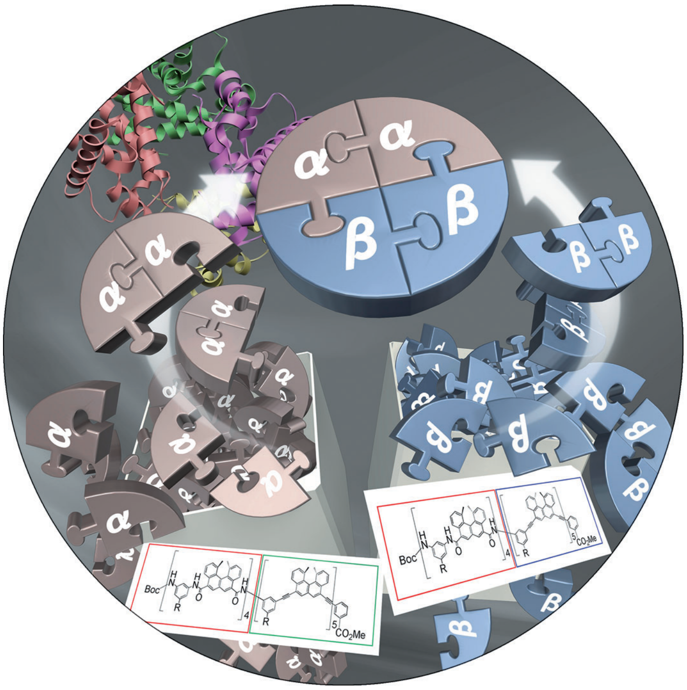
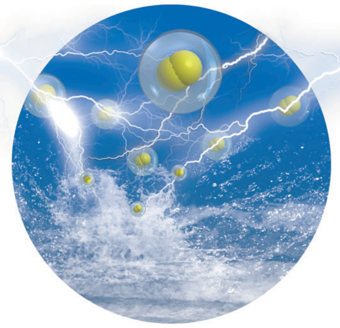
Tetrameric aggregates with ...

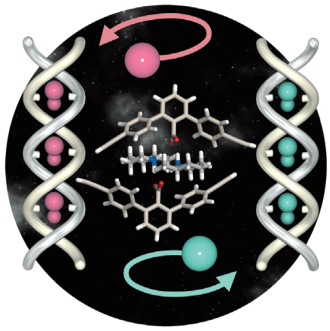


... an ααββ structure are formed by a method using stereoisomeric bidomain oligomers for the α- and β-subunits, as described in the Communication on page 5290 ff. by M. Yamaguchi et al.. Mixing the dimeric αα and ββ homoaggregates gave tetrameric ααββ heteroaggregates without formation of higher and lower aggregates. The tetrameric aggregate reversibly changed its structure between the dimeric homoaggregates on heating and cooling, and gelation occurred in toluene by polymerization.

Electrochemistry

In their Communication on page 5248 ff., X. B. Zhang and co-workers report a 3D nickel foam/porous carbon/anodized nickel electrode that was designed for the oxygen evolution reaction.



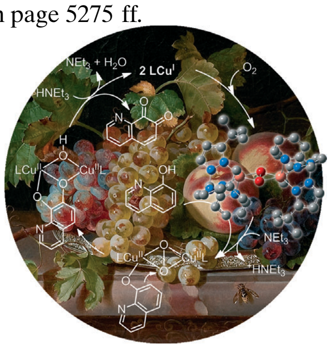


Helical Chirality

A π-conjugated polymer forms a unique one-handed double helix in the presence of chiral amines, which are sandwiched by the polymer strands, as described by E. Yashima et al. in their Communication on page 5275 ff.

Enzyme Model

In their Communication on page 5398 ff., S. Herres-Pawlis, T. D. P. Stack, and co-workers describe a copper complex that can hydroxylate various phenols by using dioxygen. The mechanism is analogous to that of the tyrosinase-catalyzed reaction.



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Service

Author Profile

"My biggest motivation is curiosity. Guaranteed to make me laugh are Charlie Chaplin movies …"
This and more about Jun-An Ma can be found on page 5212.

News

D. R. Walt

M. Behrens

F. Hollmann

H. Dietz

Books

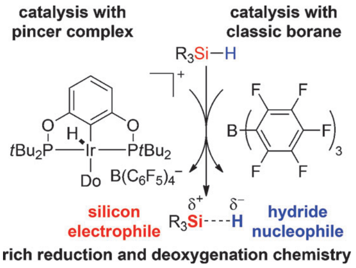
Highlights

Lewis Acid Catalysis

T. Robert, M. Oestreich* ___ **5216 – 5218**

Si–H Bond Activation: Bridging Lewis Acid Catalysis with Brookhart's Iridium(III) Pincer Complex and $B(C_6F_5)_3$

Different but almost equal: The striking similarities between Brookhart's iridium(III) pincer complex and the electron-deficient tris(pentafluorophenyl)borane in transformations involving Si–H bond activation are highlighted. Coordination of the Si–H bond to either Lewis acid enhances the electrophilicity of the silicon atom, thereby enabling its transfer to various Lewis basic groups (see scheme, Do = acetone).



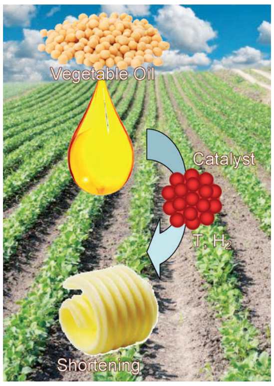
Essays

Vegetable Oil Hydrogenation

A. Philippaerts,* P. A. Jacobs, B. F. Sels ___ **5220 – 5226**

Is there still a Future for Hydrogenated Vegetable Oils?

