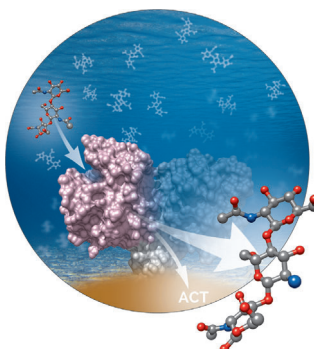


... with spatial control over the internal morphology, shape anisotropy, and stimuli-responsiveness can now be realized in a single colloidal system. C. J. Hawker et al. describe in their Communication on page 7018 ff. a facile and hierarchical self-assembly strategy driven by polystyrene-*b*-poly(2-vinylpyridine) block copolymers and surfactant blends for dynamically tuning the shape and functionality of polymer-based nanoparticles.

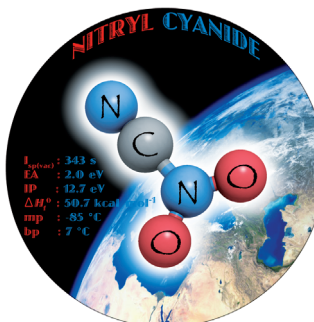
Enzyme Specificity

Through the crystal structure of *V. cholerae* chitin de-N-acetylase in relevant states of its catalytic cycle, M. E. Guerin, A. Planas, et al. show in their Communication on page 6882 ff. that significant conformational change occurs in the loop closing the active site.



Light Harvesting

D. Kim, W.-D. Jang et al. describe in their Communication on page 6925 ff. how the efficient energy-transfer process in a multiporphyrin dendrimer changes to electron transfer on guest binding.



Energetic Materials

In their Communication on page 6893 ff., M. Rahm, K. O. Christe et al. describe the synthesis and characterization of the ultra-high-energy-density compound NCNO₂.

How to contact us:

Editorial Office:

E-mail: angewandte@wiley-vch.de

Fax: (+49) 62 01-606-331

Telephone: (+49) 62 01-606-315

Reprints, E-Prints, Posters, Calendars:

Carmen Leitner

E-mail: chem-reprints@wiley-vch.de

Fax: (+49) 62 01-606-331

Telephone: (+49) 62 01-606-327

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E-mail: rights-and-licences@wiley-vch.de

Fax: (+49) 62 01-606-332

Telephone: (+49) 62 01-606-280

Online Open:

Margitta Schmitt, Carmen Leitner

E-mail: angewandte@wiley-vch.de

Fax: (+49) 62 01-606-331

Telephone: (+49) 62 01-606-315

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Courier Services:

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Regular Mail:

Postfach 101161, 69451 Weinheim

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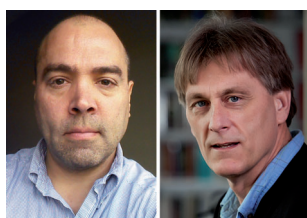
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Editorial



"... We need not merely increase the number of STEAM graduates, but we must also provide the opportunity for these students to experience a true integration across the arts, humanities, and sciences, leading to, as Leonardo da Vinci so eloquently phrased it, 'The Development of a Complete Mind' ..."

Read more in the Editorial by D. G. Lynn and J. T. Goodwin.

J. T. Goodwin, D. G. Lynn* – 6832 – 6833

Holistic Education in Times of
Specialization and Globalization

Service

Spotlight on Angewandte's Sister Journals

6850 – 6852

Author Profile



"If I were not a scientist, I would be a historian and clarify many mysteries in Japanese history."

I like refereeing because I can send more papers to journals without feeling guilty ..."

This and more about Shunichi Fukuzumi can be found on page 6854 ff.

Shunichi Fukuzumi — 6854 – 6855

News



P. A. Midgeley



G. A. Morris



J. H. Naismith



A. K. Soper



C. Bai

New Fellows and Foreign Members
of the Royal Society — 6856

Obituaries



The passing of Michael F. Lappert at the age of 85 on March 28th, 2014 has deprived organometallic chemistry of one of its greatest protagonists. In a research career spanning over six decades, he characterized a vast array of organometallic compounds from all parts of the periodic table.

Michael Lappert (1928–2014)

*P. Power** _____ **6857**

Books

Comprehensive Enantioselective Organocatalysis

Peter I. Dalko

reviewed by Y. R. Chi _____ **6858**

Modern Synthetic Methods in Carbohydrate Chemistry

Daniel B. Werz, Sebastien Vidal

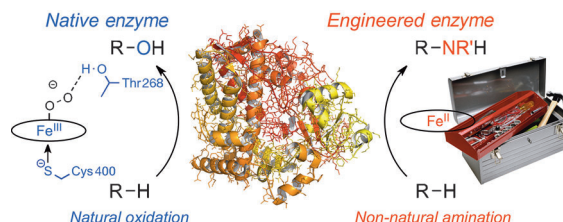
reviewed by B. Yu _____ **6858**

Highlights

Enzyme Engineering

J.-P. Mahy,* J. Ciesielski,
P. Dauban* _____ **6862 – 6864**

Catalytic C–H Amination: A Reaction Now Accessible to Engineered Natural Enzymes



Catalytic C–H aminations with natural enzymes have not been reported thus far. However, when a cytochrome P450 enzyme is modified by switching from an iron(III) to an iron(II) center and by

mutating amino acids that are critical for the catalytic activity of this monooxygenase, this modified enzyme catalyzes the intramolecular C–H amination of benzenesulfonyl azides.

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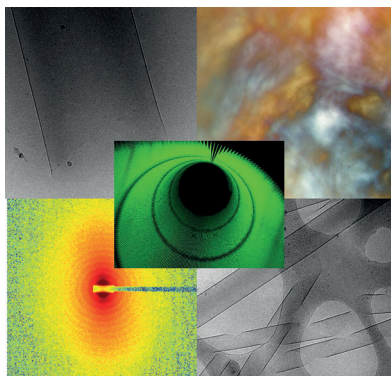
electronic delivery); for individuals who are personal members of a national chemical society prices are available on request. Postage and handling charges included. All prices are subject to local VAT/sales tax.

Reviews

Peptide Nanotubes

I. W. Hamley* _____ 6866 – 6881

Peptide Nanotubes



Building biotubes: The self-assembly of peptide nanotubes from different classes of peptide, including cyclic peptides, amyloid peptides, and surfactant-like peptides, is reviewed. Their applications in bionanotechnology and synthetic materials science are summarized.

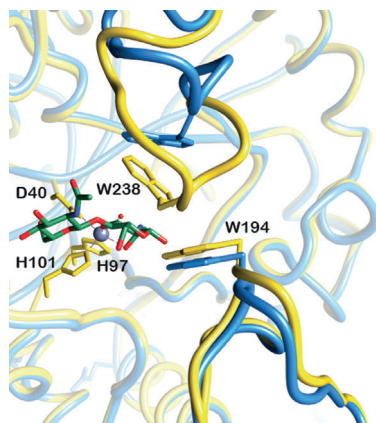
Communications

Enzyme Specificity

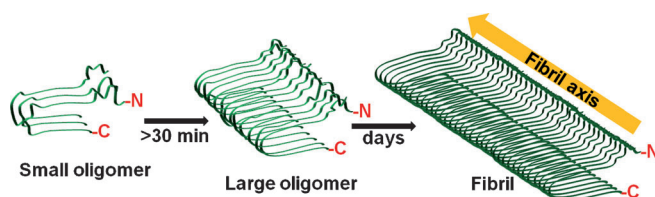
E. Andrés, D. Albesa-Jové, X. Biarnés,
B. M. Moerschbacher, M. E. Guerin,*
A. Planas* _____ 6882 – 6887

Structural Basis of Chitin Oligosaccharide
Deacetylation


Frontispiece



In the loop: The 3D structure of a chitin de-N-acetylase with its natural substrate (GlcNAc)₂ in a productive mode for catalysis has resulted in an induced-fit mechanism being identified in which a significant conformational change occurs in the loop closing the active site. In line with the proposed subsite-capping model, a complex with the longer substrate (GlcNAc)₃ shows additional loop rearrangements.



