

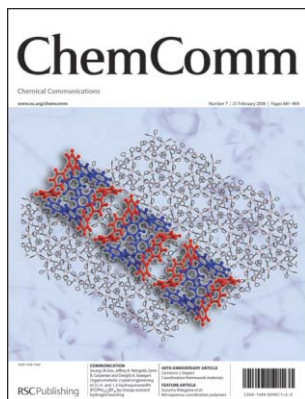
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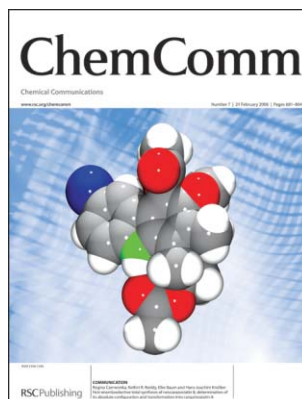
IN THIS ISSUE

ISSN 1359-7345 CODEN CHCOFS (7) 681–804 (2006)



Cover

See Dwight A. Sweigart *et al.*, page 708.
Charge assisted hydrogen bonding in the ionic complex $[(\eta^6\text{-hydroquinone})\text{Rh}(\text{P}(\text{OPh})_3)_2]^+\text{BF}_4^-$ results in a solid state structure featuring hydrophobic channels of aromatic phenyl rings. Image reproduced by permission of Seung Uk Son, Jeffrey A. Reingold, Gene B. Carpenter and Dwight A. Sweigart from *Chem. Commun.*, 2006, 708.



Inside cover

See Hans-Joachim Knölker *et al.*, page 711.
Key intermediate of the first enantioselective total synthesis of the potent lipid peroxidation inhibitor neocarazostatin B. Image reproduced by permission of Regina Czerwonka, Kethiri R. Reddy, Elke Baum and Hans-Joachim Knölker from *Chem. Commun.*, 2006, 711.

CHEMICAL SCIENCE

C9

Drawing together the research highlights and news from all RSC publications, *Chemical Science* provides a 'snapshot' of the latest developments across the chemical sciences showcasing newsworthy articles, as well as the most significant scientific advances.

Chemical Science

February 2006/Volume 3/Issue 2

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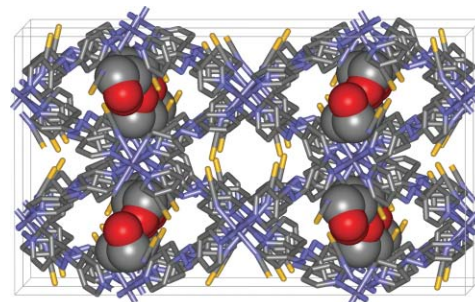
40TH ANNIVERSARY ARTICLE

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Advanced functional properties in nanoporous coordination framework materials

Cameron J. Kepert

Coordination framework materials display a rich array of host-guest properties and are notable amongst porous media for their extreme chemical versatility. This article highlights a number of areas where specific function has been incorporated into these framework host lattices.



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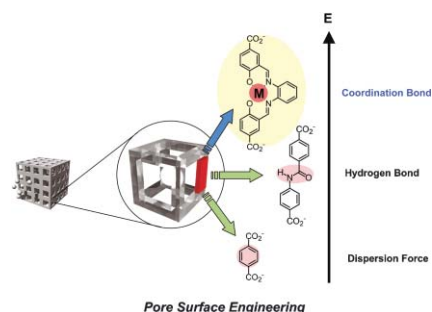
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Pore surface engineering of microporous coordination polymers

Susumu Kitagawa,* Shin-ichiro Noro and Takayoshi Nakamura

Pore surface properties of microporous coordination polymers are easily modified using metalloligands, which introduce coordinatively unsaturated metal centers onto the channel surface.



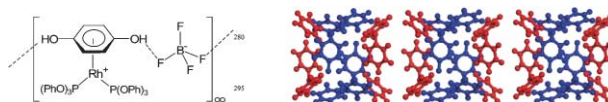
COMMUNICATIONS

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Organometallic crystal engineering of [(1,4- and 1,3-hydroquinone)Rh(P(OPh)₃)₂]BF₄ by charge assisted hydrogen bonding

Seung Uk Son, Jeffrey A. Reingold, Gene B. Carpenter and Dwight A. Sweigart*

The ionic complexes [(1,4- and 1,3-hydroquinone)Rh(P(OPh)₃)₂]-BF₄ form porous organometallic structures dictated by charge assisted hydrogen bonding.