For over a century, catalytic hydrogenation of vegetable oils has been an important process in the food industry, but today, it has lost much of its former popularity, because of the detrimental *trans* fatty acids as side products. However, the design of novel hydrogenation catalysts and the development of new strategies to remove *trans* isomers enable the production of *trans*-free hydrogenated fat products with desirable functionality.

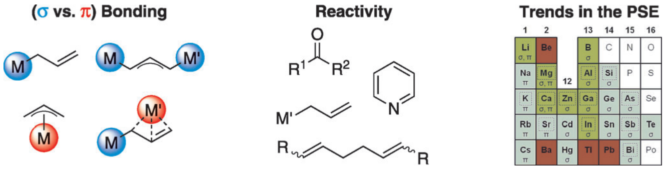


Reviews

Main Group Organometallic Chemistry

C. Lichtenberg, J. Okuda* ___ **5228 – 5246**

Structurally Defined Allyl Compounds of Main Group Metals: Coordination and Reactivity



No longer an aside: The allyl chemistry of main group metals was traditionally less investigated, but there is now a fundamental understanding of metal–allyl interactions. Furthermore, reactivity trends and new allyl-specific reaction patterns have been identified. In this Review, the coordination modes and reactivity of allyl compounds of main group metals as well as trends in the periodic system of the elements (PSE) are discussed.

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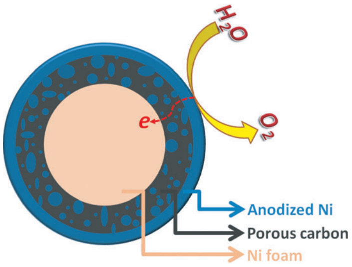
Communications



Oxygen evolution: A 3D nickel foam/porous carbon/anodized nickel electrode was designed for the oxygen evolution reaction (see picture). The conductive porous carbon membrane, which is derived from a zeolite imidazolate framework, plays a key role as an interlayer to both protect the inner instable Ni foam and support the outermost oxygen-evolving Ni catalyst layer.

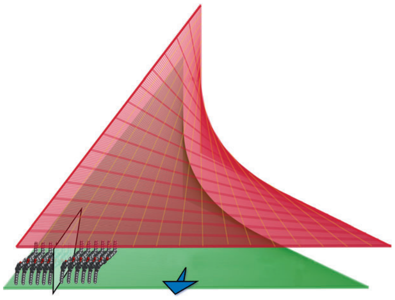
Electrochemistry

J. Wang, H. X. Zhong, Y. L. Qin,
X. B. Zhang* ______ **5248 – 5253**

An Efficient Three-Dimensional Oxygen Evolution Electrode

Frontispiece

A new liquid crystal phase, denoted modulated helical nanofilament ($HNF_{(mod)}$), is formed from a very simple class of biphenyl carboxylates lacking the benzylidene aniline moieties typically found in HNF mesogens. The $HNF_{(mod)}$ phase represents a novel kind of nanoparticle possessing stacked aromatic rings, with potential applications in organic electronics.

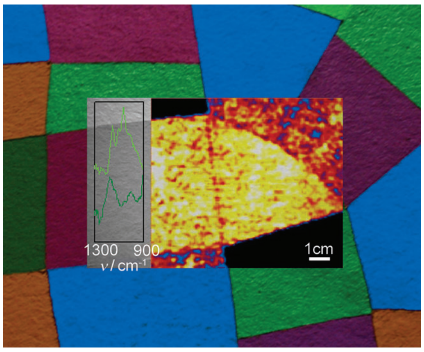


Liquid Crystals

E. Tsai, J. M. Richardson, E. Korblova,
M. Nakata, D. Chen, Y. Shen, R. Shao,
N. A. Clark, D. M. Walba* __ **5254 – 5257**

A Modulated Helical Nanofilament Phase

Cultural detective work: Remote hyperspectral imaging in the mid-infrared region enables the identification and localization of the painting materials used by artists (see brightness temperature difference image overlayed with the corresponding portion of the painting *Sestante 10* by Alberto Burri and IR reflection spectra for this area). The resulting molecular images are thus of great value for art conservation.



Molecular Imaging of Paintings

F. Rosi, C. Miliani,* R. Braun, R. Harig,
D. Sali, B. G. Brunetti,
A. Sgamellotti ______ **5258 – 5261**

Noninvasive Analysis of Paintings by Mid-infrared Hyperspectral Imaging

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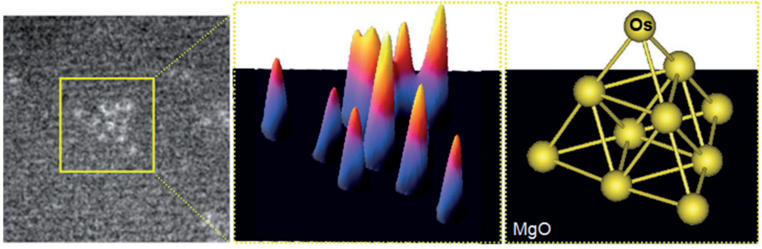


Size, shape, nuclearity: Aberration-corrected scanning transmission electron microscopy was used to determine the 3D structures of MgO-supported Os_3, Os_4, Os_5, and Os_{10} clusters, which have structures nearly matching those of osmium carbonyl compounds with known crystal structures. The samples are among the best-defined supported catalysts.

