**Cover**

The cover illustrates the complex between cucurbit[7]uril and a viologen-containing dendrimer (pp. 1677–1683).



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Chemical Science
inside this issue

contents

C57

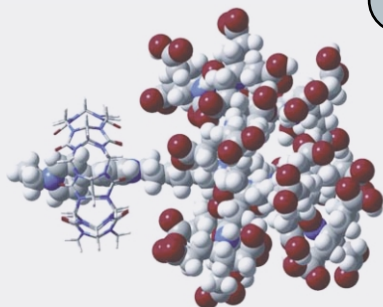
Chemical Science

August 2004/Volume 1/Issue 8
www.rsc.org/chemicalscience

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.

FEATURE ARTICLE

1677



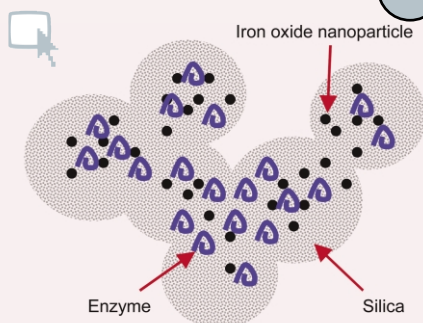
Dendrimers as guests in molecular recognition phenomena

Winston Ong, Marielle Gómez-Kaifer and Angel E. Kaifer*

Although dendrimers are often seen as molecular hosts, this review describes molecular recognition phenomena involving dendrimers with single or multiple guest residues.

COMMUNICATIONS

1684



Entrapment of enzymes and nanoparticles using biomimetically synthesized silica

Rajesh R. Naik, Melanie M. Tomczak, Heather R. Luckarift, Jim C. Spain and Morley O. Stone

A one-step method for the entrapment of enzymes and inorganic nanoparticles in biomimetically-synthesized silica under benign reaction conditions.

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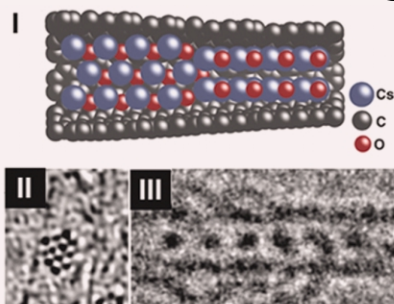
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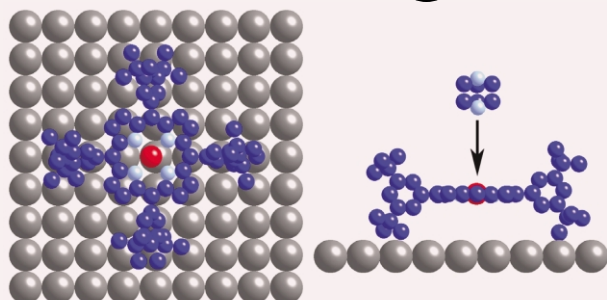


Single-walled carbon nanotubes filled with $M\text{OH}$ ($M = \text{K}, \text{Cs}$) and then washed and refilled with clusters and molecules

Narun Thamavaranakup, Henning A. Höpfe, Luisa Ruiz-Gonzalez, Pedro M. F. J. Costa, Jeremy Sloan,* Angus Kirkland* and Malcolm L. H. Green*

Treatment of and then washing out single walled carbon nanotubes with CsOH (I) makes possible their facile filling with clusters (II) and molecules (III).

1688

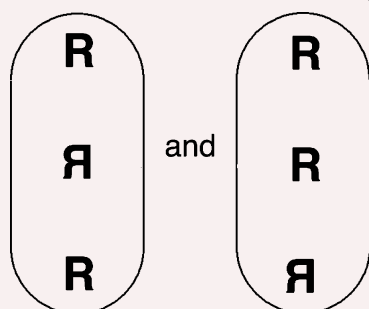


First observation of capping/uncapping by a ligand of a Zn porphyrin adsorbed on Ag(100)

Federico J. Williams, Owain P. H. Vaughan, Kerry J. Knox, Nick Bampos and Richard M. Lambert*

STM results provide the first observation of capping/uncapping by a ligand, 1,4-diazabicyclo[2.2.2]octane (DABCO), of Zn tetra[3,5-di-*t*-butylphenyl]porphyrin (Zn-TBPP) adsorbed on Ag(100).

1690

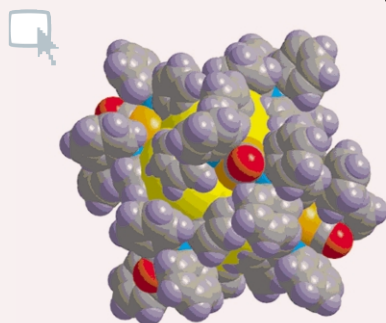


Constellational diastereomers in encapsulation complexes

Masamichi Yamanaka and Julius Rebek, Jr.*

A cylindrical host binds three small molecule guests. Restricted motion of the guests lead to isomeric arrangements; with chiral guests these arrangements are supramolecular diastereomers.

