ISSN 1359-7345 • CHCOFS (5)477-622 (2004)

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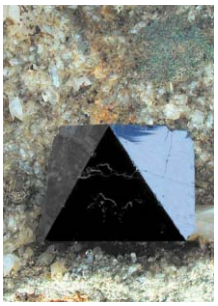

Cover
A crystal of magnetite (on an assembly of crystals of orthose) from Fiesch, Valais, Switzerland. Courtesy of Musée ENSMP. Photo copyright: J. M. Le Cléach, Musée de Minéralogie ENSMP, Paris, France (pp. 481–487).

Chemical biology articles published in this journal also appear in the *Chemical Biology Virtual Journal:* **www.rsc.org/chembiol**

contents

Chemical Communications
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Chemical Communications (print: ISSN 1359-7345; electronic: ISSN 1364-548X) is published 24 times a year by the Royal Society of Chemistry, Thomas Graham House, Science Park, Milton Road, Cambridge, UK CB4 0WF. All orders accompanied by payment should be sent directly to Turpin Distribution Services Ltd, Blackhorse Road, Letchworth, Herts, UK SG6 1HN. 2004 Annual (print + electronic) subscription price: £1045; US$1725. 2004 Annual (electronic) subscription price: £940; US$1552. Customers in Canada will be subject to a surcharge to cover GST. Customers in the EU subscribing to the electronic version only will be charged VAT. If you take an institutional subscription to any RSC journal you are entitled to free, site-wide web access to that journal. You can arrange access via Internet Protocol (IP) address at www.rsc.org/ip. Customers should make payments by cheque in sterling payable on a UK clearing bank or in US dollars payable on a US clearing bank. Periodicals postage paid at Rahway, NJ, USA and at additional mailing offices. Airfreight and mailing in the USA by Mercury Airfreight International Ltd., 365 Blair Road, Avenel, NJ 07001, USA. US Postmaster: send address changes to *Chemical Communications*, c/o Mercury Airfreight International Ltd., 365 Blair Road, Avenel, NJ 07001. All despatches outside the UK by Consolidated Airfreight.
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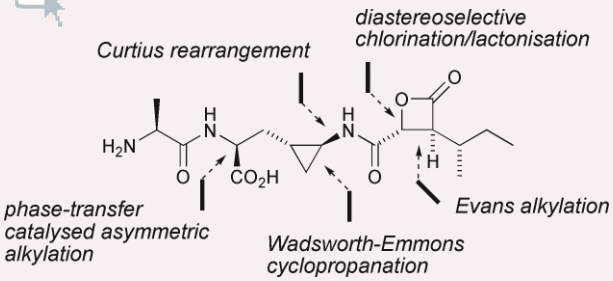
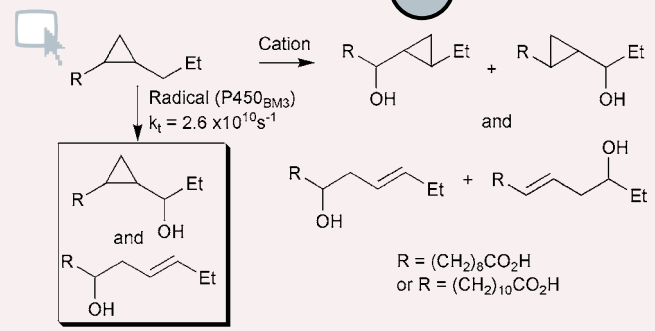


Total synthesis of (+)-belactosin A

Alan Armstrong* and James N. Scutt

A concise and stereocontrolled first total synthesis of the antitumour antibiotic belactosin A is reported, involving coupling of a β-lactone carboxylic acid to *N*-Ala-aminocyclopropyl alanine.



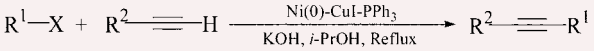
Cyclopropyl fatty acids implicate a radical but not a cation as an intermediate in $P450_{BM3}$-catalysed hydroxylations

Max J. Cryle, Julia M. U. Stuthe, Paul R. Ortiz de Montellano and James J. De Voss*

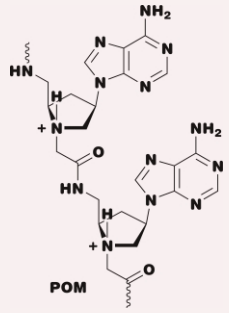
Novel cyclopropyl containing fatty acids are good substrates for $P450_{BM3}$ catalysed hydroxylation. Oxidation products indicate the presence of a radical intermediate (rebound rate ~ 2.6×10^{10} s^{-1}) but not a cationic intermediate.

