

IN THIS ISSUE

ISSN 1359-7345 CODEN CHCOFS (31) 3877-4012 (2005)

In this issue...

Chemist, Molecular Biologist, Human Rights Activist: John Meurig Thomas pays tribute to influential scientist Max Perutz. See pp. 3891–3894.



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



Cover

See Javier Vicario, Rienk Eelkema, Wesley R. Browne, Auke Meetsma, René M. La Crois and Ben L. Feringa, page 3936. The cover depicts reels of movie film overlaid by a series of movie frames showing the movement of a microparticle by means of attached motor units: a design intended to emphasize the idea of a moving object. The particle is pushed forward by the catalytic motors engaged in the decomposition of hydrogen peroxide (the fuel) into water and oxygen. Image reproduced by permission of Ben L. Feringa *et al.* from *Chem. Commun.*, 2005, 3936.

CHEMICAL TECHNOLOGY

T29

Chemical Technology highlights the latest applications and technological aspects of research across the chemical sciences.

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August 2005/Volume 2/Issue 8

www.rsc.org/chemicaltechnology

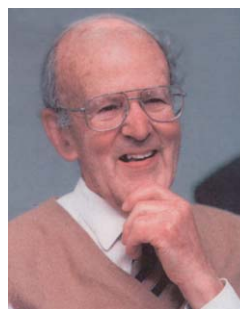
40TH ANNIVERSARY ARTICLE

3891

Max Perutz: chemist, molecular biologist, human rights activist

John Meurig Thomas

The story of Max Perutz's life and achievements is an inspiration to all chemists and scientists and to non-scientific members of the general public.



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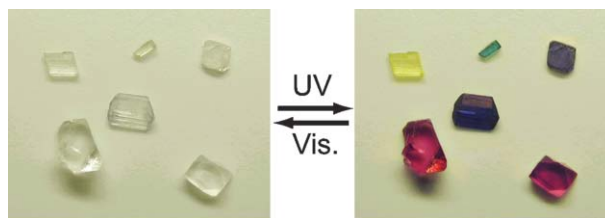
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3895

Photochromism of diarylethene single crystals: crystal structures and photochromic performance

Masakazu Morimoto and Masahiro Irie*

Diarylethenes undergo thermally irreversible and fatigue-resistant photochromic reactions in the single-crystalline phase. Photocyclization/cycloreversion quantum yields, absorption properties, and surface morphology changes are strongly dependent on the conformations and packing structures of the molecules in the crystals.



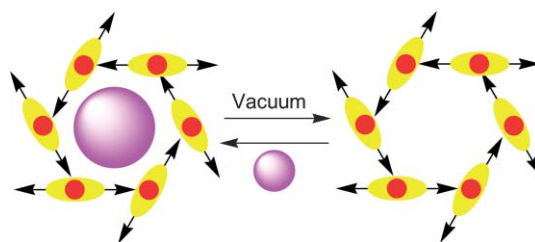
COMMUNICATIONS

3906

Reversible single-crystal-to-single-crystal guest exchange in a 3-D coordination network based on a zinc porphyrin

Emmanuel Deiters, Véronique Bulach and Mir Wais Hosseini*

A self-complementary tecton based on a Zn-porphyrin core bearing two extra pyridines self-assembles in the crystalline phase into a robust 3-D network undergoing reversible guest exchange through a single-crystal-to-single-crystal transformation.

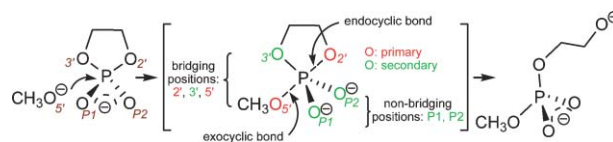


3909

Kinetic isotope effects on thio-substituted biological phosphoryl transfer reactions from density-functional theory

Yun Liu, Xabier Lopez and Darrin M. York*

Kinetic isotope effects in the in-line dianionic mechanism of ethylene phosphate methanolysis (a reverse-reaction model for RNA phosphate transesterification).

