructured Carbon for Enhanced CO₂ Capture and Supercapacitor Performance



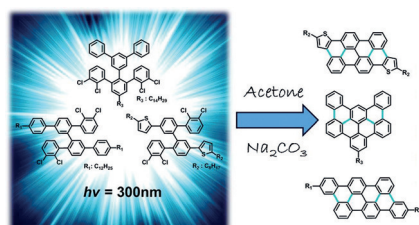
An oriented chiral DNA-silica film was fabricated using crystalline mica as both substrate and template. The connecting effect of the bridging metal ions between DNA molecules and the substrate led to vertical growth of chiral blades and align-

ment of the film in uniform horizontal direction. This coincides with the crystal orientation of the mica surface, producing a large-scale chiral film with long-range alignment.

Chiral Films

Y. Cao, K. Kao, C. Mou, L. Han,* S. Che* _____ 2037 – 2041

Oriented Chiral DNA-Silica Film Guided by a Natural Mica Substrate



Gently does it: Upon irradiation under metal-free, mild conditions, chlorinated precursors underwent up to four regioselective cyclization reactions to provide rigid π -conjugated molecules (see example). This photochemical cyclodehydrochlorination reaction was compatible with both electron-poor and electron-rich substrates, thus enabling the synthesis of pyridine- and thiophene-fused nanographenes.

Carbon Materials

M. Daigle, A. Picard-Lafond, E. Soligo, J.-F. Morin* _____ 2042 – 2047

Regioselective Synthesis of Nanographenes by Photochemical Cyclodehydrochlorination



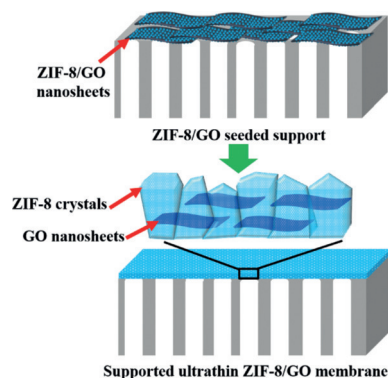
Microporous Materials

Y. X. Hu, J. Wei, Y. Liang, H. C. Zhang,
X. W. Zhang, W. Shen,
H. T. Wang* — 2048 – 2052



Zeolitic Imidazolate Framework/
Graphene Oxide Hybrid Nanosheets as
Seeds for the Growth of Ultrathin
Molecular Sieving Membranes

Thin sieves: Zeolitic imidazolate framework-8 (ZIF-8)/graphene oxide (GO) membrane with a thickness of 100 nm was prepared using 2D ZIF-8/GO hybrid nanosheets as seeds. The nanohybrid seeding facilitates uniform crystallization of ZIF-8 on the porous substrate, and the GO promotes crystal intergrowth, leading to high-quality molecular sieving membranes.

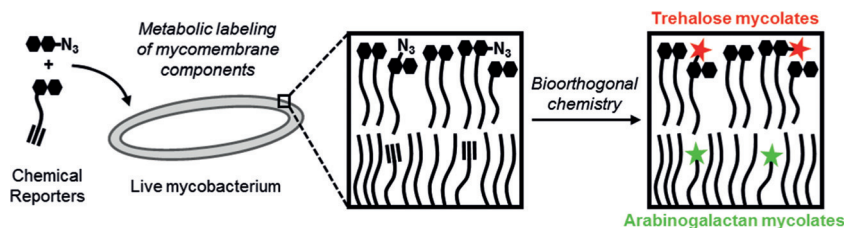


Bioimaging

H. N. Foley, J. A. Stewart, H. W. Kavunja,
S. R. Rundell, B. M. Swarts* — 2053 – 2057



Bioorthogonal Chemical Reporters for
Selective In Situ Probing of
Mycomembrane Components in
Mycobacteria



Marking the mycomembrane: Bioorthogonal chemical reporters are reported that enable sensitive, selective, and simultaneous in situ fluorescence detection of

the two major mycolic acid-containing components of the mycobacterial outer membrane, which is a crucial target for tuberculosis drug development.

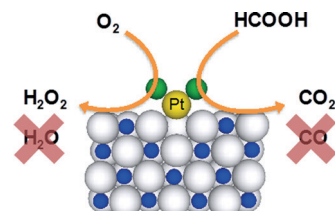
Single-Atom Catalysis

S. Yang, J. Kim, Y. J. Tak, A. Soon,
H. Lee* — 2058 – 2062



Single-Atom Catalyst of Platinum
Supported on Titanium Nitride for
Selective Electrochemical Reactions

Nanocatalysis: A single-atom platinum catalyst was successfully prepared on titanium nitride supports with the aid of chlorine ligands and applied as an electrocatalyst. Unique selectivities were observed for the oxygen reduction reaction, formic acid oxidation reaction, and methanol oxidation reaction because of the absence of Pt ensemble sites.

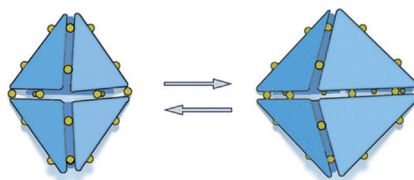


Supramolecular Chemistry

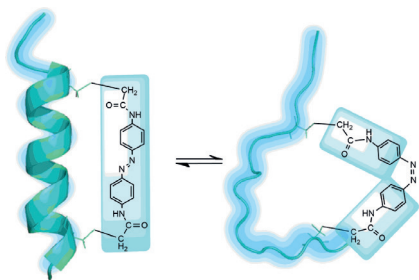
S. Wang, T. Sawada, K. Ohara,
K. Yamaguchi, M. Fujita* — 2063 – 2066



Capsule–Capsule Conversion by Guest
Encapsulation



Guests welcome: A self-assembled $M_{18}L_6$ trigonal bipyramidal capsule based on Pd^{II} units (M) and panel ligands (L) reorganizes into a larger $M_{24}L_8$ octahedral capsule, concomitant with considerable cavity expansion (381 to 943 Å³) and encapsulation of large guest(s). Although the metal–ligand interaction is labile, the capsule formed shows considerable kinetic stability.

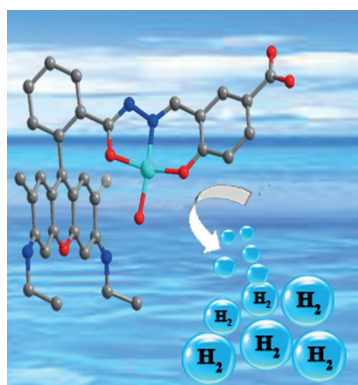


Using quantum mechanics/molecular mechanics (QM/MM) and classical MM dynamics methods the photoinduced folding and unfolding of an azobenzene cross-linked peptide was simulated. The interaction between the peptide and the cross-linker plays a key role not only in regulating the photoinduced evolution of the secondary structure of the peptide, but also in tuning the photoisomerization mechanism of the azobenzene cross-linker.