Imaging Nanoclusters in 3D

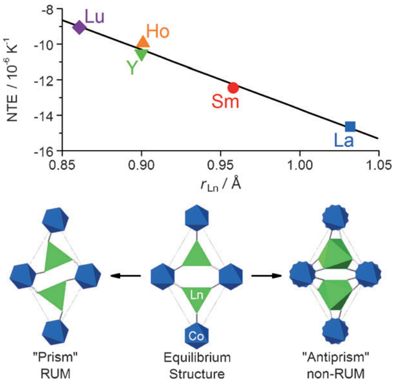
C. Aydin, A. Kulkarni, M. Chi,
N. D. Browning,
B. C. Gates* ______ **5262 – 5265**

Three-Dimensional Structural Analysis of MgO-Supported Osmium Clusters by Electron Microscopy with Single-Atom Sensitivity

Anomalous Thermal Expansion

S. G. Duyker, V. K. Peterson,* G. J. Kearley,
A. J. Ramirez-Cuesta,
C. J. Kepert* ________ 5266 – 5270

Negative Thermal Expansion in $LnCo(CN)_6$ (Ln = La, Pr, Sm, Ho, Lu, Y): Mechanisms and Compositional Trends

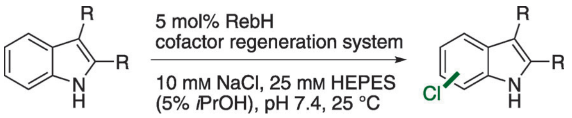


RUM with a twist: The magnitude of negative thermal expansion (NTE) in the $LnCo(CN)_6$ coordination frameworks increases with Ln ion radius r_{Ln}. The framework structure contains an unusual locally unstable trigonal prismatic LnN_6 unit that participates in an NTE-contributing vibrational mode by twisting about its axis at low energies. This contrasts with the rigid unit modes (RUMs) prevalent in other systems.

Arene Halogenation

J. T. Payne, M. C. Andorfer,
J. C. Lewis* ________ 5271 – 5274

Regioselective Arene Halogenation using the FAD-Dependent Halogenase RebH



Together we're strong: Co-expression of the halogenase RebH with GroEL/ES and fusion of the flavin reductase RebF to MBP enabled production of both enzymes on scales sufficient for preparative regioselective oxidative halogenation of arenes. The activity and selectivity of RebH contrasts with those reported for the structurally homologous halogenase PrnA, which only enabled halogenation of non-natural substrates at their most electronically activated positions.

Helical Structures

W. Makiguchi, S. Kobayashi, Y. Furusho,
E. Yashima* ________ 5275 – 5279

Formation of a Homo Double Helix of a Conjugated Polymer with Carboxy Groups and Amplification of the Macromolecular Helicity by Chiral Amines Sandwiched between the Strands

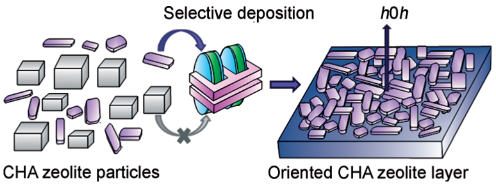
Inside Back Cover

Sandwiched amines: A π-conjugated polymer with carboxy groups self-associates to form a racemic double helix. In contrast, with chiral amines it forms a unique one-handed double helix, in which the strands sandwich pairs of chiral amines through cyclic hydrogen-bonding networks (see picture). The chiral information of the amines is transferred to the polymer backbones with remarkable amplification of the helical chirality.

Zeolite Films

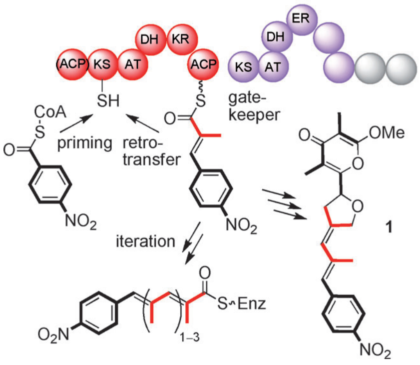
E. Kim, W. Cai, H. Baik,
J. Choi* ________ 5280 – 5284

Uniform Si-CHA Zeolite Layers Formed by a Selective Sonication-Assisted Deposition Method



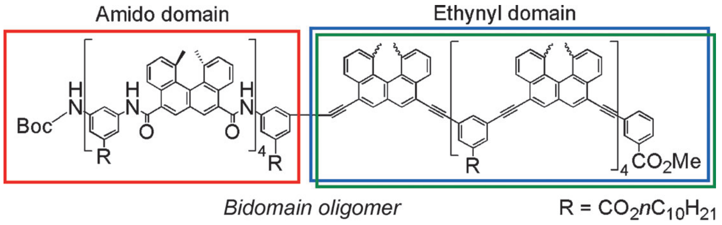
Discrimination against the majority: The broad size distribution of conventional near-cubic Si-CHA (all-silica chabazite) zeolites impedes the formation of a close-packed layer, which is critical for the manufacture of zeolite films by secondary growth. Although present in lower abundance, platelike Si-CHA particles cosynthesized with near-cubic particles were deposited selectively on an α-Al_2O_3 disk to form a uniform layer.