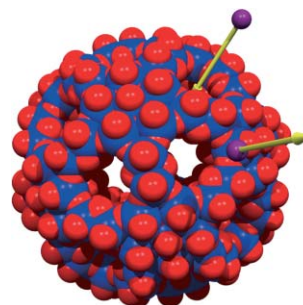


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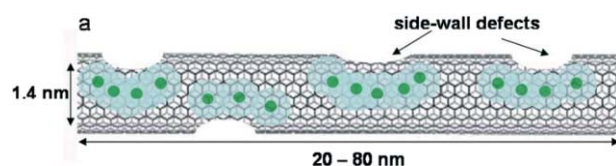
Porous inorganic capsules in action: modelling transmembrane cation-transport parameter-dependence based on water as vehicle

Erhard T. K. Haupt,* Claudia Wontorra, Dieter Rehder* and Achim Müller*

Li^+ ion trafficking between the outside and different inside sites of an artificial cell is sensitive, as shown by ^7Li NMR, to solvent influences and, in particular, the amount of water present.



3915

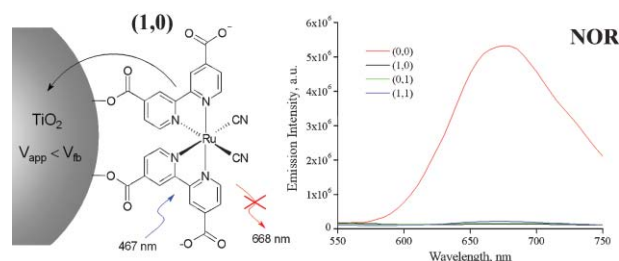


Superparamagnetic gadonanotubes are high-performance MRI contrast agents

B. Sitharaman, K. R. Kissell, K. B. Hartman, L. A. Tran, A. Baikalov, I. Rusakova, Y. Sun, H. A. Khant, S. J. Ludtke, W. Chiu, S. Laus, É. Tóth, L. Helm, A. E. Merbach and L. J. Wilson*

The nanoscale loading and confinement of aquated Gd^{3+} -ion clusters within ultra-short single-walled carbon nanotubes gives species which are linear superparamagnetic molecular magnets and high-performance MRI contrast agents.

3918

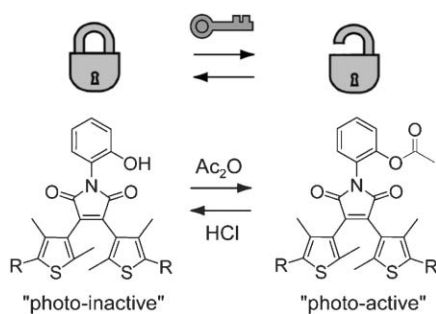


A potential and ion switched molecular photonic logic gate

Matteo Biancardo, Carlo Bignozzi,* Hugh Doyle and Gareth Redmond*

A molecular photonic logic gate is demonstrated by integrating electrical (potential) and chemical (ionic) switching functions into molecules attached at an externally addressable semiconductor substrate.

3921

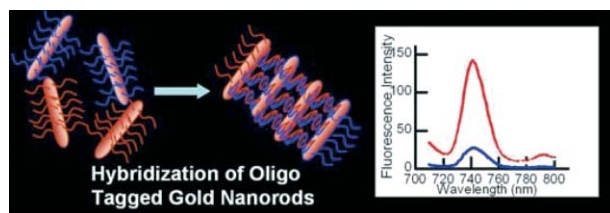


Chemical control of the photochromic reactivity of diarylethene derivatives

Masato Ohsumi, Tuyoshi Fukaminato and Masahiro Irie*

Photochemically gated performances of photochromic diarylethene derivatives were examined. The photochromic reaction of diarylethene derivatives can be controlled by the addition of acid.

3924



Fluorescence properties of gold nanorods and their application for DNA biosensing

Chen-Zhong Li, Keith B. Male, Sabahudin Hrapovic and John H. T. Luong*

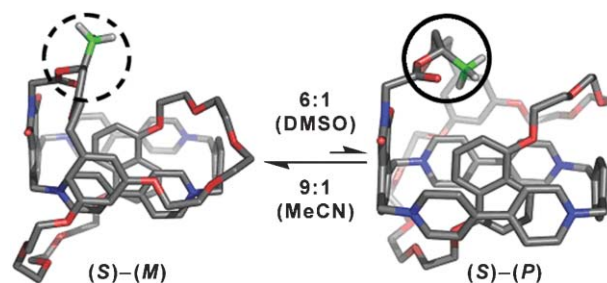
Monitoring the enhanced fluorescence of gold nanorods as they elongate is used as a novel strategy for DNA hybridization studies.

