The following Communications have been judged by at least two referees to be "very important papers" and will be published online at www.angewandte.org soon:

A. Ciesielski, S. Lena, S. Masiero, G. P. Spada, P. Samorì**
Dynamers at the Solid–Liquid Interface: Controlling the Reversible Assembly/Reassembly Process Between Two Highly Ordered Supramolecular Guanine Motifs

*M. Alcarazo, T. Stork, A. Anoop, W. Thiel, A. Fürstner**
Steering the Surprisingly Modular π-Acceptor Properties of N-Heterocyclic Carbenes: Implications for Gold Catalysis

H. Braunschweig, C.-W. Chiu, K. Radacki, T. Kupfer*
Synthesis and Structure of an Carbene-Stabilized π-Boryl Anion

S. Zhang, Y. Shao, G. Yin, Y. Lin**
Electrostatic Self-Assembly of Platinum-Around-Gold Nanocomposite with High Activity Towards Formic Acid Oxidation

*D. Bojer, A. Venugopal, B. Neumann, H.-G. Stammler, N. W. Mitzel**
Lewis Base Induced Reductions in Organolanthanide Chemistry

S.-H. Kim, Su Y. Lee, S.-M. Yang**
Janus Microspheres for Highly Flexible and Impregnable Water-Repelling Interfaces

*L. Frullano, C. Catana, T. Benner, A. D. Sherry, P. Caravan **
A Bimodal MR-PET Agent for Quantitative pH Imaging

*K. Schober, E. Hartmann, H. Zhang, R. M. Gschwind**
^{1}H DOSY Spectra of Highly Enantioselective Ligands: A Fast and Simple NMR-Specroscopy Method to Optimize Catalytic Reaction Conditions

Author Profile

Thomas Wirth ________ 1528

"I chose chemistry as a career because I like to do experiments myself. Nowadays, this is unfortunately rarely the case.
My biggest inspiration is a completely unexpected result from a seemingly straightforward experiment ..."
This and more about Anthony Thomas Wirth can be found on page 1528.

Books

Protein Engineering Handbook Stefan Lutz, Uwe T. Bornscheuer *reviewed by P. Lindner* ________ 1529

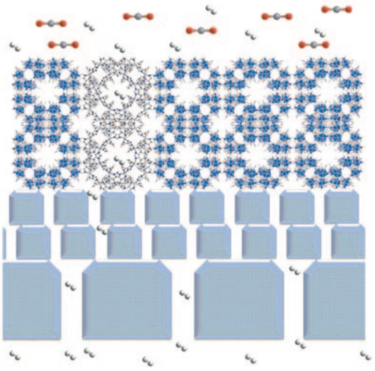
Highlights

Supported Membranes

J. Gascon, F. Kapteijn* ____ 1530 – 1532

Metal-Organic Framework Membranes—High Potential, Bright Future?

"Hard work today leads to success tomorrow" can be said for research on membranes of porous crystalline materials, which are ideal for size- and shape-selective separations with low energetic effort. The realization of the first metal-organic framework membranes may open up a plethora of applications, including functional coatings. The picture shows the separation of H_2 and CO_2 by a membrane on an alumina support.



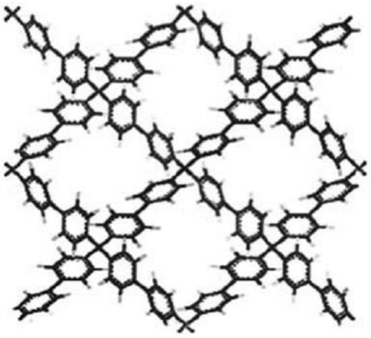
Contents

Microporous Materials

A. Trewin, A. I. Cooper* ___ **1533 – 1535**

Porous Organic Polymers: Distinction from Disorder?

An alternative explanation: The new microporous organic polymer framework PAF-1 displays exceptional physicochemical stability along with an extremely high surface area (BET surface area $5640\ m^2 g^{-1}$). The question arises whether this material displays the high degree of crystalline order presumed necessary for this high surface area.



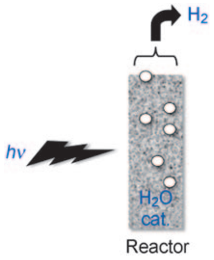
Essays

Hydrogen Generation

T. Maschmeyer,* M. Che ___ **1536 – 1539**

Catalytic Aspects of Light-Induced Hydrogen Generation in Water with TiO_2 and Other Photocatalysts: A Simple and Practical Way Towards a Normalization?

Avoiding erroneous conclusions: Only when operating under non-diffusion-limited conditions can statements be made regarding the inherent activity of a catalyst. Otherwise, such results reflect merely aspects of the reactor or reaction design. Suggestions are made as how to arrive at a more standardized method to overcome these problems.

