

IN THIS ISSUE

ISSN 1359-7345 CODEN CHCOFS (27) 2821–2928 (2006)



Cover

See Ye-Gao Yin *et al.*, page 2845. Structure and function design of eight-fold interpenetrated photoluminescent metal-organic frameworks with trimeric copper(I)-pyrazolate as secondary building block. Image reproduced by permission of Jun He, Ye-Gao Yin, Tao Wu, Dan Li and Xiao-Chun Huang, from *Chem. Commun.*, 2006, 2845.



Inside cover

See Robin J. Leatherbarrow *et al.*, page 2848. Cascade thia-*zip* reaction enables the first total synthesis of MCoTI-II; background: stylised flower of *Momordica cochinchinensis*, the plant from which MCoTI-II is derived. Image reproduced by permission of Panumart Thongyoo, Edward W. Tate and Robin J. Leatherbarrow, from *Chem. Commun.*, 2006, 2848.

CHEMICAL SCIENCE

C49

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

July 2006/Volume 3/Issue 7

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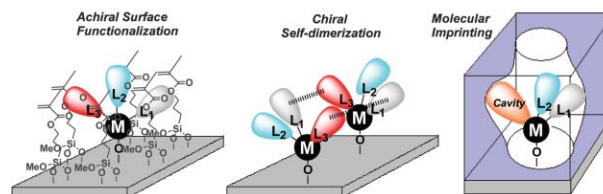
FEATURE ARTICLE

2833

Advanced chemical design with supported metal complexes for selective catalysis

Mizuki Tada and Yasuhiro Iwasawa*

Recent topics of novel surface design with supported metal complexes are reviewed: achiral surface functionalization for enantioselective catalysis, chiral self-dimerization for asymmetric coupling, and molecular imprinting for shape-selective catalysis.



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Chemical Communications (print: ISSN 1359-7345; electronic: ISSN 1364-548X) is published 48 times a year by the Royal Society of Chemistry, Thomas Graham House, Science Park, Milton Road, Cambridge, UK CB4 0WF. All orders, with cheques made payable to the Royal Society of Chemistry, should be sent to RSC Distribution Services, c/o Portland Customer Services, Commerce Way, Colchester, Essex, UK CO2 8HP. Tel +44 (0)1206 226050; E-mail sales@rscdistribution.org

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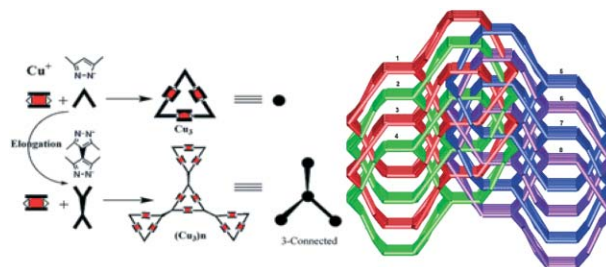
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2845

Design and solvothermal synthesis of luminescent copper(I)-pyrazolate coordination oligomer and polymer frameworks

Jun He, Ye-Gao Yin,* Tao Wu, Dan Li* and Xiao-Chun Huang

Copper(I)-pyrazolate trimer (**1**) and polymer (**2**) possessing desired structure and luminescence were synthesized, of which **2** has a 3-D framework structure showing a rare 3-connected binodal $(6^2.10)(6.10^2)$ topology and eight-fold interpenetration.

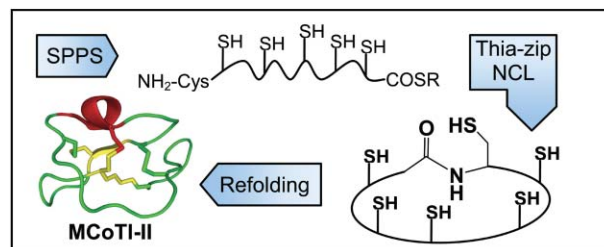


2848

Total synthesis of the macrocyclic cysteine knot microprotein MCoTI-II

Panumart Thongyoo, Edward W. Tate and Robin J. Leatherbarrow*

The first total synthesis of MCoTI-II, a cysteine knot microprotein and potent trypsin inhibitor, is described; a synthetic strategy has been developed that combines efficient backbone construction *via* optimised solid phase peptide synthesis with one-pot 'thia-zip' native chemical ligation and refolding to yield the natural product.