1692

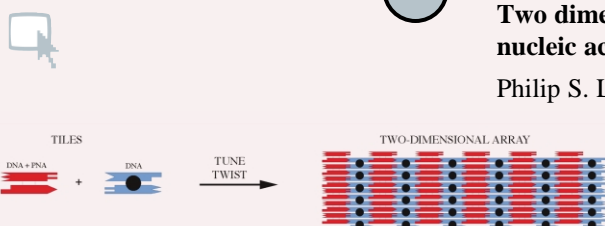


$[\text{Re}_6\text{Te}_8(\text{CN})_6][\{\text{Ir}(\text{CO})(\text{PPh}_3)_2\}_6](\text{OTf})_2$: a new Re_6 cluster-supported iridium(I) compound

Youngmee Kim, Sehye Kim, Sung-Jin Kim,* Myung Ki Lee, Mieock Kim, Hyungeui Lee and Chong Shik Chin*

A new hexanuclear rhenium cluster encapsulated by six iridium complexes, $[\text{Re}_6\text{Te}_8(\text{CN})_6][\{\text{Ir}(\text{CO})(\text{PPh}_3)_2\}_6](\text{OTf})_2$, which is effective in catalyzing the hydrogenation of $p\text{-CH}_3\text{C}_6\text{H}_4\text{C}\equiv\text{CH}$ to $p\text{-CH}_3\text{C}_6\text{H}_4\text{CH}=\text{CH}_2$ has been prepared.

1694



Two dimensional PNA/DNA arrays: estimating the helicity of unusual nucleic acid polymers

Philip S. Lukeman, Alexander C. Mittal and Nadrian C. Seeman*

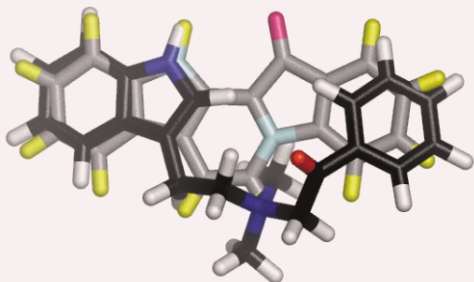
The generality of nucleic acid based structural nanotechnology is demonstrated by incorporating non-natural nucleic acids into a DNA double crossover (DX) tile; visualizing two-dimensional arrays of these tiles by Atomic Force Microscopy (AFM) enables us to measure the helical repeat of any heteroduplex sequence capable of forming the outer arms of a DX molecule.

1696

New fascaplysin-based CDK4-specific inhibitors: design, synthesis and biological activity

Carine Aubry, Paul R. Jenkins,* Sachin Mahale, Bhabatosh Chaudhuri, Jean-Didier Maréchal and Michael J. Sutcliffe

The first biologically active non-planar analogues of the toxic anti-cancer agent, fascaplysin, have been produced; we present the design, synthesis and biological activity of three tryptamine derivatives.

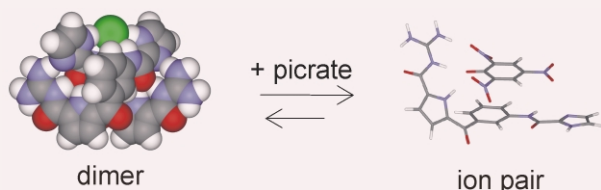


1698

Dimerization of a guanidiniocarbonyl pyrrole cation in DMSO that can be controlled by the counteranion

Carsten Schmuck* and Lars Geiger

The dimerization of cation **1** ($K_{\text{dim}} = 1080 \text{ M}^{-1}$ in DMSO for the chloride salt) can be switched off by the addition of picrate anions which form even more stable ion pairs with **1** ($K_{\text{ion}} = 2400 \text{ M}^{-1}$).

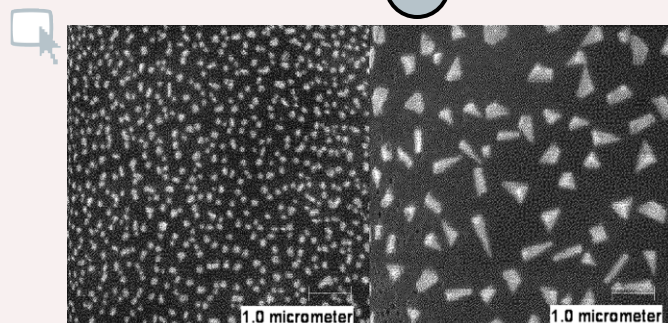


1700

Cruciform π -systems: effect of aggregation on emission

James N. Wilson, Mark D. Smith, Volker Enkelmann and Uwe H. F. Bunz*

The solid state properties of the novel cruciform pentamers **1–4** are examined in thin film preparation, in the single crystalline state and in nanoparticle formulations; emission behavior was found to vary substantially with the solid state morphology.

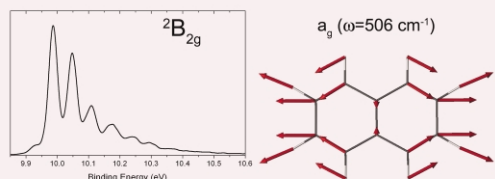


1702

Vibronic coupling in the ground and excited states of the naphthalene cation

Demetrio A. da Silva Filho, Rainer Friedlein, Veaceslav Coropceanu,* Gunnar Öhrwall, Wojciech Osikowicz, Christian Suess, Stacey L. Sorensen, Svante Svensson, William R. Salaneck and Jean-Luc Brédas*

The hole–vibrational coupling in naphthalene is studied using high-resolution gas-phase photoelectron spectroscopy and density functional theory calculations. A remarkable increase of the coupling with low-frequency vibrations is observed in the excited states.