3927

Conformational diastereoisomerism in a chiral pretzelane

Yi Liu, Scott A. Vignon, Xiyun Zhang, K. N. Houk and J. Fraser Stoddart*

The introduction of a stereogenic center by a stereospecific synthesis into an optically active, donor–acceptor pretzelane that exhibits helicity as well as fixed chirality leads to a marked preference for the (*S*)-(*M*)- over the (*S*)-(*P*)-isomer that can be understood from computational models.

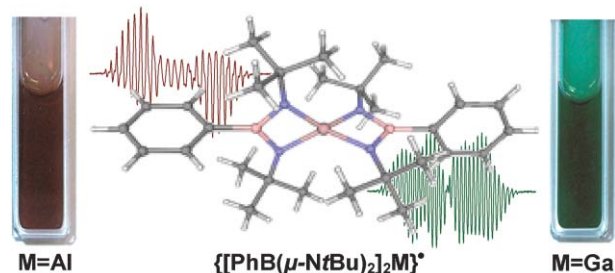


3930

Stable spirocyclic neutral radicals: aluminium and gallium boraamidinates

Tristram Chivers,* Dana J. Eisler, Chantall Fedorchuk, Gabriele Schatte, Heikki M. Tuononen and René T. Boéré

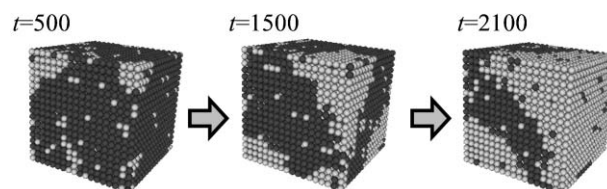
Stable dark red ($M = \text{Al}$) or dark green ($M = \text{Ga}$) neutral radicals $\{[\text{PhB}(\mu\text{-}N^t\text{Bu})_2]_2M\}^{\cdot}$ are obtained by oxidation of the corresponding anions with iodine and characterized by X-ray crystallography in the solid state and EPR spectra in solution.



3933

Thermal hysteresis loop of the spin-state in nanoparticles of transition metal complexes: Monte Carlo simulations on an Ising-like model

Tohru Kawamoto* and Shuji Abe

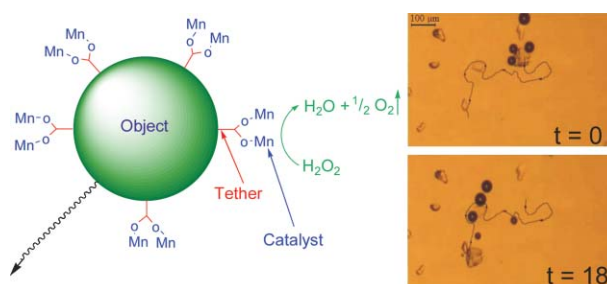


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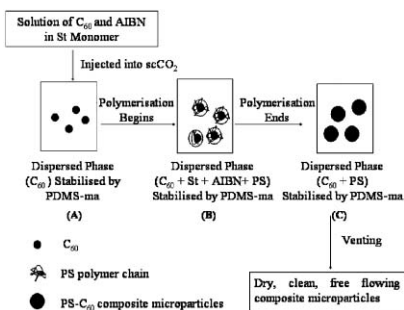
Catalytic molecular motors: fuelling autonomous movement by a surface bound synthetic manganese catalase

Javier Vicario, Rienk Eelkema, Wesley R. Browne, Auke Meetsma, René M. La Crois and Ben L. Feringa*

A synthetic catalase mimic, covalently attached to a SiO_2 microparticle, induces the catalytic disproportionation of hydrogen peroxide to oxygen and water, resulting in both translational and rotational motion of the microparticle.