The Sonogashira coupling reaction catalyzed by ultrafine nickel(0) powder

Lei Wang,* Pinhua Li and Yicheng Zhang



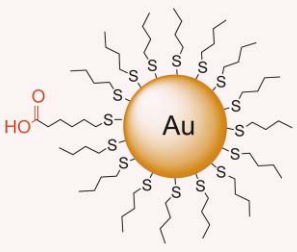
The Sonogashira coupling reaction catalyzed by ultrafine nickel(0) powder has been developed. Terminal alkynes couple with aryl, alkenyl iodide and aryl bromide in the presence of cuprous iodide, triphenylphosphine, potassium hydroxide and ultrafine particle nickel(0) to provide the corresponding cross-coupling products with high yields.



Nucleic acid binding properties of thyminyl and adeninyl pyrrolidine-amide oligonucleotide mimics (POM)

T. H. Samuel Tan, David T. Hickman, Jordi Morral, Ian G. Beadham and Jason Micklefield*

Adeninyl POM, prepared by solid-phase synthesis, exhibits higher affinity than adeninyl PNA for DNA and RNA. Moreover thyminyl POM binds much more rapidly to single stranded RNA than DNA.

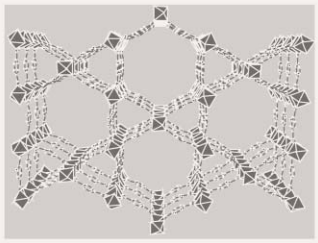


Controlled functionalization of gold nanoparticles through a solid phase synthesis approach

James G. Worden, Andrew W. Shaffer and Qun Huo*

Gold nanoparticles with controlled functionality are prepared through a solid phase synthesis technique.

520



$V^{III}(OH)\{O_2C–C_6H_4–CO_2\}.(HO_2C–C_6H_4–CO_2H)_x(DMF)_y(H_2O)_z$ (or MIL-68), a new vanadocarboxylate with a large pore hybrid topology : reticular synthesis with infinite inorganic building blocks?

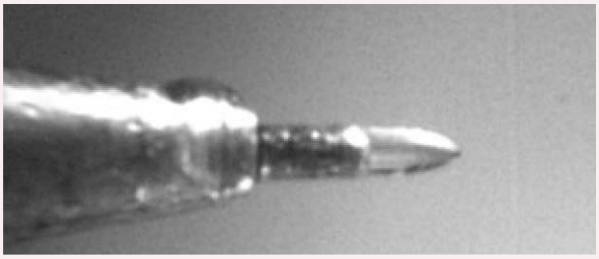
K. Barthelet, J. Marrot, G. Férey and D. Riou*

MIL-68 is a vanado(III)terephthalate compound whose porous framework is described as an 'augmented' hexagonal bronze structure. This allows the extension of the reticular synthesis concept to edifices built from infinite inorganic building blocks.

522

Electrospray mass spectrometry of undiluted ionic liquids

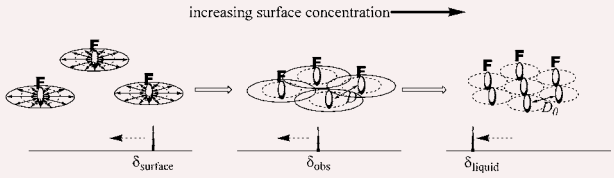
Glen P. Jackson and Douglas C. Duckworth*



An *undiluted* ionic liquid Taylor cone forms during an electrospray mass spectrometric (ESMS) analysis. ESMS of neat ionic liquids provides a new approach for analyzing reactive ionic liquids or dissolved catalysts.

524

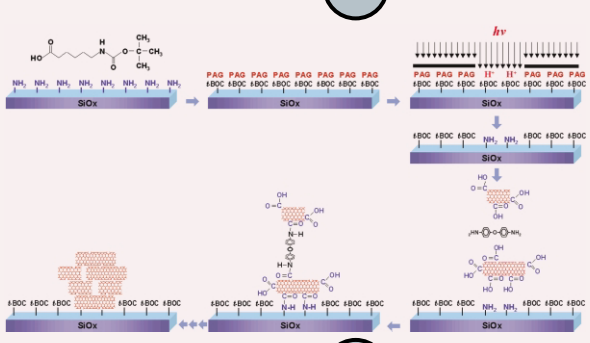
Surface energy and surface area measurements by ^{19}F MAS NMR of adsorbed trifluoroacetic acid



Vitaliy L. Budarin, James H. Clark and Stewart J. Tavener*

Trifluoroacetic acid, when adsorbed on the surface of inorganic materials, is a useful ^{19}F NMR probe molecule for studying surface properties including surface energy and surface area.