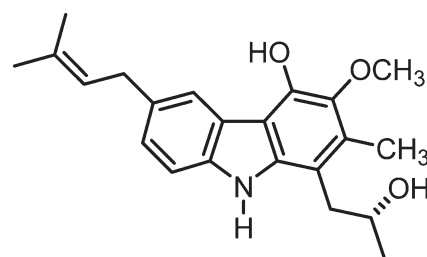


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First enantioselective total synthesis of neocarazostatin B, determination of its absolute configuration and transformation into carquinostatin A

Regina Czerwonka, Kethiri R. Reddy, Elke Baum and Hans-Joachim Knölker*

The first enantioselective total synthesis of neocarazostatin B, the determination of its absolute configuration and transformation into carquinostatin A are described.



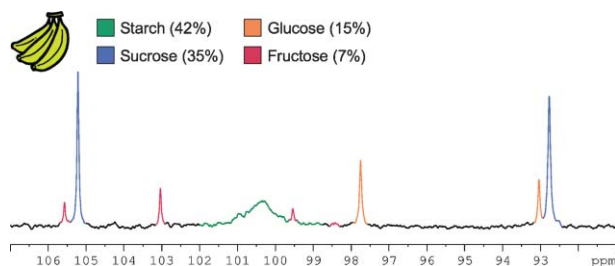
(R)-(-)-neocarazostatin B

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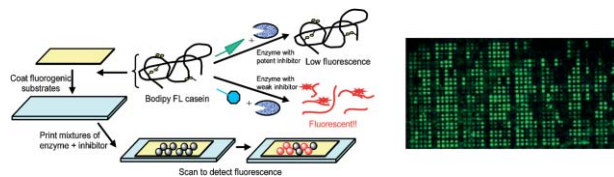
Use of ionic liquids in the study of fruit ripening by high-resolution ¹³C NMR spectroscopy: 'green' solvents meet green bananas

Diego A. Fort, Richard P. Swatloski, Patrick Moyna,* Robin D. Rogers* and Guillermo Moyna*

Banana pulps at any ripening stage can be completely dissolved in solvent systems based on the ionic liquid (IL) 1-*n*-butyl-3-methylimidazolium chloride ([C₄mim]Cl), and variations in the carbohydrate composition of the fruit analyzed directly on the resulting solutions using high-resolution ¹³C NMR spectroscopy.



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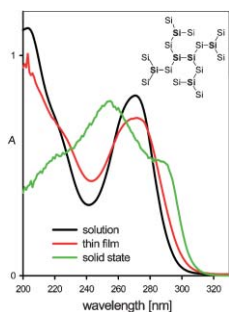


Activity-based high-throughput profiling of metalloprotease inhibitors using small molecule microarrays

Jun Wang, Mahesh Uttamchandani, Li Ping Sun and Shao Q. Yao*

A high-throughput small molecule microarray (SMM) method that enables quick and cost-effective identification of potent inhibitors of metalloproteases in an activity-dependent manner is described. This offers a rapid means for inhibitor discovery and inhibitor fingerprinting.

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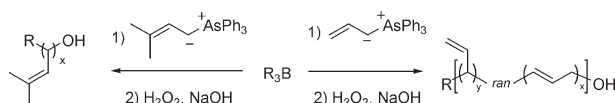


Novel double-cored oligosilane dendrimers—conformational dependence of the UV absorption spectra

C. Krempner,* M. Köckerling and C. Mamat

The X-ray data and the UV absorption behaviour of the novel double-cored oligosilane dendrimers $\{[\text{Me}(\text{Me}_3\text{Si})_2\text{Si}-\text{Me}_2\text{Si}]_2\text{SiMe}_2\}$ (**4**) and $\{[\text{Me}(\text{Me}_3\text{Si})_2\text{Si}-\text{Me}_2\text{Si}]_2\text{SiMe}-\text{SiMe}_2\}$ (**5**) reveal the extent of σ -conjugation in these molecules to be determined by steric interactions and the conformation of the longest chains.