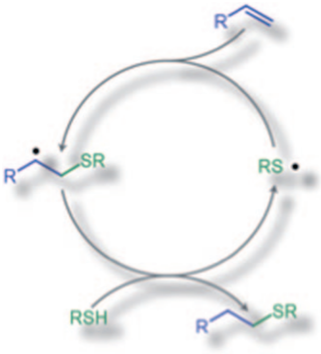


Reviews

Polymer Chemistry

C. E. Hoyle,
C. N. Bowman* ___ **1540 – 1573**

Thiol–Ene Click Chemistry



Click and easy: The radical thiol–ene reaction (see scheme) fulfills all the criteria for being a click reaction, and it has a unique potential for applications in polymer science and molecular synthesis. Particular advantages are simplicity, high yields and conversions, high reaction rates, and the possibility of photoinitiation.

For the USA and Canada:
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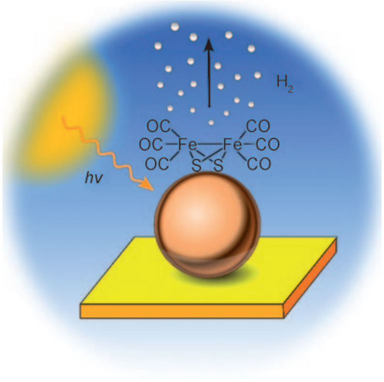
Communications

Quantum Dots

T. Nann,* S. K. Ibrahim, P.-M. Woi, S. Xu, J. Ziegler, C. J. Pickett* ____ 1574–1577

Water Splitting by Visible Light: A Nanophotocathode for Hydrogen Production

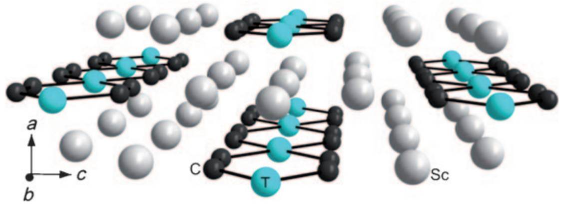


Hitting water with a 2 iron: A novel nanophotocathode for hydrogen production is based on a multilayer array of InP quantum dots activated with a synthetic diiron catalyst (see picture), which is related to the subsite of FeFe hydrogenase.

Superconducting Carbides

W. Scherer,* C. Hauf, M. Presnitz, E.-W. Scheidt, G. Eickerling,* V. Eyert, R.-D. Hoffmann, U. C. Rodewald, A. Hammerschmidt, C. Vogt, R. Pöttgen* ____ 1578–1582

Superconductivity in Quasi One-Dimensional Carbides

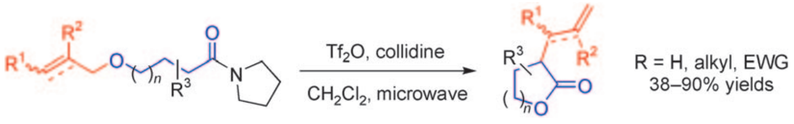


The electronic structures of the isotypic carbides Sc_3TC_4 (see picture; T = Fe, Co, Ni) are investigated by theoretical and experimental charge-density studies. Even tiny differences in the electronic band structure of these solids are reflected in the properties of the Laplacian of the experimental electron density. Only the cobalt carbide is superconducting below 4.5 K and displays a structural phase transition around 70 K.

Rearrangements

C. Madelaine, V. Valerio, N. Maulide* ____ 1583–1586

Unexpected Electrophilic Rearrangements of Amides: A Stereoselective Entry to Challenging Substituted Lactones

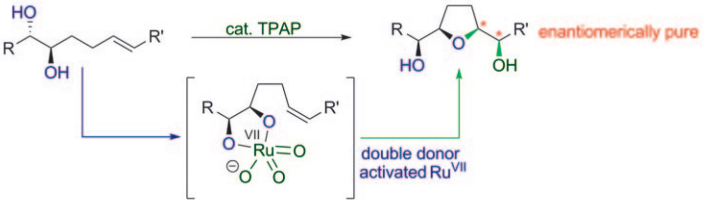


Surprise, surprise! An unexpected skeletal rearrangement was developed into a chemo- and stereoselective synthesis of α-allyl and allenyl lactones with challenging substitution patterns (see scheme; EWG = electron-withdrawing group). The generality, unique features, and synthetic potential of this reaction were probed and a mechanism was proposed.

Heterocycles

H. Cheng, C. B. W. Stark* ____ 1587–1590

A Double Donor-Activated Ruthenium(VII) Catalyst: Synthesis of Enantiomerically Pure THF-Diols



Double, double, no toil and trouble: Enantiomerically pure tetrahydrofurans are obtained with high position- and stereoselectivity through a ruthenium(VII)-catalyzed oxidative cyclization of 5,6-dihydroxy alkenes (see scheme TPAP = tetrapropylammonium perruthenate). A dual activation modifies the reactivity and increases the carbophilicity of the transition metal so that an otherwise unusual dioxygenation with perruthenate occurs.