Fast fluorescence and slow solid-state NMR techniques were combined to probe the structure of small, unstable A β ₄₀ oligomers that show a potential sign of toxicity, namely, an enhanced affinity for

cell membranes. The turn region and the N-terminal tail are strikingly different to those of the less-toxic fibrils. These results may provide specific assays for Alzheimer's therapeutics.

Transient Oligomer Structures

B. Sarkar, V. S. Mithu, B. Chandra,
A. Mandal, M. Chandrasekan,
D. Bhowmik, P. K. Madhu,*
S. Maiti* _____ 6888 – 6892

Significant Structural Differences between
Transient Amyloid- β Oligomers and Less-
Toxic Fibrils in Regions Known To Harbor
Familial Alzheimer's Mutations


Inside Cover





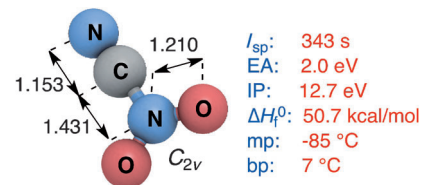
Energetic Materials

M. Rahm,* G. Bélanger-Chabot,
R. Haiges, K. O. Christe* — **6893–6897**



Nitryl Cyanide, NCNO₂

Small yet feisty: The elusive small molecule nitryl cyanide, NCNO₂, has been synthesized and characterized. It has a high kinetic stability, is extremely energetic, has a perfect oxygen balance with respect to combustion to CO₂ and N₂, and has potential as a building block for other energetic materials. Nitryl cyanide might also be of interest for atmospheric and interstellar chemistry. I_{sp} = specific impulse.



Back Cover

Nanocrystal Formation

K. Yu,* X. Liu, Q. Y. Chen, H. Yang,*
M. Yang, X. Wang, X. Wang, H. Cao,
D. M. Whitfield, C. Hu,
Y. Tao — **6898–6904**



Mechanistic Study of the Role of Primary Amines in Precursor Conversions to Semiconductor Nanocrystals at Low Temperature

Reaction $\text{Cd}(\text{OOCCH}_2\text{H}_{33})_2 + \text{Se}=\text{P}(\text{C}_8\text{H}_{17})_3 + \text{HPPH}_2 + \text{C}_{18}\text{H}_{35}\text{NH}_2$

³¹P NMR detected the P-containing compounds of

Ph₂P(Se)–NHC₁₈H₃₅ (1, 58 ppm)

Ph₂P–NHC₁₈H₃₅ (2, 42 ppm)

Ph₂P–PPh₂ (3, -14 ppm)

Ph₂P–OOCCH₂H₃₃ (4, 99 ppm)

Identifying roles: ³¹P NMR and absorption spectroscopy was used in conjunction with DFT calculations to identify the reaction pathways leading to semiconductor nanocrystals (NCs), together with compounds 1–4 (see scheme). With

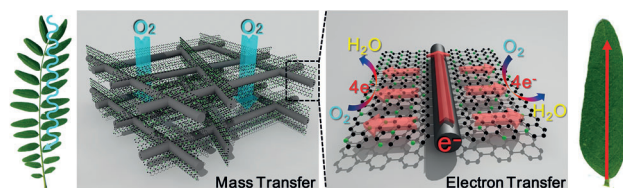
Se=PPh₂H as the Se precursor instead of Se=P(C₈H₁₇)₃, the conversion takes place at low temperature. The amine contributes to the formation of the two nitrogen compounds 1 and 2.

Electrocatalysis

T. N. Ye, L. B. Lv, X. H. Li,* M. Xu,
J. S. Chen* — **6905–6909**



Strongly Veined Carbon Nanoleaves as a Highly Efficient Metal-Free Electrocatalyst



“Green” leaves: A novel “vein-leaf”-type 3D complex of carbon nanofibers with nitrogen-doped graphene (CNF@NG) was prepared through thermolysis of biomass materials. The interconnected 3D network of CNF@NG facilitates both

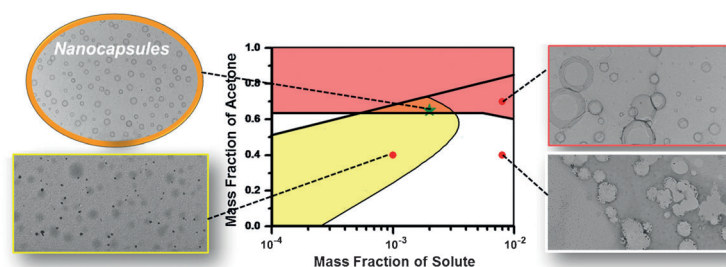
the electron transfer and mass diffusion for electrochemical reactions. As a result, the CNF@NG complexes demonstrate remarkable electrocatalytic activities toward the oxygen reduction reaction (ORR).

Functional Nanostructures

X. Yan, M. Delgado, A. Fu, P. Alcouffe,
S. G. Gouin, E. Fleury, J. L. Katz,
F. Ganachaud, J. Bernard* — **6910–6913**

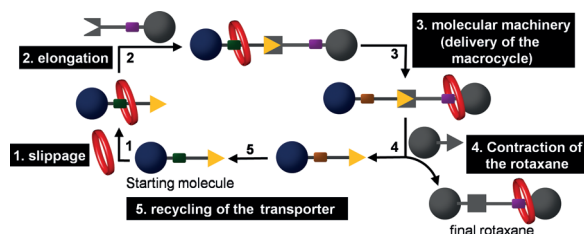


Simple but Precise Engineering of Functional Nanocapsules through Nanoprecipitation



Shift N' go! The establishment of both polymer and oil phase diagrams facilitated the generation of stable nanocapsules through nanoprecipitation (see pic-

ture). This simple methodology enabled the preparation of shell-functionalized and core-loaded nanocapsules through batch mixing.



Mode of transport: A chemical route to [2]rotaxanes, devoid of any template, is reported. The strategy relies on the preparation of an isolable [2]rotaxane building-block, bearing a macrocycle which can be delivered to another docked molecular

axle through a translational motion with subsequent contraction of the axle. It efficiently affords new triazolium rotaxanes which cannot be accessed by classical template methods.

