**Cover**

See Stuart L. James and Qiang Wei, page 1555.

This communication reports a novel use for metal-organic framework chemistry. Macroporous gels based on these structures are used as polymerisation templates. Image produced by permission of Qiang Wei and Stuart L. James from *Chem. Commun.*, 2005, 1555.

**Inside cover**

See Stephen Loeb, page 1511.

Mechanically interlocked, molecular components can be organized into one-, two-, and three dimensional arrays by utilizing them as ligand building blocks in metal-organic frameworks. Image reproduced by permission of Stephen Loeb from *Chem. Commun.*, 2005, 1511.

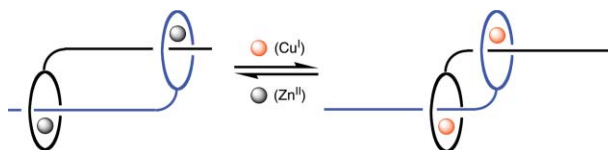
40TH ANNIVERSARY ARTICLE

1507

Transition metal-complexed catenanes and rotaxanes as molecular machine prototypes

Jean-Pierre Sauvage

Various recently prepared copper- or ruthenium(II)-complexed catenanes and rotaxanes can be regarded as molecular machine prototypes; the systems are set in motion by using redox, chemical or photonic signals.



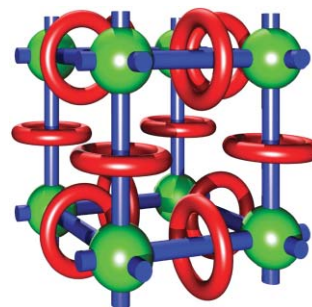
FEATURE ARTICLE

1511

Metal-organic rotaxane frameworks; MORFs

Stephen J. Loeb

Metal-organic framework (MOF) materials can be constructed which contain rotaxanes as the organic linker and transition or lanthanide metal ions as the connecting nodes. These 1-, 2- and 3D metal-organic rotaxane frameworks (MORFs) contain ordered arrays of mechanically interlocked units and may be the forerunners of unique solids which contain machine-like components.



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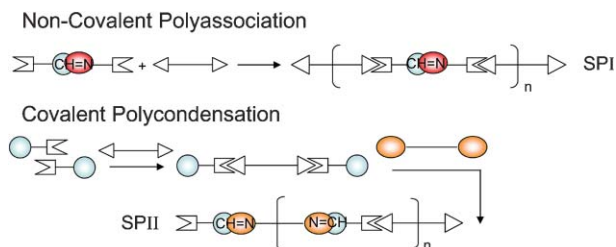
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1519

Double dynamers: molecular and supramolecular double dynamic polymers

Elena Kolomiets and Jean-Marie Lehn*

The formation of double dynamers, polymers incorporating both non-covalent and reversible covalent connections and combining polyassociation with polycondensation of components, was achieved in solution.

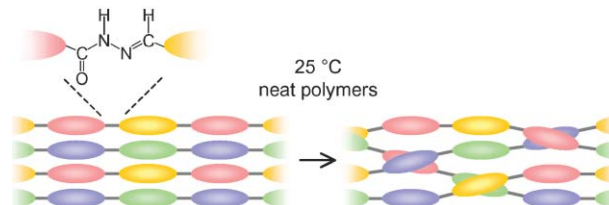


1522

Dynamic polymer blends—component recombination between neat dynamic covalent polymers at room temperature

Takashi Ono, Tadahito Nobori and Jean-Marie Lehn*

Polymers based on acylhydrazone bonds in the linear main chains are capable of exchanging their bonds mutually leading to crossover component recombination as neat polymers even at room temperature.

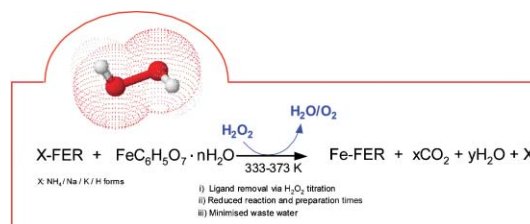


1525

Ion exchanged Fe-FER through H₂O₂-assisted decomplexation of organic salts

I. Melián-Cabrera,* S. Espinosa, F. J. García-Montelongo, F. Kapteijn* and J. A. Moulijn

A green preparation method for Fe-FER catalysts is presented. The method is based on the decomplexation of ferric citrate by redox titration using H₂O₂. Short reaction times with reduced waste water are also achieved.

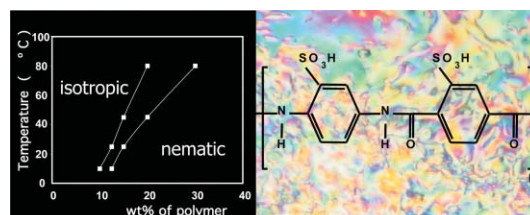


1528

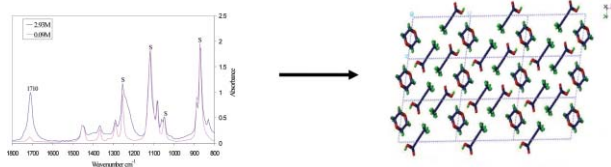
Formation of aqueous molecular nematic liquid crystal phase in poly(*p*-sulfophenylene sulfoterephthalamide)

S. Viale, A. S. Best, E. Mendes* and S. J. Picken

We report strong experimental evidence on the formation of a molecular lyotropic phase in water of a modified polyaramide. A fully sulfonated poly-*p*-phenylene terephthalamide (sulfo² PPTA) exhibits similar liquid crystalline behavior in water to its non-sulfonated counterpart (PPTA, commercially known as Kevlar® or Twaron®) in sulfuric acid.