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$$[(NH_3)_5Os^{II}(N_2)Os^{III}(NH_3)_5]^{5+} + H_2O \xrightarrow{h\nu} [Os^{VI}(NH_3)_4N]^{3+} + [Os^{V}(NH_3)_4(H_2O)]^{2+} + 3\,NH_3$$

A quantum of light for nitrogen: An intramolecular photoredox reaction leads to the reductive cleavage of dinitrogen upon metal-to-ligand charge transfer (MLCT) excitation. The primary photochemical step is assumed to proceed by formation of an Os^{VI} nitride complex (see scheme). Secondary reactions modify the product formation.

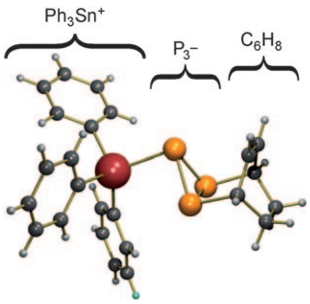
N_2 Activation

H. Kunkely, A. Vogler* ______ **1591 – 1593**

Photolysis of Aqueous $[(NH_3)_5Os(\mu\text{-}N_2)\text{-}Os(NH_3)_5]^{5+}$: Cleavage of Dinitrogen by an Intramolecular Photoredox Reaction

---

Anatomy of a P_3^- synthon: The $Ph_3SnP_3(C_6H_8)$ molecule (see picture; Sn red, P orange) has been synthesized by triphosphorus transfer from a niobium complex. This molecule was shown to be a viable source of P_3^- in a proof-of-principle reaction: loss of Ph_3SnCl and 1,3-cyclohexadiene from $Ph_3SnP_3(C_6H_8)$ upon treatment with Wilkinson's catalyst, $[ClRh(PPh_3)_3]$, gives rise to the new cyclo-P_3 complex $[(\eta^3\text{-}P_3)Rh(PPh_3)_3]$.



Phosphorus Chemistry

B. M. Cossairt,
C. C. Cummins* ______ **1595 – 1598**

Shuttling P_3 from Niobium to Rhodium: The Synthesis and Use of $Ph_3SnP_3(C_6H_8)$ as a P_3^- Synthon

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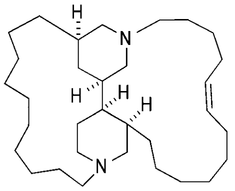


(±)-Haliclonacyclamine C

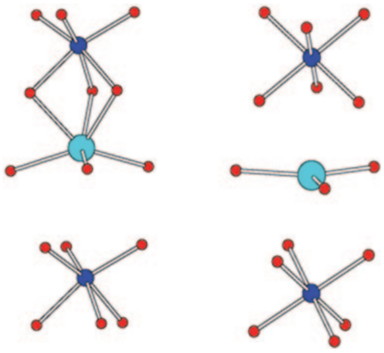
First in its class: The synthesis of the tetracyclic alkylpiperidine marine alkaloid (±)-haliclonacyclamine C has been completed, with a longest linear sequence of 24 steps. The key transformations are the stereoselective hydrogenation of an unsaturated macrocyclic bis(piperidine) and a ring-closing alkyne metathesis reaction.

Natural Product Synthesis

B. J. Smith,
G. A. Sulikowski* ______ **1599 – 1602**

Total Synthesis of (±)-Haliclonacyclamine C

The origin of the ferroelectricity in the indium-based multiferroic perovskites $(In_{1-x}M_x)MO_3$ ($M = Mn_{0.5}Fe_{0.5}$), and the question of how to lower their magnetic ordering (T_N) and the ferroelectric Curie temperatures, was investigated by density functional calculations. The analyses explain why T_N is near room temperature and why the perovskites with s^0 and s^2 A-site cations have similar ferroelectric distortions.



Solid-State Structures

VIP

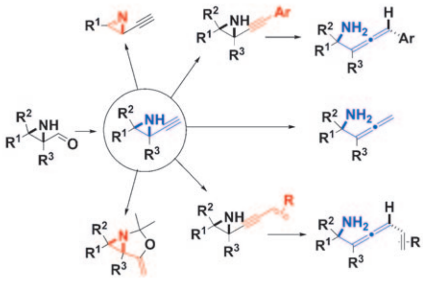
E. Kan, H. Xiang, C. Lee, F. Wu, J. Yang,
M.-H. Whangbo* ______ **1603 – 1606**

Ferroelectricity in Perovskites with s^0 A-Site Cations: Toward Near-Room-Temperature Multiferroics