Molecular Dynamics

S.-H. Xia, G. Cui,* W.-H. Fang, W. Thiel* **2067 – 2072**

How Photoisomerization Drives Peptide Folding and Unfolding: Insights from QM/MM and MM Dynamics Simulations

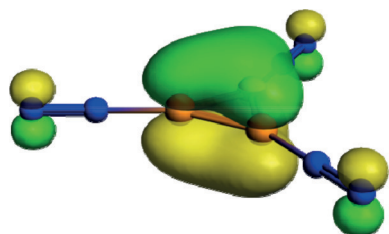


On active duty: High activity for the generation of hydrogen has been found on irradiating a crystalline coordination polymer containing redox-active copper centers and rhodamine-derived organic linkers with visible light. The ordered stacking of the linkers induced rapid transfer of photogenerated electrons to the copper nodes, which have open coordination sites and a favorable redox potential for reducing protons to dihydrogen.

Photocatalysis

X.-Y. Dong, M. Zhang, R.-B. Pei, Q. Wang, D.-H. Wei, S.-Q. Zang,* Y.-T. Fan, T. C. W. Mak **2073 – 2077**

A Crystalline Copper(II) Coordination Polymer for the Efficient Visible-Light-Driven Generation of Hydrogen

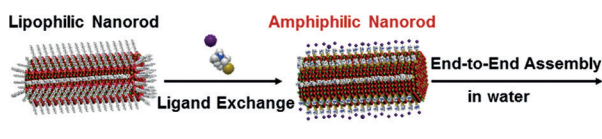


Minimal π systems: The $[B_3(NN)_3]^+$ and $[B_3(CO)_3]^+$ complexes are identified through infrared photodissociation spectroscopy. The complexes feature the smallest π -aromatic system B_3^+ . Quantum chemical bonding analysis reveals that the adducts are stabilized by $L \rightarrow [B_3L_2]^+$ σ -donation.

Boron Coordination

J. Jin, G. Wang, M. Zhou,* D. M. Andrada, M. Hermann, G. Frenking* **2078 – 2082**

The $[B_3(NN)_3]^+$ and $[B_3(CO)_3]^+$ Complexes Featuring the Smallest π -Aromatic Species B_3^+



Falling into line: The exchange of alkylphosphonic acid ligands on wurtzite CdSe nanorods for short-chained water-soluble thiols rendered the surface of the nanorods amphiphilic in nature. The amphi-

philic nanorods self-assembled into one-dimensional nanowires through hydrophobic end-to-end interactions in water. CdSe tetrapods were incorporated into the self-assemblies as branching points.

Nanostructures

Y. Taniguchi, T. Takishita, T. Kawai, T. Nakashima* **2083 – 2086**

End-to-End Self-Assembly of Semiconductor Nanorods in Water by Using an Amphiphilic Surface Design



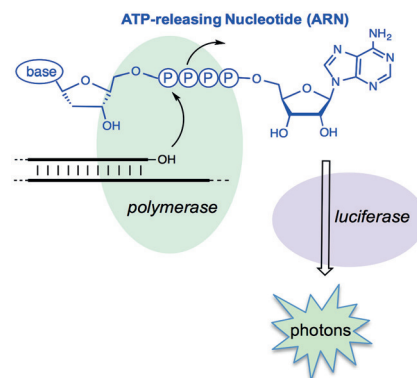
**Nucleic Acid Detection**

D. Ji, M. G. Mohsen, E. M. Harcourt,
E. T. Kool* ————— **2087–2091**



ATP-Releasing Nucleotides: Linking DNA
Synthesis to Luciferase Signaling

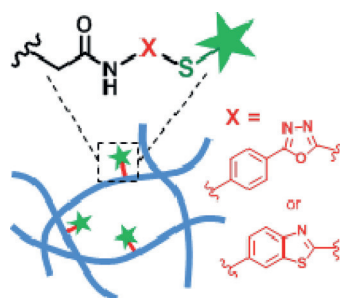
ATP lighting the way: Chimeric dinucleotides were designed to release ATP during polymerase-mediated synthesis of DNA. This ATP can be used to generate a signal with luciferase, thereby allowing the sensitive isothermal detection of both large (phage DNA) and small (miRNA) nucleic acids.

**Hydrogels**

A. Farrukh, J. I. Paez, M. Salierno,
A. del Campo* ————— **2092–2096**



Bioconjugating Thiols to Poly(acrylamide)
Gels for Cell Culture Using Methylsulfonyl
Co-monomers



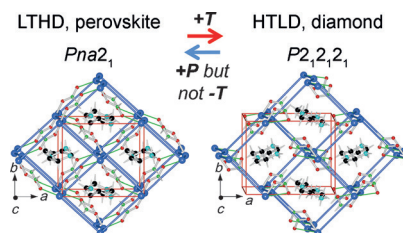
Methylsulfonyl polyacrylamide (P(AAm) gels allow easy, specific, and functional covalent coupling of thiol-containing ligands in tunable concentrations under physiological conditions, while retaining the same swelling, porosity, cytocompatibility, protein repellance, and tunable stiffness of P(AAm) gels.

Phase Transitions

R. Shang, S. Chen, B.-W. Wang,
Z.-M. Wang,* S. Gao* ————— **2097–2100**



Temperature-Induced Irreversible Phase
Transition From Perovskite to Diamond
But Pressure-Driven Back-Transition in an
Ammonium Copper Formate



[CH₃CH₂NH₃][Cu(HCOO)₃] shows a phase transition from low-temperature high-density (LTHD) perovskite to high-temperature low-density (HTLD) diamond by heating, but the backward transition can only be induced by pressure. The copper–formate bond rearrangement, a bridging-chelating switch of formate, changes in N–H···O H-bonds, and flipping of ethylammonium are involved in the transition.

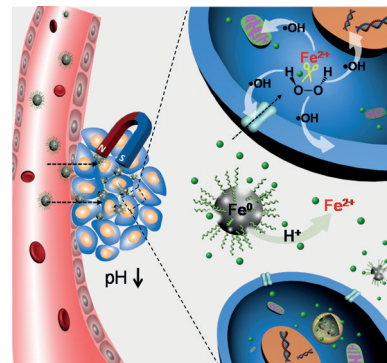
**Antitumor Agents**

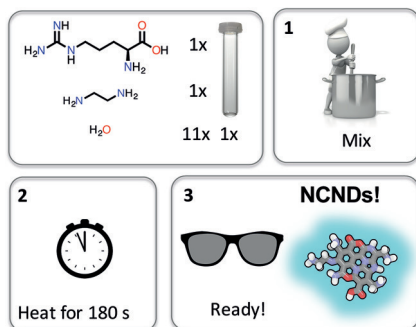
C. Zhang, W. Bu,* D. Ni, S. Zhang, Q. Li,
Z. Yao, J. Zhang, H. Yao, Z. Wang,
J. Shi* ————— **2101–2106**



Synthesis of Iron Nanometallic Glasses
and Their Application in Cancer Therapy
by a Localized Fenton Reaction

Amorphous iron nanoparticles (AFENPs) can be used for cancer theranostics. Ionization of the AFENPs in the mildly acidic tumor microenvironment promotes the release of ferrous ions in the tumor; these induce H₂O₂ disproportionation, which in turn leads to efficient $\cdot\text{OH}$ generation and significant tumor growth inhibition.