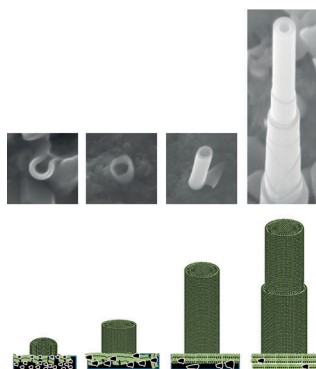
Supramolecular Chemistry

S. Chao, C. Romuald, K. Fournel-Marotte, C. Clavel, F. Coutrot* — 6914–6919

A Strategy Utilizing a Recyclable Macrocycle Transporter for the Efficient Synthesis of a Triazolium-Based [2]Rotaxane



When two systems meet: Interactions between two systems comprising different in-plane periodicities can result in misfit layered compounds even if each subsystem alone is not a layered or stable compound. A combination of two independent stimuli, namely the incommensurability of the misfit lattice (often leading to folding and scrolling) and the reactivity of the layer rim atoms, provides a new strategy to synthesize misfit nanotubes.



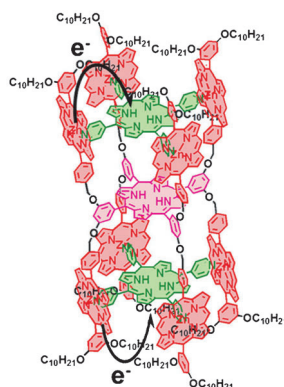
Misfit Nanotubes

L. S. Panchakarla, R. Popovitz-Biro, L. Houben, R. E. Dunin-Borkowski, R. Tenne* — 6920–6924

Lanthanide-Based Functional Misfit-Layered Nanotubes



Multiporphyrin dendrimers exhibit an excellent excitation energy transfer from the zinc porphyrin wings to the focal free-base porphyrin. The energy transfer is switched to an ultrafast electron transfer by the formation of a host–guest complex with tetrapyrrolyl porphyrin guests (green in picture). Thus, the multiporphyrin dendrimers serve as unique biomimetic models for both light-harvesting complexes LHC1 and LHC2.



Light Harvesting

Y.-H. Jeong, M. Son, H. Yoon, P. Kim, D.-H. Lee, D. Kim,* W.-D. Jang* — 6925–6928

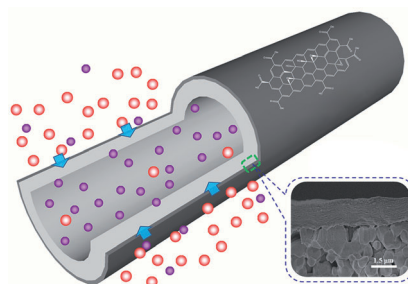
Guest-Induced Photophysical Property Switching of Artificial Light-Harvesting Dendrimers



Inside Back Cover



On a roll: A graphene oxide (GO) membrane (see picture, dark gray) was prepared on a ceramic hollow fiber (light gray) by a vacuum suction method. The resulting GO membrane exhibits excellent selective water permeation of aqueous dimethyl carbonate solution (water: purple; Me₂CO₃: red; scale bar: 1.5 μm).



Graphene Oxide Membranes

K. Huang, G. Liu, Y. Lou, Z. Dong, J. Shen, W. Jin* — 6929–6932

A Graphene Oxide Membrane with Highly Selective Molecular Separation of Aqueous Organic Solution



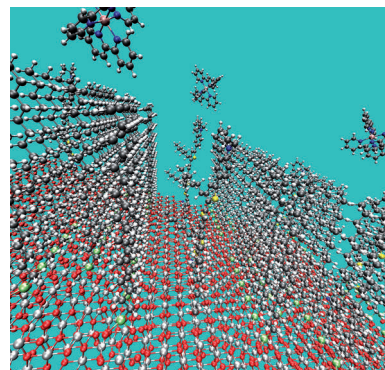
Dye-Sensitized Solar Cells

C. Dong, W. Xiang, F. Huang, D. Fu,
W. Huang, U. Bach, Y.-B. Cheng, X. Li,*
L. Spiccia* ————— **6933 – 6937**



Controlling Interfacial Recombination in
Aqueous Dye-Sensitized Solar Cells by
Octadecyltrichlorosilane Surface
Treatment

Keep them separated: Introduction of an octadecyltrichlorosilane layer on titania photoanodes is shown to be an effective strategy for suppressing electron recombination with cobalt species in dye-sensitized solar cells based on aqueous electrolytes, leading to a record PCE of 5.74 %. Color codes: Ti silvery, O red, Si green, S yellow, N blue, Co pink.



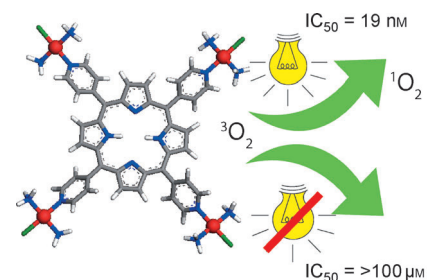
Photodynamic Therapy

A. Naik, R. Rubbiani, G. Gasser,
B. Spingler* ————— **6938 – 6941**



Visible-Light-Induced Annihilation of
Tumor Cells with Platinum–Porphyrin
Conjugates

A fatal synergy: A series of tetraplatinated porphyrins (see example; Pt red, Cl green, N blue, C gray, H white) were shown to be promising photosensitizers for photodynamic therapy. The complexes displayed excellent toxicity profiles under low-energy irradiation, with a phototoxic index of up to 5000, and high potency against a range of cancerous cell lines.

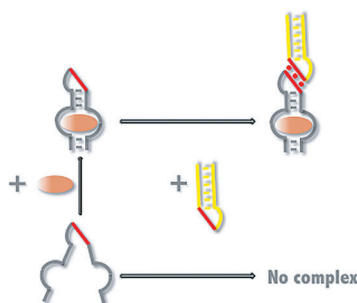


Aptamer Biosensors

G. Durand, S. Lisi, C. Ravelet, E. Dausse,
E. Peyrin,* J.-J. Toulmé* — **6942 – 6945**



Riboswitches Based on Kissing
Complexes for the Detection of Small
Ligands



Switch and kiss: Hairpin aptamers (gray) were engineered to switch between unfolded and folded conformations upon binding to their cognate ligand (orange). The folded conformation but not the unfolded one is recognized by a second hairpin termed the aptakiss (yellow) through loop–loop or kissing interactions (red). The detection of the aptakiss–aptamer–ligand ternary complex by SPR or fluorescence anisotropy signals the presence of the target ligand.

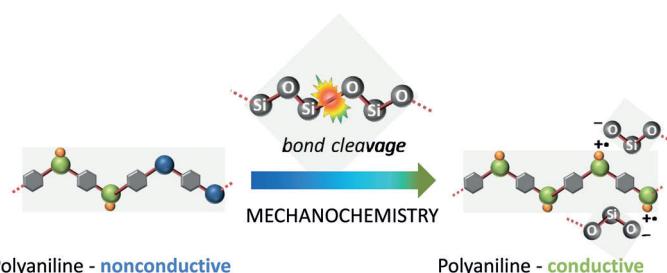


Mechanochemistry

B. Baytekin, H. T. Baytekin,
B. A. Grzybowski* ————— **6946 – 6950**



Mechanically Driven Activation of
Polyaniline into Its Conductive Form



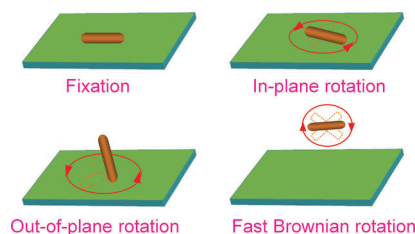
Polyaniline - nonconductive

Polyaniline - conductive

Rubbing makes conductive: Contact-charged and mechanically deformed polymers can transform nonconductive polyaniline (PANI) into its conductive form with the help of mechano-ions

formed at the polymer's surface. This phenomenon provides a convenient way for printing conductive patterns into PANI and also for rendering small polymeric objects antistatic.

