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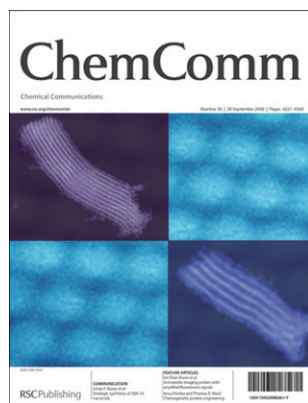
Chemical Communications

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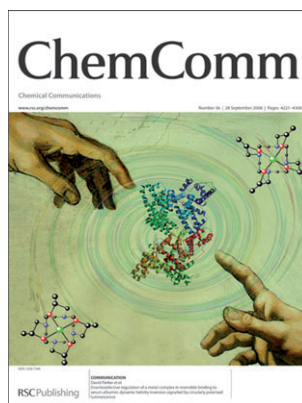
IN THIS ISSUE

ISSN 1359-7345 CODEN CHCOFS (36) 4221-4368 (2008)



Cover

See Linda F. Nazar *et al.*, pp. 4288–4290.
TEM images of nanodimensioned mesoporous silica, that typically span less than 10 porous channels in width. Image reproduced by permission of Xiulei Ji, Kyu T. Lee, Muguette Monjaube and Linda F. Nazar from *Chem. Commun.*, 2008, 4288.



Inside cover

See David Parker *et al.*, pp. 4261–4263.
A chiral lanthanide complex inverts helicity on binding to serum albumin, signalled by a change in the sign of the circularly polarised emission. Image reproduced by permission of Craig P. Montgomery, Elizabeth J. New, David Parker and Robert D. Peacock from *Chem. Commun.*, 2008, 4261.

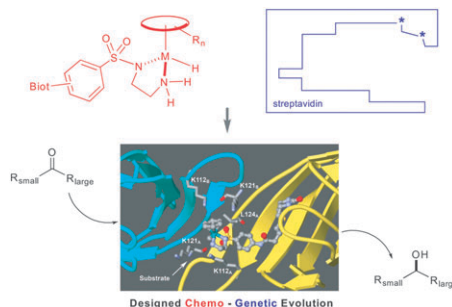
FEATURE ARTICLES

4239

Chemogenetic protein engineering: an efficient tool for the optimization of artificial metalloenzymes

Anca Pordea and Thomas R. Ward*

Artificial metalloenzymes lie at the interface between organometallic and enzymatic catalysis and have an immense optimization potential, which combines chemical and genetic methods to screen diversity space. This article describes the process of the creation and optimization of artificial transfer hydrogenases based on the biotin-streptavidin technology.

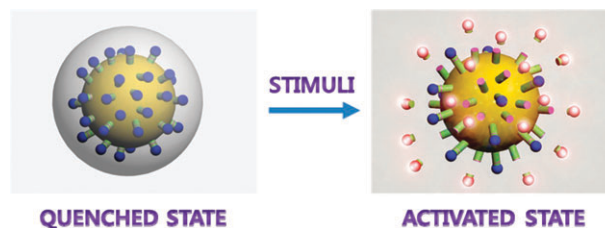


4250

Activatable imaging probes with amplified fluorescent signals

Seulki Lee, Kyeongsoon Park, Kwangmeyung Kim, Kuiwon Choi and Ick Chan Kwon*

This feature article introduces and discusses the various approaches described by the term “fluorescent signal activation methods” with respect to their unique imaging probe design strategies and applications.



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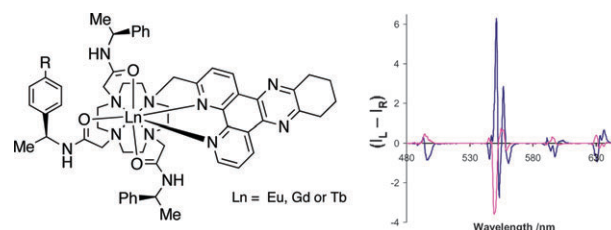
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4261

Enantioselective regulation of a metal complex in reversible binding to serum albumin: dynamic helicity inversion signalled by circularly polarised luminescence

Craig P. Montgomery, Elizabeth J. New, David Parker* and Robert D. Peacock

The helicity of the (*SSS*)- Δ enantiomer of Tb and Eu(III) complexes switches on reversible binding to 'drug site II' of serum albumin and is signalled by inversion of the circular polarisation of emitted light.

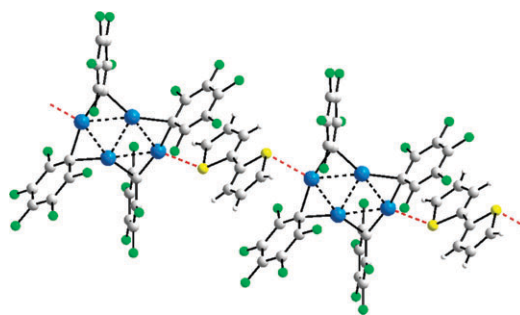


4264

Binary stacks of [Cu₆F₅]₄ with arenes

Ami Doshi, Krishnan Venkatasubbaiah, Arnold L. Rheingold and Frieder Jäkle*

The organocopper tetramer [Cu₆F₅]₄ assembles into novel luminescent binary stacks with naphthalene and 2,2'-bithiophene, respectively. Crystallographic data suggest that this supramolecular assembly process is supported by Cu- π , Cu-S, and perfluoroarene-arene π -interactions.