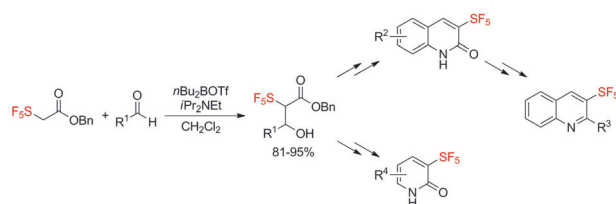


Fluorescent nanodots: A simple and controllable bottom-up route to size- and surface-controllable nitrogen-doped carbon nanodots (NCNDs) under microwave irradiation is reported. These NCNDs are among the smallest nanodots reported so far, with some of the highest quantum yields.

Carbon Nanodots

F. Arcudi,* L. Đorđević,
M. Prato* _____ 2107–2112

Synthesis, Separation, and Characterization of Small and Highly Fluorescent Nitrogen-Doped Carbon NanoDots



Easy SF₅ installation: Enolate in α -position of a pentafluorosulfanyl group was generated and underwent aldol reaction to give SF₅-heterocycles in excellent yields. Comparison of their physicochemical data

with those of their trifluoromethyl and *tert*-butyl analogues confirmed that the SF₅ group can be considered as a surrogate for CF₃ and *t*-Bu groups with potential fine-tuning effects.

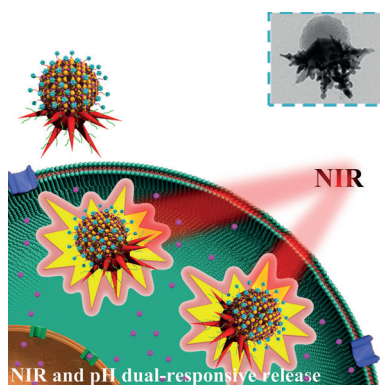
Heterocycles

A. Joliton, J.-M. Plancher,
E. M. Carreira* _____ 2113–2117

Formation of α -SF₅-Enolate Enables Preparation of 3-SF₅-Quinolin-2-ones, 3-SF₅-Quinolines, and 3-SF₅-Pyridin-2-ones: Evaluation of their Physicochemical Properties



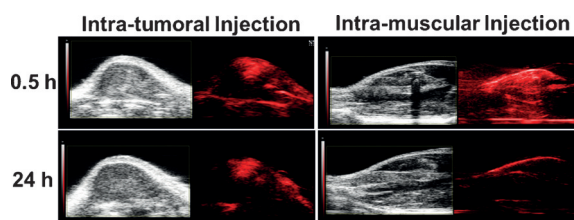
Two-faced therapeutics: Octopus-type PEG-Au-PAA/mSiO₂-LA Janus NPs are fabricated with pH and NIR light dual-stimuli responsive properties and targeting specificity for synergistic chemo-photothermal cancer therapy in vitro and in vivo.



Drug Delivery

L. Zhang, Y. Chen, Z. Li, L. Li,*
P. Saint-Cricq, C. Li,* J. Lin, C. Wang,*
Z. Su, J. I. Zink _____ 2118–2121

Tailored Synthesis of Octopus-type Janus Nanoparticles for Synergistic Actively-Targeted and Chemo-Photothermal Therapy



A photothermal nanoagent has been developed, namely degradable PEGylated molybdenum oxide, with strong near-IR absorbance, high drug loading capability, and pH-dependent degradation. It can be rapidly excreted from the body after

intravenous injection and shows no appreciable in vivo toxicity. It also shows effective accumulation and retention in the tumor, which can then be eliminated by photothermal therapy treatment.

Photothermal Therapy

G. Song, J. Hao, C. Liang, T. Liu, M. Gao,
L. Cheng, J. Hu,* Z. Liu* _____ 2122–2126

Degradable Molybdenum Oxide Nanosheets with Rapid Clearance and Efficient Tumor Homing Capabilities as a Therapeutic Nanoplatfom



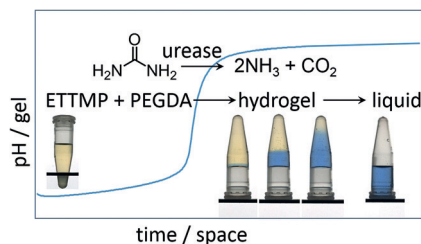
Dynamic Materials



E. Jee, T. Bánsági, Jr., A. F. Taylor,*
J. A. Pojman* 2127–2131



Temporal Control of Gelation and
Polymerization Fronts Driven by an
Autocatalytic Enzyme Reaction



In control at the front line: By coupling a base-catalyzed thiol-Michael addition with a base-producing autocatalytic enzyme reaction, it was possible to program gelation and generate propagating polymerization fronts (see picture; ETTMP is a polymeric trithiol, PEGDA is poly(ethylene glycol) diacrylate). The rate of hydrolytic degradation of the resulting hydrogel depended on the initial concentrations of the components of the system.

Photoswitches

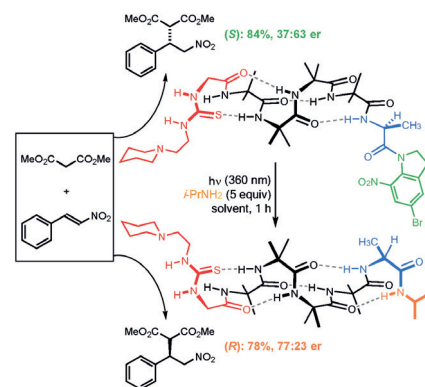


B. A. F. Le Bailly, L. Byrne,
J. Clayden* 2132–2136



Refoldable Foldamers: Global
Conformational Switching by Deletion or
Insertion of a Single Hydrogen Bond

Global refolding of a helical foldamer is induced by a change in pH or by photochemically induced transamidation. As a result, information about a single local hydrogen bond is transmitted to a remote catalytically active site, and translated into a switch in conformation that reverses the enantioselectivity of its reactions.

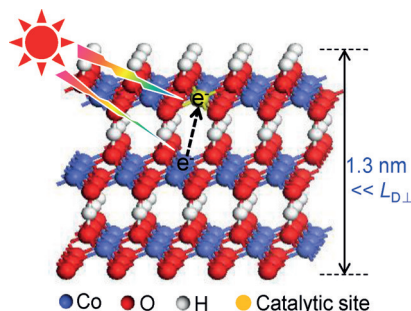


Hydrogen Evolution

J. H. Huang, Q. C. Shang, Y. Y. Huang,
F. M. Tang, Q. Zhang, Q. H. Liu,* S. Jiang,
F. C. Hu, W. Liu, Y. Luo, T. Yao,* Y. Jiang,
Z. Y. Pan, Z. H. Sun,
S. Q. Wei* 2137–2141



Oxyhydroxide Nanosheets with Highly
Efficient Electron–Hole Pair Separation for
Hydrogen Evolution



The thickness of a layered β -CoOOH semiconductor was reduced to obtain an atomically thin two-dimensional nanostructure. Electron–hole recombination is almost suppressed in the 1.3 nm thick β -CoOOH nanosheet, which leads to excellent electron–hole separation efficiencies and superior hydrogen production rates ($L_{D\perp}$: carrier diffusion length along the c axis).