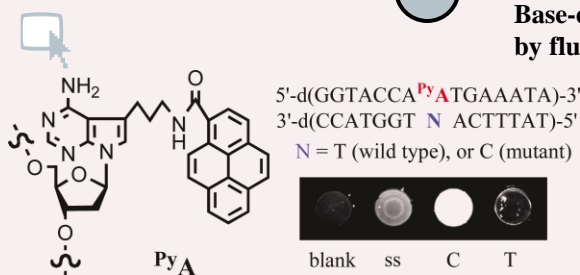


1704

Base-discriminating fluorescent (BDF) nucleoside: distinction of thymine by fluorescence quenching

Yoshio Saito, Yohei Miyauchi, Akimitsu Okamoto and Isao Saito*

A novel fluorescence BDF probe has been developed for the detection of thymine base on a target DNA.

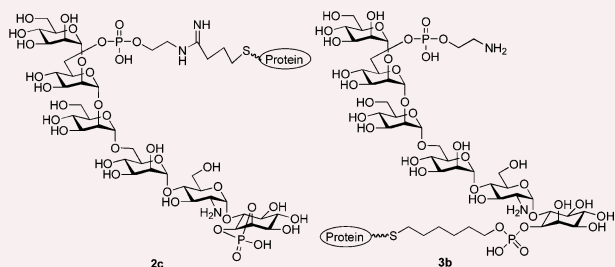


1706

A convergent, versatile route to two synthetic conjugate anti-toxin malaria vaccines

Peter H. Seeberger,* Regina L. Soucy, Yong-Uk Kwon, Daniel A. Snyder and Takuya Kanemitsu

Two molecules that serve to protect against a major menace of mankind—malaria—were synthesized *via* an efficient, scalable route.

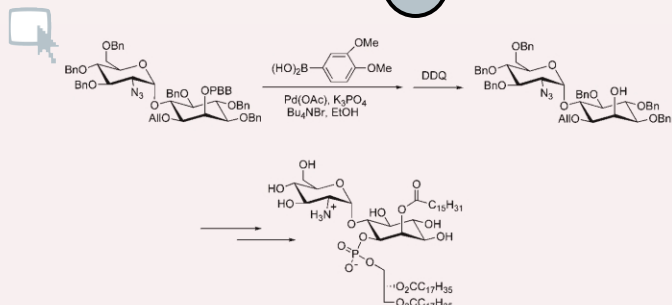


1708

A Suzuki–Miyaura coupling mediated deprotection as key to the synthesis of a fully lipidated malarial GPI disaccharide

Xinyu Liu and Peter H. Seeberger*

Ligandless palladium-catalyzed Suzuki–Miyaura coupling converted an inert *p*-bromobenzyl ether to a DDQ-labile *p*-(3,4-dimethoxyphenyl) benzyl ether in the presence of azide functionality and this strategy serves as a key step for the convergent synthesis of a fully lipidated malarial GPI disaccharide.

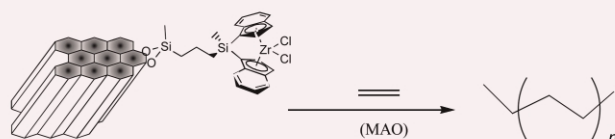


1710

Mesoporous silica-supported zirconocene catalysts for highly isotactic polypropylene

Catherine J. Miller and Dermot O'Hare*

New modified bis(indenyl)zirconocene catalysts have been grafted onto a range of mesoporous silicas. They are polymerisation catalysts for ethylene and propylene, producing polymers with very high molecular weights, low polydispersities and, in the case of polypropylene, higher levels of isotacticity than obtainable with analogous homogeneous systems.

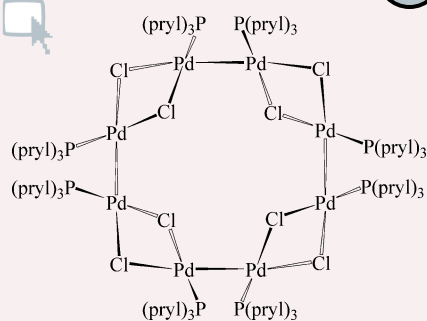


1712

Unprecedented eight-palladium(I) crown-cycle with metal–metal unsupported bonds

Inma Angurell, Isabel Martínez-Ruiz, Oriol Rossell, Miquel Seco,* Pilar Gómez-Sal and Avelino Martín

Strong π -acceptor tri(*N*-pyrrolyl)phosphine stabilizes a high nuclear Pd(I) cycle with unbridged Pd(I)–Pd(I).

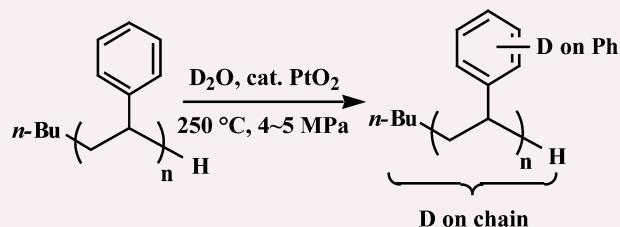


1714

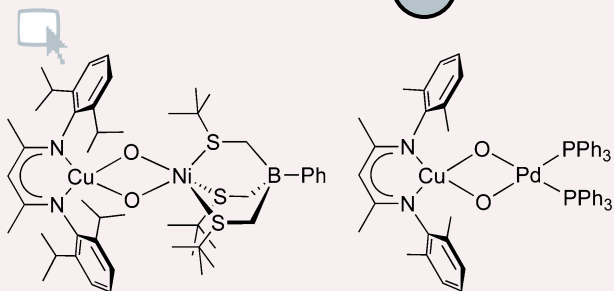
H–D exchange reaction on benzene ring of polystyrene in hydrothermal deuterium oxide with platinum(IV) oxide catalyst

Mitsuru Yamamoto, Yutaka Yokota, Koichiro Oshima and Seijiro Matsubara*

Polystyrene samples are labelled with deuterium oxide and catalytic amount of platinum(IV) oxide under hydrothermal conditions.