3939

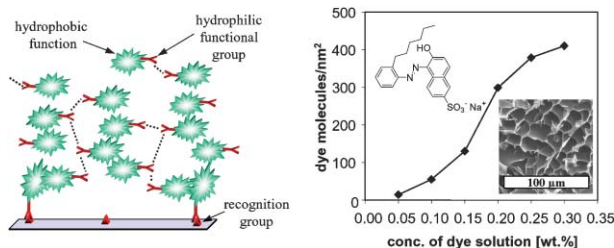


One-step seed dispersion polymerisation in supercritical carbon dioxide

Wenxin Wang, Steven M. Howdle* and Deyue Yan

We report for the first time a single step seed dispersion polymerisation. Through this simple strategy we demonstrate how scCO_2 can be used simultaneously as both an antisolvent and a polymerisation medium to create polymer- C_{60} composite microparticles.

3942

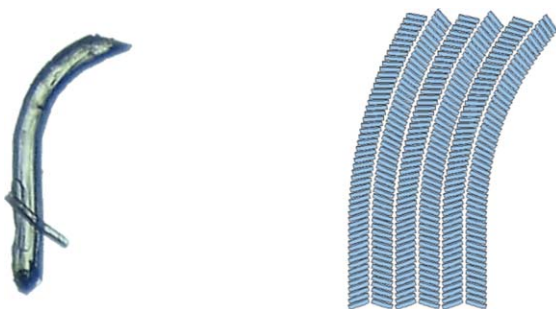


Surface-induced hydrogelation

Arno M. Bieser and Joerg C. Tiller*

A novel anionic hydrogelling azo dye is greatly concentrated on a complementary cationic surface upon forming a hydrogel even at concentrations 50 times below its minimal gelation concentration in water.

3945

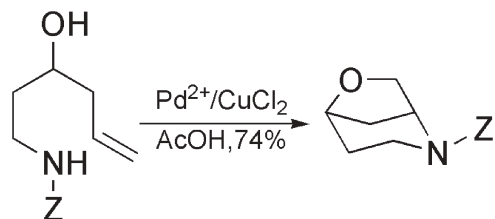


Structural basis for bending of organic crystals

C. Malla Reddy, Ravi C. Gundakaram, Srinivas Basavoju, Michael T. Kirchner, K. Anantha Padmanabhan* and Gautam R. Desiraju*

Organic molecular crystals are soft. Why some of them bend rather than break is explained.

3948



Novel Pd(II)-catalysed *N,O*-bicyclisation as an efficient route to the 6-oxa-2-azabicyclo[3.2.1]octane skeleton

Peter Szolcsányi* and Tibor Gracza

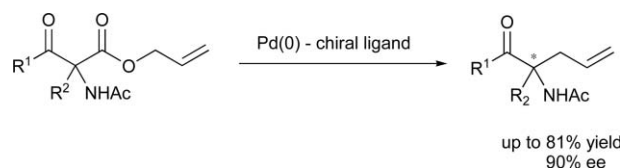
Protected aminoalkenitol undergoes a novel Pd(II)/ CuCl_2 -catalysed bicyclisation to furnish the corresponding 6-oxa-2-azabicyclo[3.2.1]octane in good yield.

3951

Asymmetric Carroll rearrangement of allyl α -acetamido- β -ketocarboxylates catalysed by a chiral palladium complex

Ryoichi Kuwano, Naoki Ishida and Masahiro Murakami*

Asymmetric decarboxylative rearrangement of allyl α -acetamido- β -ketocarboxylates is catalysed by a chiral palladium complex, creating an N-substituted quaternary carbon center with up to 90% ee. Acetamido group is crucial for chiral induction.

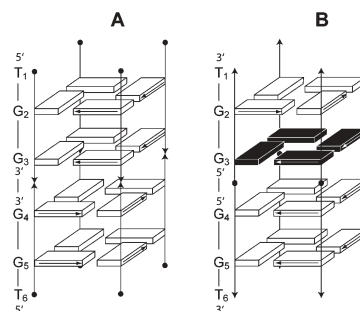


3953

A new class of DNA quadruplexes formed by oligodeoxyribonucleotides containing a 3'-3' or 5'-5' inversion of polarity site

Veronica Esposito, Antonella Virgilio, Antonio Randazzo, Aldo Galeone and Luciano Mayol*

Unprecedented DNA quadruplex structures containing a 3'-3' (A) or 5'-5' (B) inversion of polarity site in the G-tract are presented. Interestingly, in (B), one of the two central tetrads is characterized by Gs in the *syn* glycosidic conformation.

