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ISSN 1359-7345 CODEN CHCOFS (36) 4489-4620 (2005)



Cover

See Toshihiro Ihara et al., page 4523. Fluorescent labelling of the repetitive sequence through cooperative hybridization. Image reproduced by permission of Yusuke Kitamura, Toshihiro Ihara, Kenji Okada, Yusuke Tsujimura, Yoshinori Shirasaka, Masato Tazaki and Akinori Jyo, from Chem. Commun., 2005, 4523-4525.



Inside cover See Yasushi Tsuji et al., page 4526. A positive dendrimer effect was found in the hydrosilylation of ketones catalyzed by rhodium N-heterocyclic carbene complexes. Image reproduced by permission of Tetsuaki Fujihara, Yasushi Obora, Makoto Tokunaga, Hiromichi Sato and Yasushi Tsuji, from Chem. Commun., 2005, 4526-4528.

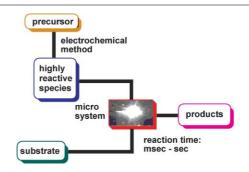
FEATURE ARTICLE

4509

Flash chemistry using electrochemical method and microsystems

Jun-ichi Yoshida

This feature article provides a brief outline of the concept of flash chemistry for conducting extremely fast reactions in organic synthesis using electrochemically generated highly reactive species and microsystems.



COMMUNICATIONS

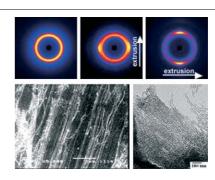
4517



A simple extrusion method for the synthesis of aligned silica nanowires using the template of a rigid surfactant mesophase

Limin Liu, Grace Tan, Vivek Agarwal, Arijit Bose, Jibao He, Gary L. McPherson and Vijay T. John*

The hexagonal domains of a rigid bis-2-ethylhexyl sodium sulfosuccinate and α-phosphatidylcholine surfactant mesophase are oriented by extrusion prior to silica precipitation in the hexagonal aqueous channels to form aligned 10-20 nm silica nanowires.



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Misleading effects of impurities derived from the extremely long-lived electron-transfer state of 9-mesityl-10-methylacridinium ion

Kei Ohkubo, Hiroaki Kotani and Shunichi Fukuzumi*

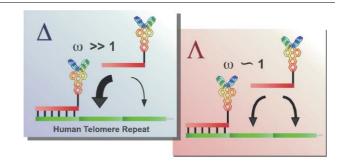
9-Mesityl-10-methylacridinium ion has an extremely long-lived electron-transfer state, as revealed from the misleading effects from some impurities in benzonitrile and synthesized acridinium ion, such as aromatic amines and acridine.

4523

Asymmetric cooperativity in tandem hybridization of enantiomeric metal complex-tethered short fluorescent DNA probes

Yusuke Kitamura, Toshihiro Ihara,* Kenji Okada, Yusuke Tsujimura, Yoshinori Shirasaka, Masato Tazaki and Akinori Jyo

The complex $[Ru(phen)_2(dppz)]^{2+}$ was attached to the 5' end of a short oligonucleotide to form conjugates, the Δ -isomer of which showed a high cooperativity during the recognition of the repetitive sequence, while the Δ -isomer did not.

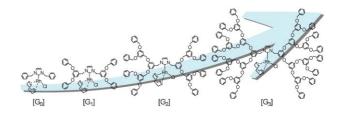


4526

Dendrimer N-heterocyclic carbene complexes with rhodium(I) at the core

Tetsuaki Fujihara, Yasushi Obora, Makoto Tokunaga, Hiromichi Sato and Yasushi Tsuji*

Rhodium(I) *N*-heterocyclic carbene complexes bearing Fréchet-type polybenzyl ether dendrimers have been synthesized and characterized. In the hydrosilylation of ketones with these complexes as a catalyst, a positive dendrimer effect was observed, in which the yields of products increased with an increase in the dendrimer generation.