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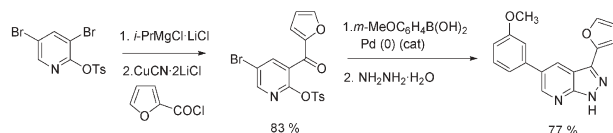


Boron-mediated polymerization of ylides derived from allylic arsonium salts: influence of the double bond substitution on the outcome

Régis Mondière, Jean-Philippe Goddard, Mickaël Huiban, Géraldine Carrot, Thierry Le Gall* and Charles Mioskowski*

The polymerization of ylides derived from arsonium salts containing either an allyl or a 3,3-dimethylallyl group in the presence of boron compounds was shown to afford new oligomers or polymers: poly(propenylene/propenyldiene) random copolymers, and polypropenyldienes, respectively.

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Regioselective functionalization of trisubstituted pyridines using a bromine–magnesium exchange

Hongjun Ren and Paul Knochel*

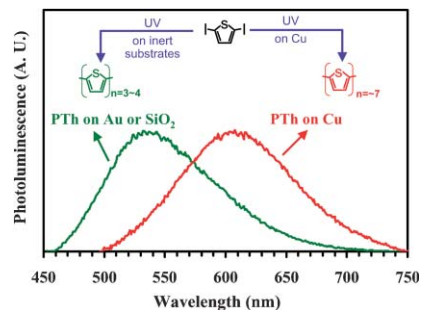
A tosyloxy substitution in position 2 allows the realisation of a highly regioselective Br/Mg exchange reaction on 3,5-dibromopyridine derivatives using *iso*-PrMgCl·LiCl. The resulting functionalized pyridylmagnesium reagents react with various electrophiles in position 3. Acylated pyridines of this type can be readily converted into pyrazolo [3,4-*b*] pyridines.

729

Synthesis-in-place of highly-conjugated oligothiophene micropatterns *via* photo-activated Ullmann coupling on copper surface

Sudarshan Natarajan and Seong H. Kim*

In contrast to the traditional thermally-activated Ullmann coupling, a photo-activated Ullmann coupling for facile synthesis of oligothiophene and polythiophene films and micropatterns is reported.

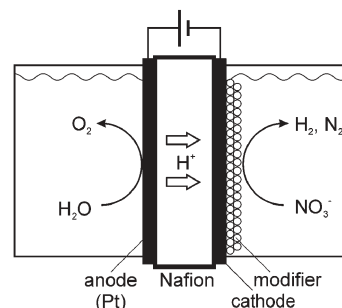


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Electrocatalytic nitrate hydrogenation over an H⁺-conducting solid polymer electrolyte membrane-modified cathode assembly

Masato Machida,* Kiwako Sato, Isao Ishibashi, Mohammad Abul Hasnat and Keita Ikeue

Selective electrocatalytic hydrogenation of NO₃⁻ to N₂ in water has successfully been achieved at room temperature using a membrane-electrode assembly (MEA) consisting of an H⁺-conducting solid polymer electrolyte (Nafion-117) and a surface-modified Pt cathode.

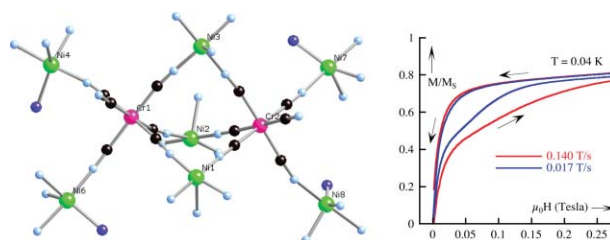


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One step assembly of a nonanuclear Cr^{III}₂Ni^{II}₇ bimetallic cyanide bridged complex

Jean-Noël Rebilly, Laure Catala, Eric Rivière, Régis Guillot, Wolfgang Wernsdorfer and Talal Mallah*

The reaction of hexacyanochromate(III) with a mononuclear Ni^{II} complex bearing a bulky tridentate macrocycle leads in one step to a nonanuclear Cr₂Ni₇ complex that presents fast tunnelling of the magnetization at low temperature.

