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IN THIS ISSUE

ISSN 1359-7345 CODEN CHCOFS (43) 4445-4552 (2006)

Cover



See J. L. Sessler et al., page 4486. Just like a rose uses both its color and its perfume to attract a bee into its cavity, an oligopyrrolic macrocycle uses bimodal recognition (i.e., both covalent and hydrogen bonding) to "attract" its targeted dioxovanadium cation. Image reproduced by permission of Bojan Bato and Elisa Tomat from Jonathan L. Sessler, Elisa Tomat and Vincent M. Lynch, Chem. Commun., 2006, 4486.



Inside cover

See Y. K. Gun'ko *et al.*, page 4474. New fluorescent magnetic nanocomposites, based on magnetite nanoparticles, a polyhedral silsesquioxane and a porphyrin derivative as potential agents for MRI and biological cell imaging. Image reproduced by permission of S. A. Corr, A. O' Byrne, Y. K. Gun'ko, S. Ghosh, D. F. Brougham, S. Mitchell, Y. Volkov and A. Prina-Mello from *Chem. Commun.*, 2006, 4474.

CHEMICAL SCIENCE

C81

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Chemical Science

November 2006/Volume 3/Issue 11

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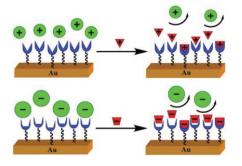
FEATURE ARTICLE

4461

Ion recognition properties of self-assembled monolayers (SAMs)

Sheng Zhang, Claudia M. Cardona and Luis Echegoyen*

In the search for new sensors, self-assembled monolayers (SAMs) have gained intensive interest due to their size, highly-ordered structures, and molecular recognition properties. This article presents an overview of ion recognition at SAM-modified electrode/solution interfaces, and brings up to date the most notable examples of ionic sensing.



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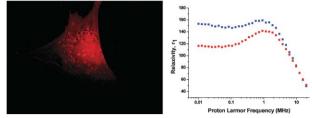
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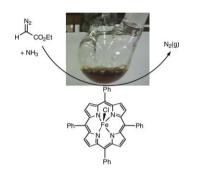
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multitasking





4477

Iron porphyrins catalyze the synthesis of non-protected amino acid esters from ammonia and diazoacetates

intracellular uptake of the nanocomposites by macrophage and bone osteoblast cells, and their potential as MRI contrast

Magnetic-fluorescent nanocomposites for biomedical

Serena A. Corr, Aisling O' Byrne, Yurii K. Gun'ko,* Swapankumar Ghosh, Dermot F. Brougham, Siobhan Mitchell, Yuri Volkov and Adriele Prina-Mello Fluorescent magnetite nanocomposites based on magnetic nanoparticles, a polyhedral octaaminopropylsilsesquioxane and a porphyrin derivative have been prepared. The

Iris Aviv and Zeev Gross*

agents, has been demonstrated.

The iron complexes of porphyrins are the first reported metal complexes to catalyze the formation of N-free glycine and alanine esters from ammonia and diazoesters.

4480

Triphasic liquid systems: generation and segregation of catalytically active Pd nanoparticles in an ammonium-based catalyst-philic phase

Alvise Perosa,* Pietro Tundo, Maurizio Selva and Patrizia Canton

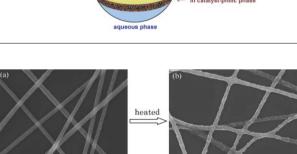
A triphasic liquid system made from organic, aqueous, and ammonium phases was used to form and stabilize Pd-nanoparticles, which remained segregated in the ammonium phase, catalysed organic reactions, and could be recycled.

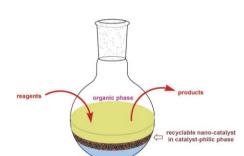
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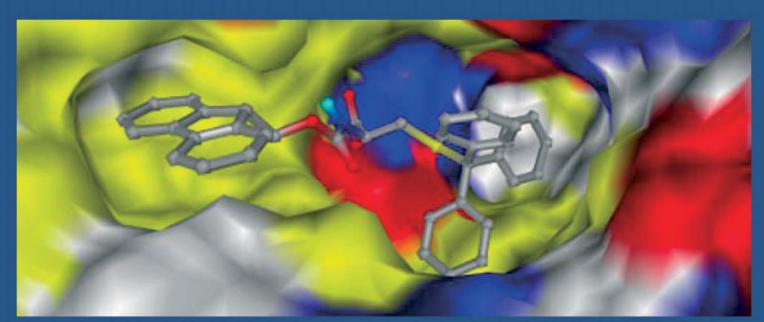
Fibrous TiO₂-SiO₂ nanocomposite photocatalyst

Ming Jin, Xintong Zhang, Alexei V. Emeline, Zhaoyue Liu, Donald A. Tryk, Taketoshi Murakami and Akira Fujishima'

The electrospinning method is employed to prepare the photocatalyst, in which Degussa P25 TiO₂ nanoparticles are embedded in the body of SiO_2 fibers with a netlike structure formed during the calcination process.