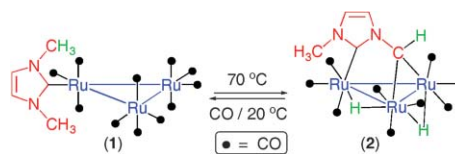


3956

Easy activation of two C–H bonds of an *N*-heterocyclic carbene *N*-methyl group

Javier A. Cabeza,* Ignacio del Río, Daniel Miguel and M. Gabriela Sánchez-Vega

The syntheses of the first examples of trinuclear cluster complexes containing NHC ligands are described. The observation of an easy activation of two C–H bonds of a methyl group attached to a nitrogen atom of an NHC ligand is also reported. Such a reaction represents an unprecedented degradation of an NHC ligand containing *N*-bonded methyl groups.

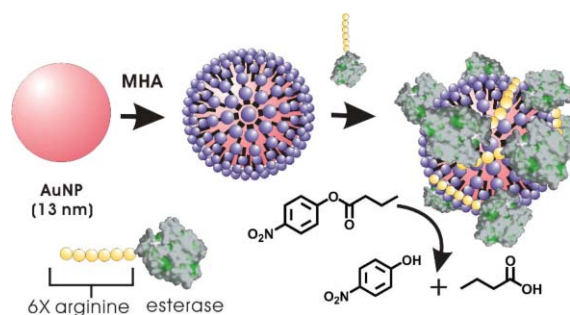


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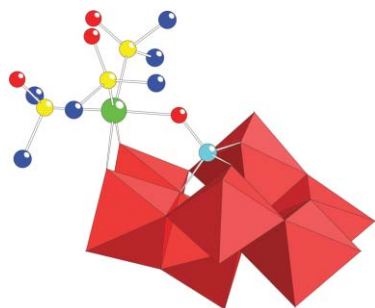
Immobilization of hexa-arginine tagged esterase onto carboxylated gold nanoparticles

Tai Hwan Ha, Jin Young Jeong and Bong Hyun Chung*

Hexa-arginine tagged esterase was efficiently immobilized onto carboxylated gold nanoparticles (AuNP-COOH) and its enzyme activity was investigated by monitoring the absorption spectrum of an enzyme substrate, *p*-nitrophenol butyrate (pNPB).



3962

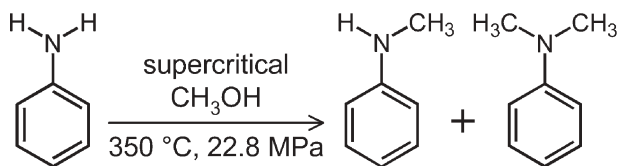


The Ru(II)-supported heptatungstates [HXW₇O₂₈Ru(dmsol)₃]⁶⁻ (X = P, As)

Li-Hua Bi, Michael H. Dickman, Ulrich Kortz* and Ina Dix

The ruthenium(II)-supported heteropolyanions [HXW₇O₂₈Ru(dmsol)₃]⁶⁻ (X = P, 1; As, 2) are composed of a Ru(dmsol)₃ group attached to an unprecedented heptatungstate fragment *via* two Ru–O–W bonds and one Ru–O–X bond, which represents a fundamentally novel mode of Ru-coordination to a polyoxoanion framework.

3965



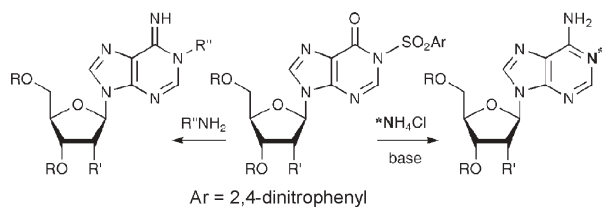
without catalyst	(2 h)	78 % yield	3 %
with 0.5 mM NaOH	(1 h)	80 %	1 %

Noncatalytic mono-*N*-methylation of aniline in supercritical methanol: the kinetics and acid/base effect

Yoshihiro Takebayashi,* Yoshinori Morita, Hideki Sakai, Masahiko Abe, Satoshi Yoda, Takeshi Furuya, Tsutomu Sugeta and Katsuto Otake

Aniline is easily *N*-methylated in supercritical methanol without catalyst to give mono-*N*-methylaniline with high selectivity, and the reaction rate is increased by a small amount of base, indicating a difference in the reaction mechanism from the ordinary acid-catalyzed one.