526



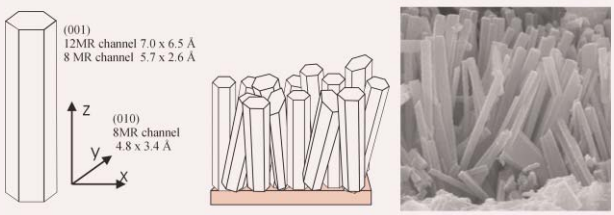
Carbon nanotube conducting arrays by consecutive amidation reactions

Dae-Hwan Jung, Myung Sup Jung, Young Koan Ko, Seung Joo Seo and Hee-Tae Jung*

Carbon nanotube conducting arrays with high-density multilayers were constructed *via* consecutive amidation reactions with the aid of a linker molecule and a condensation agent on a patterned amine-terminated glass substrate.

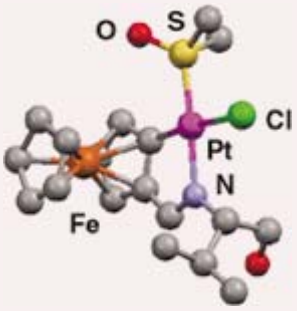
528

Preparation of highly accessible mordenite coatings on ceramic monoliths at loadings exceeding 50% by weight



M. A. Ulla, E. Miro, R. Mallada, J. Coronas and J. Santamaría*

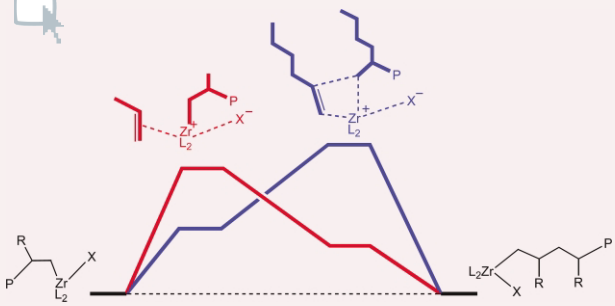
Mordenite coatings with a high accessibility have been synthesised on cordierite monolith supports at loadings exceeding 50% by weight.



Easy access to diastereomerically pure platinacycles

Concepción López,* Amparo Caubet, Sonia Pérez, Xavier Solans and Mercè Font-Bardía

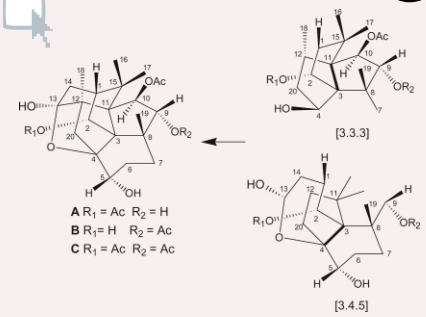
The method for the preparation and isolation of the first examples of diastereomerically pure platinacycles containing simultaneously a chiral carbon and a σ[Pt-Csp^2(ferrocene)] bond is described, and the factors affecting their formation are also discussed.



The kinetics of propene and hexene polymerisation with [(SBI)ZrR]$^+$X$^-$: evidence for monomer-dependent early or late transition states

Fuquan Song, Roderick D. Cannon and Manfred Bochmann*

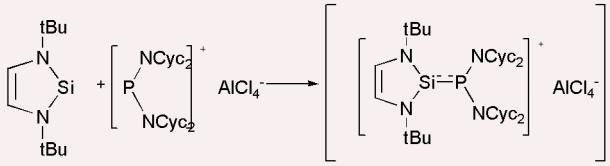
Whereas the rates of *propene* polymerisation catalysed by zirconocene ion pairs are strongly anion-dependent, *hexene* polymerisations are not: the findings demonstrate the existence of very different kinetic regimes for two closely related reactions.



First three examples of taxane-derived di-propellanes in *Taxus canadensis* needles

Qing Wen Shi, Françoise Sauriol, Alain Lesimple and Lolita O. Zamir*

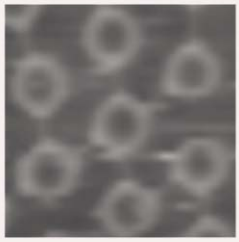
The first three examples of taxane-derived [3.3.3][3.4.5] di-propellanes isolated from the needles of a yew tree are reported. They differ in their acetylation pattern and their biogenesis from a putative taxane precursor is proposed.



Insights into the reactivity and structure of silylene phosphonium ions

Rudolf Pietschnig*

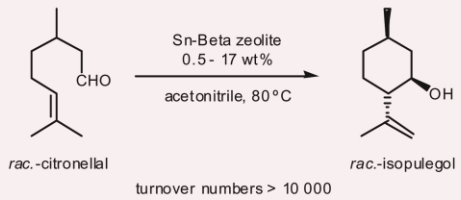
Experimental evidence for the intermediate occurrence of a so far unknown silylene phosphonium ion is presented and its reactivity and bond situation is discussed on the basis of *ab initio* calculations.