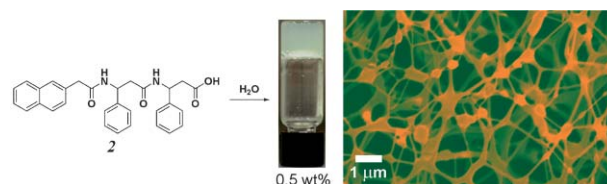


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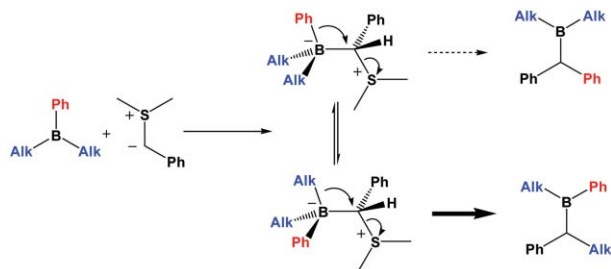
Supramolecular hydrogels based on β-amino acid derivatives

Zhimou Yang, Gaolin Liang and Bing Xu*

A new class of supramolecular hydrogels that consist of β-amino acids may confer proteolytic resistance to the hydrogels for biomedical applications.



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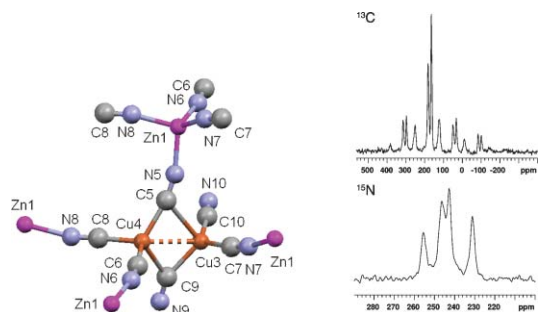


Is phenyl a good migrating group in the rearrangement of organoborates generated from sulfur ylides?

Raphaël Robiette, Guang Yu Fang, Jeremy N. Harvey* and Varinder K. Aggarwal*

We report here on the unexpected low migratory aptitude of the phenyl group observed in the titled reaction and the computational investigation we carried out in order to understand the results obtained.

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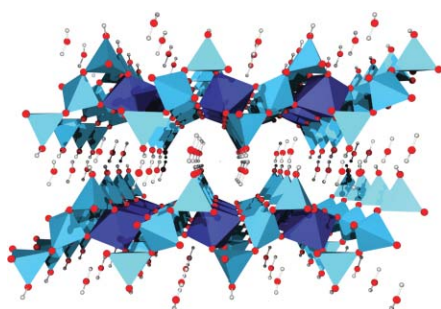


A paramagnetic Cu(I)/Cu(II)/Zn(II) coordination polymer with multiple CN-binding modes and its solid-state NMR characterization

Liang Ouyang, Pedro M. Aguiar, Raymond J. Batchelor, Scott Kroeker* and Daniel B. Leznoff*

A mixed-valent $[\text{Cu}(\text{en})_2][\text{Zn}(\text{CN})_4(\text{CuCN})_2]$ 2-D layered polymer has been prepared using tetrahedral $[\text{Zn}(\text{CN})_4]^{2-}$ as a starting material. It has six inequivalent cyanides in four bonding modes and readily observable ^{13}C and ^{15}N MAS NMR spectra.

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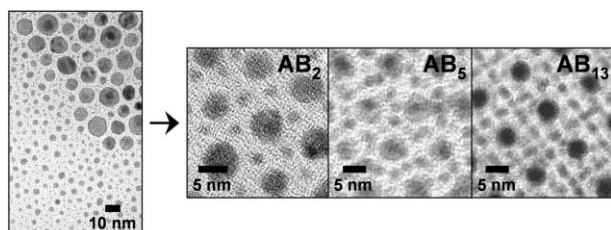


Synthesis and crystal structure of $\text{AlH}_2\text{P}_3\text{O}_{10}\cdot 2\text{H}_2\text{O}$; a new structure-type for layered acid phosphates

Steven K. Rishi, Benson M. Kariuki, Neal J. Checker, John Godber and Adrian J. Wright*

A new structure-type for a series of layered metal phosphate phases is reported following this study of $\text{AlH}_2\text{P}_3\text{O}_{10}\cdot 2\text{H}_2\text{O}$. This layered structure, with accessible hydroxyl groups, has potential for a number of important applications, including hosts for novel organic–inorganic hybrids.