1716

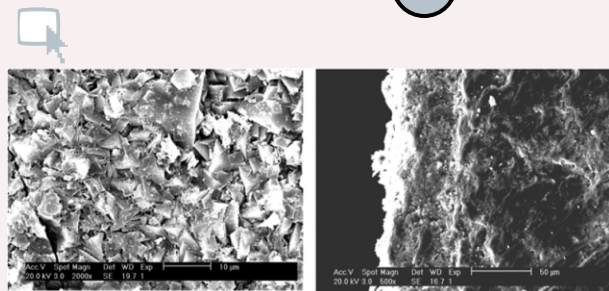


Mixed metal bis(μ -oxo) complexes with $[\text{CuM}(\mu\text{-O})_2]^{n+}$ ($\text{M} = \text{Ni(III)}$ or Pd(II)) cores

Nermeen W. Aboeella, John T. York, Anne M. Reynolds, Koyu Fujita, Christopher R. Kinsinger, Christopher J. Cramer,* Charles G. Riordan* and William B. Tolman*

Two heterodinuclear bis(μ -oxo) complexes have been prepared by combining mononuclear peroxo species with reduced metal precursors at $-80\text{ }^\circ\text{C}$ and characterized by spectroscopy and, for the CuPd system, DFT calculations.

1718

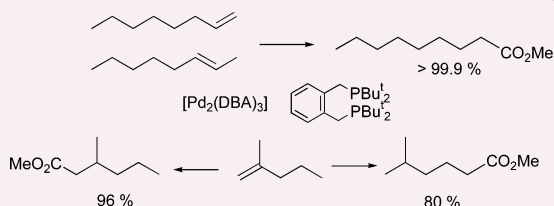


Novel synthesis of FAU-type zeolite membrane with high performance

Zhilin Cheng,* Enqing Gao and Huilin Wan

FAU-type zeolite membranes were successfully synthesized by vapor phase transformation methods with or without prior seeding on the substrate: the integrity of the seeded membrane is greater than that of the unseeded membrane.

1720

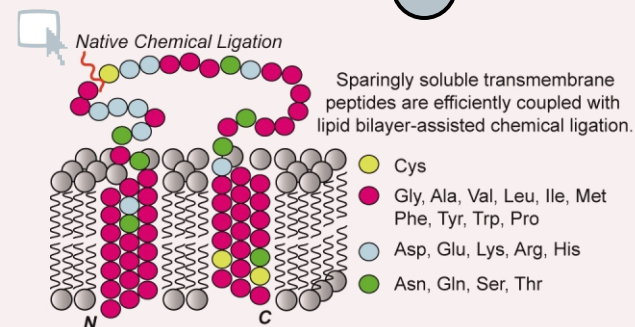


Highly selective formation of linear esters from terminal and internal alkenes catalysed by palladium complexes of bis-(di-*tert*-butylphosphinomethyl)benzene

Cristina Jimenez Rodriguez, Douglas F. Foster, Graham R. Eastham and David. J. Cole-Hamilton*

Terminal and internal alkenes are converted to linear methyl esters with almost quantitative selectivity by reaction with methanol and carbon monoxide in the presence of palladium complexes of bis-(di-*tert*-butylphosphinomethyl)benzene under very mild conditions.

1722



Facile synthesis of membrane-embedded peptides utilizing lipid bilayer-assisted chemical ligation

Akira Otaka,* Satoshi Ueda, Kenji Tomita, Yoshiaki Yano, Hirokazu Tamamura, Katsumi Matsuzaki and Nobutaka Fujii

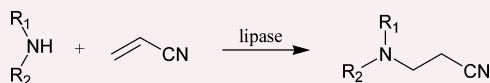
Lipid bilayer-assisted chemical ligation between thiolester and N-terminal cysteine peptides has been developed with successful application to the synthesis of membrane protein segments possessing both two transmembrane and one extracellular regions.

1724

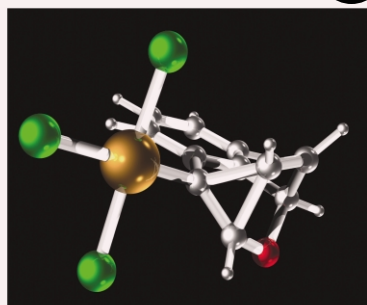
Lipase catalysed Michael addition of secondary amines to acrylonitrile

Oliver Torre, Ignacio Alfonso and Vicente Gotor*

Lipase B from *Candida antarctica* shows an aminolyase activity in the catalysis of Michael addition of secondary amines to acrylonitrile.



1726

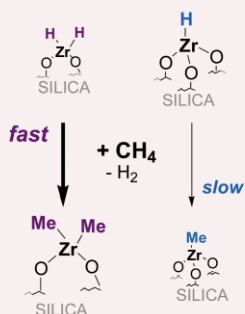


Gold(I) or gold(III) as active species in AuCl₃-catalyzed cyclization/cycloaddition reactions? A DFT study

Bernd F. Straub*

In Yamamoto's 2-alkynyl benzaldehyde plus alkyne benzannulation, a [3+2] cycloaddition of a carbonyl ylide plus ethyne with subsequent rearrangement (see picture) is predicted to occur instead of a direct [4+2] cycloaddition, for both Au(I) and Au(III).

