VIP 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:

*L. M. Fidalgo, G. Whyte, D. Bratton, C. F. Kaminski, C. Abell, W. T. S. Huck**
From Microdroplets to Microfluidics: Selective Emulsion Separation in Microfluidic Devices

I. Paterson, E. A. Anderson, S. M. Dalby, J. Ho Lim, J. Genovino, P. Maltas, C. Moessner*
Total Synthesis of Spirastrellolide A Methyl Ester. Part 1: Synthesis of an Advanced C17–C40 Bis(spiroacetal) Subunit

I. Paterson, E. A. Anderson, S. M. Dalby, J. Ho Lim, J. Genovino, P. Maltas, C. Moessner*
Total Synthesis of Spirastrellolide A Methyl Ester. Part 2: Subunit Union and Completion of the Synthesis

*X. Ning, J. Guo, M. A. Wolfert, G.-J. Boons**
Visualizing Metabolically Labeled Glycoconjugates of Living Cells by Copper-Free and Fast Huisgen Cycloadditions

S. Ghosh, A. Mukherjee, P. J. Sadler, S. Verma**
Periodic Iron Nanomineralization in Human Serum Transferrin Fibrils

M. Murata, Y. Ochi, F. Tanabe, K. Komatsu, Y. Murata**
Internal Magnetic Fields of Dianions of Fullerene and Its Cage-Opened Derivatives Studied with Encapsulated H_2 as an NMR Spectroscopic Probe

Obituary

Books

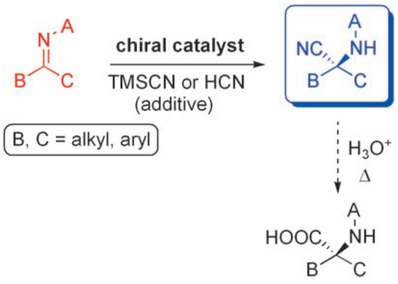
Highlights

Strecker Reaction

S. J. Connon* ________ **1176–1178**

The Catalytic Asymmetric Strecker Reaction: Ketimines Continue to Join the Fold

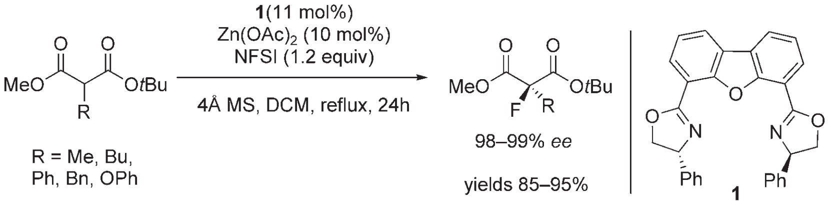
Picking up the gauntlet: The challenge of developing highly effective catalyst systems for the enantioselective Strecker reaction to convert ketimine substrates (see scheme, red; A denotes protecting group) into α-amino nitriles (blue) and ultimately α-amino acids has been met by several recent developments.



Asymmetric Fluorination

V. A. Brunet, D. O'Hagan* ________ **1179–1182**

Catalytic Asymmetric Fluorination Comes of Age



Gimme an F: Ever since the first asymmetric fluorination reagents were reported in 1988, the enantioselective introduction of a C–F bond at a stereogenic center has emerged as a major objective in organic chemistry. Newly published results on the enantioselective fluorination of malonates (see scheme) are put into context in this Highlight. NFSI = *N*-fluorodibenzenesulfonimide.

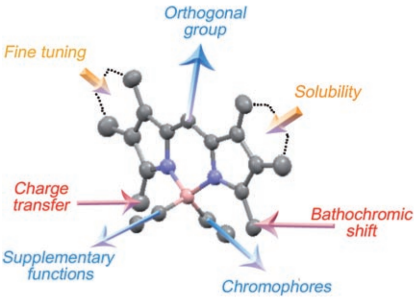
Contents

Minireviews

Fluorescent Molecular Devices

G. Ulrich, R. Ziessel,*
A. Harriman ———— **1184–1201**

The Chemistry of Fluorescent Bodipy Dyes: Versatility Unsurpassed



Unexpected adaptability: The properties of dipyrrometheneboron difluoride (F-Bodipy) dyes can be modified in many ways (see picture, B pink, N blue, C gray). This versatility has allowed new uses to be developed for this molecule. Emphasis is given to synthetic considerations, optical properties, and applications as chemical sensors, luminescent devices, and molecular materials.

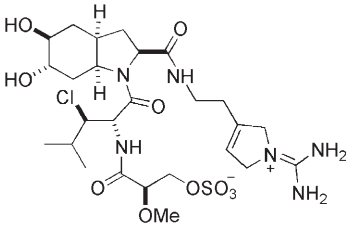
Reviews

Serine Protease Inhibitors

K. Ersmark, J. R. Del Valle,
S. Hanessian* ———— **1202–1223**

Chemistry and Biology of the Aeruginosin Family of Serine Protease Inhibitors

Oceans apart: With sources ranging from the marine sponges in North Queensland, Australia, to the cyanobacterial water-blooms of Lake Kasumigaura, Japan, and the blue-green algae found in Hula Valley, Israel, the aeruginosin family of linear peptides exhibit potent inhibition against serine proteases such as the medically relevant thrombin, as well as other proteins involved in the blood coagulation cascade.