Propargyl Amines

Z. He, A. K. Yudin* ———— **1607 – 1610**

A Versatile Synthetic Platform Based on Strained Propargyl Amines

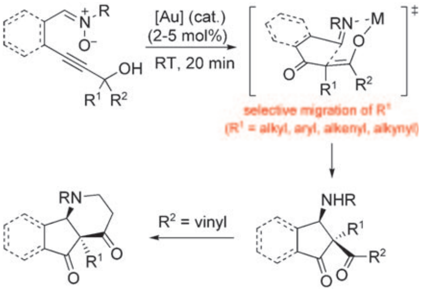


Divergent reactivity: Various ethynylaziridines behave as strained propargyl amines and can be directly converted into unprotected α-amino allenes by a highly diastereoselective S_N2' hydride delivery (see scheme). Additional reaction routes involve chemo- and regioselective transformation into either bicyclic aziridine/enol ethers or highly strained azirine alkynes.

Domino Reactions

H.-S. Yeom, Y. Lee, J. Jeong, E. So, S. Hwang, J.-E. Lee, S. S. Lee, S. Shin* ———— **1611 – 1614**

Stereoselective One-Pot Synthesis of 1-Aminoindanes and 5,6-Fused Azacycles Using a Gold-Catalyzed Redox-Pinacol-Mannich-Michael Cascade



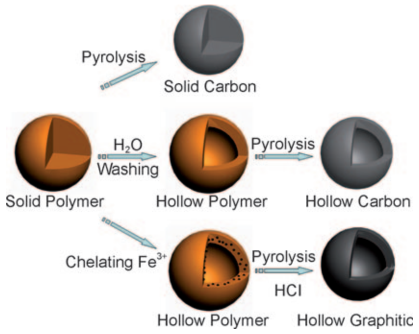
Just another Mannich Monday: A cascade intramolecular redox-pinacol-Mannich-Michael reaction sequence catalyzed by gold complexes can be used to generate a variety of structures including spirocycles, 1-aminoindanes, and 5,6-fused azabicycles that have a quaternary carbon center. The reaction is characterized by complete atom-economy, high diastereoselectivity, and remarkable efficiency through tandem reactions.

Microstructures

A.-H. Lu,* W.-C. Li, G.-P. Hao, B. Spliethoff, H.-J. Bongard, B. B. Schaack, F. Schüth ———— **1615 – 1618**

Easy Synthesis of Hollow Polymer, Carbon, and Graphitized Microspheres

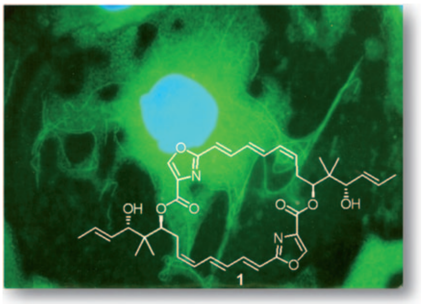
Balls galore! A new approach was developed for the easy synthesis of hollow microspheres with amorphous or graphitized microstructure. Starting from one type of solid polymer sphere, a simple water washing treatment led to the formation of hollow structures. Diverse products such as hollow carbon or graphitized spheres can be obtained, depending on subsequent treatment methods (see picture).



Natural Products

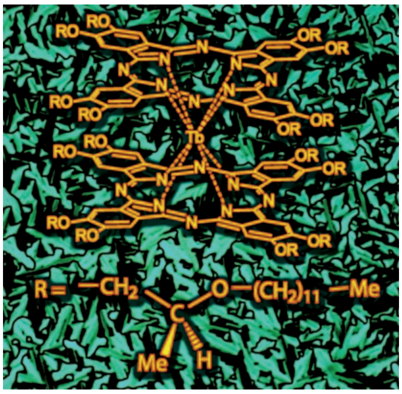
R. Schäckel, B. Hinkelmann, F. Sasse, M. Kalesse* ———— **1619 – 1622**

The Synthesis of Novel Disorazoles



Big little brother: The simplified disorazole analogue **1** exhibits its cytotoxic activity at low nanomolar concentrations and provides selectivities not observed for the parent natural product.

A terbium double-decker phthalocyanine complex has been prepared and characterized. The mesomorphism of the liquid-crystalline complex is used as a tool to reversibly adjust its magnetic properties, thus allowing the coexistence of two different relaxation processes, which can be reversibly modified by simple thermal treatments, to be observed.