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4486

Coordination of oxovanadium(v) in an expanded porphyrin macrocycle

Jonathan L. Sessler,* Elisa Tomat and Vincent M. Lynch

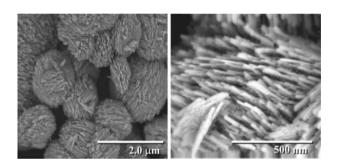
The formation of a dioxovanadium(V) complex of an expanded porphyrin-type Schiff base macrocycle is reported; the tetrapyrrolic ligand undergoes a tautomeric rearrangement which permits a bimodal recognition of the nonspherical cationic guest.

4489

Mesoporous materials with zeolite framework: remarkable effect of the hierarchical structure for retardation of catalyst deactivation

Rajendra Srivastava, Minkee Choi and Ryong Ryoo*

MFI zeolite with mesoporous/microporous hierarchical structure showed remarkably high resistance to catalyst deactivation in various acid-catalyzed reactions.



HC

2

4492

Probing the substrate specificity of the catalytically selfsufficient cytochrome P450 RhF from a *Rhodococcus sp.*

Ayhan Çelik, Gareth A. Roberts, John H. White, Stephen K. Chapman, Nicholas J. Turner and Sabine L. Flitsch*

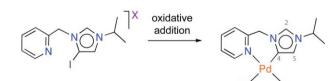
Analysis of the substrate specificity of the self-sufficient cytochrome P450 RhF revealed that the enzyme tends to catalyse the dealkylation of substituted alkyl–aryl ethers with shorter alkyl moieties more readily than equivalent compounds with longer alkyl groups.

4495

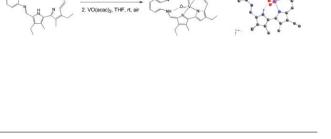
Mild and rational synthesis of palladium complexes comprising C(4)-bound *N*-heterocyclic carbenes

Evelyne Kluser, Antonia Neels and Martin Albrecht*

Oxidative addition of 4-iodoimidazolium salts to palladium(0) provides a mild and potentially general route to carbene complexes in which metal bonding occurs non-classically at C(4) rather than at C(2).



1



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Regioselectivity control of radiation-induced reaction: electron beam-induced Fries rearrangement of sulfonamide within a β-cyclodextrin inclusion complex

Jun Kato,* Hiroyuki Kakehata, Yasunari Maekawa and Takashi Yamashita

Solvent-free EB-Fries rearrangement proceeded at a shorter reaction time with reversed regioselectivity by inclusion into the β -CD, compared with that of sulfonamide crystals; the β -CD as a restricted nanospace had a large effect on the reactivity and regioselectivity of the solvent-free EB-Fries rearrangement.

4501

Rh(II)-catalysed room temperature aziridination of homoallyl-carbamates

Christopher J. Hayes,* Peter W. Beavis and Lesley A. Humphries

A rhodium(II)-catalysed method has been developed for the intramolecular aziridination of homo-allylic carbamates. Ring opening of the carbamate tethered aziridines occurs at the C4 position, providing functionalized 6-membered ring opened products.

4503

Mononuclear metavanadate catalyses gas phase oxidation of methanol to formaldehyde employing dioxygen as the terminal oxidant

Tom Waters, George N. Khairallah, Samantha A. S. Y. Wimala, Yien C. Ang, Richard A. J. O'Hair* and Anthony G. Wedd

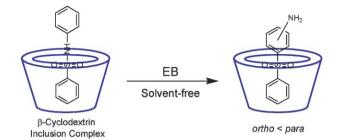
Multistage mass spectrometry experiments reveal a cycle of gas phase reactions for the oxidation of methanol to formaldehyde with a mononuclear oxo vanadate anion as the catalyst and dioxygen as the terminal oxidant.