Chlorodysinosin A

Communications

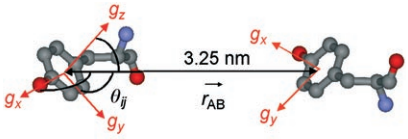
VIP

High-Field Pulsed EPR Spectroscopy

V. P. Denysenkov, D. Biglino, W. Lubitz,
T. F. Prisner, M. Bennati* —— **1224–1227**

Structure of the Tyrosyl Biradical in Mouse R2 Ribonucleotide Reductase from High-Field PELDOR

Paramagnetic centers rigidly embedded in proteins serve as a probe in structural studies of macromolecular complexes. Pulse EPR at high frequencies allows the determination of not only the distance but also the relative orientation of these centers (see tyrosyl radicals; C gray, O red, N blue). The method has considerable potential for studying the assembly of protein complexes.



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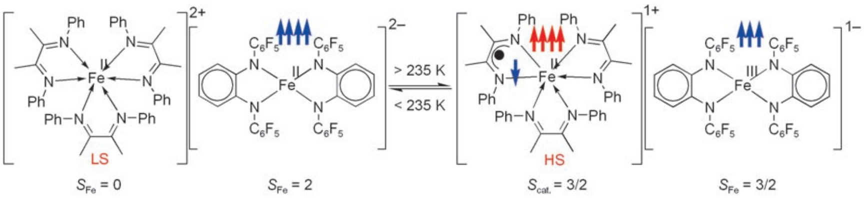
Spin Crossover

M. M. Khusniyarov,* T. Weyhermüller,
E. Bill, K. Wieghardt* ______ 1228 – 1231

Reversible Electron Transfer Coupled to Spin Crossover in an Iron Coordination Salt in the Solid State

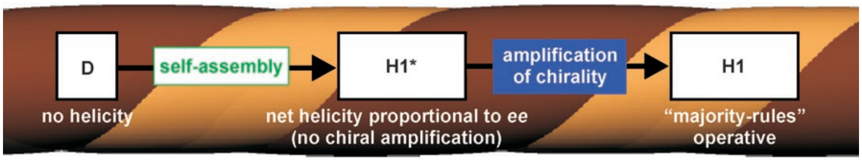


A rad. dad: The electronic structure of [Fe(dad)$_3$][Fe(pda)$_2$] was investigated by X-ray crystallography, Mössbauer spectroscopy, and magnetic susceptibility measurements as a function of the temperature. Reversible one-electron transfer from the dianion (high-spin Fe^{II}) to the dication (low-spin Fe^{II}) generates a monoanion (intermediate-spin Fe^{III}) and a monocation (high-spin Fe^{II} and α-diimine π radical anion) above 235 K.

Chiral Amplification

A. Lohr, F. Würthner* ______ 1232 – 1236

Evolution of Homochiral Helical Dye Assemblies: Involvement of Autocatalysis in the “Majority-Rules” Effect

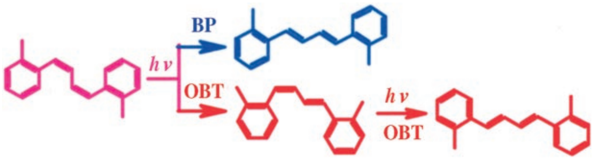


Chiral amplification in action: Kinetic studies on the self-assembly of bis(merocyanine) dyes reveal that two phenomena that have been suggested to be of importance for natural homochirogenesis, namely, autocatalysis and the “majority-rules” effect, are both involved in the evolution of homochiral helical dye nanorods (see picture, **D** = supramolecular oligomer species; **H1** = kinetically self-assembled nanorods; **H1*** = **H1**-type aggregate precursors).

Photoisomerization

J. Saltiel,* M. A. Bremer,
S. Laohhasurayotin,
T. S. R. Krishna ______ 1237 – 1240

Photoisomerization of *cis,cis*- and *cis,trans*-1,4-Di-*o*-tolyl-1,3-butadiene in Glassy Media at 77 K: One-Bond-Twist and Bicycle-Pedal Mechanisms

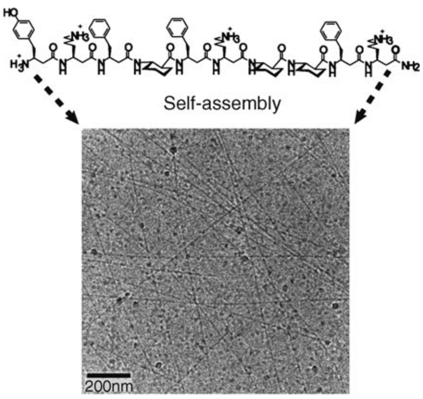


Let’s twist again: In the isomerization of *cis,cis*-1,4-di-*o*-tolyl-1,3-butadiene in isopentane glass at 77 K, hula-twist pathways are eliminated, because unstable photoproduct conformers do not form. Instead, the isomerization is found to proceed by bicycle-pedal (BP) and one-bond-twist (OBT) mechanisms (see scheme).