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One-pot synthesis of bi-disperse FePt nanoparticles and size-selective self-assembly into AB_2 , AB_5 , and AB_{13} superlattices

Amandeep K. Sra, Trevor D. Ewers, Qiang Xu, Henny Zandbergen and Raymond E. Schaak*

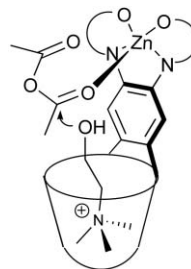
By chemically modifying the nucleation burst that generates monodisperse FePt nanocrystals, a mixture of Pt and $\text{Fe}_x\text{Pt}_{1-x}$ nanoparticles forms during a one-pot reaction; size-selective precipitation yields a bi-disperse population of $\text{Fe}_x\text{Pt}_{1-x}$ nanoparticles, which can assemble into binary superlattices.

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Cavitand templated catalysis of acetylcholine

Felix H. Zelder and Julius Rebek, Jr.*

A Zn–salen-modified cavitand templates the catalytic formation of acetylcholine from choline and acetic anhydride.

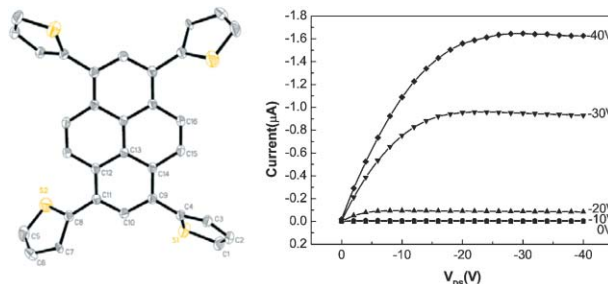


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Novel butterfly pyrene-based organic semiconductors for field effect transistors

Hengjun Zhang, Ying Wang, Kuizhan Shao, Yunqi Liu,* Shiyao Chen, Wenfeng Qiu, Xiaobo Sun, Ting Qi, Yongqiang Ma, Gui Yu, Zhongmin Su* and Daoben Zhu*

Novel butterfly pyrene derivatives functionalized with trifluoromethylphenyl and thienyl aromatic groups have been synthesized by Suzuki coupling reactions; the first example of a p-type FET based on a butterfly pyrene-type molecule is reported.

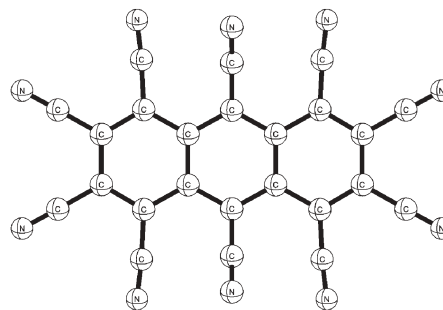


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Remarkable electron accepting properties of the simplest benzenoid cyanocarbons: hexacyanobenzene, octacyanonaphthalene and decacyanoanthracene

Xiuhui Zhang, Qianshu Li, Justin B. Ingels, Andrew C. Simmonett, Steven E. Wheeler, Yaoming Xie, R. Bruce King, Henry F. Schaefer III* and F. Albert Cotton

Electron affinities of the simplest benzenoid cyanocarbons are predicted to be significantly larger than those of the analogous benzenoid fluorocarbons and suggest their use as electron acceptors in novel magnetic materials.