Single-Molecule Magnets

M. Gonidec, F. Luis, À. Vílchez,
J. Esquena, D. B. Amabilino,*
J. Veciana* ———— 1623 – 1626

A Liquid-Crystalline Single-Molecule Magnet with Variable Magnetic Properties

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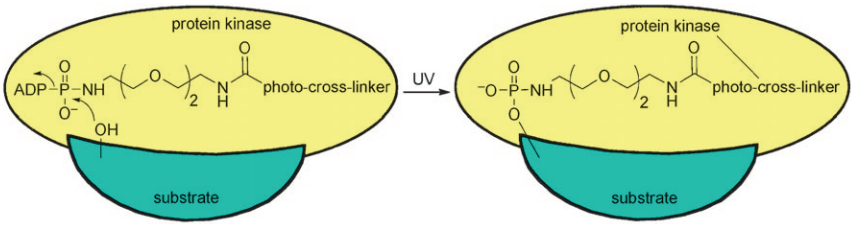
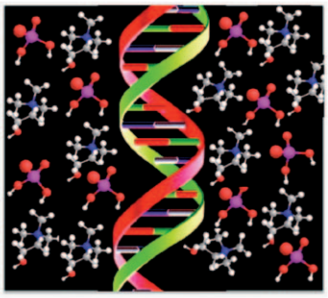


Pinning down kinase substrates: The identification of substrates of a particular kinase is fundamental to the elucidation of cell-signaling cascades. This problem has now been addressed by the title approach involving kinase-catalyzed labeling coupled with photo-cross-linking (see scheme). When coupled with mass spectrometry analysis, this strategy can be used to determine the sites of phosphorylation as well as the effector kinase. ADP = adenosine 5′-diphosphate.

Biochemical Mechanisms

S. Suwal, M. K. H. Pflum* — 1627 – 1630

Phosphorylation-Dependent Kinase–Substrate Cross-Linking



Storage solutions: DNA is shown to be soluble in a variety of choline-based, hydrated ionic liquids (ILs; see picture). The IL-stored DNA molecules have exceptional long-term stability, in excess of one year.

DNA Solutions

R. Vijayaraghavan, A. Izgorodin,
V. Ganesh, M. Surianarayanan,
D. R. MacFarlane* ———— 1631 – 1633

Long-Term Structural and Chemical Stability of DNA in Hydrated Ionic Liquids

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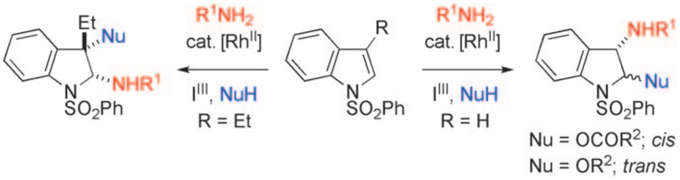


Twofold versatility: An efficient intermolecular oxyamidation of indoles relies on the application of rhodium(II)-catalyzed nitrene transfers. The *cis* or *trans* products are exclusively isolated in very good yields (up to 88 %) by changing the nucleophile. Control of the regioselectivity of the addition is also possible by modifying the substitution of the indole double bond (see scheme).

Synthetic Methods

S. Beaumont, V. Pons, P. Retailleau,
R. H. Dodd, P. Dauban* —— 1634 – 1637

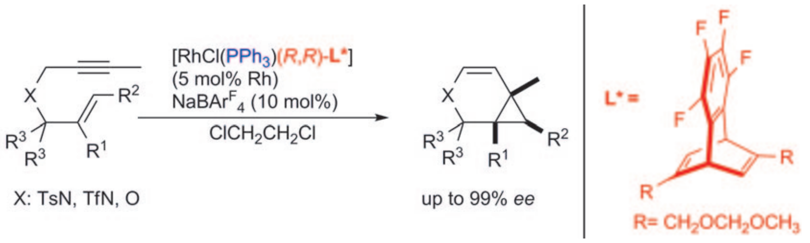
Catalytic Oxyamidation of Indoles

Contents

Asymmetric Catalysis

T. Nishimura,* T. Kawamoto,
M. Nagaosa, H. Kumamoto,
T. Hayashi* ______ **1638 – 1641**

Chiral Tetrafluorobenzobarrelene Ligands for the Rhodium-Catalyzed Asymmetric Cycloisomerization of Oxygen- and Nitrogen-Bridged 1,6-Enynes

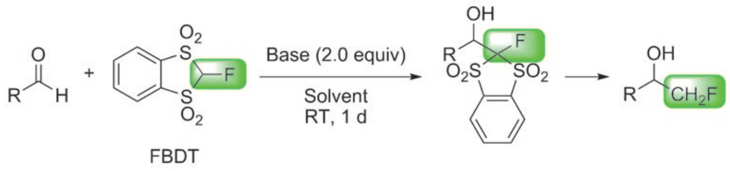


The long and winding Rhoad: The asymmetric cycloisomerization of nitrogen- and oxygen-bridged 1,6-enynes proceeded in the presence of a cationic rhodium complex that was coordinated by PPh$_3$ and a chiral diene ligand to afford high yields of chiral 3-aza- and 3-oxabicyclo-[4.1.0]heptenes and high enantioselectivities. TS = 4-toluenesulfonyl, Tf = trifluoromethanesulfonyl.

