**Cover (far left)**

A 3-D inorganic titanium phosphate (foreground) and a hexagonal mesophase with crystalline walls (background) produced, starting from titanium fluoride and fluorophosphates primary building units (pp. 2755–2765).

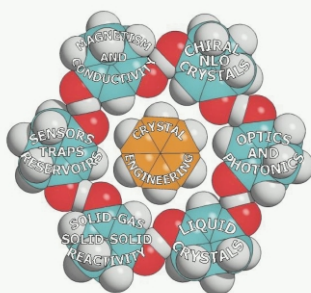
Inside cover (left)

Silica-coated pollen grains (yellow) and calcined inorganic replicas (white) against a background clover image (photo: Stephanie Summerfield) (pp. 2784–2785).

contents

FOCUS ARTICLE

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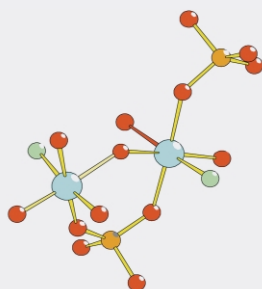
Crystal engineering, Where from? Where to?

Dario Braga

Crystal engineering is *making crystals by design*. The crystal engineer is a chemist with interests ranging from synthetic chemistry and crystallography to supramolecular solid state and materials chemistry.

FEATURE ARTICLE

2755



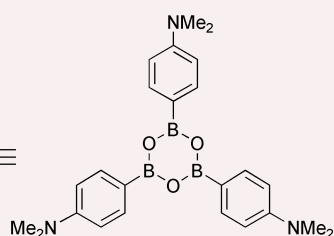
Rational design of porous titanophosphates

Christian Serre,* Francis Taulelle and Gérard Férey

A small library of titanium phosphate crystalline compounds has been set-up and investigated by diffraction and by *in-* and *ex-situ* NMR. The main trends of a rational design of porous titanium-based materials were therefore established.

COMMUNICATIONS

2766



$$\lambda_{\text{abs}} = 281 \text{ nm}$$

$$\beta_{\text{HLS}}(0) = 56 \times 10^{-30} \text{ esu}$$

Improved transparency–nonlinearity trade-off with boroxine-based octupolar molecules

Gilles Alcaraz, Lisenn Euzenat, Olivier Mongin, Claudine Katan, Isabelle Ledoux, Joseph Zyss, Mireille Blanchard-Desce* and Michel Vaultier*

A series of octupolar molecules derived from the boroxine framework were found to combine excellent transparency in the near UV-visible region and significant first-order hyperpolarisabilities (up to $\beta_{\text{HLS}}(0) = 56 \times 10^{-30}$ esu).

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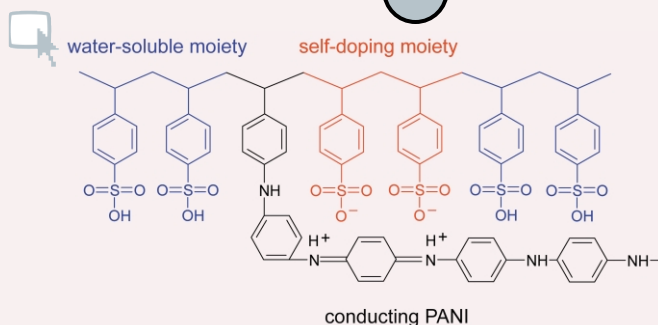
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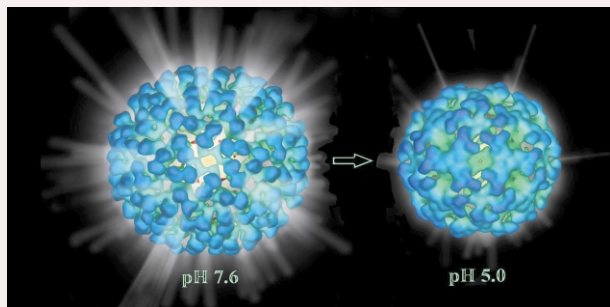


A novel water-soluble and self-doped conducting polyaniline graft copolymer

Woo Jin Bae, Keon Hyeong Kim, Yun Heum Park* and Won Ho Jo*

A novel water-soluble and self-doped conducting polyaniline graft copolymer, poly(styrenesulfonic acid-*g*-aniline) (PSSA-*g*-PANI), was first synthesized and its electrical properties were investigated.