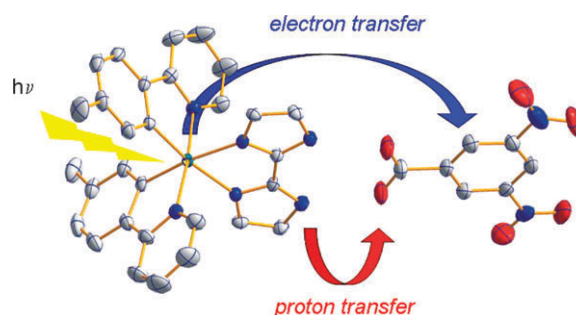


4267

Proton-coupled electron transfer from a luminescent excited state

Jonathan C. Freys, Gérald Bernardinelli and Oliver S. Wenger*

A luminescent hydrogen-bonded cation/anion pair is identified as a useful model system for light-triggered proton-coupled electron transfer reactions.

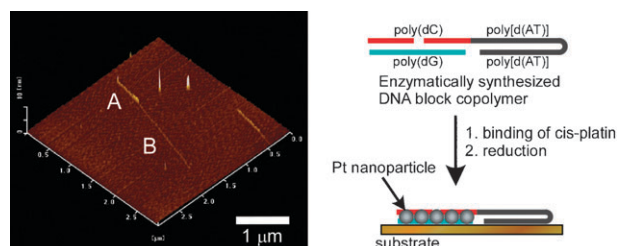


4270

Sequence-specific platinum metal deposition on enzymatically synthesized DNA block copolymer

Aya Tanaka, Yasutaka Matsuo, Yuichi Hashimoto and Kuniharu Ijiri*

Platinum metal was sequence-specifically deposited on the DNA block copolymer synthesized by the Klenow fragment of *E. coli* DNA polymerase I (3'-5' exonuclease deficient).



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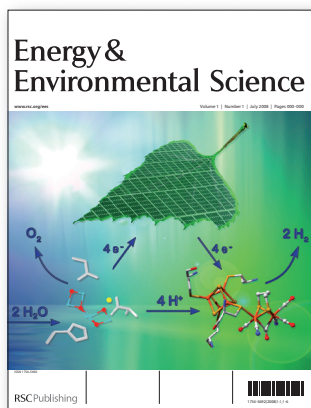
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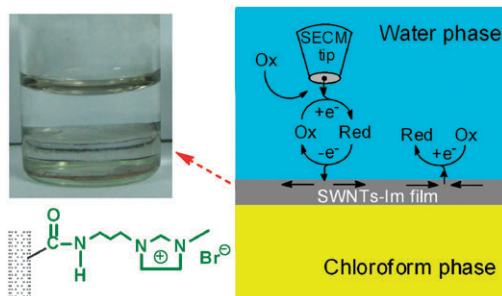
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4273

Directing single-walled carbon nanotubes to self-assemble at water/oil interfaces and facilitate electron transfer

Yuanjian Zhang, Yanfei Shen, Daniel Kuehner, Shuixing Wu, Zhongmin Su, Shen Ye and Li Niu*

Flexible SWNT films with different thicknesses at water/oil interfaces were obtained by modifying SWNTs, and facilitated electron transfer at SWNT-sandwiched water/oil interfaces was observed using scanning electrochemical microscopy.

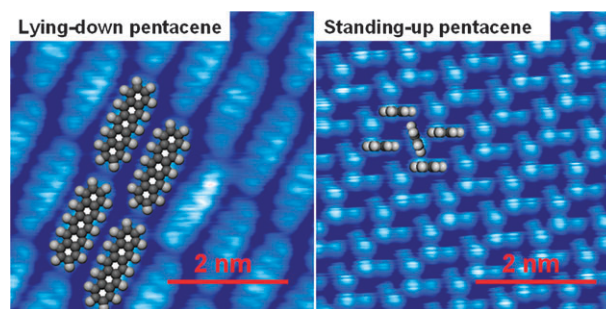


4276

Molecular orientation transition of organic thin films on graphite: the effect of intermolecular electrostatic and interfacial dispersion forces

Wei Chen,* Han Huang, Andrew Thyne and Shen Wee*

In situ low-temperature scanning tunnelling microscopy investigation reveals a molecular orientation transition of pentacene and *p*-sexiphenyl films on graphite, arising from the coverage-dependent delicate balance between the intermolecular electrostatic and interfacial dispersion forces.