1531

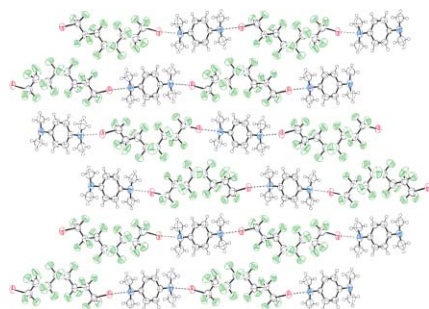


Linking solution chemistry to crystal nucleation: the case of tetrolic acid

S. Parveen, R. J. Davey,* G. Dent and R. G. Pritchard

The application of FTIR spectroscopy to concentrated solutions of tetrolic acid shows, for the first time, a direct relationship between molecular self association in solution and H-bonded motifs in the subsequently crystallised solid phases.

1534

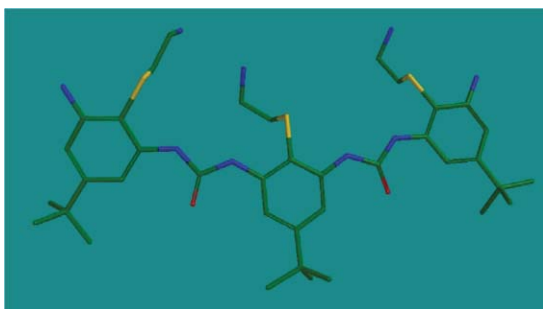


Spontaneous resolution in a halogen bonded supramolecular architecture

Hannes Neukirch, Emanuela Guido, Rosalba Liantonio, Pierangelo Metrangolo,* Tullio Pilati and Giuseppe Resnati*

The first case of spontaneous resolution under the control of halogen bonding is reported. The X-ray structure of the halogen bonded homochiral co-crystal of *N,N,N',N'*-tetramethyl-*p*-phenylenediamine with 1,8-diiodoperfluorooctane is described.

1537

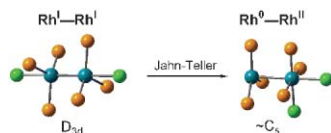


Synthesis of urea oligomers and their antibacterial activity

Haizhong Tang, Robert J. Doerksen* and Gregory N. Tew*

A series of facially amphiphilic oligomeric ureas were prepared in one-pot reactions by CDI coupling. The resulting compounds have potent antibacterial activity against both Gram-negative and Gram-positive bacteria.

1540



A model for two-electron mixed valence in metal–metal bonded dirhodium compounds

Thomas G. Gray and Daniel G. Nocera*

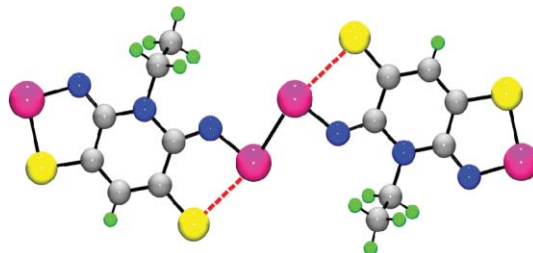
Two-electron mixed valence of bimetallic complexes possessing three bridging ligands results from a second-order Jahn–Teller instability of the redox-symmetric state.

1543

Bis-1,2,3-thiaselenazoly radicals and their σ -bonded dimers

Leanne Beer, Jaclyn L. Brusso, Robert C. Haddon, Mikhail E. Itkis, Alicea A. Leitch, Richard T. Oakley,* Robert W. Reed and John F. Richardson

The resonance stabilized *bis*-thiaselenazoly π -radical **2** associates in the solid state *via* a Se–Se σ -bond to produce a closed shell material.

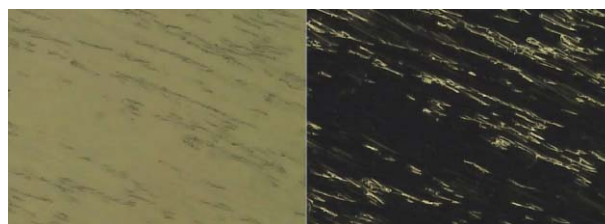


1546

A chiral electrooptic response in a racemic liquid crystal

Stephen J. Cowling, Alan W. Hall and John W. Goodby*

Little attention has been paid to the electrooptic properties of racemic modifications of liquid crystals. In this communication we report a chiral response of a racemate to an applied electrical field while in its achiral smectic C phase.

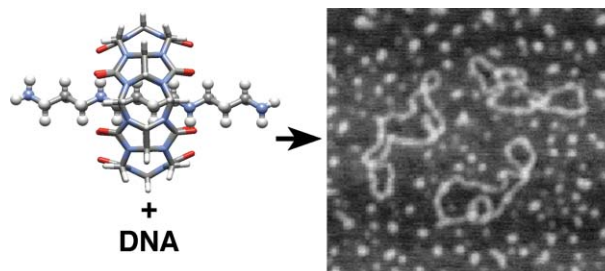


1549

Supramolecular modulation of action of polyamine on enzyme/DNA interactions

Hiroyuki Isobe, Sota Sato, Jae Wook Lee, Hee-Joon Kim, Kimoon Kim and Eiichi Nakamura*

Thread-in-hole complexation of cucurbit[6]uril (CB[6]) with spermidine and spermine either enhances or reduces the activity of the polyamine on the enzymatic reactions of DNA.