Aureothin Polyketide Synthase



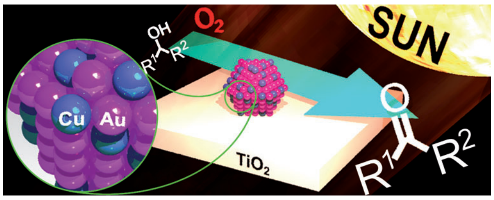
Freedom and control: First insights into the rare programmed iteration of an individual polyketide synthase (PKS) module were obtained from the analysis and mutation of aureothin (**1**) synthase. The first ketosynthase (KS) domain primes the PKS, allowing intermediate retrotransfer. Addition of a designated loading module results in a complete loss of iteration. The downstream KS functions as a gatekeeper for correct chain length.

Tetrameric Heteroaggregation



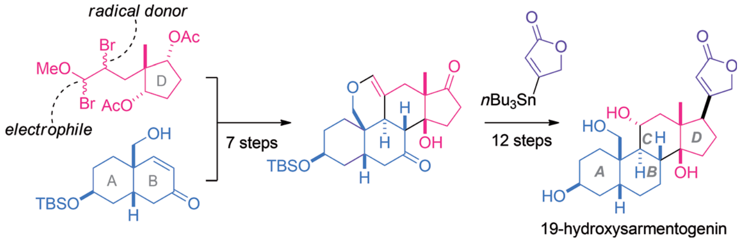
How to aggregate your domains: A method of forming tetrameric aggregates with an ααββ structure has been developed using stereoisomeric bidomain oligomers. Upon mixing the oligomers, a tetrameric ααββ aggregate was obtained by heteroaggregation at the ethynyl domain. The tetrameric aggregate reversibly changed its structure between the dimeric homoaggregates on heating and cooling, whereas gelation by polymerization occurred in toluene.

Heterogeneous Catalysis



Rejuvenating sunlight: Supported Au–Cu bimetallic alloy nanoparticles promote aerobic oxidation at room temperature under visible light ($\lambda > 450$ nm) irradiation with little deactivation by the oxidation of surface Cu atoms by oxygen. This is achieved through the reduction of oxidized surface Cu atoms by the surface Au atoms, a process which is activated by visible-light irradiation, even by sunlight.

Natural Product Synthesis

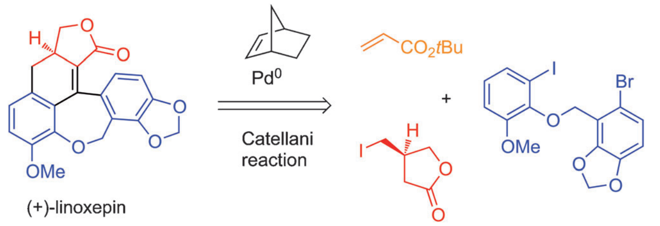


19 in 19: A convergent total synthesis of 19-hydroxysarmentogenin has been achieved, starting from three simple fragments. The desired product was synthesized in 19 steps from the AB ring system with the installation of six stereocenters and the formation of three C–C bonds.

C–H Activation

H. Weinstabl, M. Suhartono, Z. Qureshi, M. Lautens* 5305–5308

Total Synthesis of (+)-Linoxepin by Utilizing the Catellani Reaction

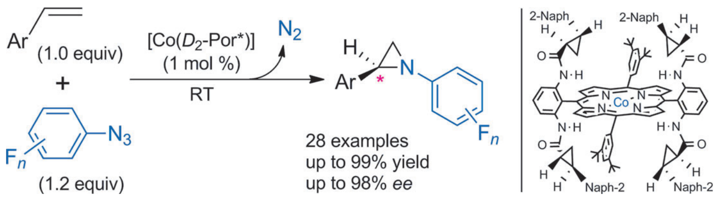


Molecular intelligence: The structurally novel lignan (+)-linoxepin is synthesized in an eight-step sequence. The enantioselective synthesis features the palladium-catalyzed Catellani reaction as the key step. In this highly convergent multicomponent reaction, two new carbon–carbon bonds are formed, one of which results from a C–H bond functionalization.

Asymmetric Catalysis

L. M. Jin, X. Xu, H. Lu, X. Cui, L. Wojtas, X. P. Zhang* 5309–5313

Effective Synthesis of Chiral *N*-Fluoroaryl Aziridines through Enantioselective Aziridination of Alkenes with Fluoroaryl Azides

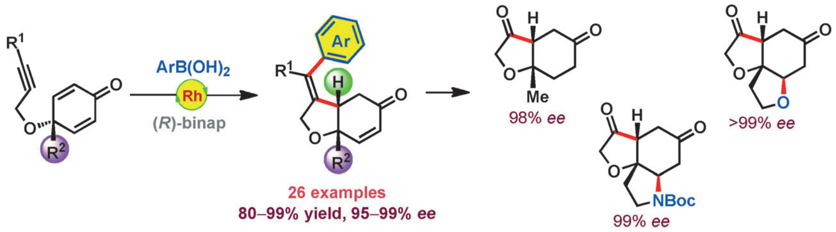


The Co^{II} complex of a D_2-symmetric chiral porphyrin ([Co(D_2-Por*)], see scheme) is a highly effective catalyst for the enantioselective aziridination of alkenes with fluoroaryl azides. The reaction can be performed at RT with low catalyst loading, and the olefin is the limiting reagent. Furthermore, the reaction is tolerant toward different combinations of aromatic olefins and fluoroaryl azides.