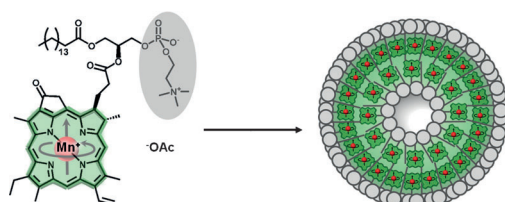
True affinity or mixed feelings? By the rotational and translational tracking of single protein-coated gold nanorods on C₁₈-modified silica surfaces in real time, four different dynamic states and their transitions were observed. The different states (see picture) reflected spatial and temporal variation of the strength of the hydrophobic interaction between individual probes and the surface.



Interfacial Dynamics

D. Xu, Y. He,* E. S. Yeung — 6951 – 6955

Direct Observation of the Orientation Dynamics of Single Protein-Coated Nanoparticles at Liquid/Solid Interfaces



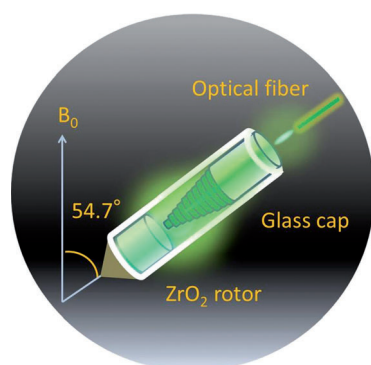
Lights, camera, action: Photothermal therapy makes use of photothermal sensitizers and laser light to thermally ablate diseased tissues. Porphyrin nanoparticles offer a nontoxic alternative to inorganic nanocrystals for the efficient

conversion of light into heat. Mn³⁺ ions were incorporated into the building blocks of porphyrin nanoparticles, thus imparting MRI sensitivity while improving photostability and maintaining high photothermal efficiency.

Theranostics

T. D. MacDonald, T. W. Liu, G. Zheng* — 6956 – 6959

An MRI-Sensitive, Non-Photobleachable Porphyrin Photothermal Agent

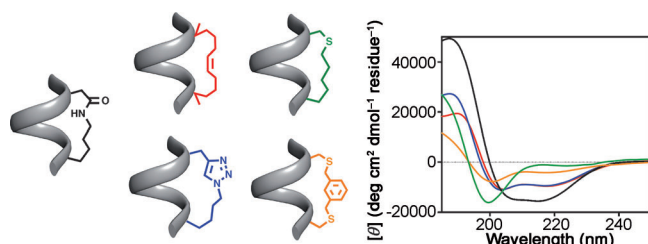


The photocycle of a sensory rhodopsin I (SrSRI) was examined using in situ photo-irradiation solid-state NMR spectroscopy. The M-intermediate was trapped by illumination (520 or 595 nm) and is responsible for the positive phototaxis. The M-intermediate was transferred to the P-intermediate (negative phototaxis) by irradiation at 365 nm. These results provide insights into the mechanism of the color-discriminating functional switch of SrSRI.

Photoreceptor Proteins

H. Yomoda, Y. Makino, Y. Tomonaga, T. Hidaka, I. Kawamura,* T. Okitsu, A. Wada, Y. Sudo,* A. Naito* — 6960 – 6964

Color-Discriminating Retinal Configurations of Sensory Rhodopsin I by Photo-Irradiation Solid-State NMR Spectroscopy



Covalent linkers between amino acid side chains in peptide sequences can induce bioactive α -helical conformations that modulate protein–protein interactions. A specific lactam linker is shown here to

confer more α -helicity than other cross-links to a cyclic pentapeptide in water, and to be a better helix nucleator when attached to longer peptides.

Helix Inducers

A. D. de Araujo, H. N. Hoang, W. M. Kok, F. Diness, P. Gupta, T. A. Hill, R. W. Driver, D. A. Price, S. Liras, D. P. Fairlie* — 6965 – 6969

Comparative α -Helicity of Cyclic Pentapeptides in Water

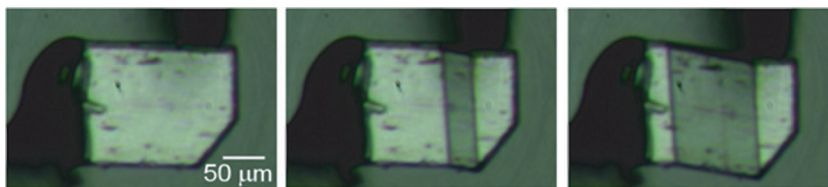


Organosuperelasticity

S. Takamizawa,*
Y. Miyamoto _____ 6970–6973



Superelastic Organic Crystals



A pure organic crystal of terephthalamide exhibits superelasticity (crystal-to-crystal transformation pseudo elasticity). A large motion is produced precisely with high repetition and high energy storage effi-

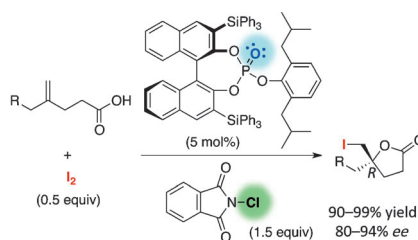
ciency. This process is driven by a small shear stress owing to the low density of strain energy, which is related to the low lattice energy.

Asymmetric Catalysis

H. Nakatsuji, Y. Sawamura, A. Sakakura,*
K. Ishihara* _____ 6974–6977



Cooperative Activation with Chiral Nucleophilic Catalysts and *N*-Haloimides: Enantioselective Iodolactonization of 4-Arylmethyl-4-pentenoic Acids



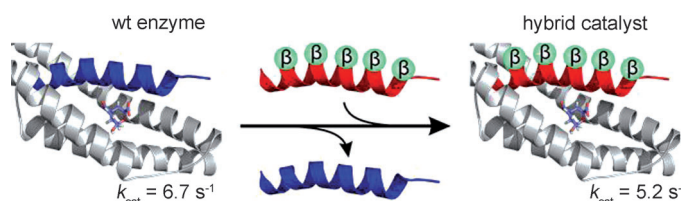
Active duty: Chiral triaryl phosphates promote the enantioselective iodolactonization of 4-substituted 4-pentenoic acids to give the corresponding iodolactones in high yields with high enantioselectivity (see scheme). *N*-Chlorophthalimide (NCP) is employed as a Lewis acidic activator and oxidant of I_2 for the present iodolactonization. In combination with 1.5 equivalents of NCP, only 0.5 equivalents of I_2 are sufficient for generating the iodinating reagent.