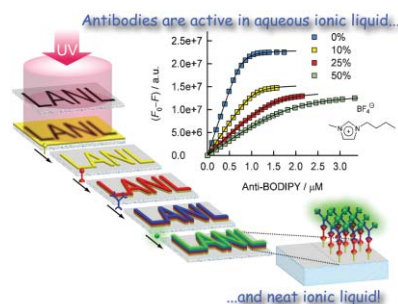


2851

Fluorescence quenching immunoassay performed in an ionic liquid

Sheila N. Baker, Eric B. Brauns, T. Mark McCleskey, Anthony K. Burrell and Gary A. Baker*

We describe the first example of immunoanalysis performed within an ionic liquid with minimal deleterious effect; our results bode well for the development of second-generation biosensors.

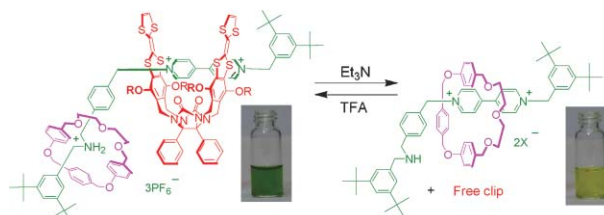


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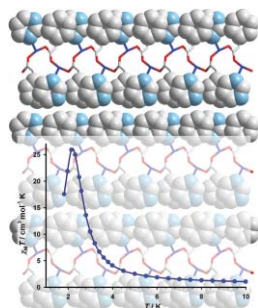
Reading the operation of an acid/base-controllable molecular switch by naked eye

Kuang-Wei Cheng, Chien-Chen Lai, Pinn-Tsong Chiang and Sheng-Hsien Chiu*

We report a pH-controllable molecular switch whose switching can be monitored by the naked eye; this system involves the formation of a complex between a [2]rotaxane—featuring dibenzylammonium and 4,4'-bipyridinium stations—and a TTF-side-walled molecular clip.



2857

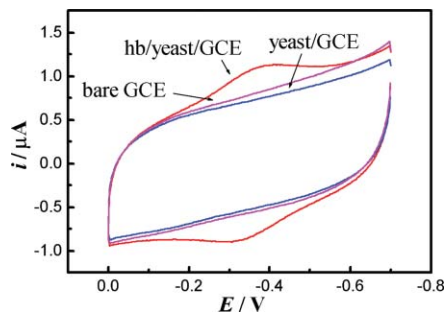


Metamagnetism in hydrophobically induced carboxylate (phenylmalonate)-bridged copper(II) layers

Jorge Pasán, Joaquín Sanchiz, Catalina Ruiz-Pérez,*
Javier Campo, Francesc Lloret and Miguel Julve

Self-assembly of copper(II) ions, phenylmalonate and pyrimidine yields the layered compound $[\text{Cu}(\text{pym})(\text{Phmal})]_n$, where intralayer ferro- and interlayer antiferromagnetic interactions occur with three-dimensional antiferromagnetic ordering at $T_c = 2.15$ K.

2860

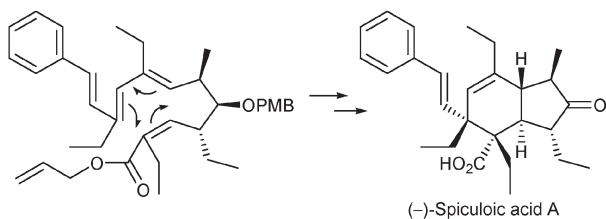


Studies on the direct electrochemistry of hemoglobin immobilized by yeast cells

Qing Lu, Junhui Xu and Shengshui Hu*

Saccharomyces cerevisiae, a commonly employed yeast cell, was utilized to immobilize hemoglobin onto the electrode through electrostatic attractions, and the direct electrochemistry of the protein was achieved.

2863

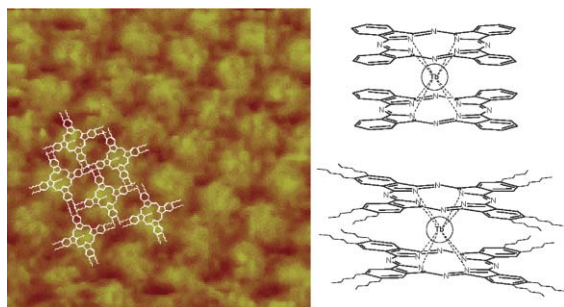


Biomimetic synthesis of marine sponge metabolite spiculoic acid A and establishment of the absolute configuration of the natural product

James E. D. Kirkham, Victor Lee* and Jack E. Baldwin*

An asymmetric biomimetic synthesis of the marine sponge polyketide spiculoic acid A is described. The key step involves the use of an intramolecular Diels–Alder reaction to secure the spiculane skeleton.