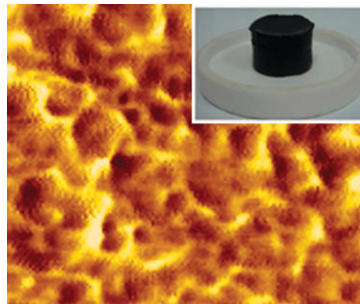


4279

Facile fabrication of conducting polymer hydrogels via supramolecular self-assembly

Tingyang Dai, Xiujuan Jiang, Shouhu Hua, Xiaoshu Wang and Yun Lu*

Conducting polymer hydrogels have been fabricated *via* supramolecular self-assembly. Multivalent cations act as both the oxidant and the ionic crosslinker. The gelation process takes place simultaneously with the polymerization process.

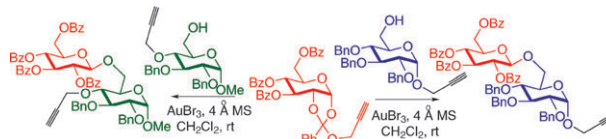


4282

Gold mediated glycosylations: selective activation of propargyl 1,2-orthoesters in the presence of aglycones containing a propargyl moiety

Gopalsamy Sureshkumar and Srinivas Hotha*

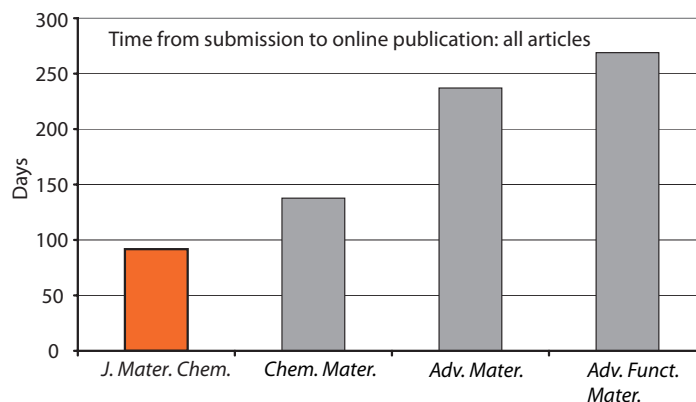
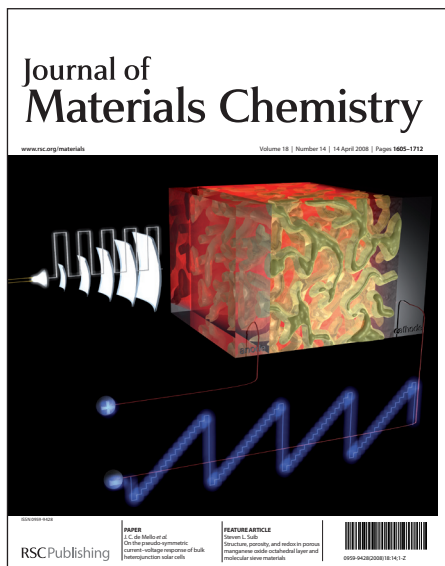
Selective activation of propargyl 1,2-orthoesters in the presence of propargyl glycosides and propargyl ethers was studied. A catalytic amount of AuBr₃ activated the propargyloxy group of the 1,2-orthoester thereby giving access to disaccharides with the propargyl group at the reducing end.



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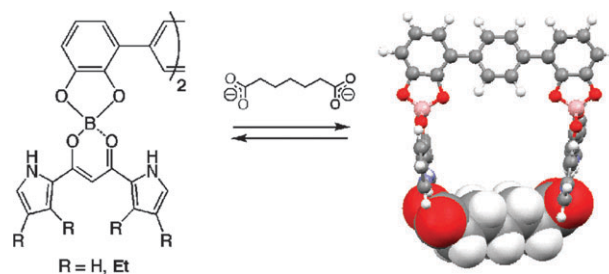
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4285

Diol-substituted boron complexes of dipyrrolyl diketones as anion receptors and covalently linked 'pivotal' dimers

Hiromitsu Maeda,* Yasunobu Fujii and Yuta Mihashi

Diol-substitution at a boron unit results in the formation of anion receptors consisting of dipyrrolyl diketones and covalently linked dimers, which exhibit selective binding for dianions with appropriate lengths.



4288

Strategic synthesis of SBA-15 nanorods

Xiulei Ji, Kyu T. Lee, Muguette Monjauze and Linda F. Nazar*

A simple synthesis of homogeneously sized, ordered mesoporous silica nanorods (SBA-15), spanning about 10 porous channels in width (<100 nm) and ranging from 400–600 nm in length is reported.



4291

Hydrous zinc halide-catalyzed aminosulfonation of hydrocarbons

Biswajit Kalita, Angus A. Lamar and Kenneth M. Nicholas*

Benzylic and allylic hydrocarbons are selectively converted to the corresponding sulfonamides by a $\text{ZnBr}_2\text{-H}_2\text{O}$ -catalyzed reaction with $\text{PhI}=\text{NTs}$. Saturated adamantane is aminosulfonated at the tertiary C–H bond.