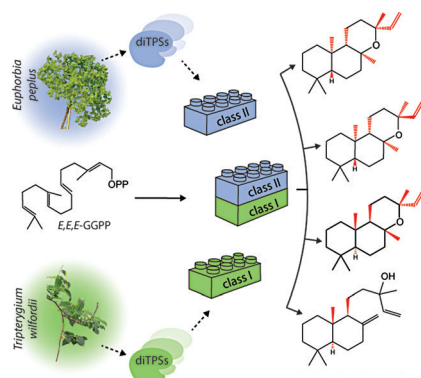
Biosynthesis



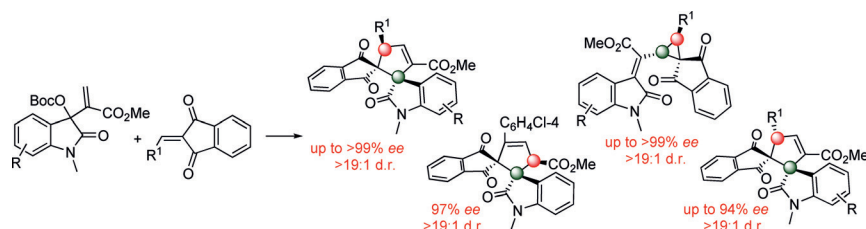
J. Andersen-Ranberg, K. T. Kongstad,
M. T. Nielsen, N. B. Jensen, I. Pateraki,
S. S. Bach, B. Hamberger, P. Zerbe,
D. Staerk, J. Bohlmann, B. L. Møller,
B. Hamberger* 2142–2146



Expanding the Landscape of Diterpene
Structural Diversity through
Stereochemically Controlled
Combinatorial Biosynthesis



Beyond the horizon: A broad range of cyclic diterpenes, including precursors for therapeutic compounds, was efficiently biosynthesized by combining class I and II diterpene synthase enzymes from diverse species. Stereospecific cyclizations were achieved and scale-up was demonstrated with efficient biotechnological production.



Switching it up: The title reactions have been accomplished by employing different chiral Lewis base catalysts, thus resulting in a broad collection of chiral compounds with high structural and stereochemical

diversity (see scheme). DFT calculations indicate that the structure of Lewis base plays a key role in controlling the chemo- and diastereoselectivity. Boc = *tert*-butoxycarbonyl.

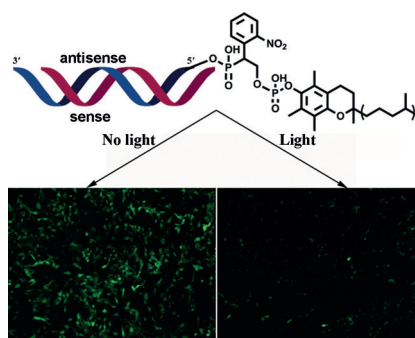
Organocatalysis

G. Zhan, M.-L. Shi, Q. He, W.-J. Lin, Q. Ouyang,* W. Du, Y.-C. Chen* — 2147–2151

Catalyst-Controlled Switch in Chemo- and Diastereoselectivities: Annulations of Morita–Baylis–Hillman Carbonates from Isatins



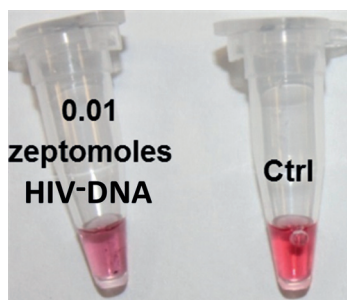
Caged siRNAs with a single photolabile linker and/or vitamin E modification at 5' terminal were rationally designed and synthesized. These virtually inactive caged siRNAs could be fully restored to native siRNAs through simple light activation with up to an 18.6-fold enhancement of gene silencing activity.



Gene Regulation

Y. Ji, J. Yang, L. Wu, L. Yu, X. Tang* — 2152–2156

Photochemical Regulation of Gene Expression Using Caged siRNAs with Single Terminal Vitamin E Modification

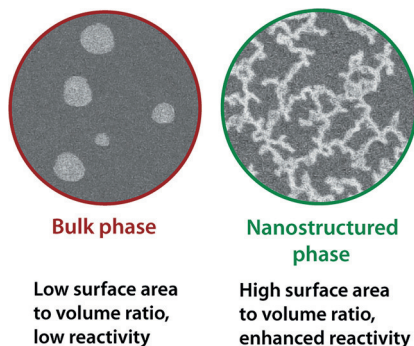


Goodbye gels! PCR developer is a rapid (5 min), highly specific, and universal naked-eye assay to replace gel electrophoretic analysis of PCR reactions.

Analytical Methods

P. Valentini, P. P. Pompa* — 2157–2160

A Universal Polymerase Chain Reaction Developer



Watered down: The formation of micro- and nanometer-scale compartments in ionic liquids, mediated by addition of water, can be directly observed by using electron microscopy. The morphologies, which include isolated droplets, aggregates, and 2D meshworks, change according to the amount of added water. The ionic liquid/water systems were applied to the formation of the platform chemical 5-(hydroxymethyl)furfural.

Ionic Liquids

A. S. Kashin, K. I. Galkin, E. A. Khokhlova, V. P. Ananikov* — 2161–2166

Direct Observation of Self-Organized Water-Containing Structures in the Liquid Phase and Their Influence on 5-(Hydroxymethyl)furfural Formation in Ionic Liquids



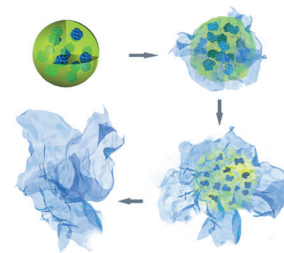
Ultrathin Nanosheets

H. Fan, X. Huang, L. Shang, Y. Cao,
Y. Zhao, L.-Z. Wu, C.-H. Tung, Y. Yin,*
T. Zhang* ————— 2167–2170



Controllable Synthesis of Ultrathin
Transition-Metal Hydroxide Nanosheets
and their Extended Composite
Nanostructures for Enhanced Catalytic
Activity in the Heck Reaction

From spheres to sheets: Ultrathin single- or multiple-component transition-metal hydroxide (TMH) nanosheets with thickness below 5 nm are prepared by the gradual decomposition of preformed metal–boron (M–B, M = Fe, Co, Ni, NiCo) composite nanospheres. The oxidation of the metal and the simultaneous release of boron species give rise to the sheets.



Back Cover

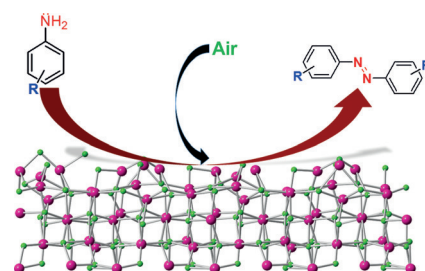
Heterogeneous Catalysis

B. Dutta, S. Biswas, V. Sharma,
N. O. Savage, S. P. Alpay,
S. L. Suib* ————— 2171–2175



Mesoporous Manganese Oxide Catalyzed
Aerobic Oxidative Coupling of Anilines To
Aromatic Azo Compounds

Cheap catalyst, free oxidant: A cost-effective and reusable mesoporous manganese oxide material catalyzed the oxidative coupling of anilines with both electron-donating and electron-withdrawing substituents under mild reaction conditions to give symmetrical and unsymmetrical azobenzene derivatives with high conversion. Air was used as the sole oxidant in this environmentally friendly process (see picture; Mn pink, O green).