1729

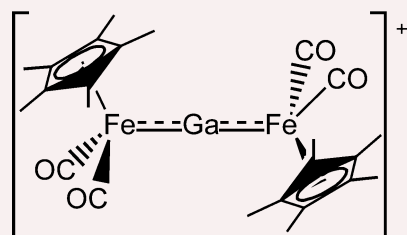


Methane activation by silica-supported Zr(IV) hydrides: the dihydride [(≡SiO)₂ZrH₂] is much faster than the monohydride [(≡SiO)₃ZrH]

Chloé Thieuleux,* Elsje Alessandra Quadrelli, Jean-Marie Basset,* Jens Döbler* and Joachim Sauer

When methane reacts with silica-supported Zr(IV) hydrides, it discriminates between monohydride and dihydride sites: despite the higher concentration of [(≡SiO)₃ZrH] in the starting material, the methylation is fast and complete only for [(≡SiO)₂ZrH₂].

1732

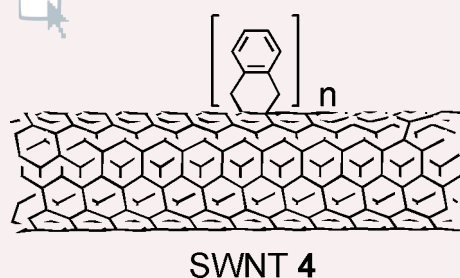


Fe–Ga multiple bonding? Synthesis, spectroscopic and structural characterization of a transition metal complex containing a cationic two-coordinate gallium centre

Natalie R. Bunn, Simon Aldridge,* Deborah L. Coombs, Andrea Rossin, David J. Willock, Cameron Jones and Li-ling Ooi

The cationic complex, [(η⁵-C₅Me₅)Fe(CO)₂]₂Ga⁺, containing a naked bridging gallium atom, can formally be regarded as a metalladiyl (L_nMGa:) complex of [(η⁵-C₅Me₅)Fe(CO)₂]⁺, but is best formulated as a delocalised Fe–Ga–Fe π system incorporating a partial Fe–Ga multiple bond character.

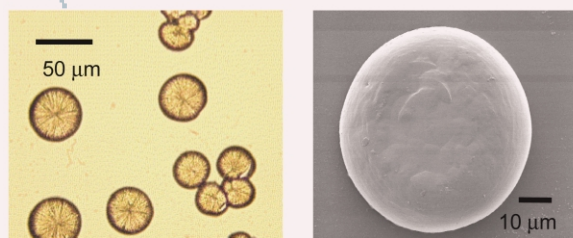
1734



Microwave-assisted sidewall functionalization of single-wall carbon nanotubes by Diels–Alder cycloaddition

Juan L. Delgado, Pilar de la Cruz, Fernando Langa,* Antonio Urbina, Juan Casado and Juan T. López Navarrete

1736

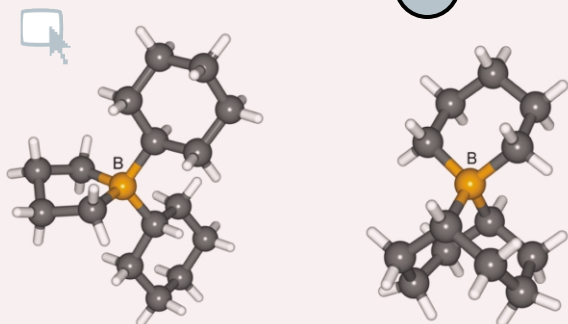


Design of a doubly-hydrophilic block copolypeptide that directs the formation of calcium carbonate microspheres

Larken E. Euliss, Tina M. Trnka, Timothy J. Deming and Galen D. Stucky*

The crystallization of calcium carbonate into microspheres has been accomplished using the rationally-designed, doubly-hydrophilic block copolypeptide poly{N_ε-2[2-(2-methoxyethoxy)ethoxy]acetyl-L-lysine}₁₀₀-b-poly(L-aspartate sodium salt)₃₀ as a structure-directing agent.

1738

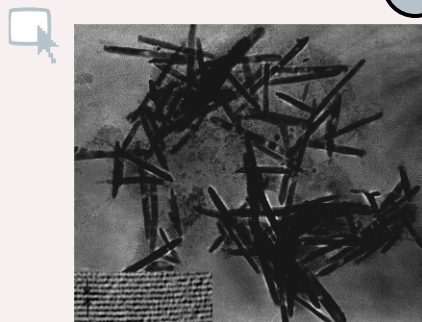


Novel organocycloborates *via* Grignard reagents

Holger Braunschweig,* Giovanni D'Andola, Tom Welton and Andrew J. P. White

An easy, versatile and unprecedented cyclisation reaction for the synthesis of novel tetraalkylborates is reported. The ring closure reaction occurs *via* an intra-molecular Grignard rearrangement at the boron centre.