3968

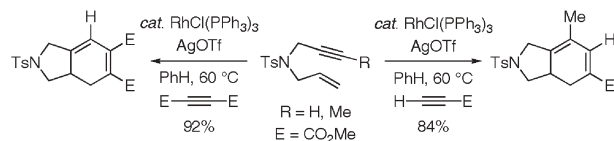


A novel nucleophilic approach to 1-alkyladenosines. A two-step synthesis of [1-¹⁵N]adenosine from inosine

Montserrat Terrazas, Xavier Ariza,* Jaume Farràs and Jaume Vilarrasa*

A novel ANRORC mechanism in the reaction of 1-(2,4-dinitrobenzenesulfonyl)inosines with amines has allowed the preparation of 1-alkyladenosines and [1-¹⁵N]adenosines in a straightforward way from inosines.

3971



Intermolecular rhodium-catalyzed [2 + 2 + 2] carbocyclization reactions of 1,6-enynes with symmetrical and unsymmetrical alkynes

P. Andrew Evans,* James R. Sawyer, Kwong Wah Lai and John C. Huffman

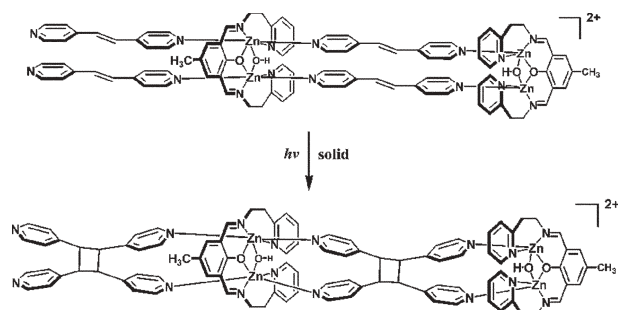
The *crossed* intermolecular rhodium-catalyzed [2 + 2 + 2] carbocyclization of carbon and heteroatom tethered 1,6-enynes can be accomplished with symmetrical and unsymmetrical alkynes, to afford the corresponding bicyclohexadienes in an efficient and highly selective manner.

3974

Directed assembly and reactivity of olefins within a one-dimensional ladder-like coordination polymer based on a dinuclear Zn(II) platform

Giannis S. Papaefstathiou, Ivan G. Georgiev, Tomislav Frišćić and Leonard R. MacGillivray*

A [2 + 2] photodimerisation is directed within a 1D ladder-like coordination polymer in the solid state.

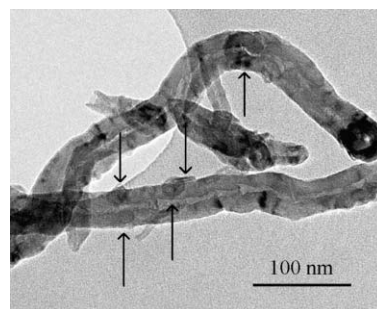


3977

Supported hydrotalcites as highly active solid base catalysts

Ferry Winter, A. Jos van Dillen and Krijn P. de Jong*

Mg–Al hydrotalcite platelets with a lateral size of 20 nm were deposited on carbon nanofibers and the resulting supported catalyst exhibited a specific activity in the condensation of acetone four times that of unsupported hydrotalcites due to the higher number of active edge sites.