2866



2-D Self-assembly of the bis(phthalocyaninato)-terbium(III) single-molecule magnet studied by scanning tunnelling microscopy

Jordi Gómez-Segura, Ismael Díez-Pérez, Naoto Ishikawa,
Motohiro Nakano, Jaume Veciana and
Daniel Ruiz-Molina*

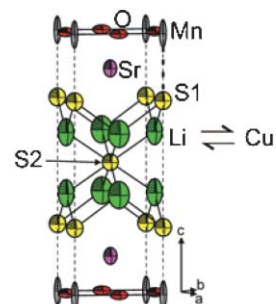
The single-molecule magnet $[(\text{Pc}')_2\text{Tb}^{\text{III}}]$ undergoes 2-D self-assembly on HOPG with the molecular plane parallel to the graphite surface, enabling us to control and/or predict the orientation of the preferential magnetization axis on the surface.

2869

Reversible lithium insertion and copper extrusion in layered oxysulfides

Oliver J. Rutt, Gareth R. Williams and Simon J. Clarke*

All of the copper in the layered oxysulfides $\text{Sr}_2\text{MnO}_2\text{Cu}_{1.5}\text{S}_2$ and $\text{Sr}_2\text{MnO}_2\text{Cu}_{3.5}\text{S}_3$ may be extruded as the element and the copper ions replaced quasi-reversibly by lithium ions in reductive topotactic ion exchange reactions; dramatic changes in magnetic properties result.

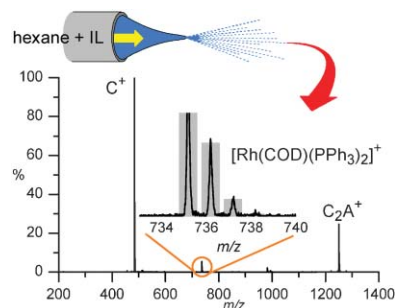


2872

Ionic liquids enable electrospray ionisation mass spectrometry in hexane

Matthew A. Henderson and J. Scott McIndoe*

Addition of a lipophilic ionic liquid to non-polar, hydrocarbon solvents (including hexane, pentane, cyclohexane, benzene and toluene) enables electrospray ionisation mass spectrometric (ESI-MS) analysis of other charged solution species, considerably broadening the scope of the ESI-MS technique. Careful selection of ionic liquid minimises the risk of coincidental overlap of ions.

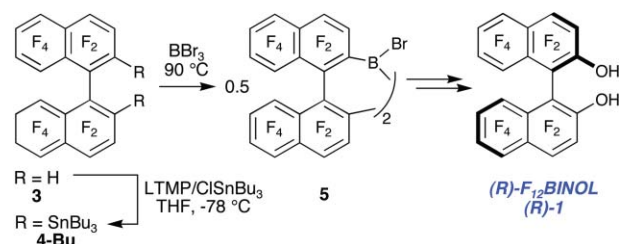


2875

2,2'-Disubstituted F_{12} binaphthyl derivatives: stannanes, boranes, and (*R*)- F_{12} BINOL

Darryl J. Morrison, Susanne D. Riegel, Warren E. Piers,* Masood Parvez and Robert McDonald

The synthesis of several F_{12} binaphthyl derivatives is described, including fully resolved (*R*)- F_{12} BINOL and a novel D_2 symmetric diborane.

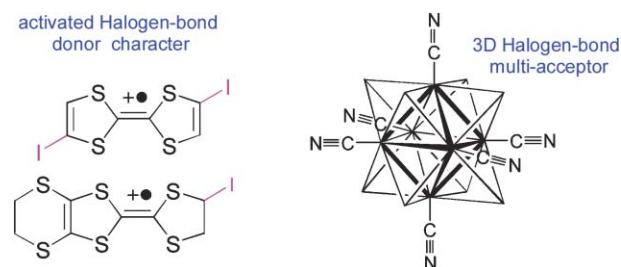


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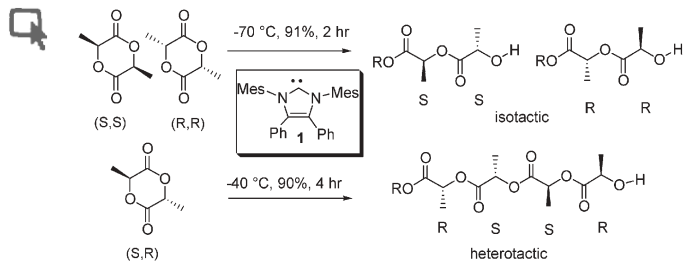
Balancing framework densification with charged, halogen-bonded- π -conjugated linkages: $[\text{PPh}_4]_2\{\text{E-TTF-I}_2\}[\text{Re}_6\text{Se}_8(\text{CN})_6]\}$ versus $[\text{PPh}_4]_2[\text{EDT-TTF-I}_2]\{\text{EDT-TTF-I}\}[\text{Re}_6\text{Se}_8(\text{CN})_6]\}$