Foldamer Catalysts

C. Mayer, M. M. Müller, S. H. Gellman,
D. Hilvert* _____ 6978–6981



Building Proficient Enzymes with Foldamer Prostheses



More than a pegleg: Foldamers (red) containing β -amino acids (green) are shown to be useful as prostheses for creating chimeric enzymes. The activity of the resulting hybrids depends on the number, type, and location of the non-

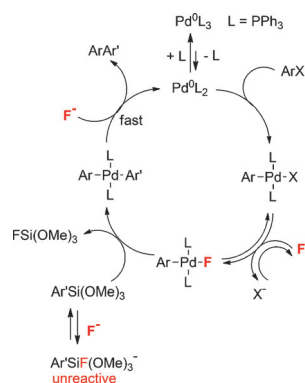
natural building blocks, but rivals that of the natural enzyme in the best cases. The unique properties of foldamers could lead to novel activities not accessible with natural enzymes.

Reaction Mechanisms

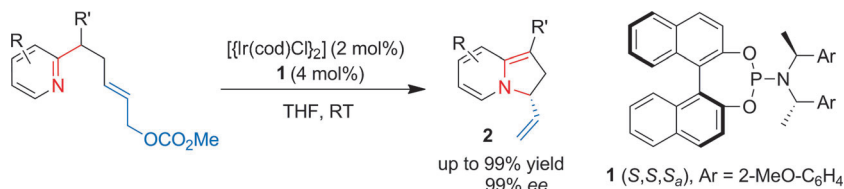
C. Amatore,* L. Grimaud,* G. Le Duc,
A. Jutand* _____ 6982–6985



Three Roles for the Fluoride Ion in Palladium-Catalyzed Hiyama Reactions: Transmetalation of $[ArPdFL_2]$ by $Ar'Si(OR)_3$



Role playing: F^- favors the Hiyama reaction by formation of $trans-[ArPdF(PPh_3)_2]$, which reacts with $Ar'Si(OMe)_3$ in the rate-determining transmetalation, and by promoting the reductive elimination from the intermediate $trans-[ArPdAr'(PPh_3)_2]$. Conversely, F^- disfavors the reaction by formation of the unreactive $[Ar'SiF(OMe)_3]^-$, thus leading to two antagonistic effects of F^- . As a consequence, $[F^-]/[Ar'Si(OMe)_3]$ has to be less than unity.



Getting 'rid' of aromaticity: An iridium-catalyzed intramolecular asymmetric allylic dearomatization reaction converts pyridines and pyrazines (not shown) into 2,3-dihydroindolizine and 6,7-dihydro-

pyrrolo[1,2-*a*]pyrazine derivatives with excellent yields and enantioselectivities. The dearomatization proceeds by direct N-allylic alkylation of pyridines or pyrazines under mild reaction conditions.

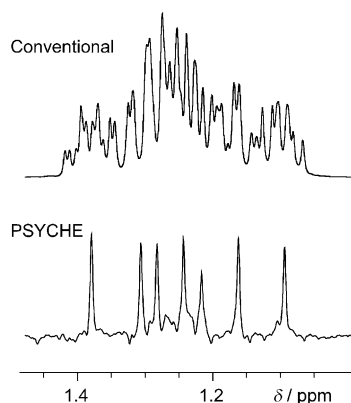
Asymmetric Catalysis

Z.-P. Yang, Q.-F. Wu,
S.-L. You* 6986–6989

Direct Asymmetric Dearomatization of Pyridines and Pyrazines by Iridium-Catalyzed Allylic Amination Reactions



All psyched up: A flexible and general pure shift experiment (PSYCHE) has been developed that offers superior sensitivity, spectral purity, and tolerance of strong coupling over existing methods for broadband homonuclear decoupling. The partial spectra of estradiol in [D₆]DMSO obtained by normal ¹H NMR spectroscopy and PSYCHE are shown for comparison.



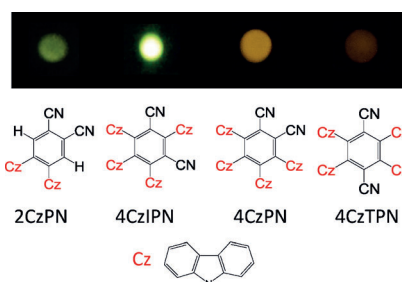
NMR Spectroscopy

M. Foroozandeh, R. W. Adams,
N. J. Meharry, D. Jeannerat, M. Nilsson,
G. A. Morris* 6990–6992

Ultrahigh-Resolution NMR Spectroscopy



Donor–acceptor molecules with thermally activated delayed fluorescence (TADF) at room temperature can emit efficient electrogenerated chemiluminescence (ECL). Efficient spin up-conversion from triplet to singlet excited states through thermal activation is required to break through the theoretical limitation according to spin statistics (25% of the quantum yield of photoluminescence). The ECL efficiency one TADF molecule reached about 50%.



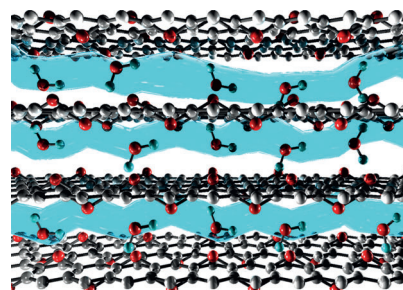
Photoelectrochemistry

R. Ishimatsu,* S. Matsunami, T. Kasahara,
J. Mizuno, T. Edura, C. Adachi,*
K. Nakano, T. Imato* 6993–6996

Electrogenerated Chemiluminescence of Donor–Acceptor Molecules with Thermally Activated Delayed Fluorescence



High proton conductivities in multilayer graphene oxide (GO) at room temperature facilitate the use of GO as a solid electrolyte in various cells. The conductivities of multilayer GO films are larger than those of single-layer films by several orders of magnitude, which implies that protons move more easily in the interlayers than on the surfaces.



Graphene Oxide

K. Hatakeyama, M. R. Karim, C. Ogata,
H. Tateishi, A. Funatsu, T. Taniguchi,
M. Koinuma, S. Hayami,*
Y. Matsumoto* 6997–7000

Proton Conductivities of Graphene Oxide Nanosheets: Single, Multilayer, and Modified Nanosheets

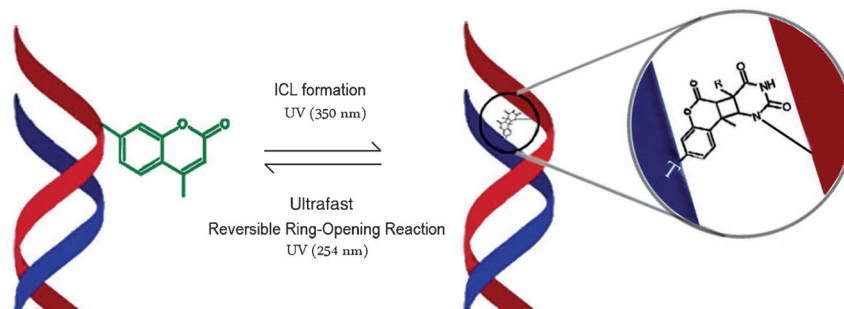


DNA Cross-Linking

M. M. Haque, H. Sun, S. Liu, Y. Wang,
X. Peng* 7001–7005



Photoswitchable Formation of a DNA Interstrand Cross-Link by a Coumarin-Modified Nucleotide



A [2+2] cycloaddition reaction between a coumarin-modified pyrimidine nucleoside and the opposing thymidine,

2'-deoxycytidine, or 2'-deoxyadenosine on a DNA strand generates photoswitchable interstrand cross-links (ICLs) in the DNA.