1740

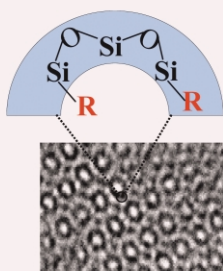


Mixed ligand system of cysteine and thioglycolic acid assisting in the synthesis of highly luminescent water-soluble CdTe nanorods

Jun Li, Xia Hong, Di Li, Kui Zhao, Lin Wang, Hongzhe Wang, Zuliang Du, Jinghong Li,* Yubai Bai* and Tiejun Li

Highly luminescent water-soluble CdTe nanorods were prepared with the assistance of the mixed ligand system of cysteine and thioglycolic acid. The aspect ratio and photoluminescence of the CdTe nanorods could be controlled by the refluxing time.

1742

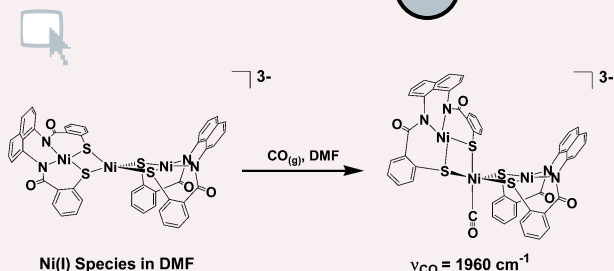


A general one-pot process leading to highly functionalised ordered mesoporous silica films

F. Cagnol, D. Grosso and C. Sanchez*

Various organic moieties are homogeneously introduced in high quantities into mesostructured porous silica films through a general co-condensation process, which influences the self assembly mechanism, depending on the physico-chemical properties of each function.

1744

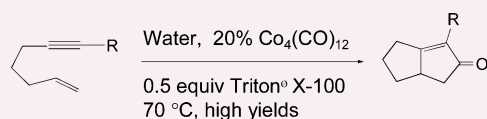


Binding of CO to structural models of the bimetallic subunit at the A-cluster of acetyl coenzyme A synthase/CO dehydrogenase

Todd C. Harrop, Marilyn M. Olmstead and Pradip K. Mascharak*

Trinuclear Ni–Cu–Ni and Ni–Ni–Ni complexes derived from an Ni(II)–dicarboxamido–dithiolato metallosynthons exhibit redox behavior and CO binding properties similar to those of the A-cluster in acetyl coenzyme A synthase/CO dehydrogenase (ACS/CODH).

1746



Catalytic activity of dodecacarbonyltetracobalt in aqueous media: a “greening” of the Pauson–Khand reaction

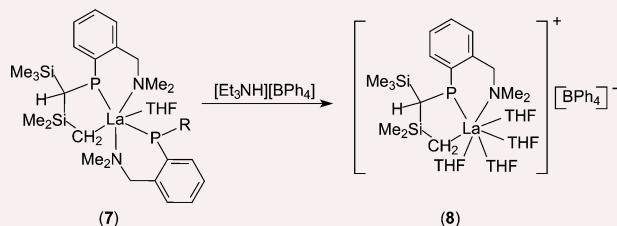
Llorente V. R. Boñaga, James A. Wright and Marie E. Krafft*

The unprecedented reactivity of $\text{Co}_4(\text{CO})_{12}$ with enynes under aqueous conditions, representing the development of a *mild and simple aqueous-phase cobalt-catalyzed* PK reaction protocol, is described.

1748

Protonation of a lanthanum phosphide-alkyl occurs at the P–La not the C–La bond: isolation of a cationic lanthanum alkyl complex

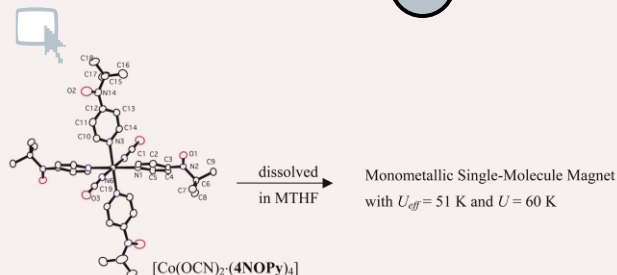
Keith Izod,* Stephen T. Liddle and William Clegg

Protonation of the heteroleptic lanthanum complex **7** yields the cationic lanthanum phosphide-alkyl complex **8**.

1750

Magnetic behavior of tetrakis[4-(*N*-*tert*-butyl-*N*-oxylamino)pyridine]bis(isocyanato-*N*)cobalt(II) in frozen solution

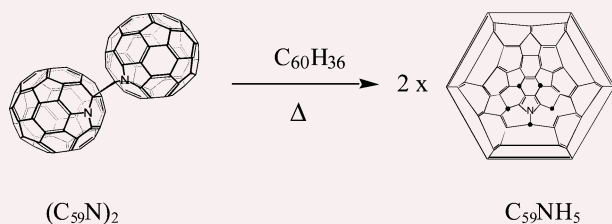
Shinji Kanegawa, Satoru Karasawa, Motohiro Nakano and Noboru Koga*

Tetrakis[4-(*N*-*tert*-butyloxylamino)pyridine]bis(isocyanato-*N*)cobalt(II), $[\text{Co}(\text{OCN})_2 \cdot (4\text{NOPy})_4]$, having four stable aminoxyl radicals in frozen MTHF solution functions as a monometallic single-molecule magnet with an activation barrier of $\Delta/k_B (=U_{\text{eff}}) = 50$ K for flipping the spin.