Nanostructured Materials

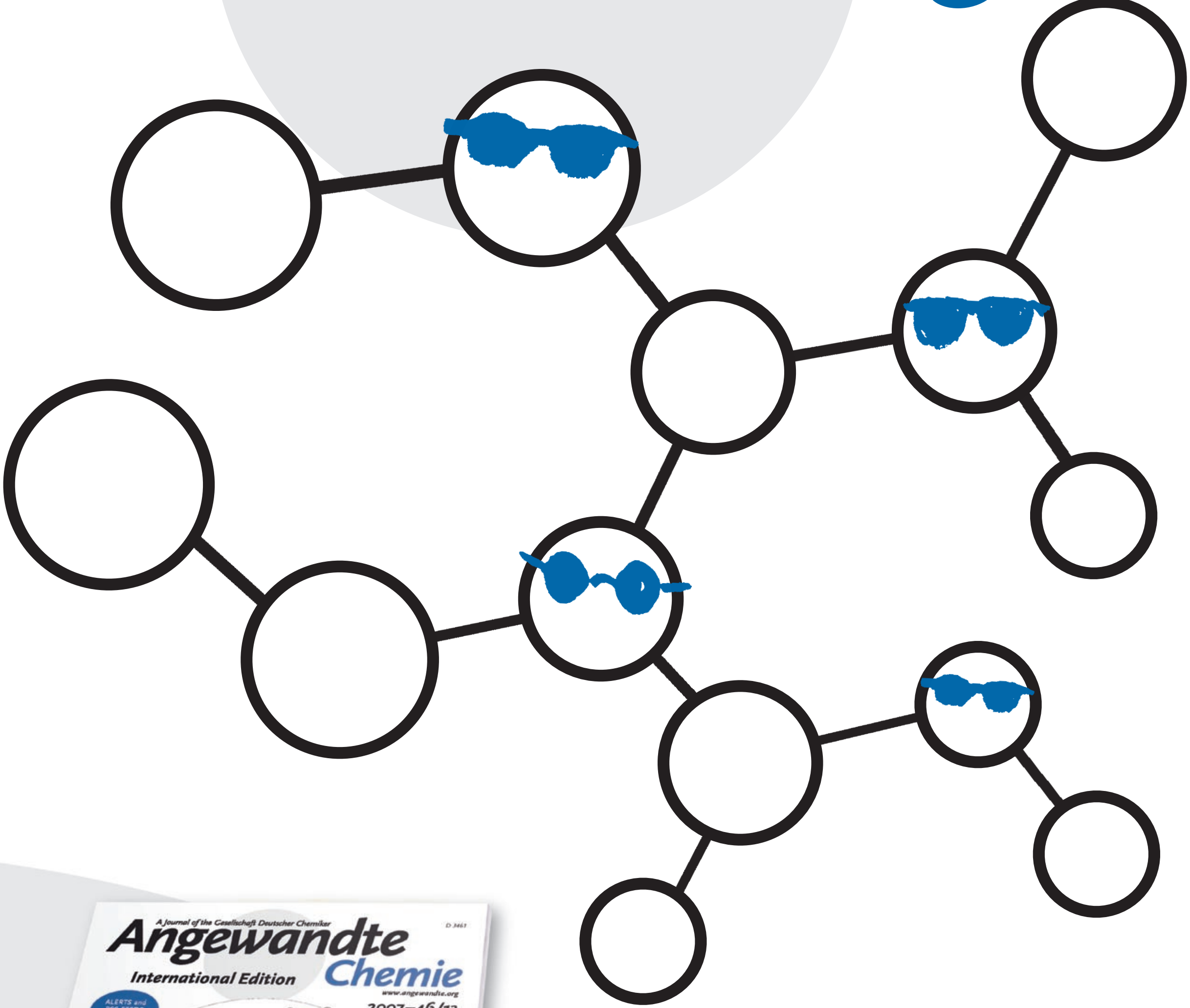
W. C. Pomerantz, V. M. Yuwono,
C. L. Pizzey, J. D. Hartgerink,*
N. L. Abbott,*
S. H. Gellman* ______ 1241 – 1244

Nanofibers and Lyotropic Liquid Crystals from a Class of Self-Assembling β-Peptides



More than a pretty phase: A class of helical β-peptides has been discovered that form lyotropic liquid-crystalline (LC) phases in water. The β-peptide sequence strongly affects the mode of assembly and ultimately whether a LC phase is formed. For a non-globally amphiphilic β-peptide that can form a LC phase, cryo-TEM revealed thin fibers several micrometers in length, consistent with Onsager theory.

Incredibly incognito!



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2007–46/13

1st April Issue

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Controlling Magnetic Properties
O. Sato et al.