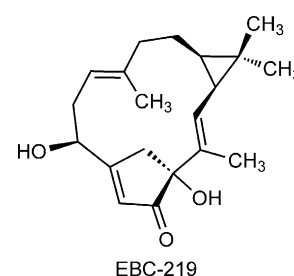
Natural Product Isolation

L. A. Maslovskaya, A. I. Savchenko,
E. H. Krenske, C. J. Pierce, V. A. Gordon,
P. W. Reddell, P. G. Parsons,
C. M. Williams* 7006–7009



EBC-219: A New Diterpene Skeleton, Crotingsulidane, from the Australian Rainforest Containing a Bridgehead Double Bond

Julius Brecht and Australia have a common history in the search for naturally occurring gems. The work on monoterpenes leading to Brecht's rule continues to develop over a hundred years later in that the search for bioactive natural products has delivered EBC-219, a bicyclo[10.2.1] bridgehead olefin with an unprecedented diterpene carbon skeleton.

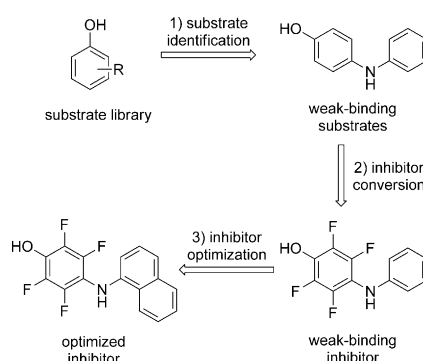


Kinase Inhibitors

M. E. Breen, M. E. Steffey, E. J. Lachacz,
F. E. Kwarcinski, C. C. Fox,
M. B. Soellner* 7010–7013



Substrate Activity Screening with Kinases: Discovery of Small-Molecule Substrate-Competitive c-Src Inhibitors



SASsy inhibitors: Small-molecule substrate-competitive inhibitors of the tyrosine kinase c-Src were discovered through the application of substrate activity screening (SAS). Characterization of the lead inhibitor demonstrates that substrate-competitive kinase inhibitors possess unique properties, including cellular efficacy that matches biochemical potency and synergy with ATP-competitive inhibitors.

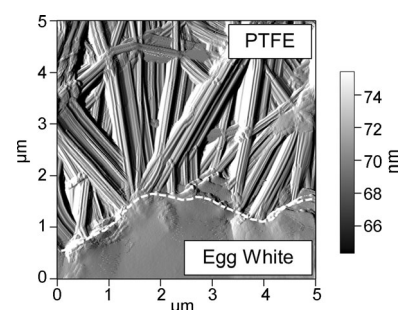
Protective Coatings

J. Imbrogno, A. Nayak,
G. Belfort* 7014–7017



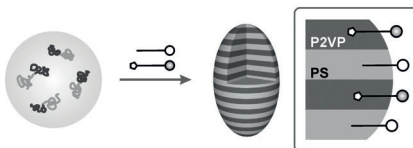
Egg White Varnishes on Ancient Paintings: A Molecular Connection to Amyloid Proteins

Egg white coatings: Why was egg white used and what were its unique properties as a varnish for paintings? These questions have puzzled the art community for hundreds of years. A molecular basis is given for the beneficial properties of egg white varnishes on paintings (see AFM amplitude image of egg white coated on a PTFE membrane) using ATR-FTIR spectra and one of its protective properties by oxygen transport.

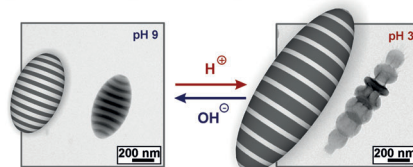


Let surfactants do the work: Controlling the shape and morphology of polystyrene-*b*-poly(2-vinylpyridine) (PS-*b*-P2VP) particles through the use of tailored, mixed surfactant systems allows for the facile and selective preparation of ellipsoidal nanoparticles. Cross-linking of the axially stacked P2VP lamellae domains allows the pH responsiveness of the 2VP moieties to be exploited with generation of unique shape- and structure-responsive particles.

surfactant-directed self-assembly



pH-induced dynamic shape change

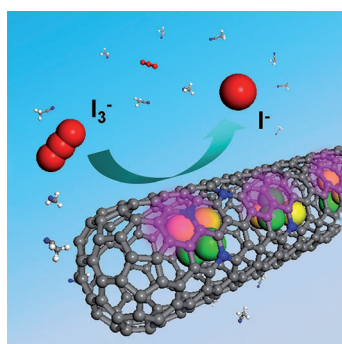


Block Copolymer Nanoparticles

D. Klinger, C. X. Wang, L. A. Connal, D. J. Audus, S. G. Jang, S. Kraemer, K. L. Killops, G. H. Fredrickson, E. J. Kramer, C. J. Hawker* – **7018–7022**

A Facile Synthesis of Dynamic, Shape-Changing Polymer Particles

Front Cover

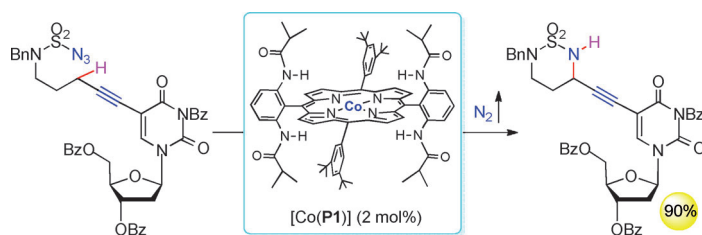


The direct pyrolysis of organometallic precursors was used to prepare podlike nitrogen-doped carbon nanotubes encapsulating FeNi alloy nanoparticles (Pod(N)-FeNi), which exhibited high electrocatalytic activity in the I_3^-/I^- redox reaction and excellent electrochemical stability. The photovoltaic performance of dye-sensitized solar cells made with the Pod(N)-FeNi counter electrode was similar to that having a Pt counter electrode.

Dye-Sensitized Solar Cells

X. J. Zheng, J. Deng, N. Wang, D. H. Deng,* W.-H. Zhang,* X. H. Bao, C. Li – **7023–7027**

Podlike N-Doped Carbon Nanotubes Encapsulating FeNi Alloy Nanoparticles: High-Performance Counter Electrode Materials for Dye-Sensitized Solar Cells



Make a ring of it: Highly chemoselective intramolecular amination of propargylic $C(sp^3)$ –H bonds has been achieved with a high degree of functional-group tolerance through the title reaction. The [Co(P1)]-catalyzed C–H amination pro-

ceeds under neutral and nonoxidative conditions without the need of any additives, thus providing a direct method for efficient synthesis of functionalized propargylamine derivatives with N_2 as the only by-product.