Anupama Ranganathan, Abdelkrim El-Ghayoury, Cécile Mézière, Etienne Harté, Rodolphe Clérac and Patrick Batail*

Charged linkages and directional C–I \cdots N halogen bonds make for two- and three-dimensional nets yet two symmetrical halogen bond acceptors remain uninvolved as a result of charge densification.



2881

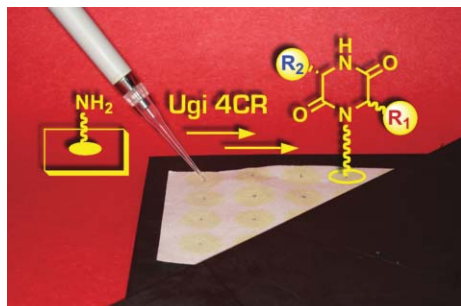


Stereoselective polymerization of *rac*- and *meso*-lactide catalyzed by sterically encumbered *N*-heterocyclic carbenes

Andrew P. Dove, Hongbo Li, Russell C. Pratt, Bas G. G. Lohmeijer, Darcy A. Culkin, Robert M. Waymouth* and James L. Hedrick*

The new sterically encumbered *N*-heterocyclic carbene **1** exhibits high activity at low temperatures for the stereospecific polymerization of *rac*- and *meso*-lactide to isotactic and heterotactic polylactide, respectively.

2884

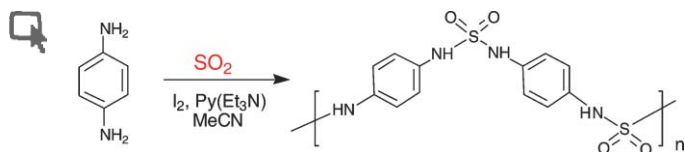


Rapid synthesis of diketopiperazine macroarrays via Ugi four-component reactions on planar solid supports

Qi Lin and Helen E. Blackwell*

Libraries of diketopiperazines have been generated using the small molecule macroarray synthesis platform. These libraries will facilitate the systematic study of the effects of synthetic diketopiperazines on bacterial quorum sensing.

2887

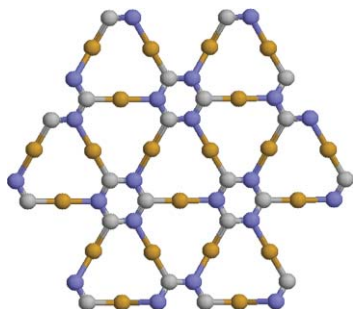


Sulfamides and sulfamide polymers directly from sulfur dioxide

Alexander V. Leontiev, H. V. Rasika Dias and Dmitry M. Rudkevich*

SO_2 gas is effectively used for the preparation of diarylsulfamides and shape-persistent sulfamide polymers. They utilize a network of intermolecular $\text{N-H}\cdots\text{O}=\text{S}$ hydrogen bonds to self-assemble into soft porous materials.

2890



Gold as intermolecular glue: a predicted planar triaurotriazine, $\text{C}_3\text{Au}_3\text{N}_3$, isomer of gold cyanide

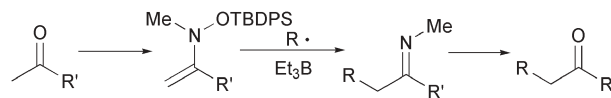
Mikko O. Hakala and Pekka Pyykkö*

It is predicted that solid gold cyanide, AuCN , could have an alternative infinite trigonal-sheet crystal structure, of almost the same energy as the known hexagonally-packed infinite-chain structure.