Asymmetric Catalysis

Z.-T. He, B. Tian, Y. Fukui, X. Tong, P. Tian,* G.-Q. Lin* 5314–5318

Rhodium-Catalyzed Asymmetric Arylative Cyclization of *meso*-1,6-Dienynes Leading to Enantioenriched *cis*-Hydrobenzofurans

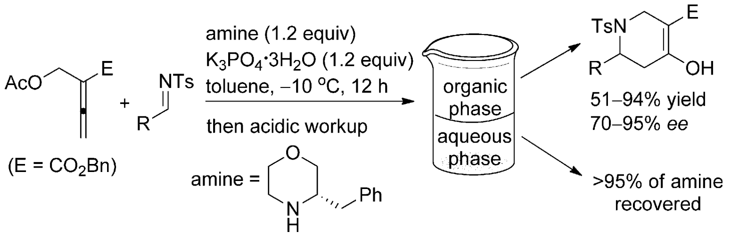


In tandem: The title reaction of cyclohexadienone-containing *meso*-1,6-dienynes with arylboronic acids through a tandem arylrhodation/conjugate addition sequence has been realized, and provides a novel approach to the enantioenriched *cis*-hydrobenzofurans with both excellent yields and enantioselectivities. binap = 2,2′-bis(diphenylphosphanyl)-1,1′-binaphthyl, Boc = *tert*-butoxycarbonyl.

Synthetic Methods

P. Hu, J. Hu, J. Jiao, X. Tong* 5319–5322

Amine-Promoted Asymmetric (4+2) Annulations for the Enantioselective Synthesis of Tetrahydropyridines: A Traceless and Recoverable Auxiliary Strategy

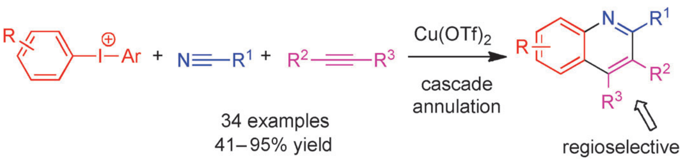


Gone, without a trace: The in situ reaction of 2-(acetoxymethyl)buta-2,3-dienoate and a secondary amine produces a 2-methylene-3-oxobutanoate equivalent that can be used in asymmetric [4+2] annulations with *N*-tosylimines to provide tetrahydropyridines in good to excellent yields and enantioselectivities. The amine is easily recovered and acts as a traceless auxiliary.

Quinoline Synthesis

Y. Wang, C. Chen,* J. Peng,
M. Li ______ **5323 – 5327**

Copper(II)-Catalyzed Three-Component Cascade Annulation of Diaryliodoniums, Nitriles, and Alkynes: A Regioselective Synthesis of Multiply Substituted Quinolines

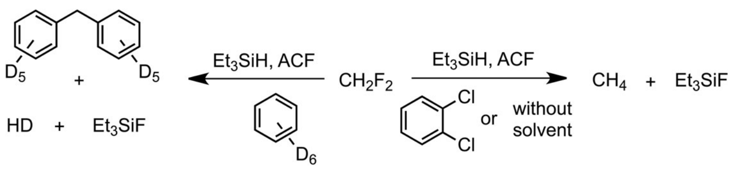


Three become one: Multiply substituted quinolines were synthesized from diaryliodoniums, alkynes, and nitriles by a Cu^{II}-catalyzed method. This cascade annulation is highly regioselective, step-economic, flexible with regard to the functional groups, and could potentially be applied to the synthesis of complex molecules.

C–F Activation

M. Ahrens, G. Scholz, T. Braun,*
E. Kemnitz* ______ **5328 – 5332**

Catalytic Hydrodefluorination of Fluoromethanes at Room Temperature by Silylium-ion-like Surface Species

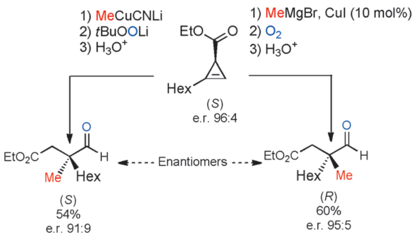


'Al'l about F: Aluminum chlorofluoride (ACF) catalyzes the hydrodefluorination, as well as Friedel–Crafts reactions of fluorinated methanes in the presence of Et_3SiH. A surface-bound silylium-ion-like species is considered to be a crucial intermediate in achieving the C–F bond cleavage.

Synthetic Methods

P.-O. Delaye, D. Didier,
I. Marek* ______ **5333 – 5337**

Diastereodivergent Carbometalation/Oxidation/Selective Ring Opening: Formation of All-Carbon Quaternary Stereogenic Centers in Acyclic Systems

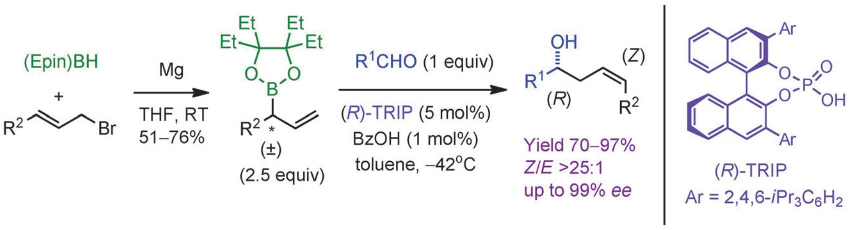


A twofer: The title reaction sequence for cyclopropenes allows the preparation of aldehydes bearing α-quaternary stereocenters in a one pot-reaction from readily available starting materials. Through a diastereodivergent carbometalation reaction, both enantiomers of the corresponding aldehyde were obtained from the same cyclopropene derivative (see scheme).