Natural Product (Bio)synthesis
H. B. Bode and R. Müller

C-H and C-C Bond Activation

ACIEFS 46 (13) 2121–2332 (2007) · ISSN 1433-7851 · Vol. 46 · No. 13

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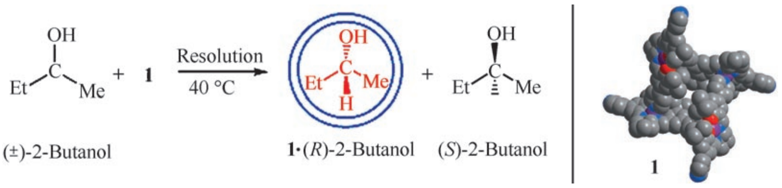
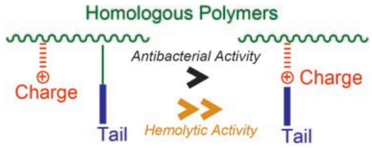
Porous Assemblies



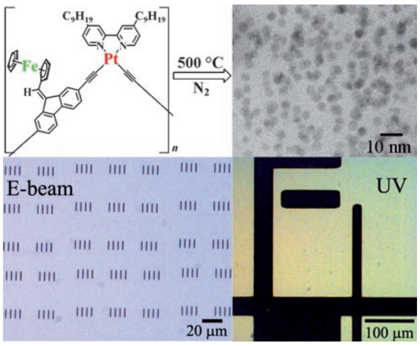
Only half may enter: A homochiral porous nanoscale metallacycle has been efficiently self-assembled from semiflexible enantiopure metallosalen complexes with complementary coordination motifs. Single crystals of the macromolecule (see structure; C gray, Zn purple, O red, N blue) show a reversible and controllable dynamic behavior. Particularly, the metallacycle can be used to resolve small racemic alcohols with high enantioselectivity (over 99.0% *ee*).

Antimicrobial Polymers



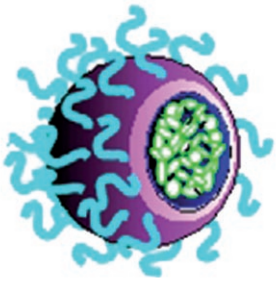
The benefits of separation: For homologous polymers having similar backbone composition and charge/tail ratios, placing the charge and alkyl tail on spatially separated centers results in a higher membrane-disrupting ability, as evident from the increased antibacterial and hemolytic activities. The spatial separation particularly amplifies mammalian cell toxicity (see picture).

Metallopolymers



Micropatterns of arrays of ferromagnetic face-centered tetragonal FePt nanoparticles can be fabricated from thin films of a novel air- and moisture-stable bimetallic polyferroplatinyne precursor, which can be utilized directly as a negative resist in both electron-beam lithography and UV photolithography.

Gene Delivery



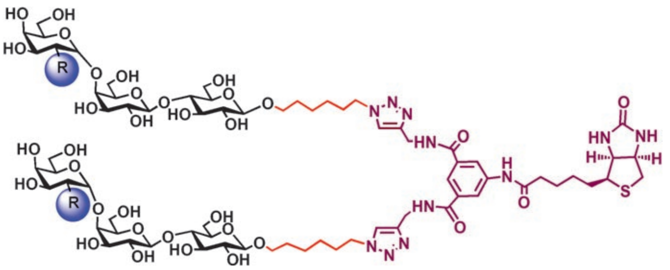
A new concept for fabricating nanocarriers carrying free DNA for gene delivery to overcome the intracellular dissociation barrier of cationic polymer/DNA complexes is presented. Free DNA plasmids (shown in green) are encapsulated in a nanocapsule core, which is protected by a hydrophobic membrane (purple) with a poly(ethylene glycol) outer layer (blue). The nanocapsules can release free DNA into the cells and have high in vitro and in vivo transfection efficiency.

Contents

Glycoconjugates

R. R. Kale, C. M. McGannon, C. Fuller-Schaefer, D. M. Hatch, M. J. Flagler, S. D. Gamage, A. A. Weiss,* S. S. Iyer* ______ **1265 – 1268**

Differentiation between Structurally Homologous Shiga 1 and Shiga 2 Toxins by Using Synthetic Glycoconjugates

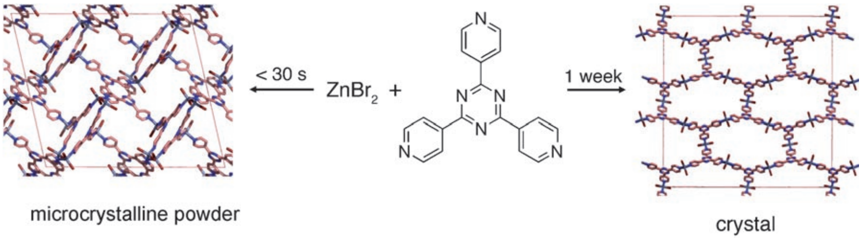


Choose your poison: Shiga toxins 1 and 2 are the major virulence factors of *E. coli* O157:H7. However, Shiga 2 is more potent than Shiga 1. Biotinylated glycoconjugates have been developed that differentiate between these structurally homologous toxins (see picture; R = OH: selective for Shiga 1; R = NHAc: selective for Shiga 2). These synthetic materials efficiently capture toxins without interference from the sample matrix.