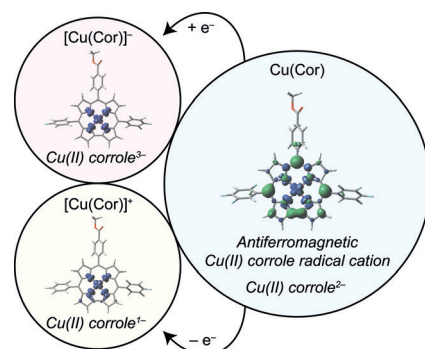
Ligand Non-Innocence

C. M. Lemon, M. Huynh, A. G. Maher,
B. L. Anderson, E. D. Bloch, D. C. Powers,
D. G. Nocera* ————— 2176–2180



Electronic Structure of Copper Corroles

Innocent until proven guilty: The electronic structure of Cu corroles is studied using a variety of techniques in conjunction with DFT calculations. Given this data, the compound is best described as an antiferromagnetically coupled Cu^{II} corrole radical cation. Electrons are added to or removed from the corrole ligand in these redox processes, preserving the Cu^{II} centre. This result underscores corrole non-innocence in these compounds.

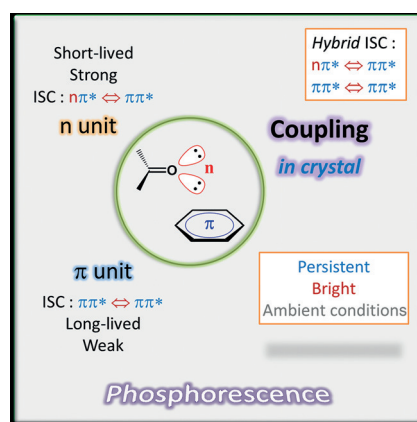


Photochemistry

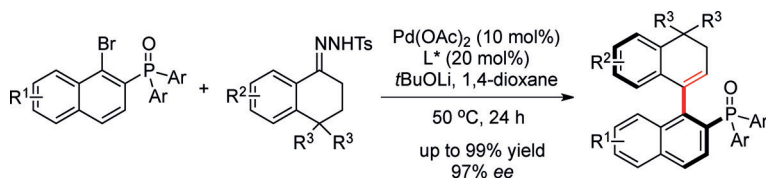
Z. Yang, Z. Mao, X. Zhang, D. Ou, Y. Mu,
Y. Zhang,* C. Zhao, S. Liu, Z. Chi,* J. Xu,
Y.-C. Wu, P.-Y. Lu, A. Lien,
M. R. Bryce* ————— 2181–2185



Intermolecular Electronic Coupling of
Organic Units for Efficient Persistent
Room-Temperature Phosphorescence



Persistence pays off: Bright persistent room-temperature phosphorescence from pure organic molecules was achieved by intermolecular electronic coupling of selected units in crystals. The combined advantages of their different excited-state configurations (i.e., the $n\pi^*$ state with a high intersystem crossing rate and the $\pi\pi^*$ state with a low radiative rate) results in a hybrid intersystem-crossing process that leads to efficient persistent room-temperature phosphorescence.



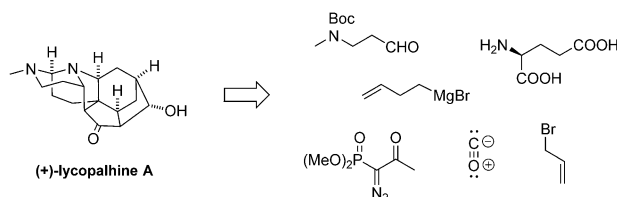
With a twist: An enantioselective palladium-catalyzed synthesis of atropisomeric vinyl arenes from aryl bromides and hydrazones is reported. The use of hydrazone precursors as coupling partners

resulted in mild reaction conditions, and thus a broad functional-group tolerance. The products were isolated with *ee* values of up to 97%. Ts = 4-toluenesulfonyl.

Biaryls

J. Feng, B. Li, Y. He, Z. Gu* **2186–2190**

Enantioselective Synthesis of Atropisomeric Vinyl Arene Compounds by Palladium Catalysis: A Carbene Strategy



Just Another Mannich Monday: By using L-glutamate and L-proline as a starting material and catalyst, respectively, the complex alkaloid lycopalhine A was assembled in only 13 steps. The synthesis

features not only an unusual Mannich reaction but also a biomimetic aldol addition that completes the intricate carbon skeleton of the natural product.

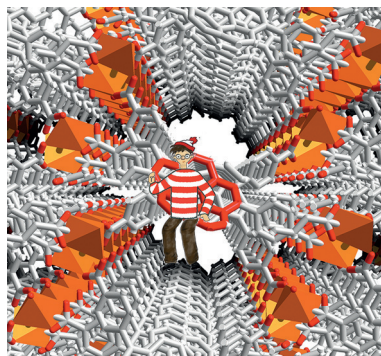
Natural Product Synthesis

B. M. Williams, D. Trauner* **2191–2194**

Expedient Synthesis of (+)-Lycopalhine A



Where is the corannulene? A porous crystalline hybrid scaffold with immobilized redox-active corannulene buckybowls has been prepared. The scaffold combines the periodicity, dimensionality, and structural modularity of hybrid frameworks with the intrinsic properties of redox-active π -bowls.



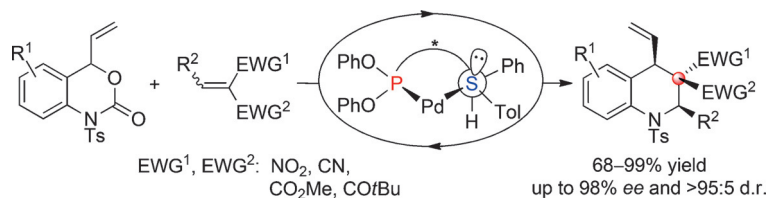
Hybrid Materials

W. B. Fellows, A. M. Rice, D. E. Williams, E. A. Dolgoplova, A. K. Vannucci, P. J. Pellechia, M. D. Smith, J. A. Krause, N. B. Shustova* **2195–2199**

Redox-Active Corannulene Buckybowls in a Crystalline Hybrid Scaffold



Front Cover



Hybrid ligands: By combining a chiral β -amino sulfide and a simple diphenyl phosphite, a new hybrid P,S ligand was used for title reaction. By doing so, multiple contiguous stereocenters and

a chiral quaternary center were rapidly constructed and highly functionalized tetrahydroquinolines were produced in up to 99% yield with 98% *ee* and greater than 95:5 d.r.