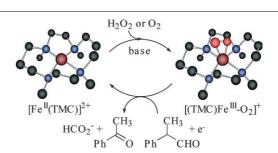


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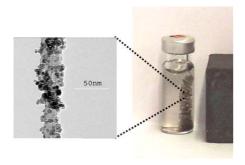
Mononuclear nonheme ferric-peroxo complex in aldehyde deformylation

Jamespandi Annaraj, Yumi Suh, Mi Sook Seo, Sun Ok Kim and Wonwoo Nam*

A mononuclear nonheme ferric-peroxo complex bearing a macrocyclic tetradentate N4 ligand, [(TMC)Fe^{III}–O₂][†], was prepared and used in mechanistic studies of aldehyde deformylation; a catalytic aldehyde deformylation by a nonheme iron(II) complex, [Fe^{II}(TMC)]²⁺, and molecular oxygen is reported as well.



4532



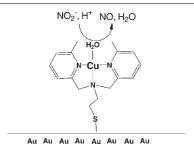
An easy and economically viable route for the decoration of carbon nanotubes by magnetite nanoparticles, and their orientation in a magnetic field

François Stoffelbach, Abdelhafid Aqil, Christine Jérôme, Robert Jérôme* and Christophe Detrembleur

A simple, cheap and tunable approach for the decoration of carbon nanotubes by magnetite nanoparticles and their orientation in a magnetic field is reported.

4534



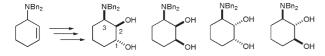


Electroreduction of nitrite on gold electrode modified with Cu-containing nitrite reductase model complex

Takamitsu Hiratsu, Shinnichiro Suzuki and Kazuya Yamaguchi*

A gold electrode modified with model complexes for the nitrite reduction centre of copper-containing nitrite reductase catalyzed efficiently the electroreduction of nitrite to nitrogen monoxide under acidic conditions.

4536



1,2-anti-2,3-syn 1,2-syn-2,3-syn 1,2-syn-2,3-anti 1,2-anti-2,3-anti

Ammonium directed dihydroxylation of N,Ndibenzylaminocyclohex-2-ene: metal-free syntheses of the diastereoisomers of 3-dibenzylamino-1,2dihydroxycyclohexane

Stephen G. Davies,* Marcus J. C. Long and Andrew D. Smith

A highly diastereoselective, metal-free, ammonium ion directed dihydroxylation protocol has been established and applied to the synthesis of all the diastereoisomers of 3-dibenzylamino-1,2-dihydroxycyclohexane.

4539









Oxidise



Asymmetric proliferation with optically active polyanilines

Yingpit Pornputtkul, Leon A. P. Kane-Maguire,* Peter C. Innis and Gordon G. Wallace*

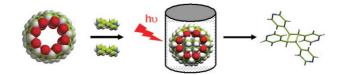
Thin optically active polyaniline layers have been shown to function as platforms to induce optical activity in polyanilines (containing achiral dopants) that are formed by subsequent electrodeposition.

4542

Template directed photodimerization of *trans*-1,2-bis(*n*-pyridyl)ethylenes and stilbazoles in water

Mahesh Pattabiraman, Arunkumar Natarajan, Raja Kaliappan, Joel T. Mague and V. Ramamurthy*

Cucurbutril serves as a versatile solution phase template to orient photoreactive olefins towards a single dimer.

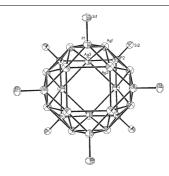


4545

Synthesis and crystal structure of [tBu₃SiPAg₂]₈: A novel Ag₁₆-cluster featuring a remarkable symmetrical structure

Hans-Wolfram Lerner,* Günter Margraf, Jan W. Bats and Matthias Wagner

The synthesis was achieved by a metathesis reaction of AgOCN with tBu₃SiPNa₂.