Fabrication of two-dimensionally ordered macroporous silica materials with controllable dimensions

Mandakini Kanungo and Maryanne M. Collinson*

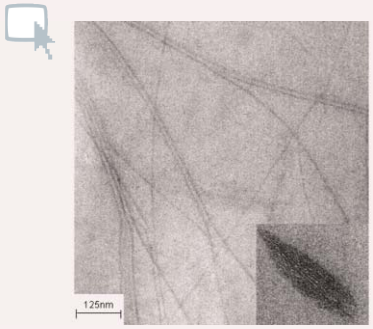
The formation of 2-D arrays of cavities of varying size and depth on an electrode surface *via* colloidal templating is described.



Sn-Beta zeolite as diastereoselective water-resistant heterogeneous Lewis-acid catalyst for carbon–carbon bond formation in the intramolecular carbonyl–ene reaction

Avelino Corma* and Michael Renz

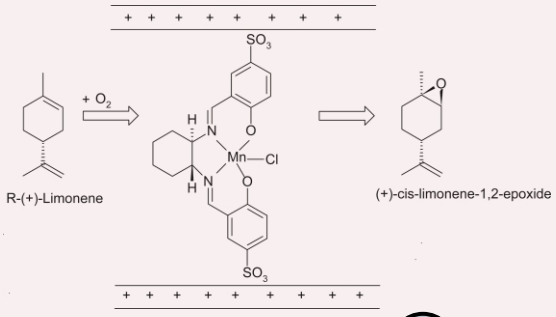
Sn-Beta is a heterogeneous water-tolerant Lewis acid that catalyses the isomerisation of citronellal to isopulegol with high diastereoselectivity. Due to its heterogeneous nature it can be employed not only in batch reactors but also in fixed bed reactors achieving very high turnover numbers.



Porphyrin–acetylene–thiophene polymer wires

Guangtao Li, Tianyu Wang, Andrea Schulz, Sheshanath Bhosale, Matthias Lauer, Pamela Espindola, Jürgen Heinze and Jürgen-Hinrich Fuhrhop*

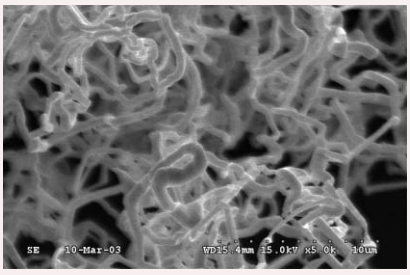
5,15-Bis[acetylene-4-(ethylenedioxy)thiophene]-10,20-bis(4-carboxyphenyl)-porphyrin was synthesized by a Sonogashira coupling and polymerized to fibres; TEM and AFM images show uniform porphyrin wires with a length of several micrometers and monomolecular thickness.



Synthesis and characterization of novel chiral sulfonato–salen–manganese(III) complex in a zinc–aluminium LDH host

Samiran Bhattacharjee and James A. Anderson*

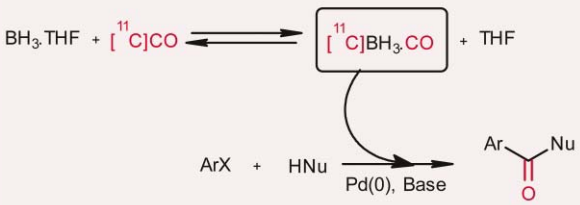
A salen–Mn(III) complex, intercalated within the layers of an LDH host acted as an effective, stable, stereoselective heterogeneous catalyst for the epoxidation of *R*-(+)-limonene using molecular oxygen at room temperature.



Template-free synthesis of single-crystalline cadmium nanotubes

Pingàn Hu, Yunqi Liu,* Lei Fu, Lingchao Cao and Daoben Zhu*

Single-crystalline cadmium nanotubes growing in the form of a heap were produced for the first time by a template-free method in high yield

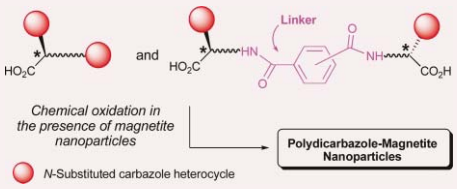


Utilisation of [^{11}C]-labelled boron carbonyl complexes in palladium carbonylation reaction

Hélène Audrain,* Laurent Martarello, Antony Gee and Dirk Bender

The use of the [^{11}C]BH_3·CO complex as a source of carbon monoxide in the carbonylation of iodobenzene catalysed by palladium(0) is described, which allows the synthesis of an amide and a lactone in a straightforward manner.