Fluoromethylation

T. Furukawa, Y. Goto, J. Kawazoe,
E. Tokunaga, S. Nakamura, Y. Yang, H. Du,
A. Kakehi, M. Shiro,
N. Shibata* ______ **1642 – 1647**

2-Fluoro-1,3-benzodithiole-1,1,3,3-tetraoxide: A Reagent for Nucleophilic Monofluoromethylation of Aldehydes

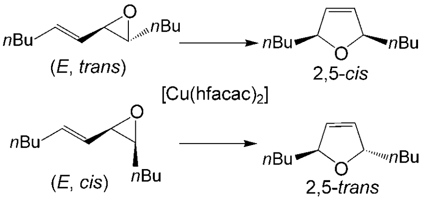


Matter of choice: The title compound (FBDT), a cyclic analogue of 1-fluorobis(phenylsulfonyl)methane (FBSM), was developed as a reagent for the nucleophilic monofluoromethylation of aldehydes. By choice of an appropriate base it is possible to achieve selective 1,2- or 1,4-addition of FBDT to conjugated aldehydes. The method was applied to the synthesis of a fluorinated isostere of osmundalactone.

Synthetic Methods

M. Brichacek, L. A. Batory,
J. T. Njardarson* ______ **1648 – 1651**

Stereoselective Ring Expansion of Vinyl Oxiranes: Mechanistic Insights and Natural Product Total Synthesis

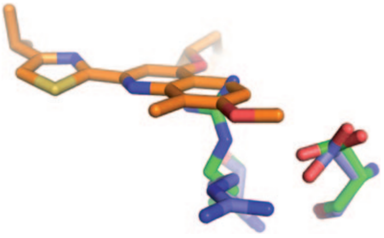


What a (strain) relief! The first broadly applicable, catalytic, and stereoselective vinyl oxirane ring expansion is described (see scheme; hfacac = hexafluoroacetylacetonate). The stereoselectivity was influenced by several reaction parameters, and kinetic studies support a mechanistic proposal involving the in situ formation of a more reactive catalytic species. This ring-expansion reaction has been employed in the asymmetric total synthesis of (+)-goniothalesdiol.

Enzyme Inhibitors

M. D. Cummings,* J. Lindberg, T.-I. Lin,
H. de Kock, O. Lenz, E. Lilja, S. Felländer,
V. Baraznenok, S. Nyström, M. Nilsson,
L. Vrang, M. Edlund, Å. Rosenquist,
B. Samuelsson, P. Raboisson,
K. Simmen ______ **1652 – 1655**

Induced-Fit Binding of the Macrocyclic Noncovalent Inhibitor TMC435 to its HCV NS3/NS4A Protease Target

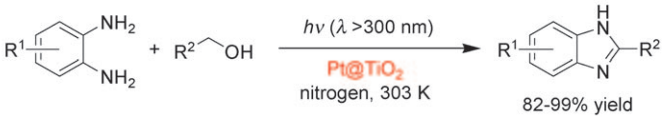


If the shoe fits: TMC435, a noncovalent small-molecule inhibitor of the hepatitis C virus (HCV) NS3/NS4A protease, is currently undergoing clinical evaluation as an HCV therapeutic. In the crystal structure of the noncovalent NS3/NS4A protease–TMC435 complex the bound inhibitor exploits induced-fit binding. The new structure is consistent with the emerging view of viral resistance to NS3/NS4A protease inhibitors.

Heterogeneous Catalysis

Y. Shiraishi,* Y. Sugano, S. Tanaka, T. Hirai ——— 1656 – 1660

One-Pot Synthesis of Benzimidazoles by Simultaneous Photocatalytic and Catalytic Reactions on Pt@TiO$_2$ Nanoparticles



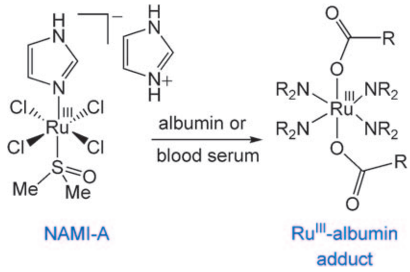
Dual platinum action: A one-pot catalytic synthesis of benzimidazoles from the photoirradiation of an alcohol solution containing an *ortho*-arylenediamine and Pt@TiO$_2$ nanoparticles is described. This reaction proceeded by the platinum-assisted photocatalytic oxidation of an alcohol and a catalytic dehydrogenation of the intermediates on the surface of platinum nanoparticles.