Porous Networks

M. Kawano,* T. Haneda, D. Hashizume, F. Izumi, M. Fujita* ______ **1269 – 1271**

A Selective Instant Synthesis of a Coordination Network and Its Ab Initio Powder Structure Determination

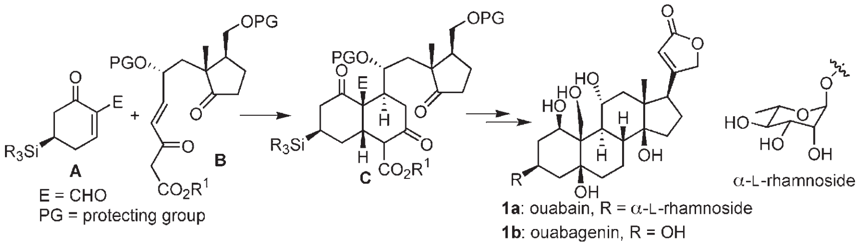


Fast powders and slow crystals: A uniform powder sample of a flexible porous coordination network is prepared in high yield by kinetically controlled synthesis and its crystal structure solved by synchrotron ab initio powder X-ray diffraction (see picture). Crystals of a totally different porous network with larger channels for guest encapsulation are also obtained by thermodynamic control in a slow crystallization process.

Steroid Synthesis

H. Zhang, M. Sridhar Reddy, S. Phoenix, P. Deslongchamps* ______ **1272 – 1275**

Total Synthesis of Ouabagenin and Ouabain

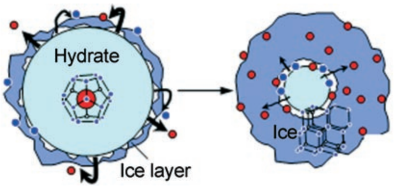


The highly oxygenated steroid ouabagenin (**1b**) and its glycoside ouabain (**1a**) were prepared by a strategy based on a polyanionic cyclization. Starting building blocks **A** and **B** were combined to give the key intermediate **C** and transformed into **1b** in 27 steps. Finally, ouabagenin (**1b**) was converted into ouabain (**1a**) in six steps (see scheme).

Gas Hydrates

S. Takeya, J. A. Ripmeester* **1276 – 1279**

Dissociation Behavior of Clathrate Hydrates to Ice and Dependence on Guest Molecules

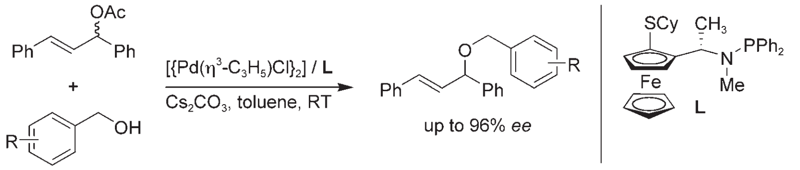


Self-preservation: The existence of gas hydrates (see picture) outside their stability zone for prolonged periods is shown to depend on the interaction strength between guest molecules (red) and water (blue), as reflected by the dissociation pressures at 273 K.

Asymmetric Catalysis

F. L. Lam, T. T.-L. Au-Yeung, F. Y. Kwong, Z. Zhou, K. Y. Wong, A. S. C. Chan* ______ 1280 – 1283

Palladium–(S_{p},R)-FerroNPS-Catalyzed Asymmetric Allylic Etherification: Electronic Effect of Nonconjugated Substituents on Benzylic Alcohols on Enantioselectivity

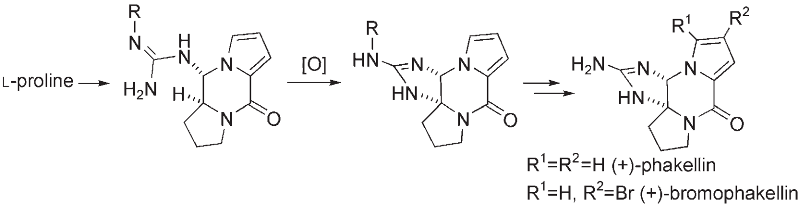


An enantioselective C—O bond-forming reaction proceeds well under palladium catalysis with newly developed N–P,S ligands with a ferrocene motif (see scheme). Nonconjugated substituents on the benzylic alcohol substrate lead to an increase in enantioselectivity with increasing electron-donating ability in the title reaction with racemic 1,3-diphenyl-2-propenyl acetate. Cy = cyclohexyl.

Alkaloid Synthesis

S. Wang, D. Romo* ______ 1284 – 1286

Enantioselective Synthesis of (+)-Monobromophakellin and (+)-Phakellin: A Concise Phakellin Annulation Strategy Applicable to Palau'amine



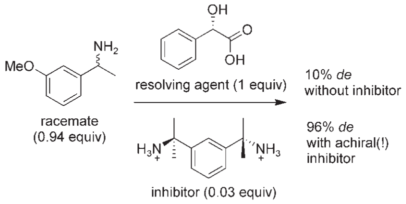
A unique oxidative cyclization of a tricycle bearing a guanidine aminal provides a concise route to the enantioselective synthesis of (+)-phakellin and (+)-monobromophakellin in nine and ten steps, respectively, starting from L-proline (see scheme). This sequence provides a simple annulation strategy applicable to the preparation of more complex members of this family of marine sponge-derived alkaloids including palau'amine.