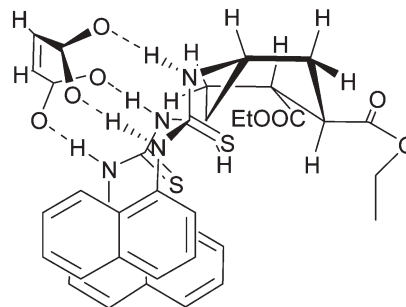


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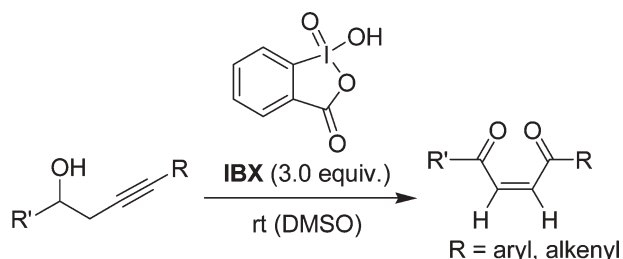
Fluorescent sensing of maleate *versus* fumarate by a neutral cyclohexane based thiourea receptor

Ana M. Costero,* Manuel Colera, Pablo Gaviña and Salvador Gil

A new cyclohexyl based fluorescent anion receptor, is able to recognize maleate *versus* fumarate both as their TMA salts in DMSO. The recognition was also observed in the presence of 5% water.



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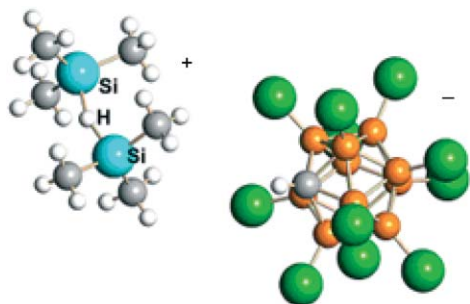


A new method for the synthesis of *Z*-enediones via IBX-mediated oxidative rearrangement of 2-alkynyl alcohol systems

Benedikt Crone and Stefan F. Kirsch*

o-Iodoxybenzoic acid (IBX) was found to mediate the selective conversion of 2-alkynyl alcohols into *Z*-enediones under notably mild conditions *via* an unprecedented rearrangement mechanism (33–65% yield, 13 examples).

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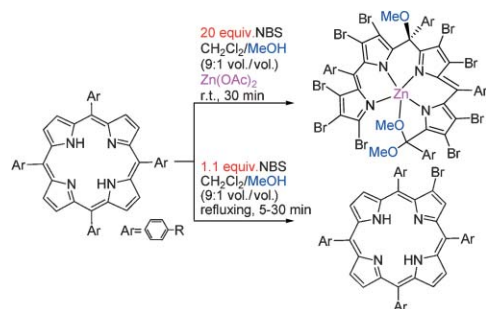


Novel weak coordination to silylium ions: formation of nearly linear Si–H–Si bonds

Stephan P. Hoffmann, Tsuyoshi Kato, Fook S. Tham and Christopher A. Reed*

The weakly coordinating carborane anion in ion-like trialkylsilyl species $R_3Si(CHB_{11}Cl_{11})$ can be displaced by nucleophiles as weak as *ortho*-dichlorobenzene, SO_2 and trialkylsilanes, the latter forming nearly linear hydride bridges in $R_3Si-H-SiR_3^+$ cations.

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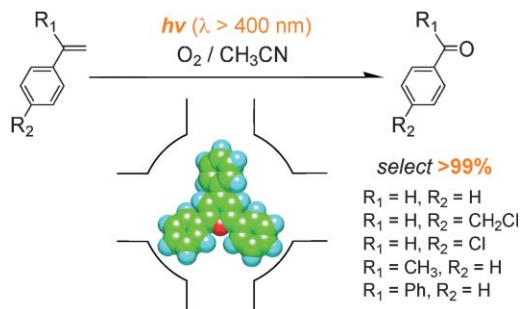


Unexpected bromination ring-opening of tetraarylporphyrins

Chao Liu, Dong-Mei Shen and Qing-Yun Chen*

An unexpected, mild, efficient bromination ring-opening method has been developed for convenient synthesis of various novel biladienes or brominated porphyrins by controlling the amounts of NBS used.

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Visible light-induced highly selective transformation of olefin to ketone by 2,4,6-triphenylpyrylium cation encapsulated within zeolite Y

Yasuhiro Shiraishi,* Naoya Saito and Takayuki Hirai

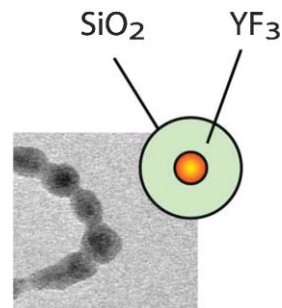
2,4,6-Triphenylpyrylium cation encapsulated within zeolite Y promotes highly selective transformation of olefins to ketones with molecular oxygen, under visible light ($\lambda > 400$ nm) irradiation at room temperature.