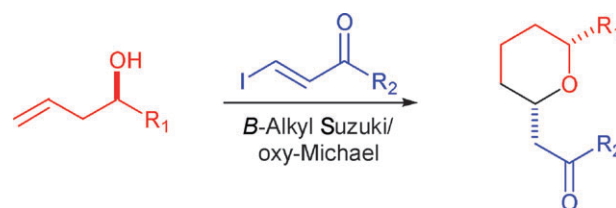


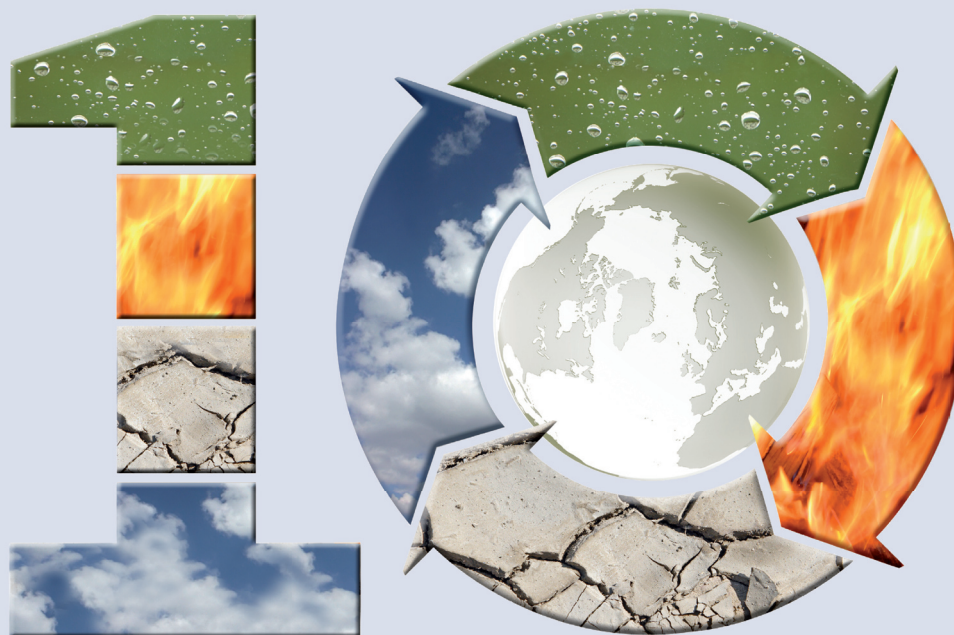
4294

B-Alkyl Suzuki couplings for the stereoselective synthesis of substituted pyrans

Gregory W. O'Neil and Alois Fürstner*

A tandem *B*-alkyl Suzuki cross-coupling/oxy-Michael reaction sequence has been described for the stereoselective synthesis of substituted tetrahydropyrans including the olfactory natural product (+)-(*S,S*)-(*cis*-6-methyltetrahydropyran-2-yl)acetic acid.





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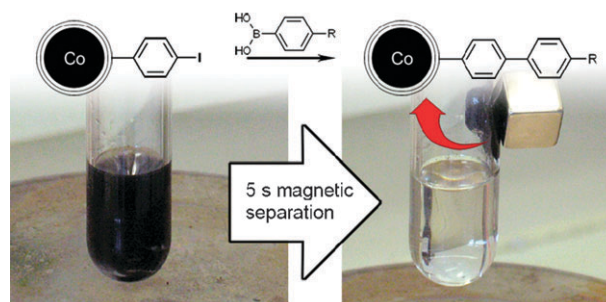
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4297

Suzuki cross-coupling reactions on the surface of carbon-coated cobalt: expanding the applicability of core-shell nano-magnets

Chun Ghee Tan and Robert N. Grass*

To develop magnetic nanomaterials applicable to organic synthesis, the Suzuki cross-coupling method was adapted to attach a range of functional groups to carbon-coated core-shell materials *via* commercially-available substituted arylboronic acids.

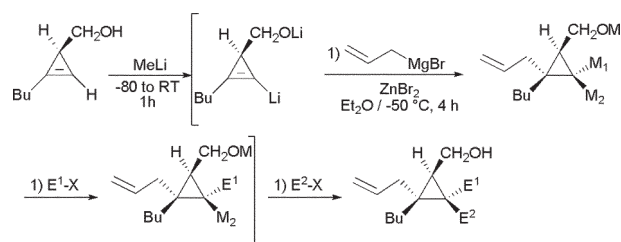


4300

Cyclopropenyllithiums as a new source of 1,1-bismetallated cyclopropyl derivatives

Anat Levin and Ilan Marek*

The allylmetalation of functionalised cyclopropenyllithium derivatives leads to the unique formation of 1,1-bismetallated cyclopropyl species that react selectively with different electrophiles.