Cyclization

H. J. Lu,* C. Li, H. Jiang, C. L. Lizardi, X. P. Zhang* – **7028–7032**

Chemoselective Amination of Propargylic $C(sp^3)$ –H Bonds by Cobalt(II)-Based Metalloradical Catalysis

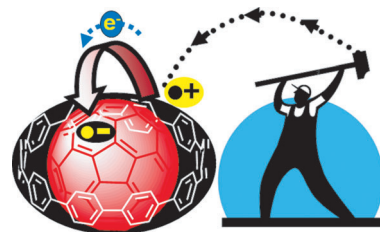
Single-Wall Nanotube Models

M. P. Alvarez, P. M. Burrezo, M. Kertesz,
T. Iwamoto, S. Yamago, J. Xia, R. Jasti,
J. T. L. Navarrete,* M. Taravillo,
V. G. Baonza,* J. Casado* — 7033 – 7037



Properties of Sizeable
[*n*]Cycloparaphenylenes as Molecular
Models of Single-Wall Carbon Nanotubes
Elucidated by Raman Spectroscopy:
Structural and Electron-Transfer
Responses under Mechanical Stress

[*n*]Cycloparaphenylenes behave as molecular templates of “perfectly chemically defined” single-wall carbon nanotubes. These [*n*]CPP molecules have electronic, mechanical, and chemical properties in size correspondence with their giant congeners. Under mechanical stress, they form charge-transfer salts, or complexes with fullerene, by one-electron concave–convex electron transfer.

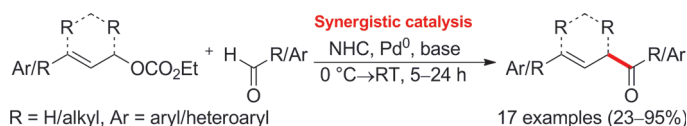


Synthetic Methods

M. M. Ahire, S. B. Mhaske* 7038 – 7042



Direct Allylation of In Situ Generated
Aldehyde Acyl Anions by Synergistic NHC
and Palladium Catalysis



Efficient access to β,γ -unsaturated ketones is achieved through synergistic NHC and Pd catalysis. The direct, regioselective allylation of in situ generated aldehyde acyl anions starts from easily accessible allylic carbonates and alde-

hydes without any preactivation and proceeds under mild reaction conditions. This synergistic catalysis method adds a new dimension to metal-mediated C allylations.

DOI: 10.1002/anie.201405644

Flashback: 50 Years Ago ...

Which elements was the milky way formed from? The chemical composition of the stars was discussed in a Review by A. Unsöld, who outlined the evolution of stars and the milky way, and the quantitative analysis of stellar spectra to determine elemental abundance in the atmospheres of various stars. The analysis showed that the first stars were formed from an almost spherical cloud of hydrogen. Topics of other Reviews included heat-resistant films and fibers (F. T. Wallenberger) and the synthesis of the corrin system (A. Eschenmoser et al.).

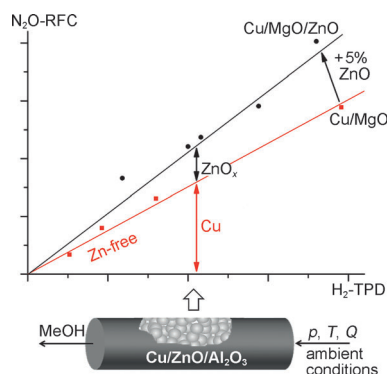
R. Köster et al. from the Max Planck Institute for Coal Research, which celebrates its centenary this year, published two Communications on boron and aluminum radicals. The first report was on the generation of boron and aluminum free radicals by dehalogenation of the corresponding chloro compounds, and the second was on the formation of organic carboranes by radical reactions.

Alan MacDiarmid, who shared the Nobel Prize in Chemistry 2000 with Alan Heeger and Hideki Shirakawa for

their work on conductive polymers, published a Communication on compounds containing the Si–N–B linkage. Compounds such as $[(\text{CH}_3)_3\text{Si}]_2\text{NBF}_2$ were prepared by reacting lithium bis(trimethylsilylamide) with BF_3 . Competition was fierce even 50 years ago: an alternative route to these compounds was published by P. Geymayer around the same time in *Angewandte Chemie*.

[Read more in Issue 7/1964.](#)

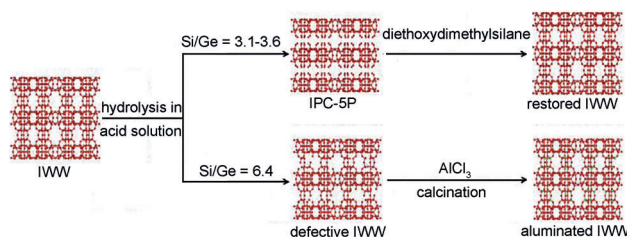
A combination of N_2O reactive frontal chromatography and H_2 temperature-programmed desorption is used to analyze the interplay of copper and zinc oxide in methanol synthesis catalysts. This method provides an easy in situ approach to quantify the direct copper–zinc interaction (SMSI effect) and offers an important possibility to rational catalyst design also for other supported metal catalysts.



Catalyst Characterization

M. B. Fichtl, J. Schumann, I. Kasatkin, N. Jacobsen, M. Behrens, R. Schlögl, M. Muhler, O. Hinrichsen* **7043 – 7047**

Counting of Oxygen Defects versus Metal Surface Sites in Methanol Synthesis Catalysts by Different Probe Molecules



ADORable zeolites: The Assembly-Disassembly-Organization-Reassembly (ADOR) mechanism is used to manipulate germanosilicate zeolites with the

IWW framework type. The outcome of the process depends critically on the Si/Ge ratio in the starting material.

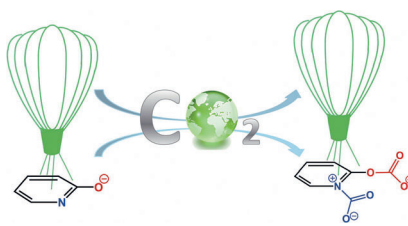
Zeolites

P. Chlubná-Eliášová, Y. Tian, A. B. Pinar, M. Kubů, J. Čejka,* R. E. Morris* **7048 – 7052**

The Assembly-Disassembly-Organization-Reassembly Mechanism for 3D-2D-3D Transformation of Germanosilicate IWW Zeolite



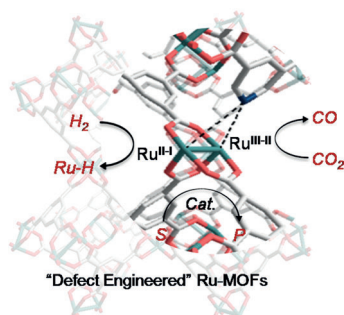
CO₂ capture was improved by several pyridine-containing anion-functionalized ionic liquids, making use of multiple-site cooperative interactions. The interactions between two kinds of sites in the anion and CO₂ resulted in superior CO₂ capacities. These novel ionic liquids open the door to develop highly efficient and reversible carbon capture processes.



Carbon Capture

X. Luo, Y. Guo, F. Ding, H. Zhao, G. Cui, H. Li, C. Wang* **7053 – 7057**

Significant Improvements in CO₂ Capture by Pyridine-Containing Anion-Functionalized Ionic Liquids through Multiple-Site Cooperative Interactions



The defect engineering in Ru-based metal–organic frameworks (MOFs) at coordinatively unsaturated metal centers (CUS) induces partial reduction of the metal nodes and leads to properties that are absent for the parent MOF, such as dissociative chemisorption of CO₂ and enhanced sorption capacity of CO. The modified MOFs offer new perspectives as multifunctional materials whose performance is controlled by design of the defects.