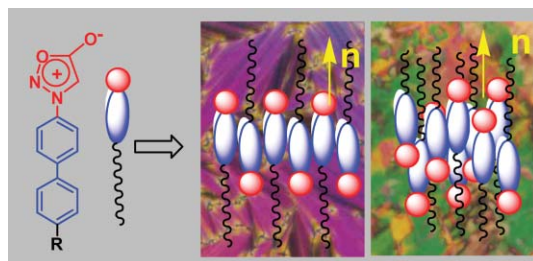


1552

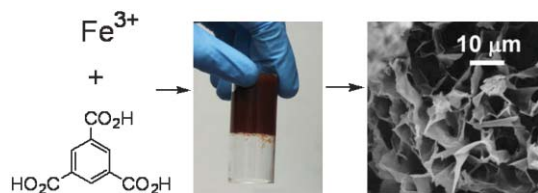
Self-organization of mesomeric–ionic hybrid heterocycles into liquid crystal phases: a new class of polar mesogens

Channabasaveshwar V. Yelamaggad,* Manoj Mathews, Uma S. Hiremath, Doddamane S. Shankar Rao and Subbarao Krishna Prasad

Sydnone, important members of a novel class of mesoionic (mesomeric + ionic) heterocycles, display mesomorphism. The nematic and/or smectic behavior of such materials has been unambiguously evidenced for the first time by optical, calorimetric and X-ray studies.



1555

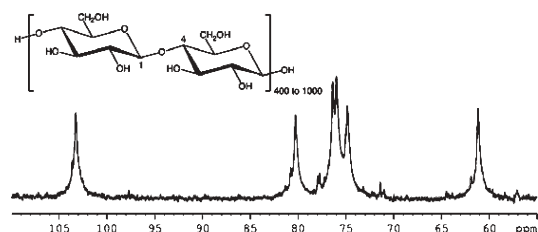


A metal–organic gel used as a template for a porous organic polymer

Qiang Wei and Stuart L. James*

Very little is known about coordination polymer gels. Here we demonstrate the novel use of a metal–organic gel to template macropores within polymethylmethacrylate.

1557

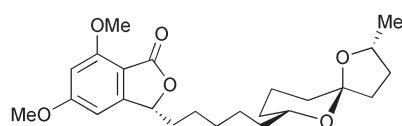


High-resolution ^{13}C NMR studies of cellulose and cellulose oligomers in ionic liquid solutions

Jason S. Moulthrop, Richard P. Swatloski, Guillermo Moyna* and Robin D. Rogers*

The β -(1 \rightarrow 4)-linked glucose oligomers are disordered in 1-butyl-3-methylimidazolium chloride and have a conformational behavior which parallels the one observed in water: the polymer is also disordered in IL solution.

1560

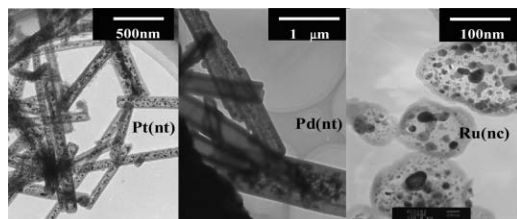


The first enantioselective total synthesis of the anti-*Helicobacter pylori* agent (+)-spiroloxine methyl ether

James E. Robinson and Margaret A. Brimble*

The first enantioselective synthesis of spiroloxine methyl ether is reported in a convergent manner using a Julia–Kocienski olefination in the key step.

1563



Unique adsorption behavior of H_2 and CO over group 8–10 metals encapsulated inside silica nanotubes and nanocapsules

Shuichi Naito,* Masato Ue, Seiko Sakai and Toshihiro Miyao

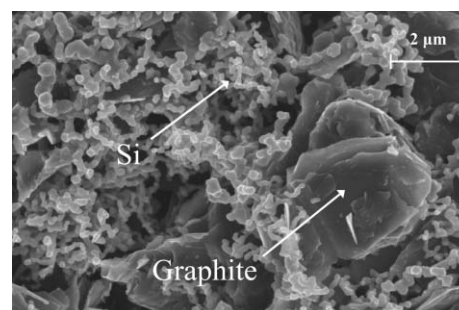
Silica nanocomposites, which encapsulate group 8–10 metal particles, reveal that adsorbed $\text{H}(\text{a})$ and $\text{CO}(\text{a})$ are markedly stabilized over the metal atoms in the network of the silica wall and inside nanoscale cavities.

1566

A new type of nano-sized silicon/carbon composite electrode for reversible lithium insertion

Michael Holzapfel,* Hilmi Buqa, Werner Scheifele, Petr Novák and Frank-Martin Petrat

A new type of nano-sized silicon/carbon composite was developed. It shows superior electrochemical cycling properties as negative electrode material for possible use in lithium-ion batteries with respect to high reversible and low irreversible capacity, and low fading.