560

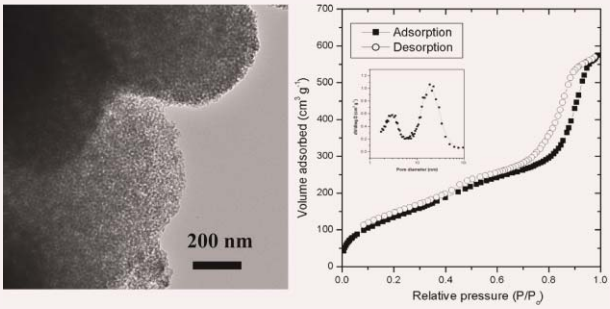


New magnetically responsive polydicarbazole-magnetite nanoparticles

Jean-Paul Lellouche,* Nurit Perlman, Augustine Joseph, Senthil Govindaraji, Ludmila Buzhansky, Aline Yakir and Ian Bruce

Magnetically responsive COOH-polydicarbazole-magnetite nanocomposites have been prepared by chemical oxidation of COOH-dicarbazole monomers in the presence of magnetite nanoparticles. Resulting nanoparticles have been tested for DNA hybridization experiments.

562

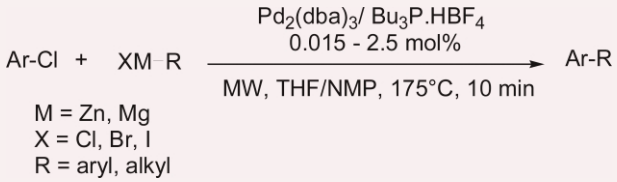


Mesocellular polymer foams with unprecedented uniform large mesopores and high surface areas

Jinwoo Lee, Jaeyun Kim, Sang-Wook Kim, Chae-Ho Shin* and Taeghwan Hyeon*

Mesocellular polymer foams with uniform ~17 nm cellular pores were fabricated using mesocellular silica foams as inorganic templates. The mesocellular polymer foams have high surface areas up to ~600 m^2g^{-1} and pore volumes of 1.6 cm^3g^{-1}

564

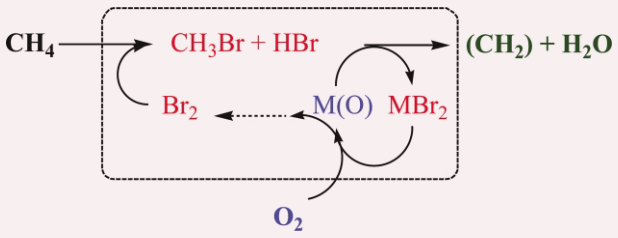


Microwave-assisted Negishi and Kumada cross-coupling reactions of aryl chlorides

Peter Walla and C. Oliver Kappe*

Fast and efficient microwave protocols for C–C coupling reactions of the Negishi and Kumada type involving aryl chlorides are described. The application to solid-phase synthesis is also reported.

566

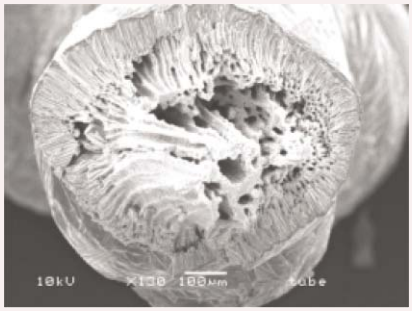


C_1 Coupling *via* bromine activation and tandem catalytic condensation and neutralization over CaO/zeolite composites

Ivan Lorkovic,* Maria Noy, Mike Weiss, Jeff Sherman, Eric McFarland, Galen D. Stucky and Peter C. Ford

Methane is partially oxidized by O_2 to olefins using Br_2 as a mediator for methane activation. Intermediate CH_xBr_{4-x} are condensed to higher hydrocarbons and HBr is neutralized over a regenerable CaO/ZSM-5 composite. Br_2 is released upon treating the spent composite with O_2.

568

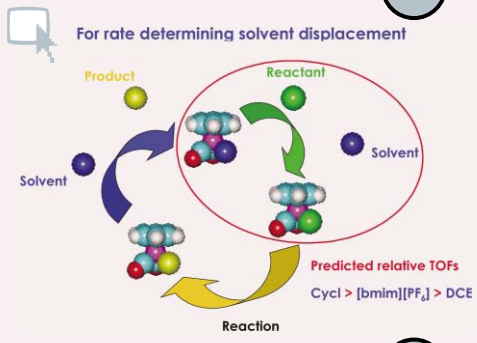


Spontaneous template-free assembly of ordered macroporous titania

Andrew Collins, Daniel Carriazo, Sean A. Davis* and Stephen Mann

Titania powders and fibres with ordered macroporosity have been produced by the facile reaction of titanium alkoxides with aqueous ammonia solution.