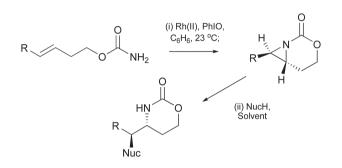
4506

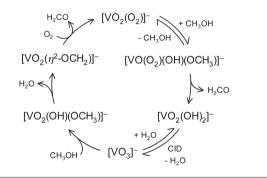
Synthesis of the spiroacetal-containing anti-*Helicobacter pylori* agents CJ-12,954 and CJ-13,014

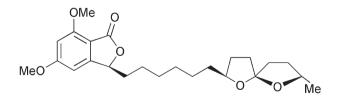
Margaret A. Brimble* and Christina J. Bryant

The first synthesis of the spiroacetal-containing anti-*Helicobacter pylori* agents CJ-12,954 and CJ-13,014 is reported, allowing assignment of the absolute stereochemistry of the natural products.









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Ring-opening polymerization of 3,6-dimethyl-2,5morpholinedione with discrete amino-alkoxybis(phenolate) yttrium initiators: mechanistic insights

Pascal M. Castro, Gang Zhao, Abderramane Amgoune, Christophe M. Thomas and Jean-François Carpentier*

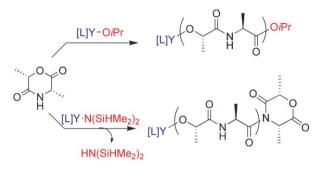
Well-defined yttrium amide and alkoxide complexes promote the ring-opening polymerization of a model 2,5-morpholinedione at 60–100 °C *via* a coordination– insertion polymerization mechanism, as evidenced by MALDI-TOF-MS.

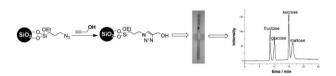
4512

Click chemistry: a new facile and efficient strategy for preparation of functionalized HPLC packings

Zhimou Guo, Aiwen Lei,* Xinmiao Liang* and Qing Xu

Click chemistry has been successfully extended into the field of preparation of functionalized HPLC packings. The potential has been demonstrated by the preparation of "Click I–IV" columns and preliminary results in the separation of sugars.





4515

Optical sculpture: controlled deformation of emulsion droplets with ultralow interfacial tensions using optical tweezers

Andrew D. Ward,* Mark G. Berry, Christopher D. Mellor and Colin D. Bain*

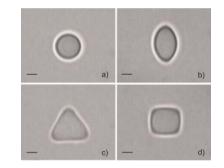
Micron-sized emulsion droplets with ultralow surface tension are deformed into a range of geometric shapes using picoNewton optical forces applied by multiple optical traps.

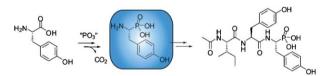
4518

Phosphonopeptide K-26 biosynthetic intermediates in Astrosporangium hypotensionis

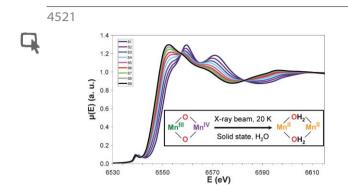
Ioanna Ntai, Vanessa V. Phelan and Brian O. Bachmann*

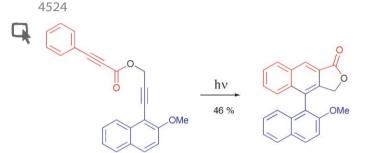
The nonproteinogenic amino acid (*R*)-1-amino-2-(4-hydroxyphenyl)ethylphosphonic acid was synthesized in ${}^{13}C_2$ -labeled form and its intermediacy in the biosynthesis of the natural phosphonopeptide K-26 was validated by incorporation studies.





COMMUNICATIONS





OH

OH

X-Ray photoreduction of a di($\mu\text{-}oxo)Mn^{\rm III}Mn^{\rm IV}$ complex occurs at temperatures as low as 20 K

Lionel Dubois, Lilian Jacquamet, Jacques Pécaut and Jean-Marc Latour*

Full reduction of the $Mn^{II}(\mu$ -O)₂ Mn^{IV} core to $Mn^{II}(\mu$ -OH₂)₂ Mn^{II} is observed upon irradiation by an X-ray beam at *ca.* 20 K.

Synthesis of 1,1'-binaphthyls by *photo-dehydro*-Diels-Alder reactions

Pablo Wessig* and Gunnar Müller

The *photo-dehydro*-Diels–Alder reaction has proved to be a powerful method for the preparation of 1,1'-binaphthyls.

Halide-free ethylation of phenol by multifunctional catalysis using phosphinite ligands

M. Carmen Carrión and David J. Cole-Hamilton*

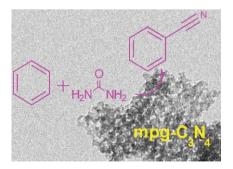
Phenol is catalytically *ortho*-alkylated by C–H activation, with significant steps occurring at the metal and at the P atom of phosphinite ligands.