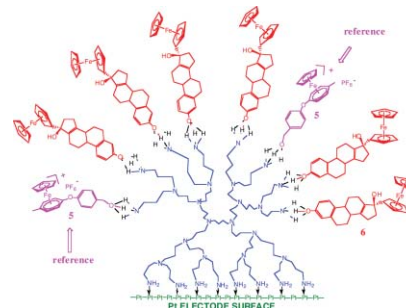


1569

Inhibition of the electrochemistry of ferrocenes by polyamine dendrimers and the key role of hydrogen-bonding with hydroxy groups

Marie-Christine Daniel, Jaime Ruiz Aranzaes and Didier Astruc*

Commercial DSM polyamine dendrimers inhibit the observation of the cyclic voltammetry (CV) of simple ferrocenes all the more as they are larger (marked dendritic effect); the CV is re-established, however, if a OH-containing group is present in the ferrocene derivative.

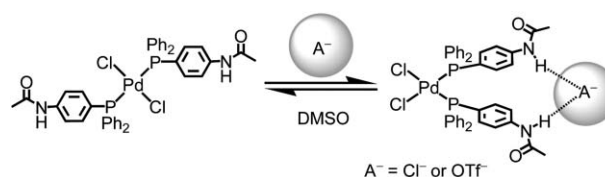


1572

Anion-assisted *trans*-*cis* isomerization of palladium(II) phosphine complexes containing acetanilide functionalities through hydrogen bonding interactions

Xiao-Xia Lu, Hau-San Tang, Chi-Chiu Ko, Jenny Ka-Yan Wong, Nianyong Zhu and Vivian Wing-Wah Yam*

The anion-assisted shift of *trans*-*cis* isomerization equilibrium of a palladium(II) complex containing acetanilide functionalities brought about by allosteric hydrogen bonding interactions has been established by UV/Vis, ^1H NMR, ^{31}P NMR and ESI-MS studies.

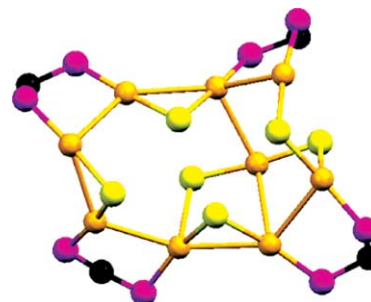


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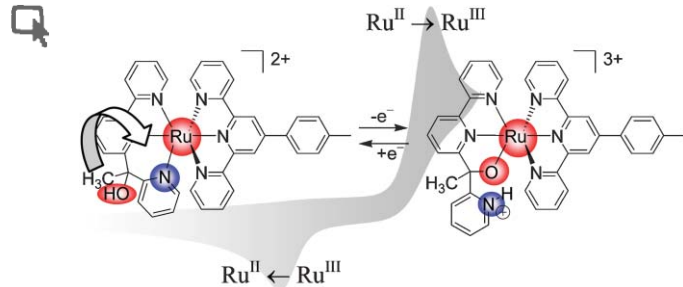
Novel metallamacrocyclic gold(I) thiolate cluster complex: structure and luminescence of $[\text{Au}_9(\mu\text{-dppm})_4(\mu\text{-p}\text{-tc})_6](\text{PF}_6)_3$

Jinhua Chen, Ahmed A. Mohamed, Hanan E. Abdou, Jeanette A. Krause Bauer, John P. Fackler, Jr.,* Alice E. Bruce* and Mitchell R. M. Bruce*

The first phosphine gold(I) thiolate cluster of high nuclearity is reported. The average $\text{Au}\cdots\text{Au}$ distance is ~ 3 Å. The metallamacrocycle is comprised of nine gold atoms linked by $\text{Au}\cdots\text{Au}$ intramolecular attractions, bridging dppm, and bridging thiolate ligands.



1578

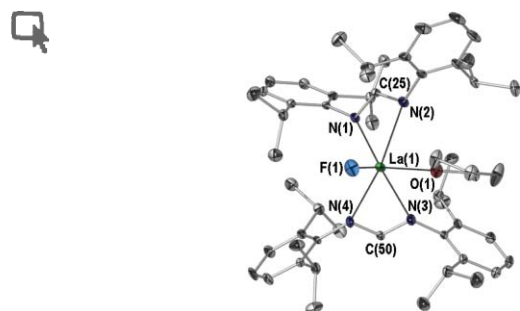


Rapid electrochemically induced linkage isomerism in a ruthenium(II) polypyridyl complex

Olof Johansson* and Reiner Lomoth*

Molecular hysteresis: In a bistridentate heteroleptic ruthenium(II) polypyridyl complex the electrochemically induced switching between the N_6 donor set and the N_5O analogue results in a hysteresis-like electrochemical behavior. The isomerization reactions are rapid and complete in both directions. The linkage isomerism of this new structural motif may eventually provide the basis for a molecular memory.

1581

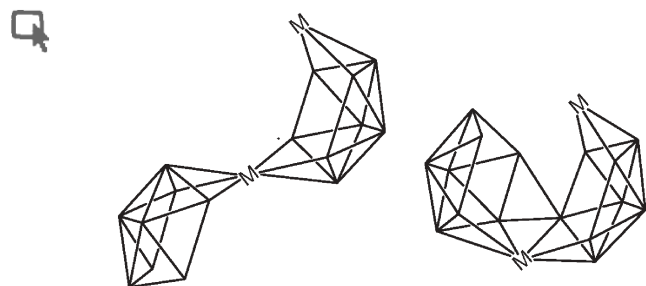


Steric engineering of C–F activation with lanthanoid formamidinates

Marcus L. Cole, Glen B. Deacon, Peter C. Junk* and Kristina Konstas

Reaction of lanthanum with $\text{Hg}(\text{C}_6\text{F}_5)_2$ and bulky N,N' -bis(2,6-diisopropylphenyl)formamidine (HDippForm) in tetrahydrofuran gives $[\text{LaF}\{\text{DippForm}\}_2(\text{THF})]$ with a rare terminal Ln–F bond, and a high yield of a novel functionalised formamidine, $\text{DippForm}((\text{CH}_2)_4\text{OC}_6\text{F}_4\text{H}-o)$.