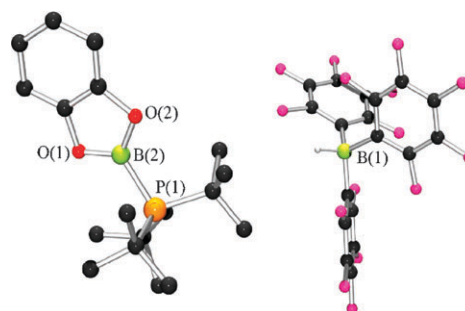


4303

B–H Activation by frustrated Lewis pairs: borenium or boryl phosphonium cation?

Meghan A. Dureen, A. Lough, Thomas M. Gilbert and Douglas W. Stephan*

Catechol borane reacts with the frustrated Lewis pairs *t*Bu₂RP (R = *t*Bu, 2-C₆H₄(C₆H₅)) and B(C₆F₅)₃ to give the species [(C₆H₄O₂)BP/*t*Bu₂R][HB(C₆F₅)₃] that can formally be described as either borenium cation or boryl-phosphonium salts; the nature of these species was probed with DFT calculations.

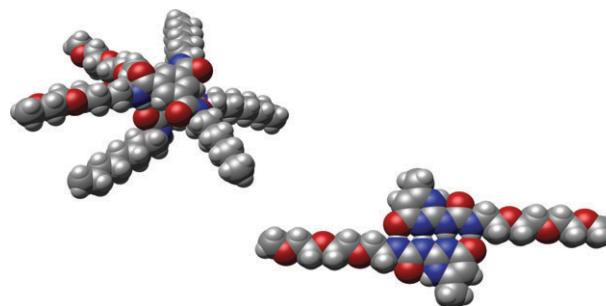


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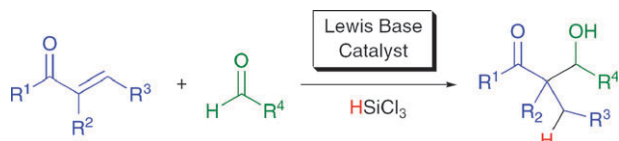
The influence of ethylene glycol chains on the thermodynamics of hydrogen-bonded supramolecular assemblies in apolar solvents

Tom F. A. de Greef, Marko M. L. Nieuwenhuizen, Patrick J. M. Stals, Carel F. C. Fitié, Anja R. A. Palmans, Rint P. Sijbesma and E. W. Meijer*

Substitution of H-bond directed supramolecular assemblies with ethylene glycol chains leads to a reduction in *K*_{ass} in apolar solvents, dependent on the length of the aliphatic spacer connecting the H-bonds and the ethylene glycol chain.



4309

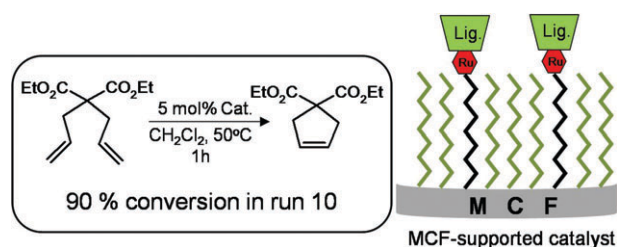


Lewis base-catalyzed conjugate reduction and reductive aldol reaction of α,β -unsaturated ketones using trichlorosilane

Masaharu Sugiura,* Norimasa Sato, Shunsuke Kotani and Makoto Nakajima*

Phosphorus oxide (P=O) Lewis bases catalyze the 1,4-reduction of α,β -unsaturated ketones with trichlorosilane and subsequent reactions with coexisting aldehydes (reductive aldol reactions).

4312

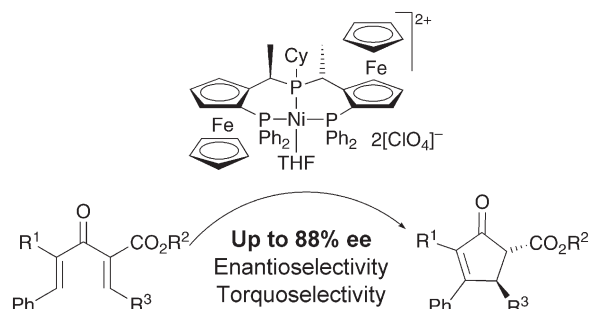


Silica-supported catalysts for ring-closing metathesis: effects of linker group and microenvironment on recyclability

Jaehong Lim, Su Seong Lee* and Jackie Y. Ying*

Recyclability was dramatically improved by modifying the linker and microenvironment of mesoporous foam-supported ruthenium catalysts for ring-closing metathesis (RCM).

4315

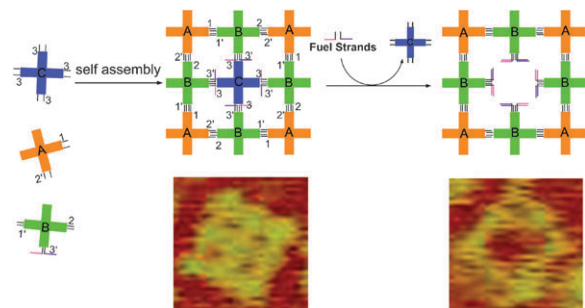


Ni(II)-catalyzed enantioselective Nazarov cyclizations

Irene Walz and Antonio Togni*

A dicationic Ni(II) complex containing the chiral ferrocenyl triphosphine Pigiphos catalyzes Nazarov cyclizations with a high degree of torquo- and enantioselectivity up to 88% ee.