590

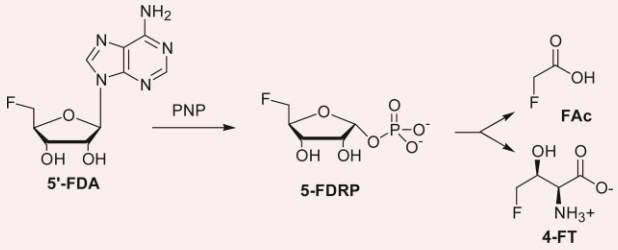


Modelling catalytic turnover frequencies in ionic liquids: the determination of the bimolecular rate constant for solvent displacement from $[(C_6H_6)Cr(CO)_2Solv]$ in 1-*n*-butyl-3-methylimidazolium hexafluorophosphate

Konrad Swiderski, Andrew McLean,* Charles M. Gordon* and D. H. Vaughan

The bimolecular rate constant for solvent displacement from $[(C_6H_6)Cr(CO)_2Solv]$ by an incoming ligand has been determined in the room temperature ionic liquid, [bmim][PF_6], and is compared to that in cyclohexane and dichoroethane.

592

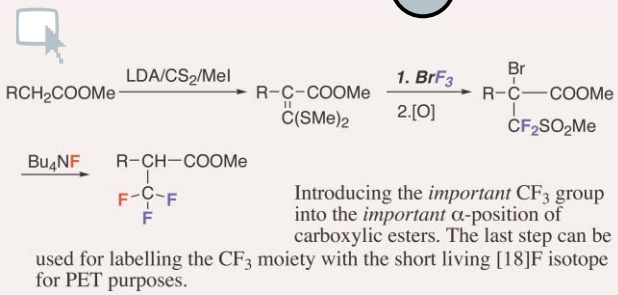


Identification of 5-fluoro-5-deoxy-D-ribose-1-phosphate as an intermediate in fluorometabolite biosynthesis in *Streptomyces cattleya*

Steven L. Cobb, Hai Deng, John T. G. Hamilton, Ryan P. McGlinchey and David O'Hagan*

5′-Fluoro-5′-deoxy-D-ribose-1-phosphate (FDRP) has been identified as a biochemical intermediate during fluorometabolite biosynthesis in *Streptomyces cattleya.*

594



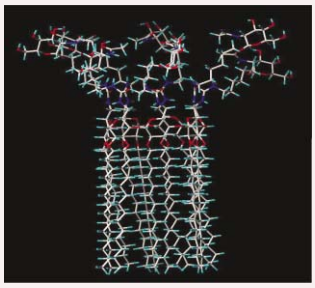
Introducing the *important* CF_3 group into the *important* α-position of carboxylic esters. The last step can be used for labelling the CF_3 moiety with the short living [18]F isotope for PET purposes.

The first general method for α-trifluoromethylation of carboxylic acids using BrF_3

Aviv Hagooly and Shlomo Rozen*

2-Carbomethoxy-1,1-bis(methylsulfide)-1-alkenes, treated with BrF_3, produce the desired α-trifluoromethyl carboxylate derivatives – $RCH(CF_3)COOR'$ – in good yields.

596

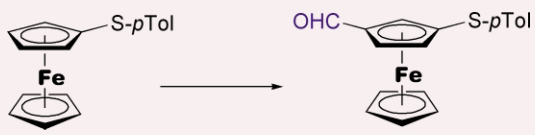


A practical synthesis of amphiphilic cyclodextrins fully substituted with sugar residues on the primary face

Florence Sallas, Kenichi Niikura and Shin-Ichiro Nishimura*

The first synthesis of a fully glycosylated cyclodextrin amphiphile has been achieved: the primary rim is substituted with 7 *N*-acetyl-D-glucosamine units while the secondary rim is substituted with 14 palmitoyl ester chains.

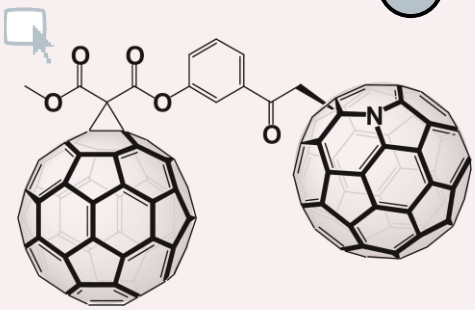
598



A direct *meta*-lithiation route to 1,3-disubstituted ferrocenes

Christophe Pichon, Barbara Odell and John M. Brown*

A non-directed lithiation of ferrocenyl sulfides affords easy access to *meta*-disubstituted ferrocenes.