1584

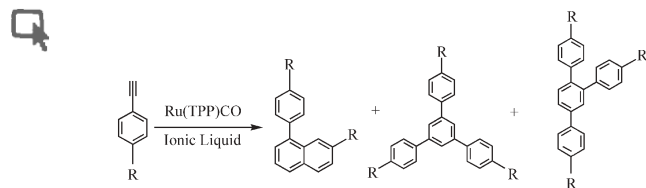


Macropolyhedral boron-containing cluster chemistry. Synchrotron X-ray structural analysis of $[(\text{PMe}_2\text{Ph})_2\text{Pd}_2\text{B}_{16}\text{H}_{20}(\text{PMe}_2\text{Ph})_2]$ and $[(\text{PMe}_2\text{Ph})_3\text{Pt}_2\text{B}_{16}\text{H}_{18}(\text{PMe}_2\text{Ph})]$: models of intermediates to more condensed metallaboranes from the $[(\text{PMe}_2\text{Ph})_2\text{PtB}_8\text{H}_{12}]$ thermolysis system

Michael G. S. Londesborough, Elizabeth J. MacLean, Simon J. Teat, Mark Thornton-Pett and John D. Kennedy

The title compounds are snapshot cluster models for initial steps in mechanisms towards more condensed macropolyhedral species.

1587



The cyclooligomerization of arylothyne in ionic liquids catalysed by ruthenium porphyrins: a case of real catalyst recycling

Valeria Conte,* Elfituri Elakkari, Barbara Floris, Valentina Mirruzzo and Pietro Tagliatesta*

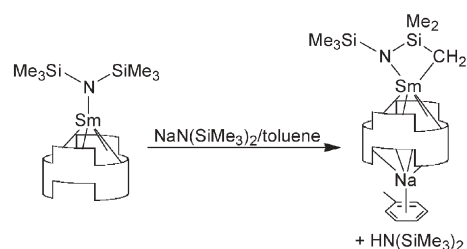
The first cyclooligomerization of arylothyne catalysed by metalloporphyrins in ionic liquids to give benzene and naphthalene derivatives with real catalyst recycling.

1589

C–H Activation (γ -deprotonation) of a Sm(III) bis(trimethylsilyl)amide complex *via* macrocyclic stabilisation of the sodium counter ion

Jun Wang and Michael G. Gardiner*

The divergent receptor characteristics of the dimetallated *trans*-dioxaporphyrinogen allows deprotonation of a samarium(III) bis(trimethylsilyl)amide complex through stabilisation of the Na counter cation within the macrocyclic cavity.

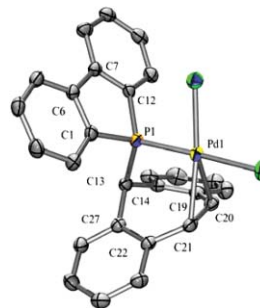


1592

Dibenzo[*a,d*]cycloheptenyl dibenzophosphole palladium dichloride: Synthesis, X-ray-crystal structure and application in the Suzuki–Miyaura coupling

Claire Thoumazet, Louis Ricard, Hansjörg Grützmacher and Pascal Le Floch*

Synthesis of a highly rigid dibenzo[*a,d*]cycloheptenyl dibenzophosphole ligand featuring both a phosphole and an olefin as binding sites is presented. This new bidentate works as an efficient ligand in the catalyzed Suzuki–Miyaura cross-coupling process (TON up to 9.9×10^5).



1595

Syntheses and molecular structures of η^3 -[(η^5 -Cp*) $(\text{CO})_2\text{Fe-AsPC}(\text{SiMe}_3)_2$]M(CO) $_2$ (η^5 -Cp) (M = Mo, W): the first complexes featuring η^3 -arsaphosphaallyl ligands

Lothar Weber,* Philipp Bayer, Hans-Georg Stammer and Beate Neumann

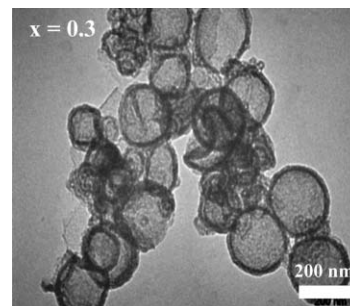
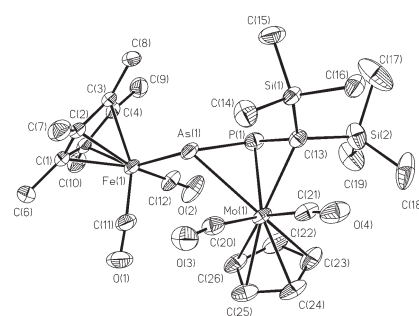
Reaction of the phosphavinylidene complexes (η^5 -C₅H₅)(CO)₂M=P=C(SiMe₃)₂ (M = Mo, W) with metalloarsaalkene (η^5 -C₅Me₅)(CO)₂FeAs=C(NMe₂)₂ afforded the novel η^1 -1-arsa-2-phosphaallyl complexes [(η^5 -C₅Me₅)(CO)₂FeAsPC(SiMe₃)₂]M(CO)₂(C₅H₅), the molecular structures of which were determined by X-ray analyses.