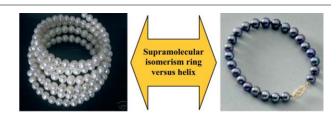


4548

Concomitant crystallization of two polymorphs—a ring and a helix: concentration effect on supramolecular isomerism

Katharina M. Fromm,* Jorge L. Sagué Doimeadios and Adeline Y. Robin

For the first time in Ag coordination chemistry, two supramolecular isomers were isolated from the same reaction mixture, one forming a [Ag₂L₂]-ring, the other a helical single-stranded structure [...L-Ag-L-Ag-L...].



4551

A ligand-free solid-supported system for Sonogashira couplings: applications in nucleoside chemistry

Neil K. Garg, Carolyn C. Woodroofe, Christopher J. Lacenere, Stephen R. Quake* and Brian M. Stoltz*

A mild heterogeneous, ligand-free protocol for Sonogashira and Heck couplings has been developed. Using these methods, the facile preparation of several biologically important alkyne-appended deoxynucleoside derivatives is described.

4554

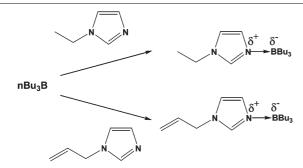


Nickel-catalyzed coupling of isocyanates with 1,3-iodoesters and halobenzenes: a novel method for the synthesis of imide and amide derivatives

Jen-Chieh Hsieh and Chien-Hong Cheng*

Substituted imide and amide derivatives were conveniently prepared from the reaction of isocyanates with o-iodobenzoates and haloarenes catalyzed by the Ni(dppe)Br₂/dppe/Zn system in moderate to good yields with excellent tolerance of functional groups.

4557



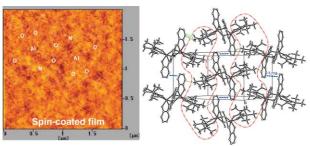
Liquid imidazole-borane complex

Noriyoshi Matsumi, Ayano Mori, Kenji Sakamoto and Hiroyuki Ohno*

Liquid imidazole–alkylborane complex was prepared as a new class of electrolyte and designer solvent.

4560





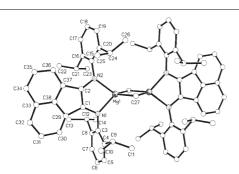
Strongly luminescent binuclear aluminium chelate with polymer-like molecular packing and solution-processibility

Juan Qiao, Li. D. Wang, Jun. F. Xie, Gang. T. Lei, Guo. S. Wu and Yong Qiu*

A novel binuclear aluminium heteroleptic chelate exhibits polymer-like molecular packing and solution-processibility, as well as high photoluminescence, which showed encouraging performance as the active layer in the multilayer small-molecule OLEDs prepared *via* low-cost spin-coating.

4563





Structural and EPR characterisation of single electron and alkyl transfer products from reaction of dimethyl magnesium with bulky α -diimine ligands

Philip J. Bailey,* Robert A. Coxall, Caroline M. Dick, Sylvie Fabre, Simon Parsons and Lesley J. Yellowlees

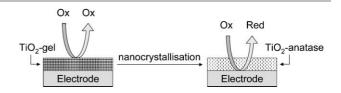
Treatment of dimethylmagnesium with bulky α -diimine ligands provides either the biradical methyl-bridged complexes $[(\alpha\text{-diimine}^-)Mg^+(\mu\text{-CH}_3)]_2$ via single electron transfer, or the product of methyl transfer to an imine carbon atom, depending upon conditions.

4566

Electrochemical evidences of morphological transformation in ordered mesoporous titanium oxide thin films

Mathieu Etienne,* David Grosso, Cédric Boissière, Clément Sanchez and Alain Walcarius

We provide the first electrochemical evidence of permeability changes in continuous mesoporous TiO₂ thin film electrodes induced by nanocrystallisation.