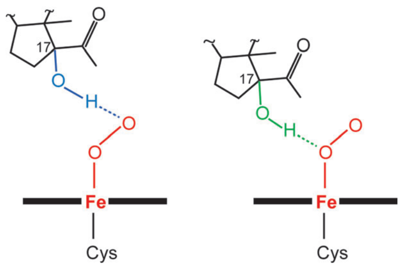
Asymmetric Allylation

C. A. Incerti–Pradillos, M. A. Kabeshov,
A. V. Malkov* ______ **5338 – 5341**

Highly Stereoselective Synthesis of *Z*-Homoallylic Alcohols by Kinetic Resolution of Racemic Secondary Allyl Boronates

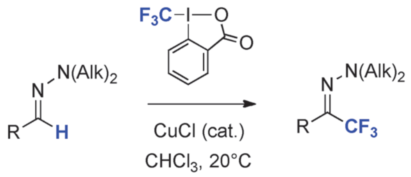


α to Z: Racemic α-chiral allyl boronates, which are readily synthesized from the respective primary allyl halides, undergo a highly efficient kinetic resolution in a face- and *Z*-selective allylation of aldehydes catalyzed by the chiral Brønsted acid (*R*)-TRIP (see scheme; Epin = tetraethylethylene glycol).



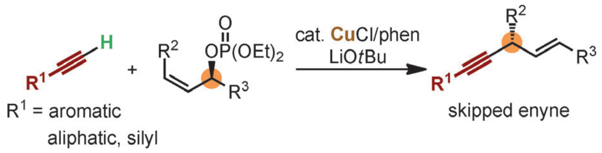
Consequences of alternative H-bonding: Raman spectra of oxygenated intermediates of Nanodisc-incorporated human CYP17 in the presence of natural substrates (pregnenolone and progesterone) directly confirm that substrate structure effectively alters hydrogen-bonding interactions with the critical Fe–O–O fragment and dictates its predisposition for one of two alternative reaction pathways. Such substrate control has profound physiological implications.

Hydrogen Bonding Control



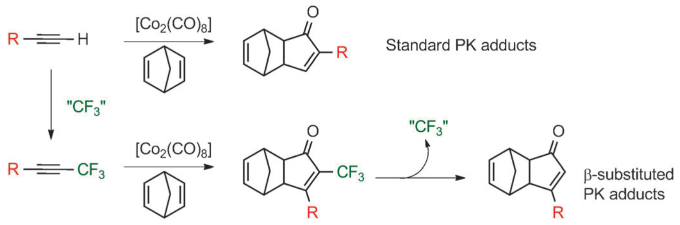
Mild and practical: Trifluoromethylation of (hetero)aromatic aldehyde *N*,*N*-dialkylhydrazones was achieved at room temperature by using Togni's trifluoromethylation reagent under CuCl catalysis (see scheme). This simple reaction is believed to occur by a CF_3-radical-transfer mechanism and yields useful trifluoromethylated building blocks.

Trifluoromethylation



Skipping out: The title reaction using internal secondary allylic phosphates proceeded with excellent γ regioselectivity and *E* stereoselectivity to give skipped enynes. Enantioenriched secondary allylic phosphates proceeded with 1,3-*anti* stereochemistry to afford the corresponding chiral 1,4-enynes, which were used for various derivatizations and the formal total synthesis of a GnRH antagonist. phen = 1,10-phenanthroline.

Homogeneous Catalysis

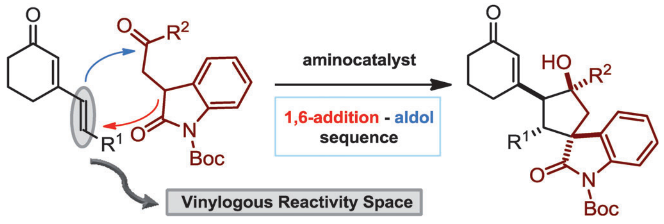


Against the rules: The synthesis of the previously unknown β-substituted regioisomers of the intermolecular Pauson–Khand reaction of terminal alkynes is reported. This regiochemistry was achieved by using the trifluoromethyl group as a removable directing group on the alkyne.

Synthetic Methods

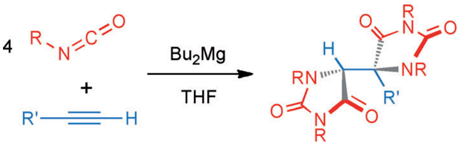
Inside Cover

Synthetic Methods



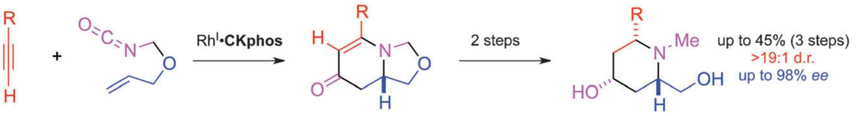
Remote control: The title reaction facilitates the synthesis of complex chiral molecules while selectively forging multiple stereocenters at distant positions, namely five and six bond lengths away from the catalyst chiral fragment (see scheme; Boc = *tert*-butoxycarbonyl). The potential of the strategy is demonstrated through the one-step preparation of spirocyclopentane oxindoles having four contiguous stereocenters.