2893

Tin-free radical alkylation of ketones via *N*-silyloxy enamines

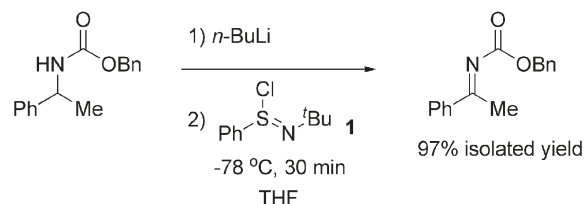
Hyun-Ji Song, Che Jo Lim, Sunggi Lee and Sunggak Kim*

A new tin-free radical alkylation of aldehydes and ketones via *N*-silyloxy enamine intermediates is developed.

2896

A mild and convenient synthesis of *N*-carbobenzyloxy ketimines

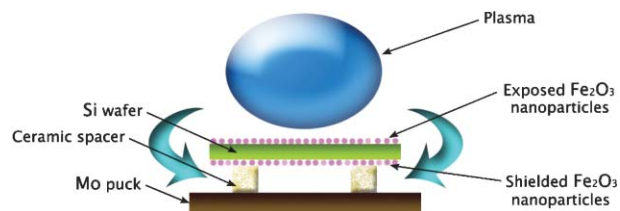
Jun-ichi Matsuo,* Yumi Tanaki, Aimi Kido and Hiroyuki Ishibashi*

N-Carbobenzyloxy (Cbz) ketimines were prepared conveniently from *N*-Cbz amines by oxidation with *N*-*tert*-butylbenzenesulfinimidoyl chloride under mild conditions.

2899

Dendrimer-assisted low-temperature growth of carbon nanotubes by plasma-enhanced chemical vapor deposition

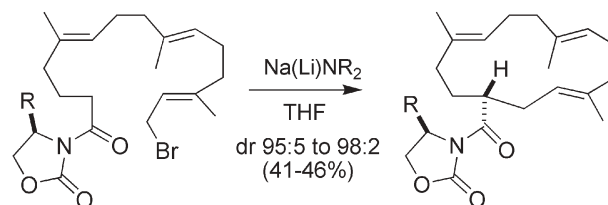
Placidus B. Amama,* Oluwaseyi Ogebole, Matthew R. Maschmann, Timothy D. Sands and Timothy S. Fisher

Using a shielded growth approach and N_2 -annealed, nearly monodispersed Fe_2O_3 nanoparticles synthesized by interdendritic stabilization of Fe^{3+} species within poly(amidoamine) dendrimers, carbon nanotubes and nanofibers were successfully grown at low substrate temperatures.

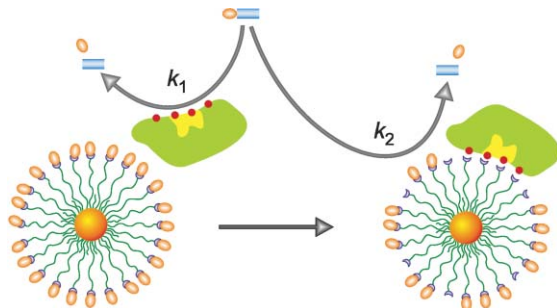
2902

Enantioselective synthesis of α -terpineol and nephtenol by intramolecular acyloxazolidinone enolate alkylations

Yinghua Jin and Robert M. Coates*

The acyloxazolidinone method for enantioselective alkylation was adapted to asymmetric cyclizations forming 6- and 14-membered rings as key steps in novel synthetic routes to the monocyclic terpenes (+)- α -terpineol and (+)-nephtenol.

2905

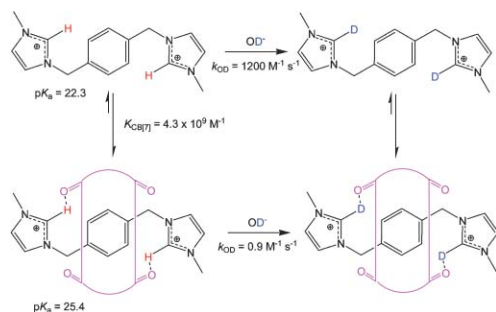


Regulation of α -chymotrypsin activity on the surface of substrate-functionalized gold nanoparticles

Chang-Cheng You, Rochelle R. Arvizo and Vincent M. Rotello*

A gold nanoparticle functionalized with substrates for α -chymotrypsin was fabricated to afford an enzyme modulator that exhibited enzyme-specific activation coupled with general inhibition of other proteases.