Resolution of Diastereomers

M. Leeman, G. Brasile, E. Gelens, T. Vries, B. Kaptein, R. Kellogg* ____ 1287 – 1290

Structural Aspects of Nucleation Inhibitors for Diastereomeric Resolutions and the Relationship to Dutch Resolution

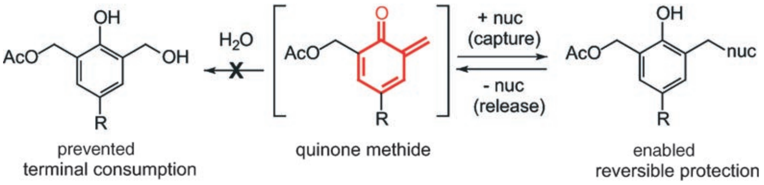
You say you want a resolution: Nucleation inhibitors for the resolution of diastereomers by selective crystallization have been designed and tested. The resolution of racemic 3-methoxyphenylethylamine was optimized with (*S*)-mandelic acid as the resolving agent (see scheme). Multifunctional inhibitors are particularly effective.



Quinone Methides

H. Wang, M. S. Wahi, S. E. Rokita* ______ 1291 – 1293

Immortalizing a Transient Electrophile for DNA Cross-Linking

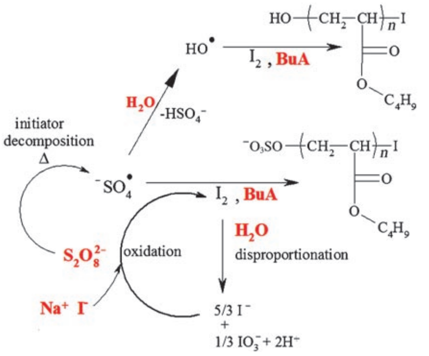


Who wants to live forever? Compounds that act as strong nucleophiles and good leaving groups repeatedly capture and release a transient quinone methide to extend its effective lifetime under aqueous conditions and promote DNA cross-linking (see scheme). This finding may expand the biological and therapeutic activity of quinone methides.

Living Radical Polymerization

J. Tonnar,
P. Lacroix-Desmazes* ______ **1294 – 1297**

Use of Sodium Iodide as the Precursor to the Control Agent in Ab Initio Emulsion Polymerization

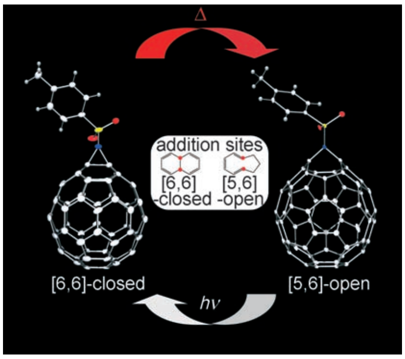


Iodide does the job: Water-soluble, harmless, cheap, and nonhazardous NaI was used in combination with $K_2S_2O_8$ instead of I_2 itself to produce an uncolored living poly(butyl acrylate) latex of controlled molecular weight by a single-step polymerization process (see scheme for one polymerization pathway). Reactivation of the polymer then yielded a block-copolymer latex. BuA = butyl acrylate.

Fullerene Chemistry

T. Nakahodo, M. Okada, H. Morita,
T. Yoshimura, M. O. Ishitsuka, T. Tsuchiya,
Y. Maeda, H. Fujihara, T. Akasaka,* X. Gao,
S. Nagase* ______ **1298 – 1300**

[2+1] Cycloaddition of Nitrene onto C_{60} Revisited: Interconversion between an Aziridinofullerene and an Azafulleroid

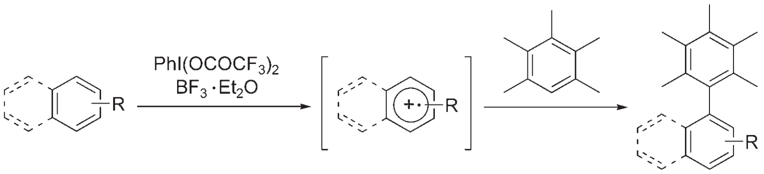


An open and closed case: The reversible interconversion of a [1,2]aziridinofullerene and a [1,6]azafulleroid represents the first such example for a monosubstituted fullerene. While the [1,2]aziridinofullerene rearranges thermally to the [1,6]azafulleroid, the [1,6]azafulleroid rearranges photochemically to the [1,2]aziridinofullerene in high yield (see picture).

Cross-Coupling

T. Dohi, M. Ito, K. Morimoto, M. Iwata,
Y. Kita* ______ **1301 – 1304**

Oxidative Cross-Coupling of Arenes Induced by Single-Electron Transfer Leading to Biaryls by Use of Organoiodine(III) Oxidants



Cross-coupling goes green: The direct oxidative cross-coupling reaction of naphthalenes and other electron-rich arenes with mesitylenes has been achieved in high yields using hypervalent iodine(III) reagents. The key for reaction success is the exclusive generation of cation radical intermediates of naphthalenes and the use of mesitylenes as the reactive and less dimerizable nucleophiles. R = alkyl, aryl, halogen, ester, alkoxy, etc.