1598

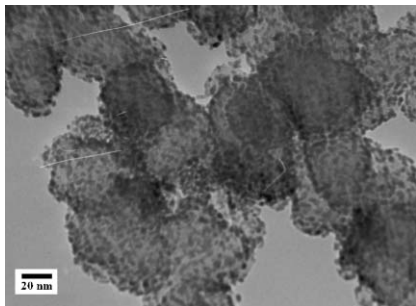
Thiol-functionalized mesostructured silica vesicles

Jainisha Shah and Thomas J. Pinnavaia*

The direct supramolecular assembly of organofunctional mesostructures with a vesicular hierarchical morphology is reported for the first time for (SiO₂)_{1-x}(LSiO_{1.5})_x compositions, where L is a mercaptopropyl group and $x = 0.10$ – 0.30 .



1601

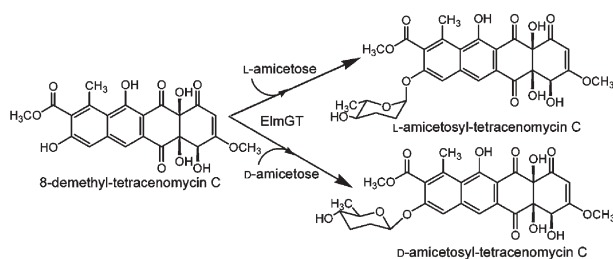


Simple and controllable synthesis of highly dispersed Pt–Ru/C catalysts by a two-step spray pyrolysis process

Xinzhong Xue, Tianhong Lu, Changpeng Liu and Wei Xing*

A carbon supported Pt–Ru catalyst with uniform distribution and small average size of the Pt–Ru particles was synthesized using a two-step spray pyrolysis (SP) method. The electrocatalytic activity of the prepared catalyst for methanol oxidation is better than that of the standard commercial catalyst.

1604

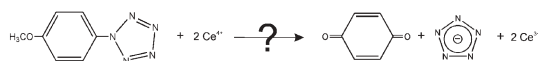


Combining sugar biosynthesis genes for the generation of L- and D-amictose and formation of two novel antitumor tetracenomycins

María Pérez, Felipe Lombó, Lili Zhu, Miranda Gibson, Alfredo F. Braña, Jürgen Rohr, José A. Salas and Carmen Méndez*

By combining sugar biosynthesis genes from different antibiotic pathways L- and D-amictose were produced and transferred by the sugar flexible glycosyltransferase ElmGT to the elloramycin aglycone.

1607

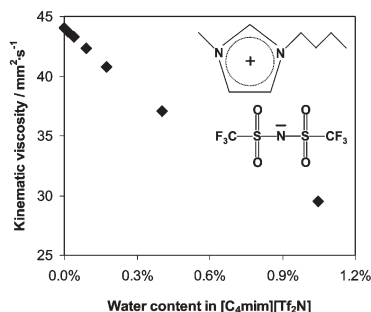


The race for the first generation of the pentazolone anion in solution is far from over

Thorsten Schroer,* Ralf Haiges, Stefan Schneider and Karl O. Christe*

The previous claim for the first generation of the pentazolone anion in solution was carefully reexamined. No evidence for the formation of *cyclo*-N₅[−] was found under the reported conditions. The ¹⁵N NMR signal at $\delta = -10 \pm 2$ ppm, previously assigned to *cyclo*-N₅[−], belongs to NO₃[−], and the one at $\delta = -147.2$ ppm, previously attributed to the crucial decomposition product, N_β-labeled N₃[−], is due to 4-MeOC₆H₄N₃, one of the decomposition products of the starting material.

1610



The effect of dissolved water on the viscosities of hydrophobic room-temperature ionic liquids

Jason A. Widegren, Arno Laesecke and Joseph W. Magee*

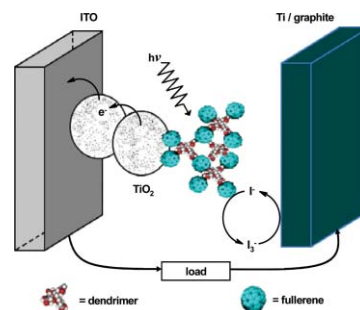
The viscosities of room-temperature ionic liquids (RTILs) are found to be strongly dependent on the amount of dissolved water. The presence of 100 ppm water can lower the viscosity of an RTIL by 1%. These results call for a reassessment of previously reported viscosities for RTILs.

1613

Preparation, characterization and photoelectrochemical study of mixed C₆₀–Starburst[®] PAMAM G0.0 dendrimer films anchored on the surface of nanocrystalline TiO₂ semiconductor electrodes

Erika Bustos, Juan Manríquez, Luis Echegoyen and Luis A. Godínez*

Nanocrystalline TiO₂ silanized electrodes were modified in a sequential fashion with C₆₀ and Starburst[®] PAMAM G0.0 dendrimers, resulting in a novel photoelectrochemical sensitization film that showed particularly high photocurrent (IPCE) and global photoconversion efficiencies (η).