Ligand Design

Y. Wei, L.-Q. Lu,* T.-R. Li, B. Feng, Q. Wang, W.-J. Xiao,*
H. Alper **2200–2204**

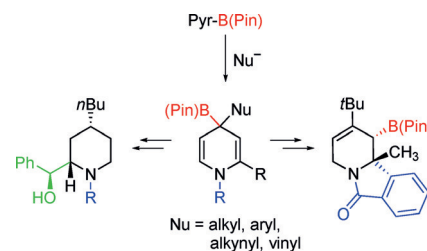
P,S Ligands for the Asymmetric Construction of Quaternary Stereocenters in Palladium-Catalyzed Decarboxylative [4+2] Cycloadditions



Heterocycle Synthesis

S. Panda, A. Coffin, Q. N. Nguyen,
D. J. Tantillo, J. M. Ready* — 2205–2209Synthesis and Utility of Dihydropyridine
Boronic Esters

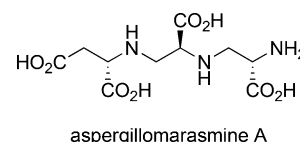
A whole lot o' rings: Dearomatization of pyridyl boronic esters leads to highly substituted heterocycles. Thus, addition of organozinc, organomagnesium, or organolithium reagents to PyB(Pin) (Pin = pinacol) yields dihydropyridine boronic esters, which participate in allylation, radical cyclizations, oxidations, and reductions.



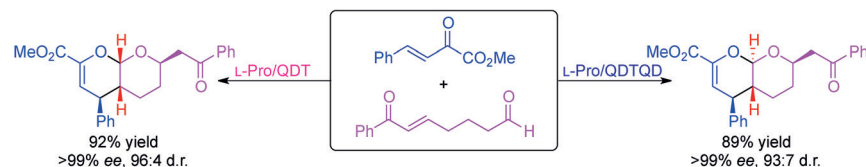
Natural Products Synthesis

K. Koteva, A. M. King, A. Capretta,*
G. D. Wright* — 2210–2212Total Synthesis and Activity of the Metallo- β -lactamase Inhibitor
Aspergillomarasmine A

Arming to fight the resistance: Metallo- β -lactamases (MBLs) are hydrolytic enzymes that cleave β -lactam rings. There is a growing need for inhibitors of MBLs that can be given as codrugs with β -lactam antibiotics. Aspergillomarasmine A (see structure) has been identified as a potential codrug. Its total synthesis confirms its stereochemical configuration and offers a route for the synthesis of derivatives.



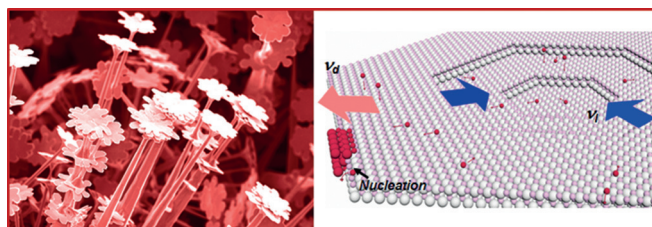
Asymmetric Catalysis

H. Huang, S. Konda,
J. C.-G. Zhao* — 2213–2216Diastereodivergent Catalysis Using
Modularly Designed Organocatalysts:
Synthesis of both *cis*- and *trans*-Fused
Pyrano[2,3-*b*]pyrans

Designer cat.: Both enantiomers of the title compounds are obtained with high diastereo- and enantioselectivities from the same starting materials by using an inverse-electron-demand hetero-Diels–Alder/oxa-Michael reaction catalyzed by

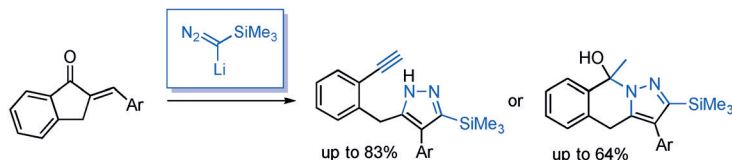
modularly designed organocatalysts (MDOs). Diastereodivergence was achieved using MDOs self-assembled from both enantiomers of proline and cinchona alkaloid thiourea derivatives.

Crystal Growth

X. Yin, D. L. Geng,
X. D. Wang* — 2217–2221Inverted Wedding Cake Growth Operated
by the Ehrlich–Schwoebel Barrier in Two-
Dimensional Nanocrystal Evolution

Nanoflowers and cake: Crystal growth was transformed from 1D nanowires to 2D nanoplates when the Ehrlich–Schwoebel barrier became tangible. Nucleation of

new atomic layers occurred along the edge and propagated toward the center resulted in the unique inverted wedding cake growth phenomenon.



The sequential 1,4-/1,2-addition of lithium(trimethylsilyl)diazomethane to cyclic α,β -unsaturated ketones induces various subsequent transformations, such as Grob-type C–C fragmentation or alkyl-

dene carbene mediated Li–N insertion, depending on the ring size and the position of the double bond. Various nitrogen heterocycles were thus obtained in good yields.

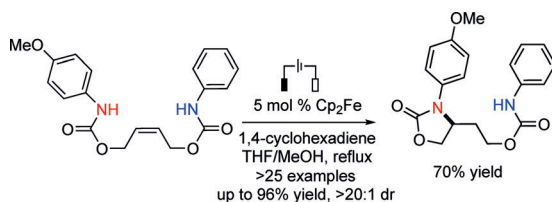
C–C Bond Fragmentation

M. J. O'Connor, C. Sun, X. Guan,
V. R. Sabbasani, D. Lee* — 2222–2225

Sequential 1,4-/1,2-Addition of Lithium(trimethylsilyl)diazomethane onto Cyclic Enones to Induce C–C Fragmentation and N–Li Insertion



Inside Cover



Easy access to N-arylamidyl radicals: The first electrocatalytic method for the generation of amidyl radicals from anilides has been developed using ferrocene (Cp_2Fe) as a highly reactive, yet chemo-

selective redox catalyst. Based on this radical-generating method, a highly chemo- and diastereoselective olefin hydroamidation reaction has been developed.

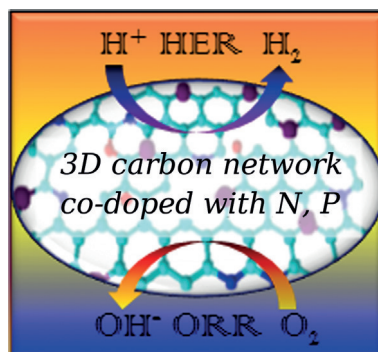
Organic Electrochemistry

L. Zhu, P. Xiong, Z.-Y. Mao, Y.-H. Wang,
X. Yan, X. Lu, H.-C. Xu* — 2226–2229

Electrocatalytic Generation of Amidyl Radicals for Olefin Hydroamidation: Use of Solvent Effects to Enable Anilide Oxidation



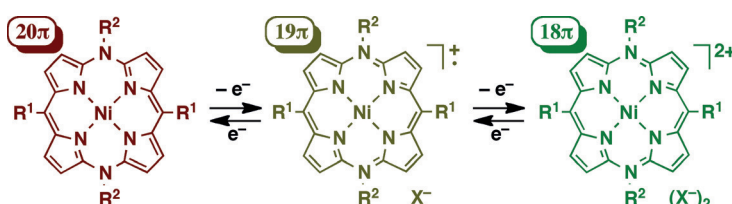
Double function: Nitrogen- and phosphorus-codoped 3D graphitic carbon networks were generated by self-assembling melamine and phytic acid into supermolecular aggregates in the presence or absence of graphene oxide, followed by pyrolysis. They display remarkably high bifunctional electrocatalytic activities for both oxygen reduction and hydrogen evolution reactions.