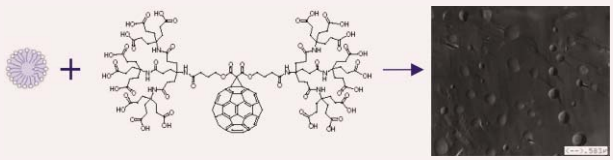
The first fullerene–heterofullerene dyad

Frank Hauke, M. Ángeles Herranz, Luis Echegoyen,* Dirk Guldi,* Andreas Hirsch* and Stefan Atalick

In this paper we present the first example of a fullerene–heterofullerene dyad, synthesized *via* a highly regioselective addition reaction. This molecule represents a very interesting functional architecture that allows for a uni-directional transduction of energy.

Well-defined self-assembling supramolecular structures in water containing a small amount of C_{60}

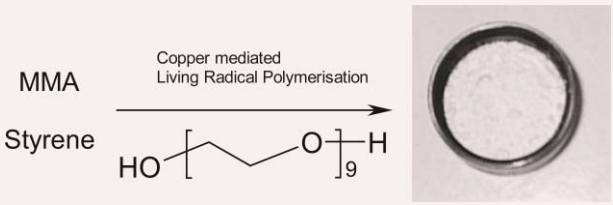
Jingcheng Hao,* Hongguang Li, Weimin Liu and Andreas Hirsch*



The well-defined spherical bilayer vesicles of the dendritic C_{60}-amphiphile–surfactant hybrids were prepared in aqueous solutions containing a small amount of C_{60}.

Poly(ethylene glycol) as solvent for transition metal mediated living radical polymerisation

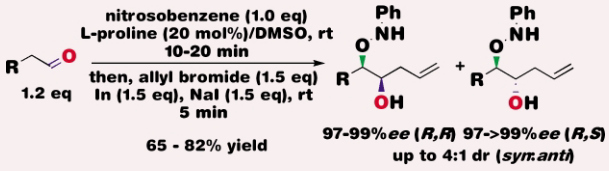
Sébastien Perrier,* Hesna Gemici and Song Li



The use of low molecular weight poly(ethylene glycol) (PEG) as novel solvent for the copper mediated living radical polymerisation of methyl methacrylate and styrene leads to well-defined polymers with low residual copper content.

Tandem aminoxylation–allylation reactions: a rapid, asymmetric conversion of aldehydes to mono-substituted 1,2-diols

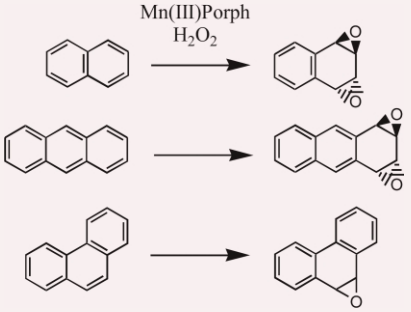
Guofu Zhong*



Enantiopure mono-substituted 1,2-diols were easily made by a rapid one-pot aminoxylation–allylation reaction. The discovery of *in situ* allylation extends the utility and importance far beyond the initial discovery of the asymmetric α-aminoxylation.

An efficient approach for aromatic epoxidation using hydrogen peroxide and Mn(III) porphyrins

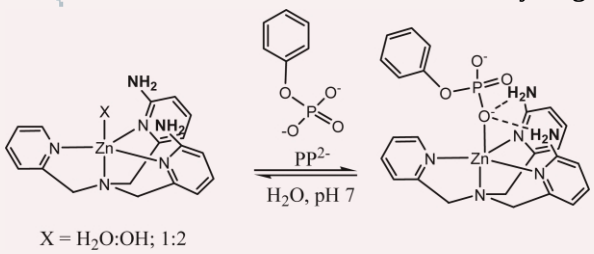
Susana L. H. Rebelo, Mário M. Q. Simões, M. Graça P. M. S. Neves, Artur M. S. Silva and José A. S. Cavaleiro*



In the presence of Mn(III)porphyrins and H_2O_2, naphthalene and anthracene afford the *anti*-1,2:3,4-arene dioxides whereas phenanthrene gives rise to the 9,10-oxide, all in very high conversions and selectivities.