1752

Hydrogen storage on fullerenes: hydrogenation of C_{59}N^+ using $\text{C}_{60}\text{H}_{36}$ as the source of hydrogen

Yury V. Vasil'ev, Andreas Hirsch, Roger Taylor and Thomas Drewello*

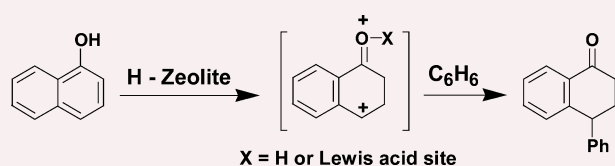
 $\text{C}_{60}\text{H}_{36}$ has been used as the source of hydrogen for the *in situ* hydrogenation of $(\text{C}_{59}\text{N})_2$, resulting in C_{59}NH_5 and providing evidence of the usage of C_{60} as a storage device for hydrogen.

1754

Superelectrophilic activation of polyfunctional organic compounds using zeolites and other solid acids

Konstantin Yu. Koltunov,* Stéphane Walspurger and Jean Sommer*

Solid acids instead of superacids may be successfully used to carry out some reactions proceeding through dicationic or dicharged intermediates.

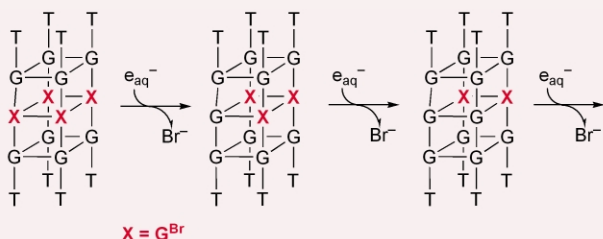


1756

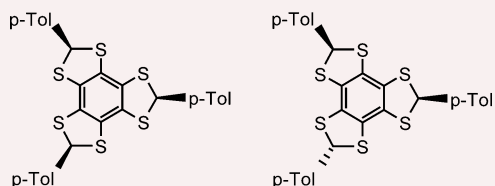
Excess electron transfer in G-quadruplex

Marcella de Champdoré, Lorenzo De Napoli, Daniela Montesarchio, Gennaro Piccialli, Clara Caminal, Quinto G. Mulazzani, Maria Luisa Navacchia and Chrystostomos Chatgililoglu*

The excess electron transfer in a G-quadruplex is successfully probed by using the reaction of hydrated electrons with quadruplex complex of pentamers and the 8-bromoguanine moieties as the detection system.



1758

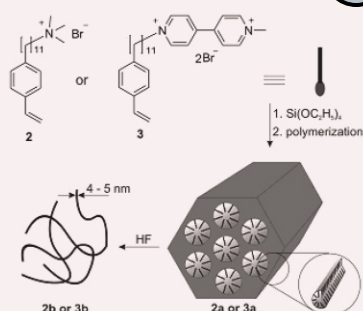


Tris(thioacetals) from benzene hexathiol: towards covalent self-assembly

Liam R. Sutton,* Wolfgang A. Donaubaer, Frank Hampel and Andreas Hirsch*

Lewis acid catalysis enables the synthesis of model compounds for covalent self-assembly using thioacetals – but is the system truly reversible?

1760

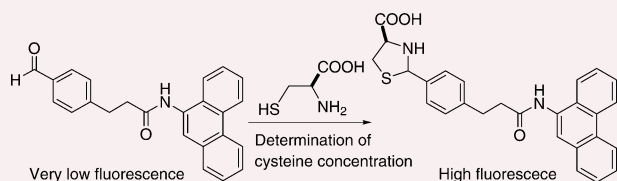


Template synthesis of functionalized polystyrene in ordered silicate channels

Guangtao Li,* Sheshanath Bhosale, Sidhanath Bhosale, Fengting Li, Yihe Zhang, Ruirong Guo, Hesun Zhu and Jurgen-Hinrich Fuhrhop*

A mesostructured nanocomposite was fabricated by using a novel electroactive, polymerizable surfactant as a template in a sol-gel process, and separated polystyrene wires with redoxactive functional groups were synthesized in silicate matrices.

1762

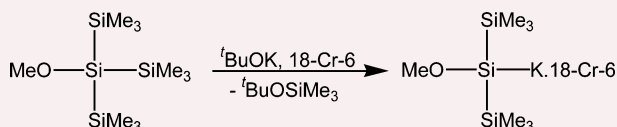


Determination of cysteine concentration by fluorescence increase: reaction of cysteine with a fluorogenic aldehyde

Fujie Tanaka,* Nobuyuki Mase and Carlos F. Barbas III*

A fluorogenic method for the determination of cysteine concentration based on the reaction of cysteine with a fluorogenic aldehyde is described. This method selectively detected cysteine and not other amino acids and thiols.

1764

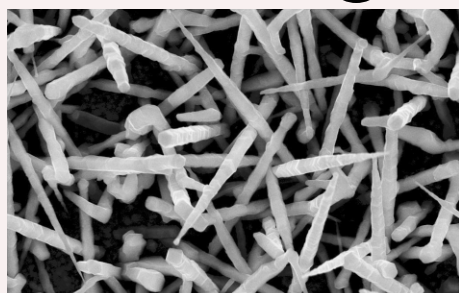


Preparation and structural characterisation of methoxybis(trimethylsilyl)silyl potassium and its condensation product

Pravin R. Likhar, Michaela Zirngast, Judith Baumgartner and Christoph Marschner*

Reaction of tris(trimethylsilyl)methoxysilane with potassium *tert*-butoxide gives the silylenoid compound $\text{MeO}(\text{Me}_3\text{Si})_2\text{SiK}$ which either can be isolated as the crown-ether adduct, or undergoes self-condensation to give a β -methoxysilyl anion.