2908

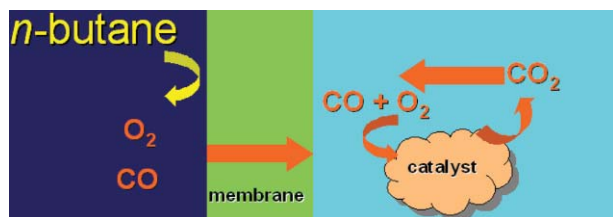


Inhibition of C(2)-H/D exchange of a bis(imidazolium) dication upon complexation with cucurbit[7]uril

Ruibing Wang, Lina Yuan and Donal H. Macartney*

Inclusion of a bis(imidazolium) dication by cucurbit[7]uril in D_2O reduces the acidity and lability of the C(2)-protons as a result of multipoint C–H \cdots O=C hydrogen bonding with the carbonyl oxygens of the host portals.

2911

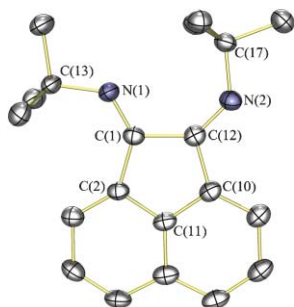


Fabrication of zeolite-4A membranes on a catalyst particle level

Yijun Zhong,* Liang Chen, Mengfei Luo, Yunlong Xie and Weidong Zhu

Absolute reactant selectivity for CO (O_2) over *n*-butane via defect-free zeolite-4A membranes coated onto Pt/ γ - Al_2O_3 catalyst particles.

2913



Facile routes to Alkyl-BIAN ligands

Jennifer A. Moore, Kalyan Vasudevan, Nicholas J. Hill, Gregor Reeske and Alan H. Cowley*

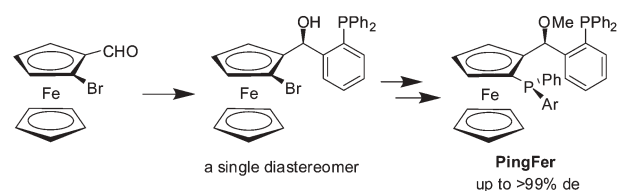
The new ligands *tert*-Butyl-BIAN and Adamantyl-BIAN have been prepared by treatment of acenaphthenequinone with iminoalane and aminoalane transfer agents, respectively.

2916

An efficient and highly stereoselective synthesis of new *P*-chiral 1,5-diphosphanylferrocene ligands and their use in enantioselective hydrogenation

Weiping Chen,* Stanley M. Roberts, John Whittall and Alexander Steiner

A novel type of *P*-chiral 1,5-diphosphanylferrocene ligands has been synthesised by two highly stereoselective reactions, and the introduction of *P*-chirality in ferrocene-based phosphine ligands enhances the enantioselective discrimination produced by the corresponding catalyst.

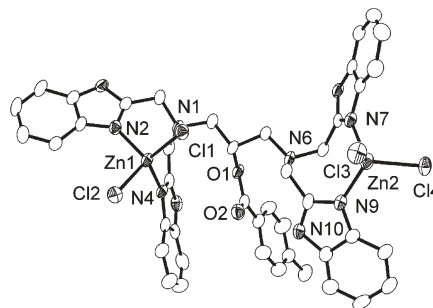


2919

Rapid phosphorus triester hydrolysis catalyzed by bimetallic tetrabenzimidazole complexes

Kortney L. Klinkel, Lauren A. Kiemele, Douglas L. Gin and John R. Hagadorn*

Bimetallic complexes based on the binucleating ligand *N,N,N',N'*-tetrakis[(2-benzimidazolyl)methyl]-2-hydroxy-1,3-diaminopropane (¹L) and its new toluoyl ester derivative (²L) catalyze the hydrolysis of phosphorus triesters at ambient temperature with activities rivalling the fastest known systems.

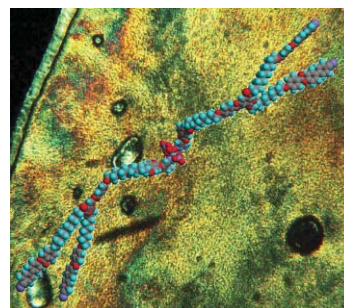


2922

Encoding calamitic mesomorphism in thermotropic lanthanidomesogens

Emmanuel Terazzi, Bernard Bocquet, Stéphane Campidelli, Bertrand Donnio, Daniel Guillon, Robert Deschenaux* and Claude Piguet*

Peripheral cyanobiphenyl dendrimers impose a microphase organization compatible with smectic mesomorphism, in which the bulky nine-coordinate lanthanide core is located between the decoupled mesogenic sublayers made up of parallel cyanobiphenyl groups.




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