610

The affinity of phosphates to zinc(II) complexes can be increased with hydrogen bond donors

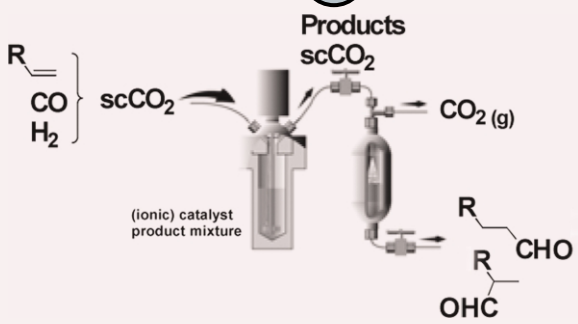


Juan C. Mareque-Rivas,* Rafael Torres Martín de Rosales and Simon Parsons

Non-coordinating amino H-bond donors adjacent to a zinc(II)-centre enhance the affinity of phosphates to the zinc(II) centre

612

Continuous flow homogeneous catalysis using supercritical fluids



Paul B. Webb and David J. Cole-Hamilton*

1-Octene is hydroformylated in a continuous flow process by retaining the catalyst within a reactor dissolved in the steady state mixture of reactants and products and transporting the substrates and products into and out of the reactor dissolved in $scCO_2$.

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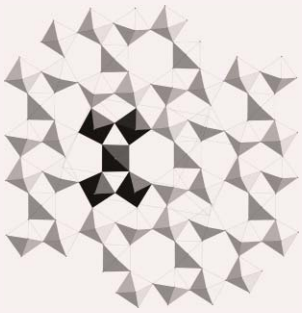
A novel scandium fluoride, $[C_2N_2H_{10}]_{0.5}[ScF_4]$, with an unprecedented tungsten bronze-related layer structure.

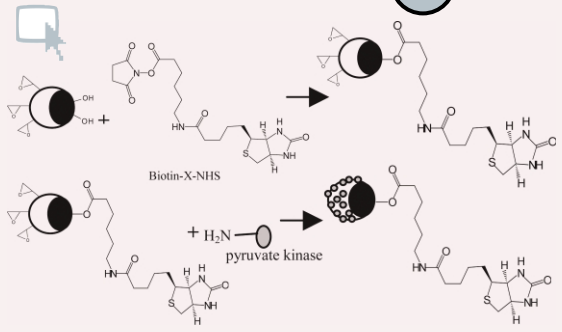


Nicholas F. Stephens, Alexandra M. Z. Slawin and Philip Lightfoot*

$[C_2N_2H_{10}]_{0.5}[ScF_4]$ exhibits a novel structure that consists of single octahedrally connected scandium fluoride layers and has features similar to those of the tungsten bronzes and layered perovskites. It may represent the first in a family of new layered structure types based on these general structural and compositional features.

616

Biotinylated and enzyme-immobilized carrier prepared by hetero-bifunctional latex beads

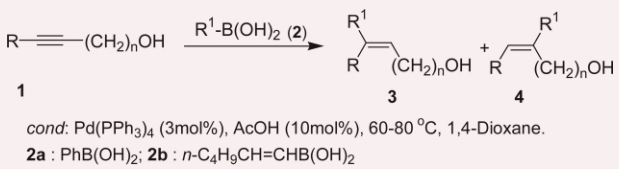


Yong-Zhong Du, Takenori Tomohiro, Gao Zhang, Kazuhiko Nakamura and Masato Kodaka*

Biotinylated and pyruvate kinase immobilized nano-bio element have been prepared using hetero-bifunctional latex beads, where the enzyme activity is roughly half of the free enzyme.

618

On the regioselectivity of Pd-catalyzed additions of organoboronic acids to unsymmetrical alkynes



Nakjoon Kim, Ki Seong Kim, Aruna Kumar Gupta and Chang Ho Oh*

The Pd-catalyzed reaction of unsymmetrical alkynes **1** with organoboronic acids **2** gave a mixture of products **3** and **4**, whose ratios were controlled by the electronic as well as steric effects of the substrates **1**.

COMMUNICATIONS

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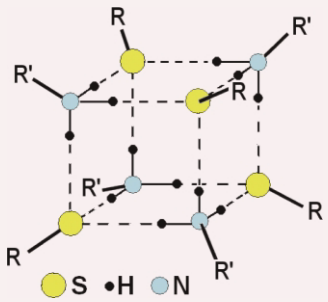

Cubane-like structure of a silanethiol – primary amine assembly – a novel, unusual hydrogen bond pattern

Barbara Becker,* Katarzyna Baranowska, Jarosław Chojnacki and Wiesław Wojnowski

Four molecules of a silanethiol RSH and four molecules of a primary amine $R'NH_2$ combine giving a 'cubane-like' supermolecule, where a somewhat idealized picture shows how its twelve N–H···S hydrogen bonds form a novel, previously unknown
pattern.

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