N-Heterocycles



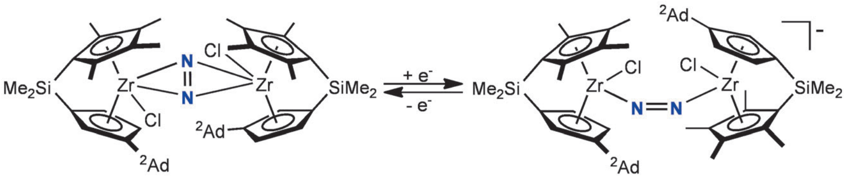
Bicycle race: Structurally complex bis(imidazolidine-2,4-dione) molecules may be synthesized with complete atom-efficiency from simple building blocks by a kinetically controlled magnesium-mediated cascade of intermolecular isocyanate insertion and intramolecular alkyne hydroamination reaction steps.

Asymmetric Synthesis



Break the cycle: The title reaction proceeds with a variety of alkyne substrates in good yield and high enantioselectivity. Upon reduction of the vinylogous amide in high diastereoselectivity (>19:1) and cleavage of the tether, *N*-methylpiperidine products with functional group handles can be accessed (see scheme).

Dinitrogen Hapticity

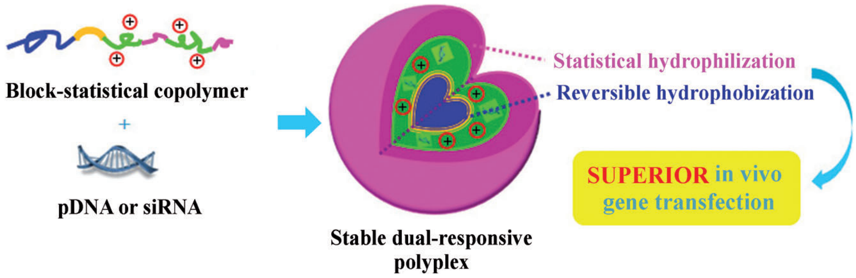


N$_2$ shuffle: One-electron reduction of the *ansa*-zirconocene complex with a side-on dinitrogen ligand furnished the corresponding anion in which the hapticity of the N$_2$ ligand changed to end-on. Oxidation of the anion regenerates the neutral complex with concomitant change in N$_2$ hapticity. The zirconocene complexes with unusual (N$_2$)$^{3-}$/(N$_2$)$^-$ ligands have been fully characterized. Ad = adamantyl.

DNA Nanoparticles

H. Wei, L. R. Volpatti, D. L. Sellers,
D. O. Maris, I. W. Andrews,
A. S. Hemphill, L. W. Chan, D. S. H. Chu,
P. J. Horner, S. H. Pun* ____ **5377 – 5381**

Dual Responsive, Stabilized Nanoparticles for Efficient In Vivo Plasmid Delivery

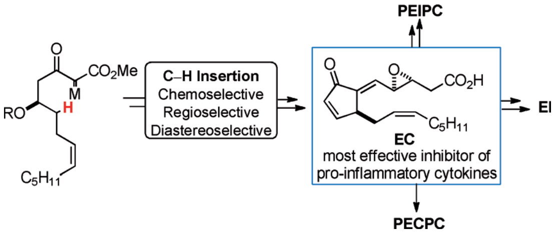


A small case of love and hate: A block-statistical copolymer combining reversible hydrophobization and statistical hydrophilization allows the preparation of pH value- and reduction-responsive nanoparticles (polyplexes) for efficient in vivo plasmid delivery.

Oxidized Phospholipids

J. Egger, P. Bretscher, S. Freigang, M. Kopf,
E. M. Carreira* ____ **5382 – 5385**

Synthesis of Epoxyisoprostanes: Effects in Reducing Secretion of Pro-inflammatory Cytokines IL-6 and IL-12

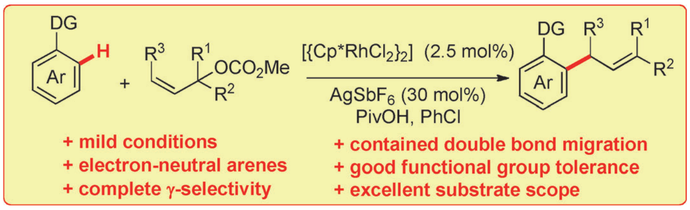


Anti-inflammatory: The efficient and general synthetic route to the elusive epoxyisoprostanoid phospholipids PECPC and PEIPC, along with the isoprostanoids EC and EI, relies on a number of stereo- and chemoselective steps, including a C–H insertion for the rapid construction of the cyclopentanone ring. The synthesized compounds display unprecedented biological activity in reducing the secretion of pro-inflammatory cytokines.

C–H Activation

H. Wang, N. Schröder,
F. Glorius* ____ **5386 – 5389**

Mild Rhodium(III)-Catalyzed Direct C–H Allylation of Arenes with Allyl Carbonates

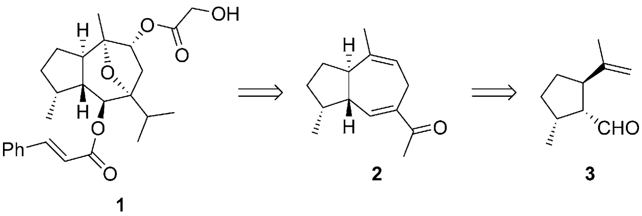


All(yl) possible! A rhodium(III)-catalyzed intermolecular direct C–H allylation reaction utilizing readily accessible allyl carbonates was developed. This method allows the allylation of electron-neutral arenes, providing complete γ-selectivity, high isomeric ratio, good substrate scope, and excellent functional group compatibility.