4318



Subtractive assembly of DNA nanoarchitectures driven by fuel strand displacement

Zhe Li, Yonggang Ke, Chenxiang Lin, Hao Yan and Yan Liu*

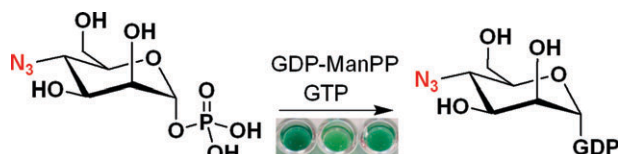
We report a new subtractive assembly strategy to generate DNA tile arrays with large central cavities, which are difficult to obtain using only additive assembly. AFM images clearly demonstrate the feasibility of this approach.

4321

Chemoenzymatic synthesis of GDP-azidodeoxymannoses: non-radioactive probes for mannosyltransferase activity

Silvia Marchesan and Derek Macmillan*

GDP-2-, 3-, 4- or 6-azidomannoses can be successfully prepared from the corresponding azidomannose-1-phosphates and GTP using the enzyme GDP-Mannosepyrophosphorylase (GDP-ManPP) from *Salmonella enterica* and may serve as useful probes for mannosyltransferase activity.

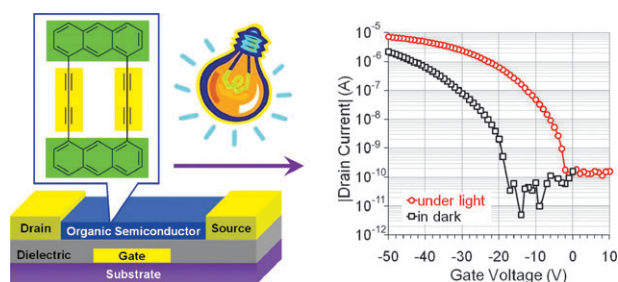


4324

Transistors from a conjugated macrocycle molecule: field and photo effects

Wei Zhao, Qin Tang, Hoi Shan Chan, Jianbin Xu, Ka Yuen Lo and Qian Miao*

This study explores a conjugated macrocycle molecule and details its synthesis, molecular structure, assemblies in the solid state and application in phototransistors.

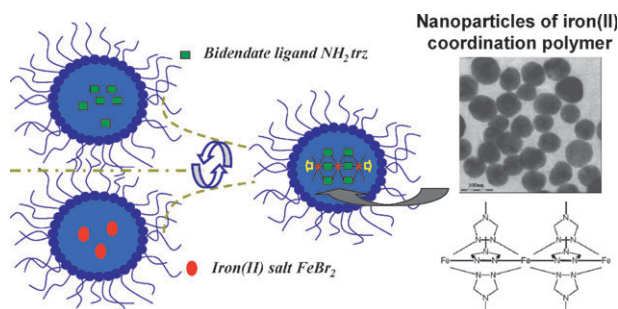


4327

Nanoparticles of iron(II) spin-crossover

Thibaut Forestier, Stéphane Mornet, Nathalie Daro, Taishi Nishihara, Shin-ichiro Mouri, Koichiro Tanaka, Olivier Fouché, Eric Freysz and Jean-François Létard*

We report the synthesis of spin crossover 69 nm spherical nanoparticles of $[\text{Fe}(\text{NH}_2\text{-trz})_3](\text{Br})_2 \cdot 3\text{H}_2\text{O} \cdot 0.03(\text{surfactant})$ ($\text{NH}_2\text{-trz}$ = 4-amino-1,2,4-triazole, surfactant = Laupopal), prepared by the reverse micelle technique, which exhibit at room temperature a thermal hysteresis characterized by magnetic, diffuse reflectivity and Raman studies.

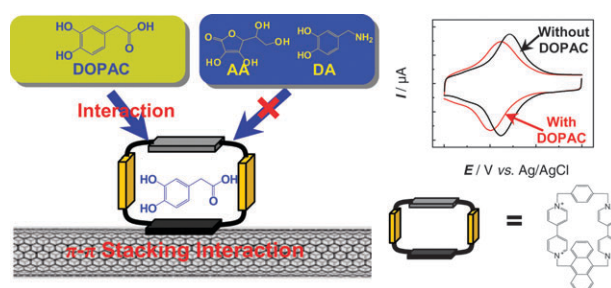


4330

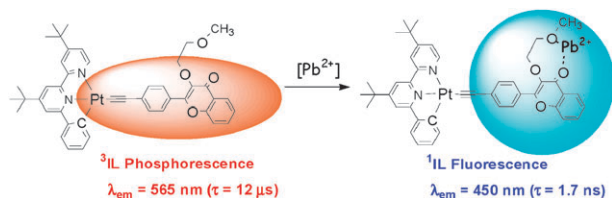
An electrochemical sensor for 3,4-dihydroxyphenylacetic acid with carbon nanotubes as electronic transducer and synthetic cyclophane as recognition element