Metal–Organic Frameworks

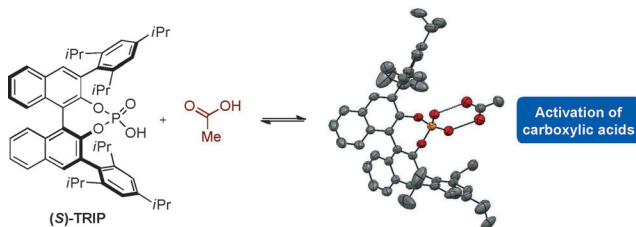
O. Kozachuk, I. Luz, F. X. Llabrés i Xamena, H. Noei, M. Kauer, H. B. Albada, E. D. Bloch, B. Marler, Y. Wang,* M. Muhler, R. A. Fischer* **7058 – 7062**

Multifunctional, Defect-Engineered Metal–Organic Frameworks with Ruthenium Centers: Sorption and Catalytic Properties



Organocatalysis

M. R. Monaco, B. Poladura,
M. Diaz de Los Bernardos, M. Leutzsch,
R. Goddard, B. List* — 7063 – 7067



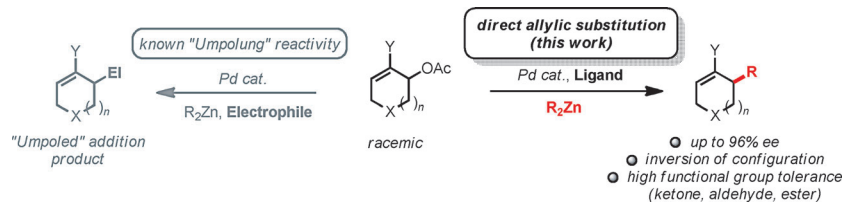
Activation of Carboxylic Acids in
Asymmetric Organocatalysis

Activation by dimerization: There is still no general activation mode for carboxylic acids in organocatalysis. The formation of heterodimers between chiral phosphoric acid diesters and carboxylic acids can be used to activate and direct reactivity of the

latter in asymmetric reactions. This novel principle has been applied to the ring-opening desymmetrization and kinetic resolution of aziridines leading to valuable amino alcohols.

Asymmetric Catalysis

A. Misale, S. Niyomchon, M. Luparia,
N. Maulide* — 7068 – 7073



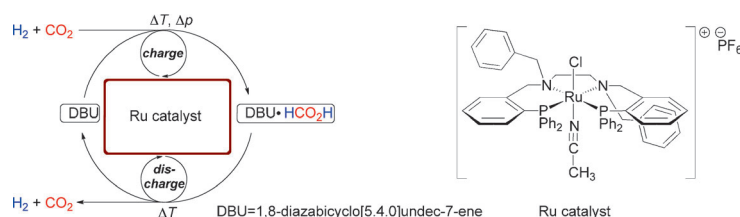
Asymmetric Palladium-Catalyzed Allylic
Alkylation Using Dialkylzinc Reagents: A
Remarkable Ligand Effect

Umpole-me-not: A serendipitously discovered palladium-catalyzed asymmetric allylic alkylation reaction with diorganozinc reagents displays broad functional group compatibility. This novel transfor-

mation hinges on a remarkable ligand effect which overrides the standard “umpolung” reactivity of allyl-palladium intermediates in the presence of dialkylzinc compounds.

Ru Catalysis

S.-F. Hsu, S. Rommel, P. Eversfield,
K. Muller, E. Klemm, W. R. Thiel,
B. Plietker* — 7074 – 7078



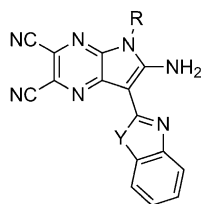
A Rechargeable Hydrogen Battery Based
on Ru Catalysis

Simple but efficient: A readily accessible Ru catalyst is the basis for a reversible H_2/CO_2 -driven battery. At elevated temperatures both the reduction of CO_2 to formic acid and the decomposition of formic acid were realized with 0.075 mol % of the Ru

complex. Several charging and discharging cycles were performed with comparable storage–release efficiency. Furthermore, the partial removal of defined gas amounts is possible under pressure.

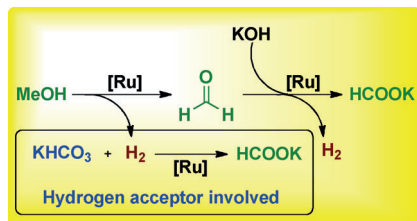
Medicinal Chemistry

D. Reker, M. Seet, M. Pillong, C. P. Koch,
P. Schneider, M. C. Witschel,
M. Rottmann, C. Freymond, R. Brun,
B. Schweizer, B. Illarionov, A. Bacher,
M. Fischer, F. Diederich,
G. Schneider* — 7079 – 7084



Deorphaning Pyrrolopyrazines as Potent
Multi-Target Antimalarial Agents

Pyrrolopyrazines show strong activity against *Plasmodium falciparum* parasites and *Plasmodium berghei* liver schizonts. In-depth bioinformatical analysis and target panel screening, suggesting IspD and multi-kinase inhibition, revealed their likely mode of action. The composite computational approach provides a unique access to deciphering polypharmacological effects of new bioactive chemical agents.



Get rid of CO and H₂: An efficient route for the industrial synthesis of formate salts without the utilization of carbon monoxide is highly desirable. A catalytic reaction combining methanol dehydrogenation and bicarbonate hydrogenation has been developed, which provides a green and cost-efficient process for the synthesis of formate salts with excellent turnover numbers and yields.

Homogeneous Catalysis

Q. Liu, L. Wu, S. Güllak, N. Rockstroh, R. Jackstell, M. Beller* — 7085 – 7088

Towards a Sustainable Synthesis of Formate Salts: Combined Catalytic Methanol Dehydrogenation and Bicarbonate Hydrogenation



Supporting information is available on www.angewandte.org (see article for access details).



A video clip is available as Supporting Information on www.angewandte.org (see article for access details).



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This article is accompanied by a cover picture (front or back cover, and inside or outside).

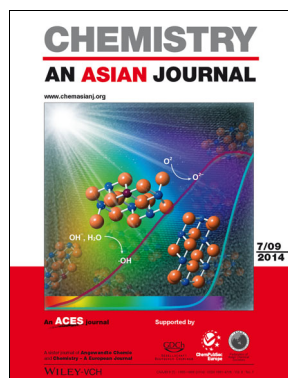


The Very Important Papers, marked VIP, have been rated unanimously as very important by the referees.



The Hot Papers are articles that the Editors have chosen on the basis of the referee reports to be of particular importance for an intensely studied area of research.

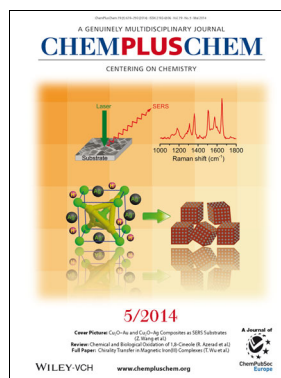
Check out these journals:



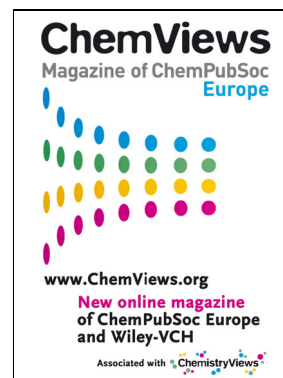
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