Terpenes

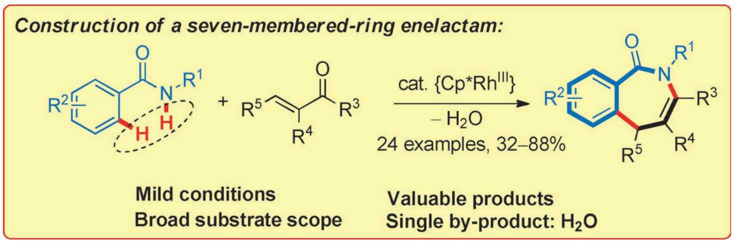
M. Zahel, A. Keßberg,
P. Metz* ____ **5390 – 5392**

A Short Enantioselective Total Synthesis of (−)-Englerin A



Selective oxidations of dienone **2** as well as a ring-closing metathesis to give the hydroazulene framework enabled the 12-step preparation of title compound **1** from (−)-photocitral A (**3**), which is in turn rapidly available from (−)-isopulegol through dual catalysis.

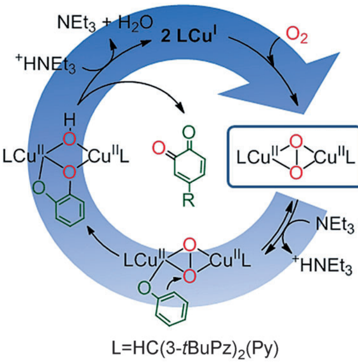
C–H Activation



Seven! The title reaction can be described as an intermolecular annulation involving tandem C–H activation, cyclization to give the seven-membered ring, and condensation steps. Biologically interesting azepinone derivatives can be prepared in this way. The synthetic potential of this method was demonstrated by the construction of the homoprotoberberine ring system.

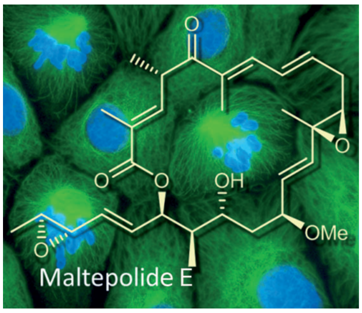
Tyrosinase Model

A new catalyst (see structure) hydroxylates phenols with O_2 via a stable side-on peroxide complex, which is similar to the active site of tyrosinase in terms of the ligand environment and its spectroscopic properties. The catalytic oxidation of phenols to quinones proceeds at room temperature in the presence of NEt_3 and even non-native substrates can be oxidized catalytically. The reaction mechanism is analogous to that of the enzyme-catalyzed reaction.



Natural Products

Epoxide "swing": A family of polyketide macrolactones originating from maltepolide E was discovered in *Sorangium cellulosum* So ce1485. Their structure was established by NMR experiments, molecular modeling, and X-ray crystallography. Maltepolides induce rather rare morphological changes in the dividing transformed cell lines (see picture in the background).



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).

Angewandte Corrigendum

In this Communication, the name of the third author is misspelled. The correct name is "*Prof. Dr. Liangcheng Du*".

Elucidating the Biosynthetic Pathway for Vibralactone: A Pancreatic Lipase Inhibitor with a Fused Bicyclic β-Lactone

P.-J. Zhao,* Y.-L. Yang, L. Du, J.-K. Liu, Y. Zeng* ______ **2298–2302**

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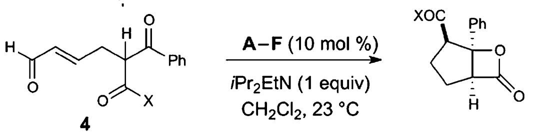
DOI: 10.1002/anie.201208182

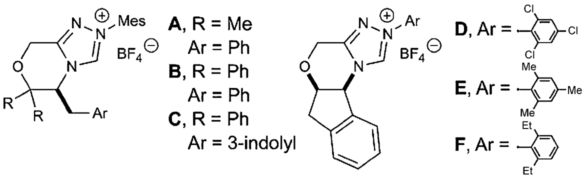
Angewandte Corrigendum

Table 1 of this Communication contains wrong data for the *ee* values of entries 1–7. The corrected entries are shown here.

Table 1: Reaction optimization.[a]



Entry	Variation of the standard conditions	P:SM[b]	d.r.[c]	*ee* [%][d]
1	**A**, X = OEt	2:3	4:1	n.d.
2	**B**, X = OEt	1.2:1	5:1	n.d.
3	**C**, X = OEt	1:20	–[e]	–
4	**D**, X = OEt	1:3	4:1	n.d.
5	**E**, X = OEt	> 20:1 (31)[f]	7:1	90
6	**F**, X = OEt	> 20:1 (72)	5:1	99
7	**F**, X = NBn_2	> 20:1	1:1	n.d.
…	…	…	…	…



[a] See the Supporting Information for details. [b] Ratio of product (P) to starting material (SM) determined by ^1H NMR spectroscopy (500 MHz). Number in parentheses is the isolated yield of both diastereomers (on a 0.4 mmol scale). [c] Ratio determined by ^1H NMR spectroscopy (500 MHz) prior to purification. [d] Enantiomeric excess of the major diastereomer determined by HPLC. [e] Starting material recovered. [f] Yield of major diastereomer only. ... n.d. = not determined.

Catalytic Dynamic Kinetic Resolutions with N-Heterocyclic Carbenes: Asymmetric Synthesis of Highly Substituted β-Lactones

D. T. Cohen, C. C. Eichman, E. M. Phillips, E. R. Zarefsky, K. A. Scheidt* ______ **7309–7313**

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