4569

Visible light-induced selective oxidation of cyclohexane to cyclohexanone on Cr-Si binary oxide with molecular oxygen

Yasuhiro Shiraishi,* Yugo Teshima and Takayuki Hirai

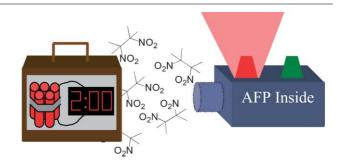
Cr–Si binary oxide prepared by a sol–gel method, when activated by visible light, catalyzes highly selective oxidation of cyclohexane to cyclohexanone with molecular oxygen.

4572

Amplifying fluorescent polymer sensors for the explosives taggant 2,3-dimethyl-2,3-dinitrobutane (DMNB)

Samuel W. Thomas III, John P. Amara, Rebekah E. Bjork and Timothy M. Swager*

Structural and electronic effects on the detection of vapor of the explosives taggant DMNB by solid-state fluorescent conjugated polymers are described.



4575

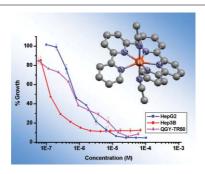
Design and synthesis of artificial phospholipid for selective cleavage of integral membrane protein

Takumi Furuta,* Minatsu Sakai, Hiroyasu Hayashi, Tomohiro Asakawa, Fumi Kataoka, Satoshi Fujii, Takashi Suzuki, Yasuo Suzuki, Kiyoshi Tanaka, Nathan Fishkin and Koji Nakanishi*

An artificial phospholipid, possessing saturated alkyl chains as a membrane anchor and protein recognition site as well as an Fe(III)–EDTA moiety as a protein cleavable polar head group, was designed and synthesized for the purpose of examination of cleavage of integral membrane proteins.

4578

4

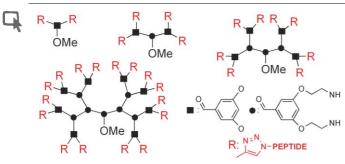


Highly cytotoxic iron(II) complexes with pentadentate pyridyl ligands as a new class of anti-tumor agents

Ella Lai-Ming Wong, Guo-Su Fang, Chi-Ming Che* and Nianyong Zhu

Iron(II) pentadentate pyridyl complexes have high potency toward human liver cancer and its drug resistant cell lines. Biological activities of iron(II) chelates could be modulated through variation of the polypyridyl ligands.

4581



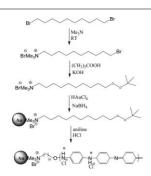
Efficient microwave-assisted synthesis of multivalent dendrimeric peptides using cycloaddition reaction (click) chemistry

Dirk T. S. Rijkers, G. Wilma van Esse, Remco Merkx, Arwin J. Brouwer, Hans J. F. Jacobs, Roland J. Pieters and Rob M. J. Liskamp*

Multivalent dendrimeric peptides were synthesized *via* a microwave-assisted Huisgen 1,3-dipolar cycloaddition between azido peptides and dendrimeric alkynes in yields ranging from 46 to 96%.

4584

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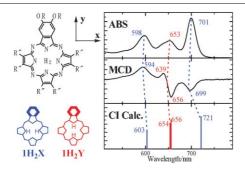


Synthesis of polyaniline-gold nanocomposites using "grafting from" approach

Sunil K. Pillalamarri, Frank D. Blum and Massimo F. Bertino*

Au nanoparticles were synthesized with 10-bromodecylperoxide as a surfactant. Aniline was oxidized by the peroxide groups of the surfactant to yield high aspect ratio polyaniline attached to the Au nanoparticles.

4586



Selective detection of minor prototropic tautomers in low-symmetry tetraazaporphyrin derivatives by the combined use of electronic absorption, MCD, and CI calculations

Kazuyuki Ishii, Hatsumi Itoya, Hideya Miwa and Nagao Kobayashi*

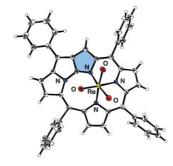
A series of low-symmetry metal-free benzo-substituted tetraazaporphyrin derivatives has been investigated by electronic absorption, MCD, and CI calculations, proving the existence of two prototropic tautomers.