4530

4527 ОН

 C_2H_4

Rh/Ph₂POPh



Metal-free catalysis of sustainable Friedel–Crafts reactions: direct activation of benzene by carbon nitrides to avoid the use of metal chlorides and halogenated compounds

Frederic Goettmann, Anna Fischer, Markus Antonietti and Arne Thomas*

The use of mesoporous C_3N_4 for the activation of benzene enables more sustainable Friedel–Crafts type reaction by using alcohols, formic acid and even quaternary ammoniums or urea as electrophiles.

Solvent-free, heterogeneous photooxygenation of hydrocarbons by Hyflon[®] membranes embedding a fluorous-tagged decatungstate

Mauro Carraro, Martino Gardan, Gianfranco Scorrano, Enrico Drioli, Enrica Fontananova and Marcella Bonchio*

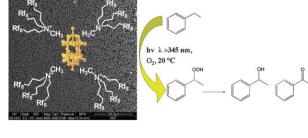
Hybrid fluoropolymeric membranes with 25% loading of the fluorous-tagged $(R_{\rm f}N)_4W_{10}O_{32}$ effect the solvent-free photooxygenation of benzylic C–H bonds with up to 6100 TONs in 4 hours.

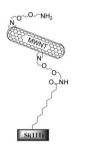
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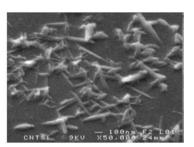
Carbon nanotube-functionalized silicon surfaces with efficient redox communication

Fanny Hauquier, Giorgia Pastorin, Philippe Hapiot, Maurizio Prato,* Alberto Bianco* and Bruno Fabre*

Functionalized multi-walled carbon nanotubes (MWNTs) were covalently bound to a silicon surface and such MWNT assemblies allowed an efficient electrical communication between the underlying semiconductor and a redox probe in solution.







4539

Organic–inorganic hybrid nanomaterial as a new fluorescent chemosensor and adsorbent for copper ion

Soo Jin Lee, Shim Sung Lee, Myoung Soo Lah, Jae-Min Hong and Jong Hwa Jung*

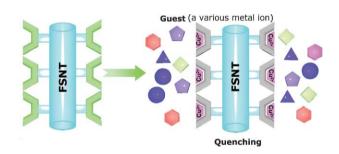
Functionalized silica nanotube (FSNT) possessing the phenanthroline moiety as a fluorescent receptor and adsorbent for Cu^{2+} was fabricated by sol–gel reaction. The FSNT showed the high selective binding ability of FSNT for only Cu^{2+} .

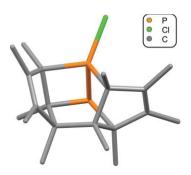
4542

The surprising and stereoselective formation of P_2C_{10} cages by the reduction of Cp^*PCl_2

Oliver C. Presly, Michael Green, John C. Jeffery, Eva Leiner, Martin Murray, Christopher A. Russell,* Manfred Scheer* and Ulf Vogel

Reactions of Cp*PCl₂ with Group 13 reducing agents Ga, GaI on InCl, result in a cascade of P–C, P–P and C–C bond forming reactions and the stereoselective formation of P_2C_{10} cages.





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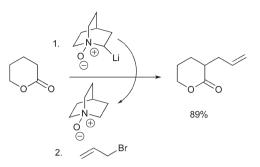
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α-Lithio quinuclidine *N*-oxide (Li-QNO): A new base for synthetic chemistry

Ian A. O'Neil,* Inder Bhamra and Peter D. Gibbons

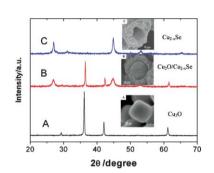
In a tandem process, α -lithio quinuclidine *N*-oxide (Li-QNO) behaves as a base then an HMPA mimetic in a range of synthetically useful reactions.

4548

Conversion of Cu_2O nanocrystals into hollow $Cu_{2-x}Se$ nanocages with the preservation of morphologies

Hongliang Cao, Xuefeng Qian,* Jiantao Zai, Jie Yin and Zikang Zhu

 Cu_{2-x} Se hollow cages were fabricated by the reaction of shape-controlled Cu₂O single-crystal templates with suitable selenium source based on the Kirkendall effect and "Ostwald ripening" process at room temperature.



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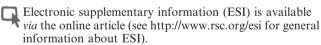
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