Antitumor Agents

M. Liu, Z. J. Lim, Y. Y. Gwee, A. Levina, P. A. Lay* ——— 1661 – 1664

Characterization of a Ruthenium(III)/NAMI-A Adduct with Bovine Serum Albumin that Exhibits a High Anti-Metastatic Activity

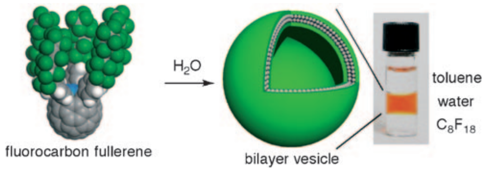
An exchange for the better: X-ray absorption spectroscopic studies show that treatment of an Ru^{III} drug, NAMI-A, with bovine serum albumin (BSA) results in complete replacement of the Cl$^-$ and S-donor ligands with N-donor and carboxylato residues of the protein (see scheme). The Ru^{III}–BSA adduct is as active in enhancing the cell–substrate interactions as the original NAMI-A complex (which converts into Ru^{III}–BSA within minutes in the cell-culture medium).



Artificial Vesicles

T. Homma, K. Harano, H. Isobe, E. Nakamura* ——— 1665 – 1668

Nanometer-Sized Fluorous Fullerene Vesicles in Water and on Solid Surfaces



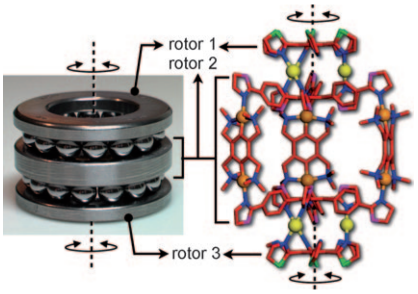
A new twist for bucky balls: A fluorous fullerene amphiphile lacking the conventional polar head / nonpolar tail motif forms a vesicle in water (see picture, F green, C gray), mediated by the cohesive power of fullerene. The fluorous moiety covers the vesicle surface, and vesicle solutions dissolve perfluorooctane. Unlike lipid vesicles, the fluorous vesicle is very robust and retains its spherical shape even on a solid substrate under high vacuum.

Molecular Rotors

S. Hiraoka,* Y. Hisanaga, M. Shiro, M. Shionoya* ——— 1669 – 1673

A Molecular Double Ball Bearing: An Ag^I–Pt^{II} Dodecanuclear Quadruple-Decker Complex with Three Rotors

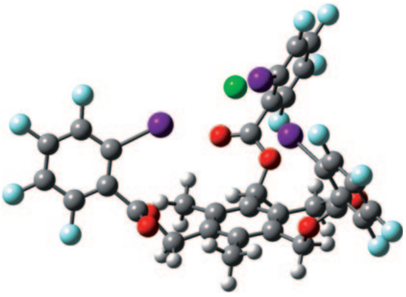
A marvelous machine: A 2.5 nm sized molecular machine that behaves like a double ball bearing with three rotors was constructed from two kinds of disk-shaped ligands, Ag^I ions, and dinuclear Pt^{II} bridging complexes (see structure: C red, N blue, O purple, S green, Ag yellow, Pt brown). Motion of the three rotors was found to depend greatly on the degree of interlocking between the two ligands in rotor 2 through helix inversion.



Anion Recognition

M. G. Sarwar, B. Dragisic, S. Sagoo,
M. S. Taylor* ______ **1674 – 1677**

A Tridentate Halogen-Bonding Receptor for Tight Binding of Halide Anions

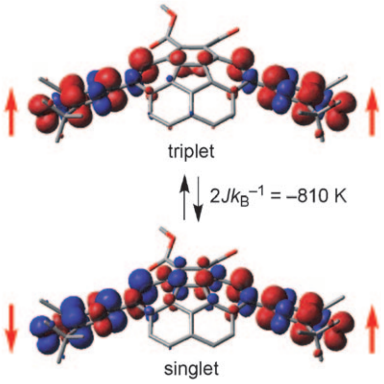


It's in the I's: Three suitably oriented electron-deficient iodoaryl groups form the basis for the first anion receptor (see structure; white H, gray C, red O, blue F, purple I, green Cl) that employs the halogen-bonding interaction alone to achieve high-affinity molecular recognition in dilute solution. The anion selectivity of this tridentate host differs from those of similar receptors based on hydrogen bonding.

Curved π Diradical

A. Ueda, S. Nishida, K. Fukui, T. Ise,
D. Shiomi, K. Sato, T. Takui,* K. Nakasuji,
Y. Morita* ______ **1678 – 1682**

Three-Dimensional Intramolecular Exchange Interaction in a Curved and Nonalternant π-Conjugated System: Corannulene with Two Phenoxyl Radicals



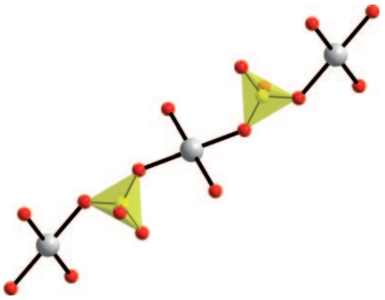
A neutral diradical based on corannulene with curved and nonalternant π conjugation has been synthesized and isolated as air-stable crystals. Sizable spin delocalization and strong intramolecular exchange interaction in the intrinsically three-dimensional π network (see picture), as well as significant diradical contributions in the ground state, were experimentally disclosed.