4589

Re(VII) complex of N-fused tetraphenylporphyrin

Motoki Toganoh, Shinya Ikeda and Hiroyuki Furuta*

The ability of the N-fused porphyrin ligand to stabilize metal complexes in high oxidation states was demonstrated through the synthesis of its Re(VII) trioxo complex.



4592

Ligand-accelerated vanadium-catalysed epoxidation in water

Zaïnaba Bourhani and Andrei V. Malkov*

Vanadium-catalysed epoxidation of allylic alcohols, a classical example of ligand-decelerated catalysis, in water it turns into a ligand-accelerated process.

1:1 ee 60%

Ligand*/Vanadium

4595

Direct aerobic epoxidation of alkenes catalyzed by metal nanoparticles stabilized by the $H_5PV_2Mo_{10}O_{40}$ polyoxometalate

Galia Maayan and Ronny Neumann*

Ag and Ru nanoparticles stabilized by $H_5PV_2Mo_{10}O_{40}were$ prepared by a sequence of redox reactions. These particles supported on $\alpha\text{-alumina}$ showed catalytic activity for the direct aerobic epoxidation of alkenes.

$$R \stackrel{\text{R'}}{\longrightarrow} R' + O_2$$
 $\stackrel{\text{M}_n - \text{H}_5 PV_2 MO_{10} O_{40}/\alpha - \text{Al}_2 O_3}{\longrightarrow} R \stackrel{\text{O}}{\longrightarrow} R'$
 $R = \text{alkyl}; R' - \text{alkyl}, H \qquad M = \text{Ru}, \text{Ag}, \text{Rh}, \text{Ir}$

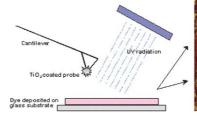


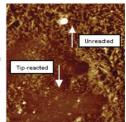
4598

Current-less photoreactivity catalyzed by functionalized AFM tips

Vasiliki Zorbas, Mandakini Kanungo, Sukhmine A. Bains, Yuanbing Mao, Tirandai Hemraj-Benny, James A. Misewich and Stanislaus S. Wong*

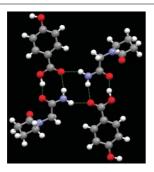
Schematic of the experimental protocol used for an AFM tip-initiated, highly-localized surface photocatalytic reaction in air. Height images presented show clear differences in surface roughness between reacted *vs.* unreacted regions.





4601



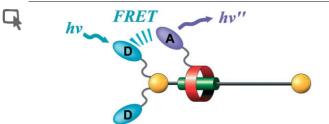


Crystal engineering of pharmaceutical co-crystals from polymorphic active pharmaceutical ingredients

Peddy Vishweshwar, Jennifer A. McMahon, Matthew L. Peterson, Magali B. Hickey, Tanise R. Shattock and Michael J. Zaworotko*

Crystal engineering of pharmaceutical co-crystals represents a recent and potentially broad strategy for formulation of active pharmaceutical ingredients (APIs). We report herein that piracetam forms pharmaceutical co-crystals with two other APIs and that these new compositions appear to be less prone to polymorphism than the parent APIs.

4604



Fluorescence resonance energy transfer across a mechanical bond of a rotaxane

Hideki Onagi and Julius Rebek, Jr.*

Fluorescence transfer across a donor-acceptor tagged rotaxane was studied and a small conformational change of the rotaxane observed using fluorescent spectroscopy and ROESY NMR.

4607

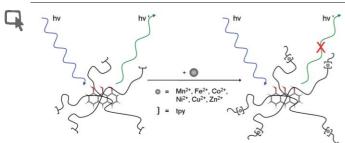


A novel route for waste water treatment: photo-assisted Fenton degradation of dye pollutants accumulated in natural polyelectrolyte microshells

Xia Tao,* Jingmei Su, Jianfeng Chen* and Jincai Zhao

The efficient accumulation of dyes in constructed natural polyelectrolyte microshells under moderate conditions, combined with the photo-assisted Fenton reagent, opens up a new route for the effective elimination of dye pollutants from waste water.