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One-pot synthesis of $\text{YF}_3@$ silica core/shell nanoparticles

Masih Darbandi and Thomas Nann*

The present paper describes a new one-pot synthetic method for core/shell $\text{YF}_3@$ SiO₂ nanoparticles. The morphology of the resulting particles could be tuned from spherical to elongated structures (“pearl necklace”); absorbance and photoluminescence spectroscopy revealed intrinsic but no extrinsic defects in the YF_3 .

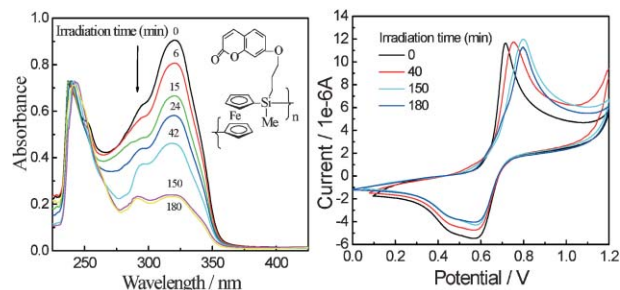


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A novel photoreversible poly(ferrocenylsilane) with coumarin side group: synthesis, characterization, and electrochemical activities

Dongli Zhao, Biye Ren,* Shanshan Liu, Xinxing Liu and Zhen Tong*

A novel photoreversible poly(ferrocenyl(3-(7-hydroxycoumarin)propyl)methylsilane) exhibiting interesting electrochemical activities with almost reversible photodimerization and photocission was synthesized and characterized.

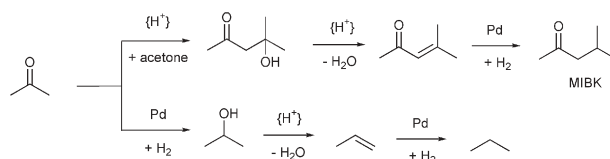


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Multifunctional catalysis by Pd-polyoxometalate: one-step conversion of acetone to methyl isobutyl ketone

Robert D. Hetterley, Elena F. Kozhevnikova and Ivan V. Kozhevnikov*

Pd metal supported on $\text{Cs}_{2.5}\text{H}_{0.5}\text{PW}_{12}\text{O}_{40}$ is an efficient bifunctional catalyst for the one-step conversion of acetone to methyl isobutyl ketone (MIBK) in gas and liquid phase.

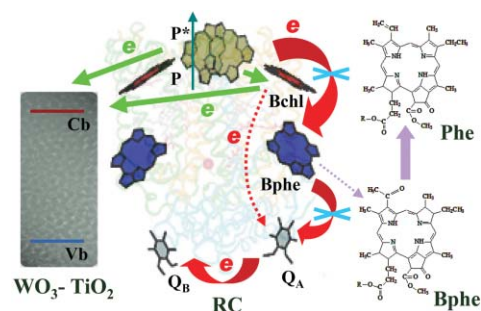


785

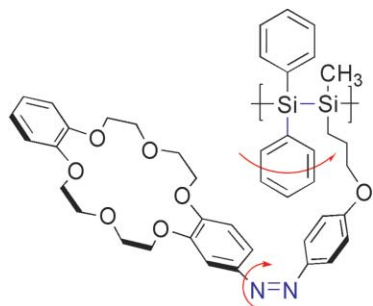
Manipulated photocurrent generation from pigment-exchanged photosynthetic proteins adsorbed to nanostructured WO_3 -TiO₂ electrodes

Yidong Lu, Jingjing Xu, Yuan Liu, Baohong Liu, Chunhe Xu, Dongyuan Zhao and Jilie Kong*

Control of the excitation relaxation pathway of reaction center proteins trapped on tailored WO_3 -TiO₂ electrodes.