Jie Yan, Yucheng Zhou, Ping Yu, Lei Su, Lanqun Mao,* Deqing Zhang* and Daoben Zhu

By taking advantage of the stronger binding affinity of the synthetic cyclophane towards 3,4-dihydroxyphenylacetic acid than ascorbic acid and dopamine, we have successfully developed a new electrochemical sensor with a promising application for the selective determination of DOPAC.



4333

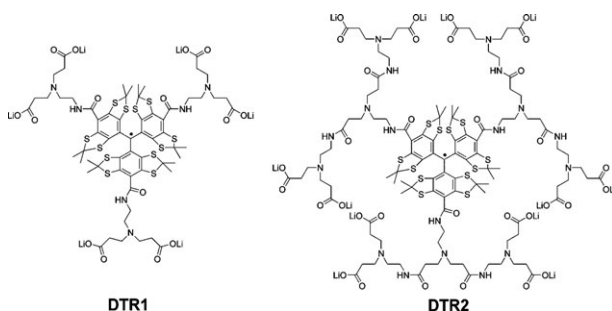


Cyclometallated platinum(II) complexes incorporating ethynyl-flavone ligands: switching between triplet and singlet emission induced by selective binding of Pb^{2+} ions

Pierre-Henri Lanoë, Jean-Luc Fillaut,* Loïc Toupet, J. A. Gareth Williams,* Hubert Le Bozec and Véronique Guerschais*

Platinum-ethynylflavone complexes display ^3IL phosphorescence associated with the appended flavone perturbed by the platinum centre, but switch dramatically to flavone-localised ^1IL fluorescence upon selective binding of Pb^{2+} .

4336

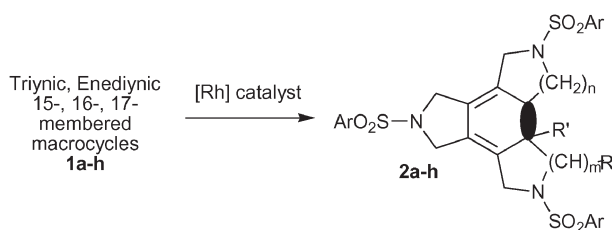


Highly stable dendritic trityl radicals as oxygen and pH probe

Yangping Liu, Frederick A. Villamena and Jay L. Zweier*

Novel dendritic trityl radicals (DTR1 and DTR2) with a TAM radical core, PAMAM branching and carboxylate exterior surface exhibit high stability towards oxidoreductants as evidenced by their electrochemical and EPR properties, offering potential application as dual oxygen and pH probe.

4339

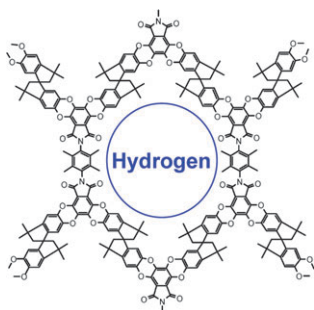


Fused tetracycles with a benzene or cyclohexadiene core: [2 + 2 + 2] cycloadditions on macrocyclic systems

Sandra Brun, Lidia Garcia, Iván González, Anna Torrent, Anna Dachs, Anna Pla-Quintana, Teodor Parella and Anna Roglans*

Rh(I)-catalyzed [2 + 2 + 2] cycloaddition reactions of macrocyclic systems afford high yields of fused tetracycles with benzene and cyclohexadiene cores.

4342



Hydrogen adsorption in microporous organic framework polymer

Saad Makhseed* and Jacob Samuel

A microporous organic framework polymer (OFP) based on a polyimide framework exhibits a high surface area ($1159 \text{ m}^2 \text{ g}^{-1}$) and shows a reversible H_2 storage capacity of 3.94 wt% at 10 bar and 77 K, the highest yet reported for an organic polymer.

4345

An easy entry to optically active α -amino phosphonic acid derivatives using phase-transfer catalysis (PTC)

Francesco Fini, Gabriele Micheletti, Luca Bernardi,*
Daniel Pettersen, Mariafrancesca Fochi and Alfredo Ricci*

The unprecedented use of phase-transfer catalysis (PTC) in an asymmetric hydrophosphonylation reaction allows the obtainment of a range of optically active α -amino phosphonic acid derivatives directly from α -amido sulfones.