Electrocatalysis

J. Zhang, L. Qu, G. Shi, J. Liu, J. Chen,
L. Dai* — 2230–2234

N,P-Codoped Carbon Networks as Efficient Metal-free Bifunctional Catalysts for Oxygen Reduction and Hydrogen Evolution Reactions



N makes all the difference: Redox-switchable 20 π -, 19 π -, and 18 π -electron 5,10,15,20-tetraaryl-5,15-diazaporphyrinoids are prepared through metal-templated annulation of Ni^{II} bis(dipyrrin) complexes followed by oxidation. The

meso N atoms in these diazaporphyrinoids give rise to characteristic redox and optical properties for the compounds that are not typical of isoelectronic 5,10,15,20-tetraarylporphyrins.

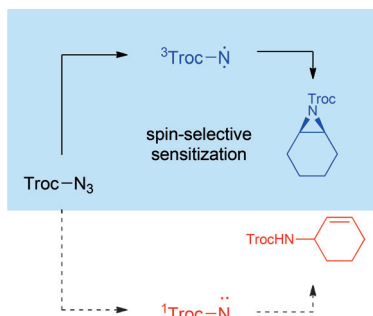
Porphyrinoids

T. Satoh, M. Minoura, H. Nakano,
K. Furukawa, Y. Matano* — 2235–2238

Redox-Switchable 20 π -, 19 π -, and 18 π -Electron 5,10,15,20-Tetraaryl-5,15-diazaporphyrinoid Nickel(II) Complexes

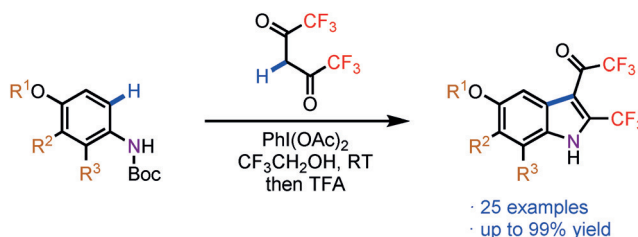


Photochemistry

S. O. Scholz, E. P. Farney, S. Kim,
D. M. Bates, T. P. Yoon* — 2239–2242Spin-Selective Generation of Triplet
Nitrenes: Olefin Aziridination through
Visible-Light Photosensitization of
Azidoformates

A fresh spin on heterocycles: Synthetically useful photochemical aziridinations can be achieved by using a visible-light-activated transition-metal photosensitizer to produce nitrenes exclusively in their triplet spin state. A wide range of aliphatic and aromatic alkenes can be readily aziridinated without competitive allylic insertion reactions. TrocN₃ = 2,2,2-trichloroethyl azidoformate.

Synthetic Methods

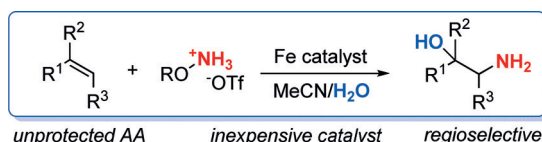
E. Vitaku, D. T. Smith,
J. T. Njardarson* — 2243–2247Metal-Free Synthesis of Fluorinated
Indoles Enabled by Oxidative
Dearomatization

A telescoped synthesis: In the title reaction, N-Boc anilines are reacted with hexafluoroacetylacetone in presence of phenyliodine(III) diacetate followed by trifluoroacetic acid (TFA) to yield fluori-

nated indoles in a single pot. Additionally, the versatility of the trifluoroacetyl group is exploited through ketone-like and ester-like chemistry.

Aminohydroxylation

L. Legnani, B. Morandi* — 2248–2251

Direct Catalytic Synthesis of Unprotected
2-Amino-1-Phenylethanols from Alkenes
by Using Iron(II) Phthalocyanine

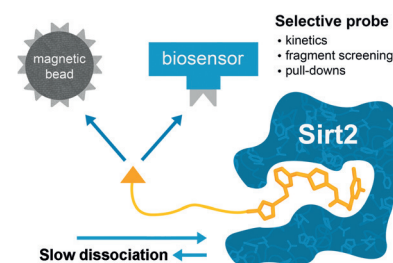
With an iron hand: Aryl-substituted amino alcohols are privileged scaffolds in medicinal chemistry and natural products. An exceptionally simple and inexpensive Fe^{II} complex can efficiently catalyze the direct transformation of simple

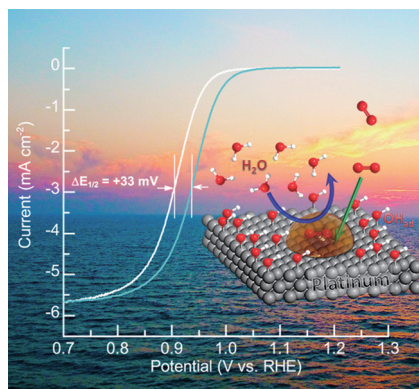
alkenes into unprotected amino alcohols in good yield and perfect regioselectivity. This new catalytic method was applied in the expedient synthesis of bioactive molecules and could be extended to amino-etherification.

Enzyme Ligands

M. Schiedel, T. Rumpf, B. Karaman,
A. Lehotzky, S. Gerhardt, J. Ovádi,
W. Sippl, O. Einsle,
M. Jung* — 2252–2256Structure-Based Development of an
Affinity Probe for Sirtuin 2

A new probe for drug design: An affinity probe for Sirt2 has been developed that shows excellent selectivity and potency. The slow dissociation rate of the enzyme–ligand complex enables new applications, such as biolayer interferometry, and pull-down assays for sirtuin rearranging ligands.





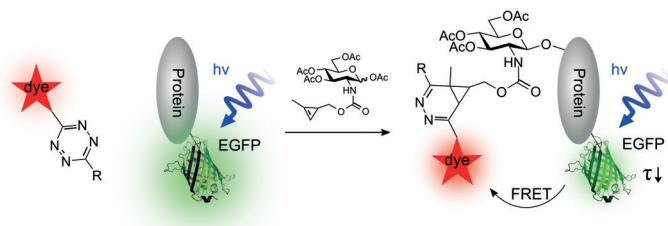
Keep my place: A hydrophobic ionic liquid phase helps protect the Pt sites from being poisoned by nonreactive oxygenated species, leading to dramatically improved kinetics of the oxygen reduction reaction (ORR) on platinum catalyst.

Oxygen Reduction

G.-R. Zhang, M. Munoz,
B. J. M. Etzold* 2257 – 2261

Accelerating Oxygen-Reduction Catalysts through Preventing Poisoning with Non-Reactive Species by Using Hydrophobic Ionic Liquids

Inside Back Cover



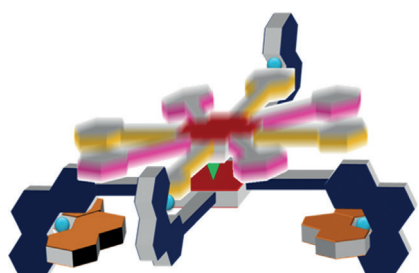
Living proof: Protein-specific glycosylation inside living cells was imaged by detecting FRET between an EGFP fused to the protein of interest and a fluorophore ligated to a metabolically incorporated *N*-acetylglucosamine derivative through an

inverse-electron-demand Diels–Alder reaction. FRET can be detected with high contrast even in presence of a large excess of acceptor fluorophores by fluorescence lifetime imaging microscopy (FLIM).