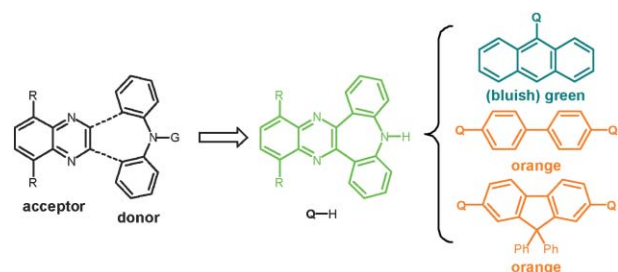


3980

Doubly *ortho*-linked quinoxaline/triarylamine hybrid as a bifunctional, dipolar electrochromic template for optoelectronic applications

Chien-Tien Chen,* Jin-Sheng Lin, Murthy V. R. K. Moturu, Yi-Wen Lin, Wei Yi, Yu-Tai Tao* and Chin-Hsiung Chien

The titled hybrid (Q-H) works as a clippable dipolar optoelectronic unit. Q-spacer-Qs function as efficient orange emitters for 4,4'-biaryl-centered systems and Q-An acts as an efficient (bluish) green emitter.

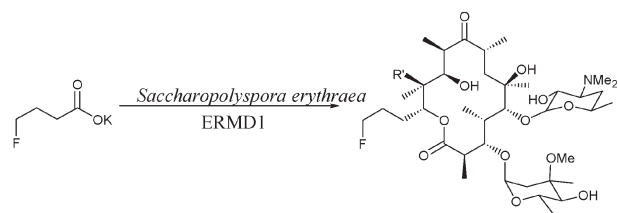


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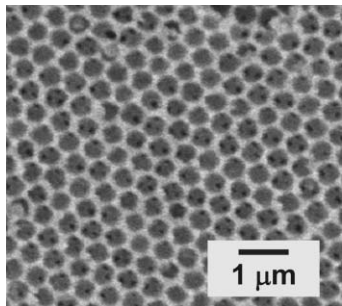
A novel fluorinated erythromycin antibiotic

Rebecca J. M. Goss* and Hui Hong

A novel fluorinated erythromycin (16-fluoroerythromycin A) has been produced by *Saccharopolyspora erythraea* ERMD1, using precursor-directed biosynthesis.



3986

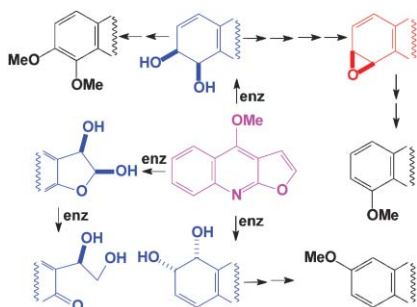


Properties of composite proton-conducting membranes prepared from three-dimensionally ordered macroporous polyimide matrix and polyelectrolyte

Hirokazu Munakata, Dai Yamamoto and Kiyoshi Kanamura*

A new proton-conducting composite membrane has been prepared by use of a three-dimensionally ordered macroporous matrix of polyimide and a proton-conducting gel polymer; the resulting composite membrane exhibited very high conductivity of $1.7 \times 10^{-1} \text{ S cm}^{-1}$ at 60°C under 90% relative humidity.

3989

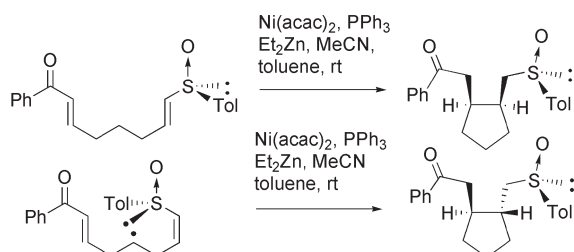


cis-Dihydrodiol, arene oxide and phenol metabolites of dictamnine: key intermediates in the biodegradation and biosynthesis of furoquinoline alkaloids

Derek R. Boyd,* Narain D. Sharma, Colin R. O'Dowd, Jonathan G. Carroll, Pui L. Loke and Christopher C. R. Allen

The first examples of *cis*-dihydrodiol bacterial metabolites from an alkaloid and their application in the chemoenzymatic synthesis of an arene oxide and other furoquinoline alkaloids are presented.