4610



Fluorescent sensing of transition metal ions based on the encapsulation of dithranol in a polymeric core shell architecture

Michael A. R. Meier and Ulrich S. Schubert*

A 5-arm star-shaped poly(ethylene glycol) polymer has been end-group functionalized with terpyridine ligands. This new polymer is capable of binding a variety of transition metal ions. Moreover, it can encapsulate functional molecules. Combining these two concepts it was possible to develop a novel kind of fluorescent sensor for transition metal ions.

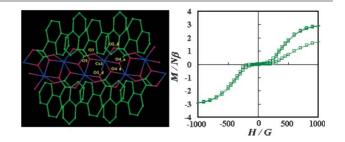
4613



Diversity in magnetic properties of 3D isomorphous networks of Co(II) and Mn(II) constructed by napthalene-1,4-dicarboxylate

Tapas Kumar Maji, Wakako Kaneko, Masaaki Ohba and Susumu Kitagawa*

Two novel 3D isomorphous frameworks, $[M(1,4-napdc)]_n$ $(M = Co(II) \text{ and } Mn(II), 1,4-napdc^2 = naphthalene-1,4$ dicarboxylate), have been synthesized and structurally characterized. The variable temperature magnetic measurements exhibit metamagnetic behaviour ($T_{\rm C} = 5.5 \, {\rm K}$) for Co(II), whereas weak antiferromagnetic interaction for Mn(II).



4616



Interfused semiconductor nanocrystals: brilliant blue photoluminescence and electroluminescence

Shinae Jun and Eunjoo Jang*

We describe a method for producing blue light-emitting interfused CdSe//ZnS (QE up to 60%) nanocrystals and report the good performance of an electroluminescent device which uses them (external quantum efficiency ~ 1.5 cd A^{-1}).



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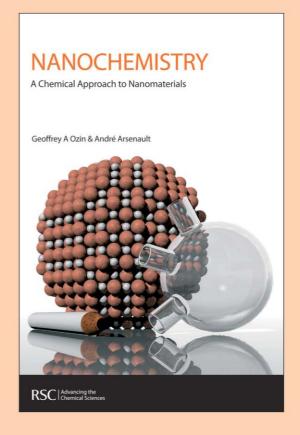
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DIANANE- Salon Ligand -for Asymmetric Nozaki-Hiyama-Kishi Reaction-

(1R,2R,4R,5R)-2,5-Bis(3,5-di-tert-butyl-2-hydroxybenzylideneamino)-

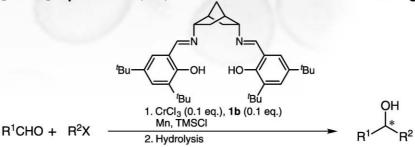
bicyclo[2.2.1]heptane (1a)

100mg [B2652]

(15,25,45,55)-2,5-Bis(3,5-di-tert-butyl-2-hydroxybenzylideneamino)-

bicyclo[2.2.1]heptane (1b)

100mg [B2653]



DIANANE-Salen ligand 1 is a useful reagent for the asymmetric Nozaki-Hiyama-Kishi (NHK) Reaction as a ligand for transition metal complexes. Such complexes can be used as highly enantioselective catalysts to obtain chiral secondary alcohols in high enantiopurity.

A. Berkessel, D. Menche, C. A. Sklorz, M. Schröder, I. Paterson, *Angew. Chem. Int. Ed.*, **42**, 1032 (2003); I. Paterson, H. Bergmann, D. Menche, A. Berkessel, *Org. Lett.*, **6**, 1293 (2004).



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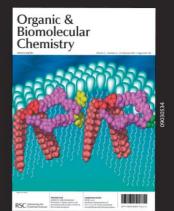
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