2770

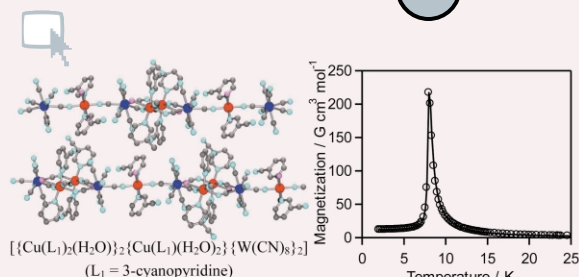


Correlation of chemical reactivity of *Nudaurelia capensis* ω virus with a pH-induced conformational change

Derek J. Taylor, Qian Wang, Brian Bothner, Padmaja Natarajan, M. G. Finn* and John E. Johnson*

An RNA virus capsid dramatically changes morphology between pH 7.6 and 5.0. The figure depicts the change by showing the hypothetical transmitted light from a source within the particle. The article compares the chemical reactivity of the two forms of the particle.

2772

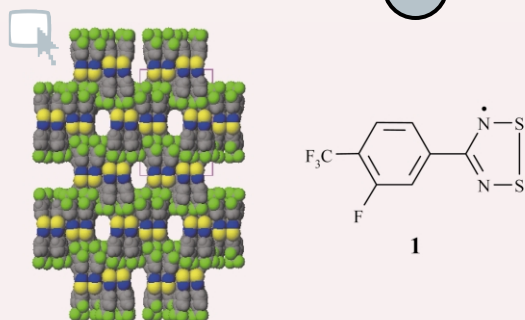


Two-dimensional metamagnet composed of cyano-bridged Cu^{II} - W^V bimetallic assembly

Shin-ichi Ohkoshi,* Yoichi Arimoto, Toshiya Hozumi, Hidetake Seino, Yasushi Mizobe and Kazuhito Hashimoto*

New types of two-dimensional cyanide-bridged copper(II) octacyanotungstates(V), $[\{Cu(L_1)_2(H_2O)\}_2\{Cu(L_1)(H_2O)_2\}\{W(CN)_8\}_2]$ ($L_1 = 3$ -cyanopyridine) and $[\{Cu(L_2)_2\}_2\{Cu(L_2)(H_2O)_2\}\{W(CN)_8\}_2]\cdot 6H_2O$ ($L_2 = 4$ -cyanopyridine), were prepared and these compounds exhibited metamagnetic behavior.

2774

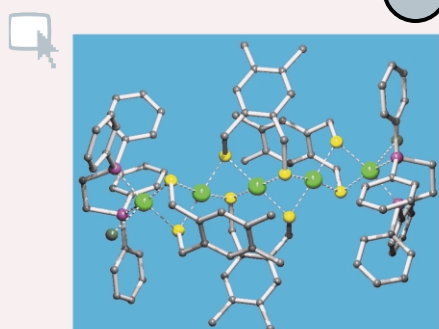


Small molecule fixation by a dithiadiazolyl radical: X-ray crystal structures of $(CF_3C_6H_3FCN_2SSN_2)_2$ and $(CF_3C_6H_3FCN_2SSN_2)_2\cdot G$ ($G = N_2, Ar, CO_2$ and SO_2)

Caroline S. Clarke, Delia A. Haynes, Jeremy M. Rawson* and Andrew D. Bond

The dithiadiazolyl radical, **1**, forms channel-type inclusion complexes when sublimed under N_2 , Ar, CO_2 or SO_2 .

2776

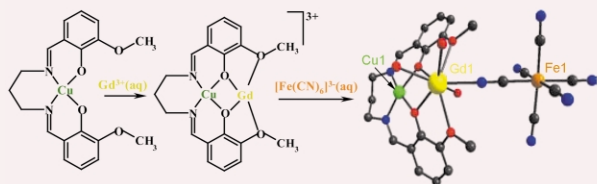


Formation of nickel-thiolate aggregates *via* reaction with CH_2Cl_2

Qiang Wang, Andrew C. Marr, Alexander J. Blake, Claire Wilson and Martin Schröder*

Reaction of the mononuclear nickel-thiolate complex $[Ni(L^1)(dppe)]$ with CH_2Cl_2 affords the novel pentanuclear complex $[Ni_5Cl_2(L^1)_4(dppe)_2]$, while $[Ni(L^1)(dppe)]$ reacts with CH_2Cl_2 to give the binuclear species $[Ni_2Cl_2(L^2)(dppe)_2]$ in which two L^1 units are linked by a methylene group derived from CH_2Cl_2 .

2778

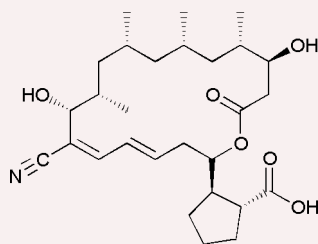


A rational synthetic route leading to 3d–3d′–4f heterospin systems: self-assembly processes involving heterobinuclear 3d–4f complexes and hexacyanometallates

Ruxandra Gheorghe, Marius Andruh,* Jean-Pierre Costes* and Bruno Donnadieu

A novel 1-D coordination polymer containing three different paramagnetic metal ions, Cu(II), Gd(III) and Fe(III), with a unique ladder topology of the spin carriers, has been obtained by using binuclear [CuGd]³⁺ cations as nodes, and [Fe(CN)₆]³⁻ ions as linkers.