3992

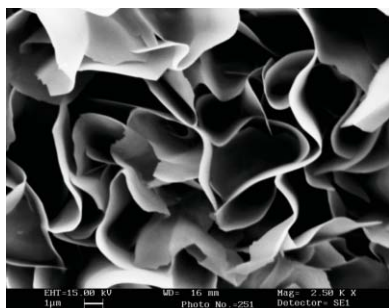


Diastereoselective Ni(0)-catalyzed carbocyclization to chiral vinylic sulfoxide

Naoyoshi Maezaki, Hiroaki Sawamoto, Hiroyuki Ishihara and Tetsuaki Tanaka*

Diastereoselective Ni(0)-catalyzed carbocyclization of enone to chiral vinylic sulfoxide has been accomplished.

3995



GaS and GaSe nanowalls and their transformation to Ga₂O₃ and GaN nanowalls

Ujjal K. Gautam, S. R. C. Vivekchand, A. Govindaraj and C. N. R. Rao*

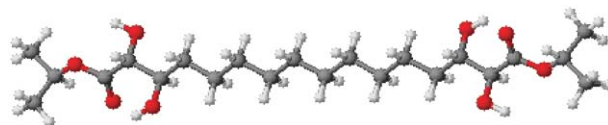
2D nanowalls of GaS and GaSe are obtained by thermal exfoliation and can be converted to Ga₂O₃ and GaN nanowalls by reaction with air and NH₃ respectively.

3998

Gelation of fluorinated liquids by non-fluorinated low-molecular-mass molecules

P. C. Griffiths,* M. Côte, R. James, Ph. G. Rogueda, I. R. Morgan and D. W. Knight

Organogelators are small molecules that spontaneously self-assemble to create a three-dimensional network capable of entrapping the solvent and creating gels.

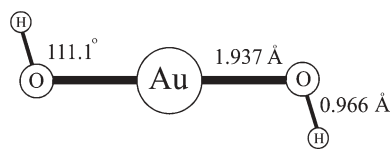


4001

Infrared spectrum and structure of the gold dihydroxide molecule

Xuefeng Wang and Lester Andrews*

Reactions of laser-ablated gold atoms with H_2O_2 and $\text{H}_2 + \text{O}_2$ mixtures give four new infrared absorptions, which match the four most intense vibrational frequencies calculated for $\text{Au}(\text{OH})_2$ using density functional theory. The calculations find a C_{2h} structure and substantial covalent bonding character for the $\text{Au}(\text{OH})_2$ molecule, which is probably due to the high electron affinity of gold.

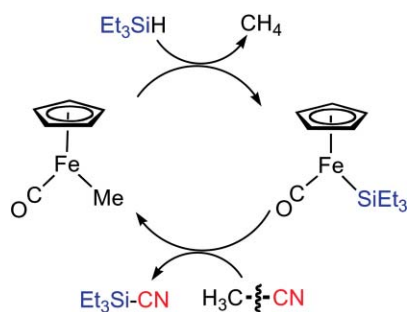


4004

Catalytic C–C bond cleavage and C–Si bond formation in the reaction of RCN with Et_3SiH promoted by an iron complex

Hiroshi Nakazawa,* Kouji Kamata and Masumi Itazaki

Catalytic C–C bond cleavage of acetonitrile and C–Si bond formation have been attained in the photoreaction of MeCN with Et_3SiH in the presence of an iron complex, $\text{Cp}(\text{CO})_2\text{FeMe}$. This catalytic system can be applied for aryl nitrile C–C bond cleavage.

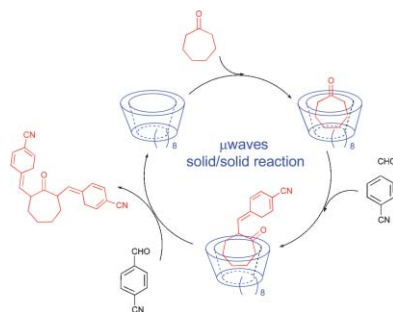


4007

New synthesis of (Z,E)-2,7-bis(4-cyanobenzylidene)cycloheptan-1-one under stereospecific constraints induced by host–guest interactions

Arnaud Grandeury, Samuel Petit, Servane Coste, Gérard Coquerel, Cécile Perrio and Géraldine Gouhier*

A selective, efficient and fast access to a precursor of the synthetic antagonist of tissue-plasminogen activator was developed using a solid/solid reaction on a supramolecular complex under MW irradiation.




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