Homogeneous Catalysis

C. M. Rao Volla, P. Vogel* __ **1305 – 1307**

Iron-Catalyzed Desulfinylative C–C Cross-Coupling Reactions of Sulfonyl Chlorides with Grignard Reagents

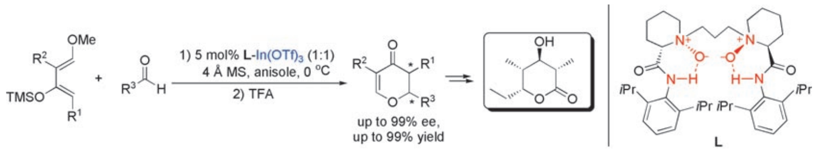
$$\text{R-SO}_2\text{Cl} + \text{R'MgX} \xrightarrow[\text{THF/NMP, 80 °C}]{[\text{Fe(acac)}_3]\text{ (cat.)}} \text{R-R'} + \text{SO}_2\uparrow + \text{Mg(Cl)X}$$

A friendly couple: Conditions have been uncovered that allow the desulfinylative C–C cross-coupling reaction of inexpensive sulfonyl chlorides and Grignard reagents (see scheme, acac = acetylacetonate, NMP = *N*-methylpyrrolidone). The reactions rely on environmentally friendly iron catalysts and do not require expensive and/or toxic ligands.

Asymmetric Catalysis

Z. Yu, X. Liu, Z. Dong, M. Xie, X. Feng* ——— 1308 – 1311

An N,N'-Dioxide/In(OTf)$_3$ Catalyst for the Asymmetric Hetero-Diels–Alder Reaction Between Danishefsky's Dienes and Aldehydes: Application in the Total Synthesis of Triketide

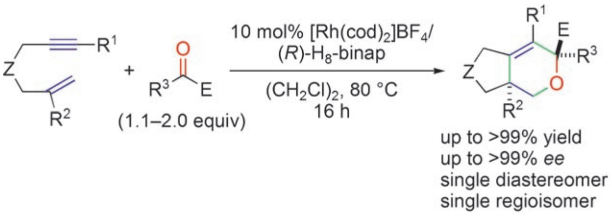


In-teresting catalyst: An asymmetric hetero Diels–Alder reaction between Danishefsky's dienes and various aldehydes using an N,N'-dioxide/In(OTf)$_3$ complex affords highly substituted chiral dihydropyranones with up to 99% *ee* (see scheme). The catalyst was applied to the synthesis of triketide from propionaldehyde in 72% yield and with 97% *ee*.

Asymmetric Catalysis

K. Tanaka,* Y. Otake, H. Sagae, K. Noguchi, M. Hirano ——— 1312 – 1316

Highly Regio-, Diastereo-, and Enantioselective [2+2+2] Cycloaddition of 1,6-Enynes with Electron-Deficient Ketones Catalyzed by a Cationic Rh^I/H$_8$-binap Complex

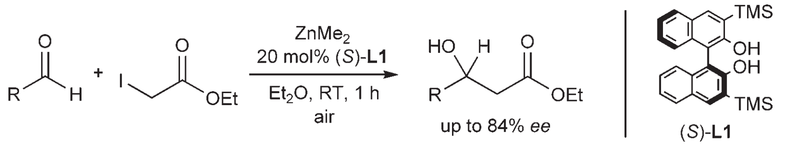


Two quaternary carbon centers are created in the title reaction to form fused dihydropyrans (see scheme). The same catalyst promotes the *ortho* functionalization of aryl ketones with 1,6-enynes with excellent regio- and enantioselectivity. Z = amide, C(CO$_2$Me)$_2$, O; E = CO$_2$Et, Ac; R^1 = Me, aryl, CO$_2$Me; R^2 = Me; R^3 = Me, CO$_2$Et.

Asymmetric Catalysis

M. Á. Fernández-Ibáñez, B. Maciá, A. J. Minnaard, B. L. Feringa* ——— 1317 – 1319

Catalytic Enantioselective Reformatsky Reaction with Aldehydes

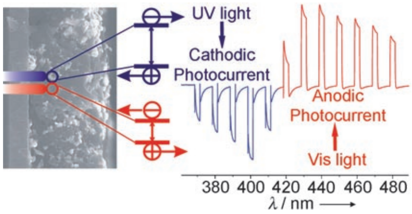


120 years after the discovery of the Reformatsky reaction, the first effective catalytic enantioselective Reformatsky reaction with aldehydes using a binol derivative as a chiral catalyst is presented (see scheme; TMS = trimethylsilyl). The reaction is performed with ethyl iodoacetate and Me$_2$Zn. The presence of air is found to be crucial to reach high conversions and selectivities.