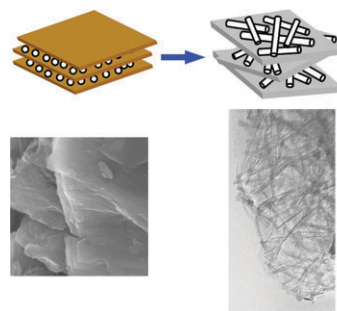


4348

Lamellar carbon nanosheets function as templates for two-dimensional deposition of tubular titanate

Wenqin Peng, Zhengming Wang,* Noriko Yoshizawa,
Hiroaki Hatori and Takahiro Hirotsu

A novel composite composed of tubular titanate–two dimensionally deposited carbon nanosheets was prepared with carbon nanosheets as templates; this nanotube-based composite and its calcined products exhibit both excellent adsorptivity and high photocatalytic activity toward organic molecules.

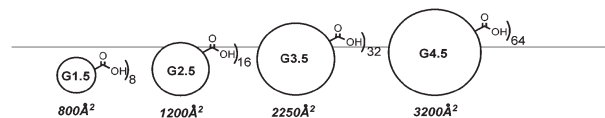


4351

Dendrimers as size selective inhibitors to protein–protein binding

Fumiko Chiba, Ting-Chou Hu, Lance J. Twyman* and
Mark Wagstaff

The “quantized” size effect of dendrimers can be exploited towards a size selective binding mechanism for the inhibition of protein–protein binding.

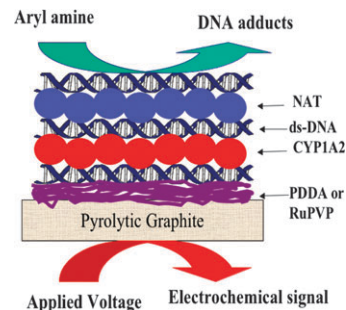


4354

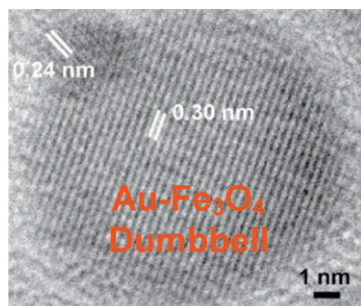
Electrochemical biosensor featuring a two-enzyme pathway and DNA for screening toxic reactive metabolites of arylamines

Minjeong So, John B. Schenkman and James F. Rusling*

The first biosensor featuring a sequential two-enzyme pathway and DNA to screen toxic reactive metabolites generated during metabolism of arylamines is reported.



4357

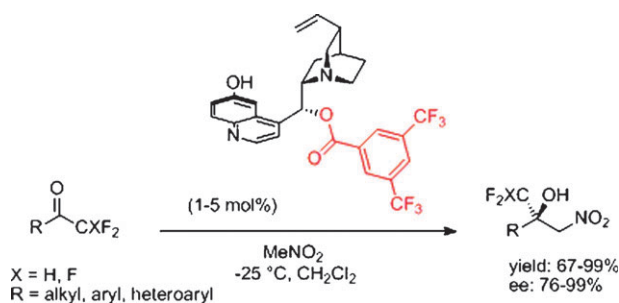


Colloidal deposition synthesis of supported gold nanocatalysts based on Au-Fe₃O₄ dumbbell nanoparticles

Hongfeng Yin, Chao Wang, Haoguo Zhu, Steven H. Overbury, Shouheng Sun* and Sheng Dai*

A new colloidal deposition methodology is used to introduce Au catalysts on supports *via* dumbbell-structured Au-Fe₃O₄ nanoparticles. Highly active and stable Au catalysts are prepared using oxide and even carbon supports.

4360

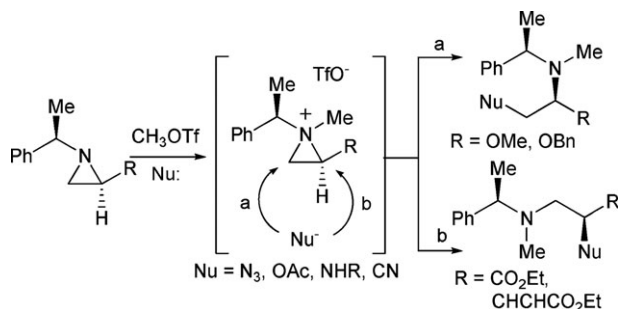


Enantioselective organocatalyzed Henry reaction with fluoromethyl ketones

Marco Bandini,* Riccardo Sinisi and Achille Umani-Ronchi*

Fluoro calls for fluoro. An highly enantioselective organocatalyzed nitroaldol condensation of fluoro-substituted ketones is presented. The designed fluoro-containing cupreine **3g** allowed the synthesis of a range of β -nitroalcohols in high yields and excellent *ees* (up to 99%).

4363



The preparation of stable aziridinium ions and their ring-openings

Yongun Kim, Hyun-Joon Ha,* Sae Young Yun and Won Koo Lee*

The reaction of enantiomerically pure 2-substituted 1-phenylethyl-aziridine with methyl trifluoromethanesulfonate generated a stable methylaziridinium ion, which was reacted with various nucleophiles to yield acyclic amine derivatives in a completely regio- and stereoselective manner.

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