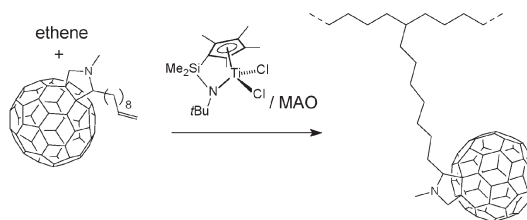


1616

Polyethene with pendant fullerene moieties

Xiaochun Zhang, Alexander B. Sieval, Jan C. Hummelen and Bart Hessen*

A novel polyethene copolymer bearing pendant fullerene groups, with up to 25 wt% net fullerene content, was prepared by catalytic copolymerisation of ethene with a fullerene-containing vinylic comonomer.

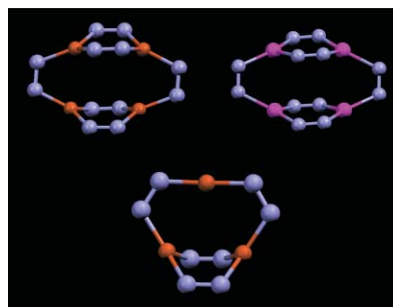


1619

Neutral Cu₄N₁₂ and Ag₄N₁₂ metallacycles with a *para*-cyclophane framework assembled from copper(I) and silver(I) pyrazolates and pyridazine

H. V. Rasika Dias,* Himashinie V. K. Diyabalanage and Chammi S. Palehepitiya Gamage

Novel copper–nitrogen and silver–nitrogen based *para*-cyclophanes have been synthesized using the corresponding metal(I) pyrazolates and pyridazine. The use of benzo[*c*]cinnoline leads to partially assembled metalla-aza-cyclophane systems.

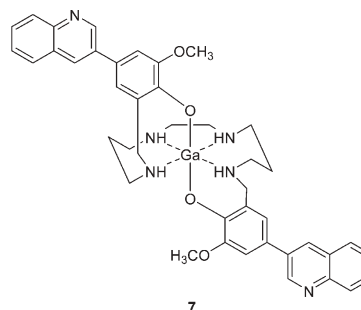


1622

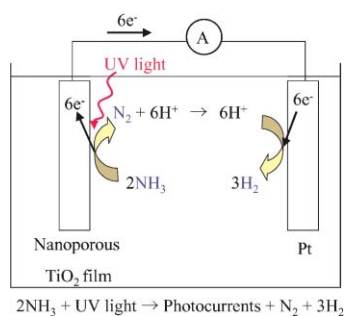
Metalloantimalarials: synthesis and characterization of a novel agent possessing activity against *Plasmodium falciparum*

Joseph A. Ocheskey, Scott E. Harpstrite, Anna Oksman, Daniel E. Goldberg and Vijay Sharma*

A novel gallium(III) complex **7** was synthesized and characterized. While equipotent in Dd2 lines, **7** demonstrated enhanced efficacy against HB3 lines compared with its predecessor antimalarial suggesting feasibility of rational drug design in metallotherapeutics.



1625

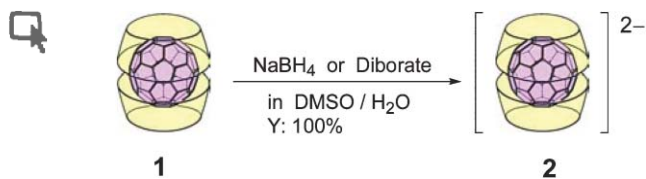


Artificial photochemical nitrogen cycle to produce nitrogen and hydrogen from ammonia by platinumized TiO_2 and its application to a photofuel cell

M. Kaneko,* N. Gokan, N. Katakura, Y. Takei and M. Hoshino

Photocatalytic decomposition of aqueous ammonia to H_2 and N_2 was achieved, and its application to a photofuel cell producing electricity and H_2/N_2 was reported.

1628



Reducing ability of supramolecular C_{60} dianion toward $\text{C}=\text{O}$, $\text{C}=\text{C}$ and $\text{N}=\text{N}$ bonds

Shin-ichi Takekuma, Hideko Takekuma and Zen-ichi Yoshida*

Supramolecular C_{60} dianion (**2**) generated from γ -cyclodextrin-bicapped C_{60} (**1**) is able to reduce $\text{N}=\text{N}^+$, $\text{C}=\text{C}$ -EWG and $\text{C}=\text{O}$ linkages; **1**-mediated reduction of acetophenone with NaBH_4 in the presence of $(\text{Me}_2\text{N})_2\text{CH}_2$ and EtONa gives TOF (h^{-1}) of 400.

1631

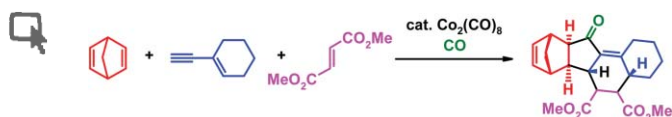


Direct and continuous synthesis of concentrated hydrogen peroxide by the gaseous reaction of H_2/O_2 non-equilibrium plasma

Juncheng Zhou, Hongchen Guo,* Xiangsheng Wang, Mingxing Guo, Jiangli Zhao, Lixing Chen and Weimin Gong

Under ambient conditions, H_2O_2 has been synthesized with 32.51% yield and 56.25% selectivity *via* the gas-phase reaction of H_2/O_2 non-equilibrium plasma.