Glycoproteins

F. Doll, A. Buntz, A.-K. Späte, V. F. Schart,
A. Timper, W. Schimpf, C. R. Hauck,
A. Zumbusch,*
V. Wittmann* 2262 – 2266

Visualization of Protein-Specific Glycosylation inside Living Cells



The magic self-assembly of supramolecular machinery! When five components are mixed, supramolecular rotors form, which depending on the fifth component (= brake block) exhibit different rotational frequencies that are quantitatively predictable.

Nanorotors

S. K. Samanta, A. Rana,
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Conformational Slippage Determines Rotational Frequency in Five-Component Nanorotors



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Angewandte Corrigendum

Rhodium-Catalyzed Asymmetric
Synthesis of Spirosilabifluorene
Derivatives

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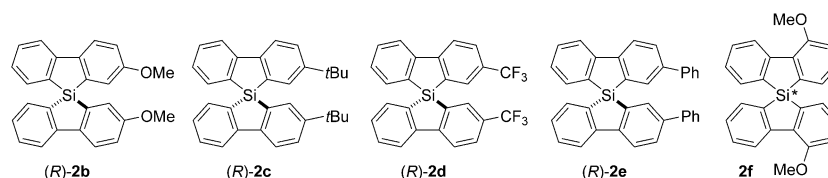
Angew. Chem. Int. Ed. 2013, 52

DOI: 10.1002/anie.201207723

The *R* and *S* designations for the absolute configuration of the chiral spiroilabifluorenes were incorrectly assigned in the Communication. As shown by the single-crystal X-ray analysis results in Figure 1, the absolute configuration of the major enantiomers from the reaction with $[\{\text{RhCl}(\text{cod})\}_2]$ and (*R*)-binap were “*R* form” not “*S* form”. Table 1 and the sentences in the original manuscript should be revised as follows:

page 1520, right column, Table 1: “*R*” as the major enantiomer for entries 1 and 4
page 1520, line 11 of the right column: “... the *R* form (Figure 1 and the Supporting ...”
page 1521, line 2 of the left column: “... preparation of **2e** from (*R*)-**2b** (See ...”
page 1521, line 6 of the left column: “compound derived from (*R*)-**2b** on”
page 1522, Ref. [8]: “... gave (–)-(*S*)-**2b** in 95% yield, 81% *ee*.”
page 1522, Ref. [9]: “... eluent)]; time: *S* form, 23 min; *R* form, 24.5 min}”
page 1522, Ref. [12]: “... by preparation of (*R*)-**2e** from (*R*)-**2b**. The details ...”
page 1522, Ref. [13]: “CCDC 914745 [(*R*)-**2b**] contains the ...”

The absolute configurations for spiroilabifluorenes **2b** and **2e** in the Supporting Information (page S6) should be corrected to “*R*” in the same way. In addition, graphics for spiroilabifluorenes **2b–2f** in the Supporting Information (pages S4 and S5) contain several errors as well. The corrected figures are shown below:



Although the absolute configuration of **2c**, **2d**, and **2f** was not determined in 2012, the *R* configuration has recently been confirmed for **2c** and **2d** using single-crystal X-ray diffraction.

The mistakes regarding the absolute configurations do not affect the validity of any of the reported yields, data, ORTEP drawings, or information provided in the experimental sections of the original manuscript and the Supporting Information. In addition, the results and conclusions of the Communication also remain valid.

The authors sincerely apologize for the errors in reporting the configurations.

Angewandte Addendum

In this Communication, the authors reported the crystal structure of the glyoxal bis(amidiniumhydrazone) (GBAH) sulfate salt, whose synthesis was first reported by Thiele and Dralle in 1898.^[1] The structure showed the sulfate crystallized as extended $[\text{SO}_4(\text{H}_2\text{O})_5]^{2-}$ clusters. As an important part of this study, it was demonstrated for the first time that competitive crystallization of GBAH sulfate from aqueous solutions can be used as a basis for selective sulfate separation. The effectiveness of this crystallization-based sulfate separation method stems from the relatively low aqueous solubility of GBAH sulfate ($K_{\text{sp}} = 3.2 \times 10^{-7}$),^[2] comparable with that of SrSO_4 . In the interest of completeness, the authors note that observations of the relatively insoluble nature of the sulfate salts of GBAH or other analogs had been previously made,^[3] though no solubility values had been reported. Single-crystal X-ray structural studies of GBAH analogues showed that the sulfate anion can crystallize with variable numbers of water molecules (from zero to six).^[4] However, due to the limited structural and solubility data available, it is not clear at this point the extent to which the included water molecules contribute to the low solubility of these salts and to the sulfate crystallization selectivity. Future structure–solubility relationship studies of bis(amidiniumhydrazone) sulfate salts offer the prospect for understanding the various factors controlling the crystallization efficiency of this intriguing class of compounds.^[5]

[1] J. Thiele, E. Dralle, *Justus Liebigs Ann. Chem.* **1898**, 302, 275.

[2] The solubility product was calculated from the measured solubility of GBAH sulfate of $7.2 \times 10^{-4} \text{ M}$, and taking into account the corresponding activity coefficient of 0.78, estimated by the Debye–Hückel limiting law.

[3] a) P. Seppanen, R. Fagerstrom, L. Alhonen-Hongisto, H. Elo, P. Lumme, J. Janne, *Biochem. J.* **1984**, 221, 483; b) H. Elo, *Spectroscopy Lett.* **1989**, 22, 123; c) H. Elo, *Spectroscopy Lett.* **1989**, 22, 161.

[4] a) P. O. Lumme, I. Mutikainen, H. O. Elo, *Acta Cryst.* **1986**, C42, 1209; b) H. Elo, I. Mutikainen, L. Alhonen-Hongisto, R. Laine, J. Jänne, P. Lumme, *Z. Naturforsch.* **1986**, 41c, 851; c) H. Elo, I. Mutikainen, *Z. Naturforsch.* **1988**, 43c, 601; d) M. Koskinen, I. Mutikainen, H. Elo, *Z. Naturforsch.* **1996**, 51b, 1161; e) M. Koskinen, I. Mutikainen, J. T. Koskinen, H. Elo, *Z. Naturforsch.* **1997**, 52b, 1114.

[5] R. Custelcean, N. J. Williams, C. A. Seipp, A. S. Ivanov, V. S. Bryantsev, *Chem. Eur. J.* DOI: 10.1002/chem.201504651.

Aqueous Sulfate Separation by
Crystallization of Sulfate-Water Clusters

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Angewandte Corrigendum

In this Communication, an affiliation needs to be added for one of the authors.

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Evidence for a Boat Conformation at the
Transition State of GH76 α -1,6-
Mannanases—Key Enzymes in Bacterial
and Fungal Mannoprotein Metabolism

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