788

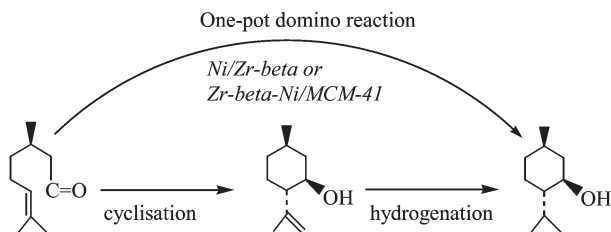


Polydiphenylsilanes bearing photosensitive azocrown groups

Liviu Sacarescu,* Rodinel Ardeleanu, Gabriela Sacarescu, Mihaela Simionescu and Nicolae Hurduc

A polydiphenylsilane bearing azocrown side groups was synthesized and studied by UV irradiation to investigate the influence of the photoisomerisation process on the σ - π conjugation effects.

790

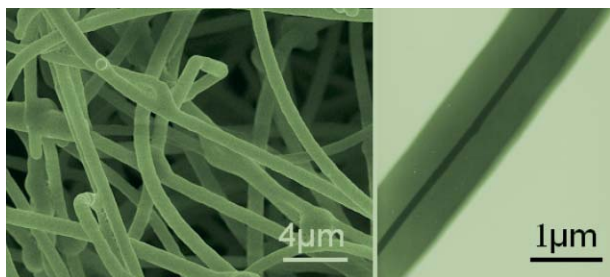


Domino-cyclisation and hydrogenation of citronellal to menthol over bifunctional Ni/Zr-Beta and Zr-beta/Ni-MCM-41 catalysts

Yuntong Nie, Gaik-Khuan Chuah* and Stephan Jaenicke

Bifunctional Ni/Zr-beta or dual catalytic system of Zr-beta and Ni/MCM-41 are highly active and selective in the cyclisation/reduction of citronellal to menthol. The diastereoselectivity to (\pm)-menthol in this one-pot reaction is between 90–94%.

793

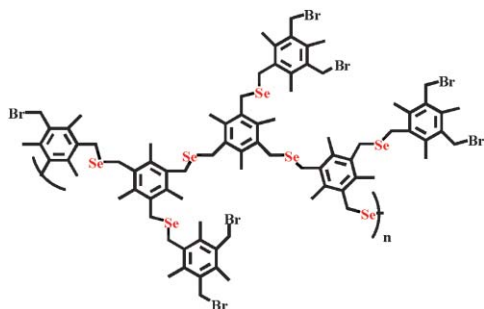


Large scale synthesis of uniform silver@carbon rich composite (carbon and cross-linked PVA) sub-microcables by a facile green chemistry carbonization approach

Lin-Bao Luo, Shu-Hong Yu,* Hai-Sheng Qian and Jun-Yan Gong

A facile green chemistry carbonization method has been discovered for the synthesis of uniform silver@carbon rich composite (carbon and cross-linked polyvinyl alcohol) core-shell microcables in large quantities.

796



Hyperbranched polyselenides as glutathione peroxidase mimics

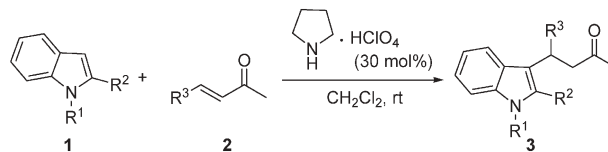
Huaping Xu, Jian Gao, Yapei Wang, Zhiqiang Wang, Mario Smet,* Wim Dehaen and Xi Zhang*

The synthesis of hyperbranched polymers (HBPs) with numerous catalytic sites (selenium) located at the branching units, which provide a novel model for glutathione peroxidase (GPx) mimics is reported.

Organocatalytic C3-selective Friedel–Crafts alkylations of indoles with α,β -unsaturated ketones

Dong-Ping Li, Ying-Cen Guo, Yu Ding and Wen-Jing Xiao*

The organocatalytic C3-selective Friedel–Crafts alkylations of indoles with simple ketone electrophiles in the presence of pyrrolidine (30 mol%) and HClO₄ (30 mol%) in CH₂Cl₂ at an ambient temperature resulted in β -indolylketones in high yields.



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