1634



Tandem Pauson-Khand reaction and Diels-Alder reaction for access to polycyclics in a one-pot reaction

Do Han Kim and Young Keun Chung*

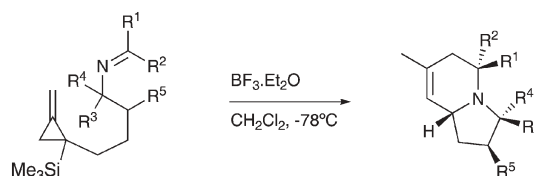
Starting from easily available cyclic alkenes, enynes, and dienophiles, a tandem intermolecular Pauson-Khand reaction and Diels-Alder reaction yields polycyclic compounds in high yields.

1637

Lewis acid mediated *endo*-cyclisation of trimethylsilylmethylenecyclopropyl imines—a stereoselective route to indolizidines

Suvi Rajamaki and Jeremy D. Kilburn*

Treatment of trimethylsilylmethylenecyclopropyl imines with a Lewis acid provides a novel route to indolizidine products *via* an unexpected mechanistic pathway involving three cationic intermediates.

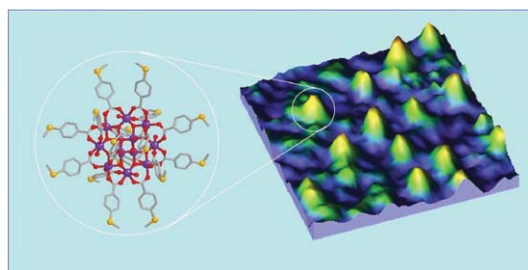


1640

Isolated single-molecule magnets on native gold

Laura Zobbi, Matteo Mannini, Mirko Pacchioni, Guillaume Chastanet, Daniele Bonacchi, Chiara Zanardi, Roberto Biagi, Umberto Del Pennino, Dante Gatteschi, Andrea Cornia* and Roberta Sessoli*

STM addressing of isolated single-molecule magnets has been achieved by grafting a new sulfur-functionalized Mn₁₂ derivative, [Mn₁₂O₁₂(O₂C-C₆H₄-SCH₃)₁₆(H₂O)₄], on a gold surface resulting in a sub-monolayer coverage by well separated molecules on a large scale.

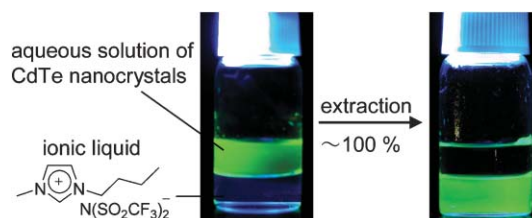


1643

Quantum dots–ionic liquid hybrids: efficient extraction of cationic CdTe nanocrystals into an ionic liquid

Takuya Nakashima* and Tsuyoshi Kawai*

Water soluble CdTe nanocrystals covered with cationic thiol derivatives are efficiently transferred into a hydrophobic ionic liquid, in which they show enhanced photoluminescence.

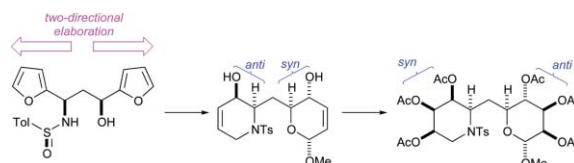


1646

A general, two-directional approach to aza-C-(1 → 1)-linked disaccharide mimetics

Andrew Kennedy, Adam Nelson* and Alexis Perry

The Upjohn and Donohoe dihydroxylations were exploited in divergent syntheses of aza-C-(1 → 1)-linked disaccharides.




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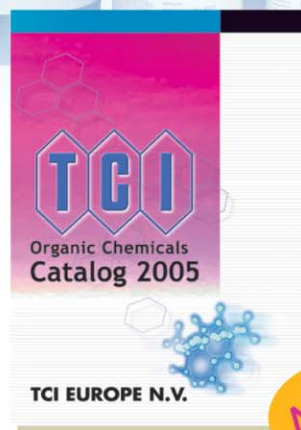
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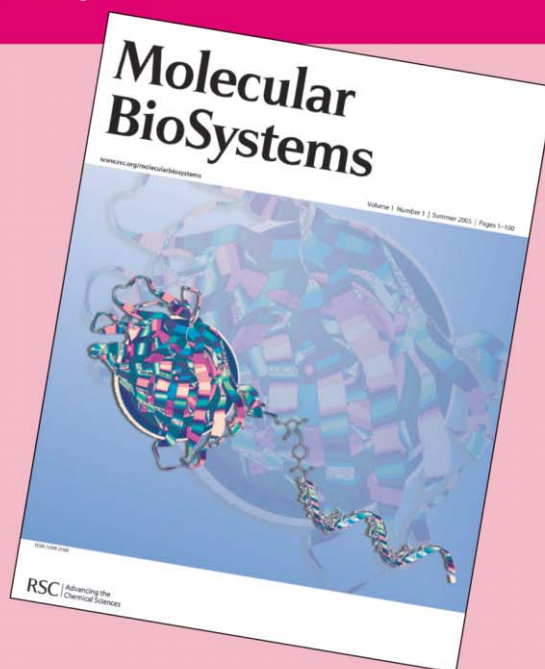
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