Photoelectrochemistry

R. Beranek, H. Kisch* ——— 1320 – 1322

A Hybrid Semiconductor Electrode for Wavelength-Controlled Switching of the Photocurrent Direction



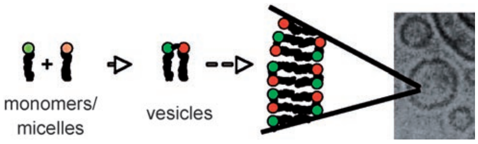
A light switch: A nanocrystalline hybrid photoelectrode consisting of nitrogen-modified titanium dioxide (an n-type semiconductor) and copper(I) iodide (a p-type semiconductor) on conducting glass shows wavelength-controlled switching of the photocurrent in the narrow range from 410 to 420 nm (see picture).

Contents

Membranes for Artificial Cells

H. H. Zepik,* P. Walde,*
T. Ishikawa **1323 – 1325**

Vesicle Formation from Reactive Surfactants



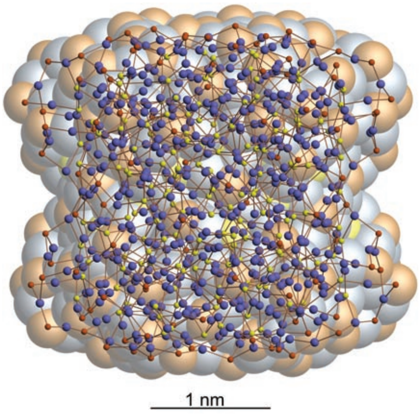
Zwitterionic gemini vesicles: Alkylphosphothioates and *N*-(2-bromoethyl)-*N*,*N*-dimethylalkylammonium surfactants react quantitatively in aqueous buffer to form zwitterionic gemini surfactants. The reaction products transform spontaneously into lipid vesicles that behave similarly to phospholipid vesicles. The zwitterionic gemini vesicles encapsulate water-soluble molecules; when fed with the single-chain precursors, they grow quantitatively.

Cluster Compounds

C. E. Anson, A. Eichhöfer, I. Issac,
D. Fenske,* O. Fuhr, P. Sevillano,
C. Persau, D. Stalke,
J. Zhang **1326 – 1331**

Synthesis and Crystal Structures of the Ligand-Stabilized Silver Chalcogenide Clusters $[Ag_{154}Se_{77}(dppxy)_{18}]$, $[Ag_{320}(S\textit{t}Bu)_{60}S_{130}(dppp)_{12}]$, $[Ag_{352}S_{128}(S\textit{t}C_5H_{11})_{96}]$, and $[Ag_{490}S_{188}(S\textit{t}C_5H_{11})_{114}]$

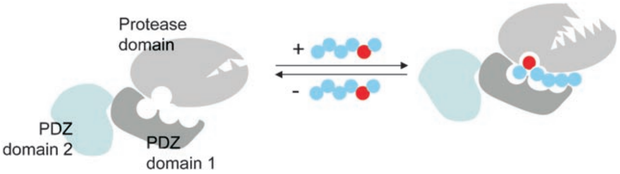
The reaction of silver thiolates with $Se(SiMe_3)_2$ or $S(SiMe_3)_2$ in the presence of bidentate phosphanes leads to the formation of very large cluster molecules with distorted spherical metal–chalcogenide cores with diameters of 2–4 nm. The surfaces of these cores are protected by thiolate and phosphane ligands. Structural analyses show a higher degree of disorder with increasing number of Ag atoms. The compound with the highest silver content (see figure) has an idealized formulation $[Ag_{490}S_{188}(S\textit{t}C_5H_{11})_{114}]$.



Allosteric Protease Regulation

M. Meltzer, S. Hasenbein, P. Hauske,
N. Kucz, M. Merdanovic, S. Grau, A. Beil,
D. Jones, T. Krojer, T. Clausen,
M. Ehrmann,* M. Kaiser* **1332 – 1334**

Allosteric Activation of HtrA Protease DegP by Stress Signals during Bacterial Protein Quality Control



Unleashing the proteolytic activity of DegP: A study with synthetic mimics of cellular stress signals reveals an allosteric (chemical) activation mechanism of the bacterial HtrA protease DegP, leading to a fine-tuned amplification of protein degradation during bacterial protein quality control (see scheme: binding of a peptide sequence (circles) increases activation. The nature of the penultimate residue (red) is shown to be decisive).

Supporting information is available on the WWW (see article for access details).

A video clip is available as Supporting Information on the WWW (see article for access details).

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Service

Corrigendum

There was a small error in the second sentence on page 8576 of this Communication. This sentence should read: "The ratio of intensities of acetylene to aromatic peaks was calculated using variable-contact-time ^{1}H–^{13}C CP/MAS NMR spectra with the following results: CMP-1 0.27 (expected value 0.40); CMP-2 0.18 (expected value 0.25); CMP-3 0.10 (expected value 0.18)."

The authors apologize for this error and wish to note that none of the interpretations in the paper are affected by this change.

Conjugated Microporous Poly(aryleneethynylene) Networks

J.-X. Jiang, F. Su, A. Trewin, C. D. Wood, N. L. Campbell, H. Niu, C. Dickinson, A. Y. Ganin, M. J. Rosseinsky, Y. Z. Khimyak, A. I. Cooper* **8574–8578**

Angew. Chem. Int. Ed. **2007**, *46*

DOI 10.1002/anie.200701595

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