1766

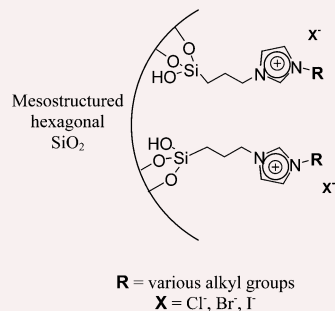


Glow discharge growth of SnO₂ nano-needles from SnH₄

Chun-Fang Wang, Su-Yuan Xie,* Shui-Chao Lin, Xuan Cheng, Xian-Hua Zhang, Rong-Bin Huang* and Lan-Sun Zheng

Needle-shaped semiconductor SnO₂ with lateral size of ~300 nm and length up to 6–7 μm are grown in a glow discharge process from gaseous SnH₄ fed at a decreasing mode.

1768

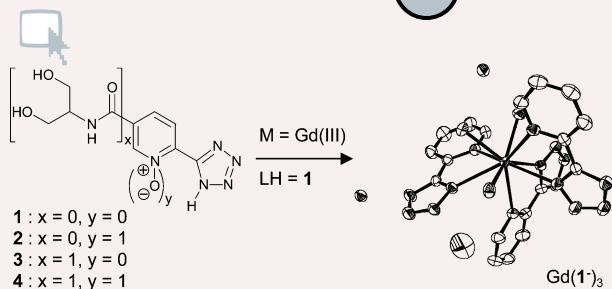


Supported ionic liquids: ordered mesoporous silicas containing covalently linked ionic species

Benoit Gadenne, Peter Hessemann and Joël J. E. Moreau*

Ordered mesoporous silicas with hexagonal or lamellar architectures incorporating covalently bound ionic species were synthesized *via* a template directed hydrolysis–polycondensation of TEOS with triethoxysilylated imidazole [(EtO)₃Si(CH₂)₃–Im] or alkylimidazolium halides [(EtO)₃Si(CH₂)₃–Im⁺–R Hal⁻].

1770

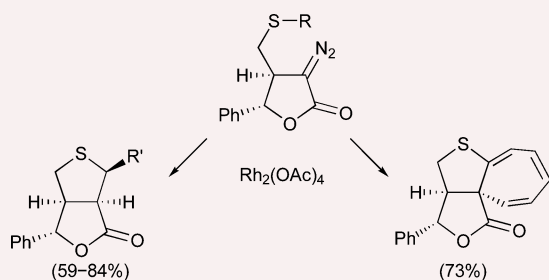


Novel coordinating motifs for lanthanide(III) ions based on 5-(2-pyridyl)tetrazole and 5-(2-pyridyl-1-oxide)tetrazole. Potential new contrast agents

Antonio Facchetti,* Alessandro Abboto, Luca Beverina, Silvia Bradamante, Palma Mariani, Charlotte L. Stern, Tobin J. Marks, Alberto Vacca and Giorgio A. Pagani*

Water-soluble Gd(III) complexes of pyridine- and (pyridine-1-oxide)tetrazole ligands **1–4** have been synthesized and have great potential as high-relaxivity MRI contrast agents.

1772

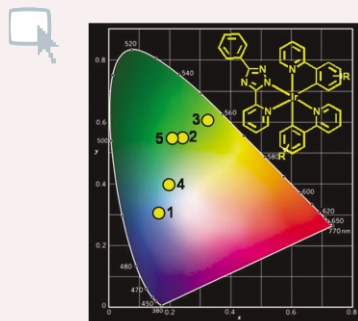


Intramolecular C–H insertions adjacent to sulfur for the diastereoselective synthesis of thienofuranones

Paul S. Skerry, Nigel A. Swain, David C. Harrowven,* Donald Smyth, Gordon Bruton and Richard C. D. Brown*

A new approach to the diastereoselective synthesis of thienofuranones is described in which an intramolecular 1,5-carbenoid C–H insertion adjacent to sulfur features as a key step.

1774

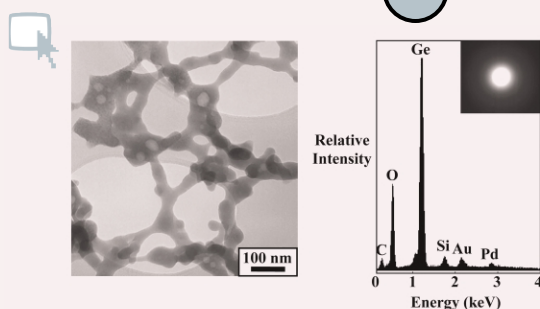


Tuning iridium(III) phenylpyridine complexes in the “almost blue” region

Paolo Coppo, Edward A. Plummer and Luisa De Cola*

A facile way to prepare hetero-leptic iridium(III) phenylpyridine complexes and rationally tune the light emission in the “almost blue” region of the spectrum is discussed.

1776



Identification of peptides that promote the rapid precipitation of germania nanoparticle networks *via* use of a peptide display library

Matthew B. Dickerson, Rajesh R. Naik, Morley O. Stone, Ye Cai and Kenneth H. Sandhage

The rapid room-temperature precipitation of amorphous germania nanoparticles, that may be used in a host of optical or chemical applications, has been achieved through the activity of specific peptides isolated from a phage display library.

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