Silver(II) Compounds

P. J. Malinowski, M. Derzsi, Z. Mazej,
Z. Jagličić, B. Gaweł, W. Łasocha,
W. Grochala* ______ **1683 – 1686**

$Ag^{II}SO_4$: A Genuine Sulfate of Divalent Silver with Anomalously Strong One-Dimensional Antiferromagnetic Interactions

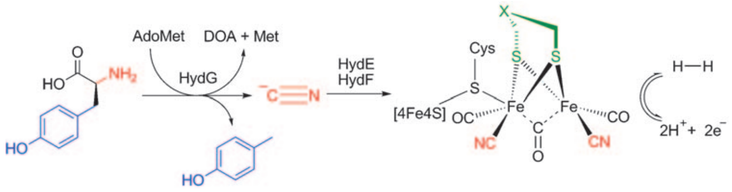
Black $AgSO_4$, synthesized for the first time by a displacement reaction, differs considerably from anhydrous sulfates of its Group 11 congeners: it has a very small electronic band gap (ca. 0.2 eV) and an anomalously strong one-dimensional antiferromagnetic ordering ($J \approx 10$ meV per Ag), which persists up to the onset of its thermal decomposition at about 120 °C. Ag gray, O red, S yellow.



Cyanide Biosynthesis

R. C. Driesener, M. R. Challand,
S. E. McGlynn, E. M. Shepard, E. S. Boyd,
J. B. Broderick, J. W. Peters,
P. L. Roach* ______ **1687 – 1690**

[FeFe]-Hydrogenase Cyanide Ligands Derived From *S*-Adenosylmethionine-Dependent Cleavage of Tyrosine



What's your poison? Hydrogenases catalyze the reversible formation of dihydrogen from two electrons and two protons. The maturation of the [FeFe]-hydrogenase active-site cofactor (H cluster) requires three gene products, HydE, HydF, and HydG. Cyanide has been characterized as one of the products of tyrosine cleavage by the *S*-adenosylmethionine-dependent enzyme HydG, clarifying its role in H-cluster biosynthesis. DOA = deoxyadenosine.

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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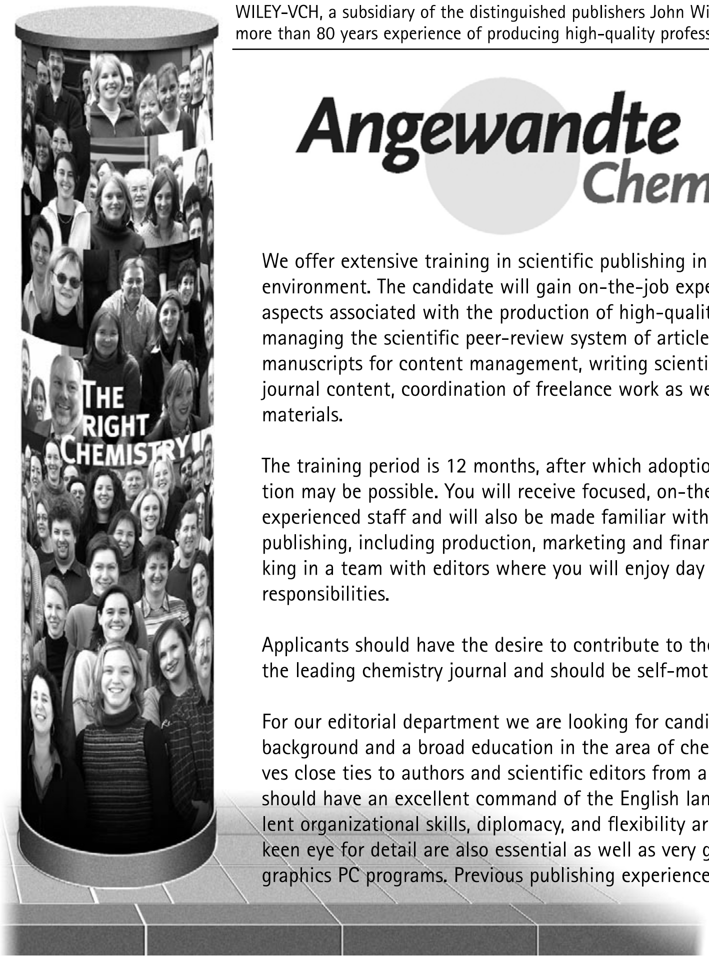

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