2780

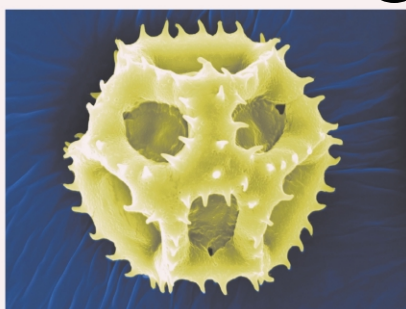


Evidence from engineered gene fusions for the repeated use of a module in a modular polyketide synthase

Carlos Olano, Barrie Wilkinson, Steven J. Moss, Alfredo F. Braña, Carmen Méndez, Peter F. Leadlay and José A. Salas*

Functional evidence is provided for the repeated reuse of a single module of the modular type-I polyketide synthase involved in the biosynthesis of the potent anti-angiogenic macrolide borrelidin.

2784

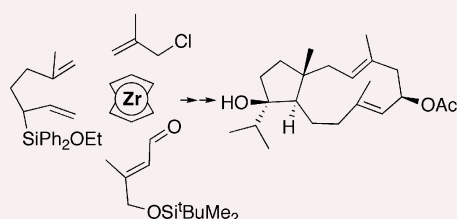


Morphosynthesis of complex inorganic forms using pollen grain templates

Simon R. Hall, Helen Bolger and Stephen Mann*

Morphosynthesis of silica, calcium carbonate and calcium phosphate with complex form is achieved by the use of pollen grains as a bio-template. The mineral particles exhibit the intricate morphology of the original pollen grains.

2786

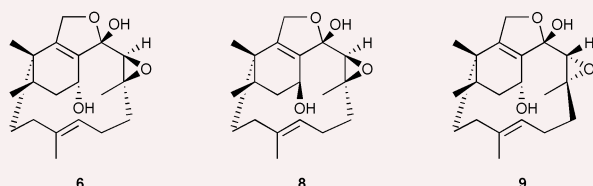


Total synthesis of (±)-acetoxydontoschismenol using zirconium chemistry

Ian R. Baldwin and Richard J. Whitby*

Tandem zirconocene induced co-cyclisation, allyl carbenoid insertion, aldehyde addition and iodolysis is used to assemble most of the carbons needed in the total synthesis of the dollabelane diterpene (±)-acetoxydontoschismenol.

2788



Synthetic studies towards congeners of phomactin A. Re-examination of the structure of Sch 49028

John W. C. Cheing, William P. D. Goldring and Gerald Pattenden*

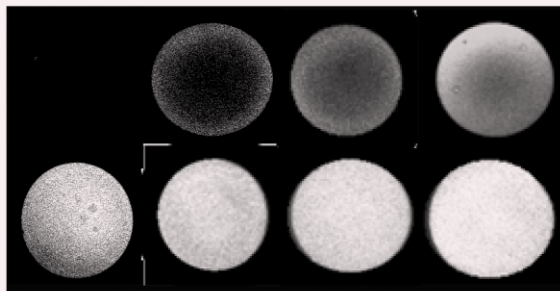
Synthesis of the epoxy cyclic hemiacetal structures **8** and **9** supports the suggestion that Sch 49028, isolated from the marine fungus *Phoma* sp., has been incorrectly assigned the structure **6** and is phomactin A.

2790

Using two photon microscopy to quantify enzymatic reaction rates on polymer beads

Annie Y. Bosma, Rein V. Ulijn, Gail McConnell, John Girkin, Peter J. Halling and Sabine L. Flitsch*

Two photon fluorescence microscopy can be used to spatially quantify enzyme reactions in solid phase biocatalysis.

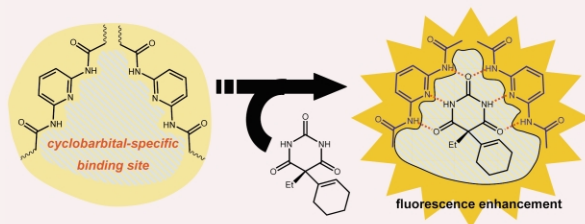


2792

Multiple hydrogen bonding-based fluorescent imprinted polymers for cyclobarbital prepared with 2,6-bis(acrylamido)pyridine

Hiroyuki Kubo, Hiroyuki Nariai and Toshifumi Takeuchi*

A cyclobarbital-imprinted polymer was prepared using a fluorescent functional monomer, 2,6-bis(acrylamido)pyridine, and the polymer showed not only selective binding of cyclobarbital but also enhancement of fluorescence intensity, suggesting that the polymer could be utilized as a selective fluorescence probe.

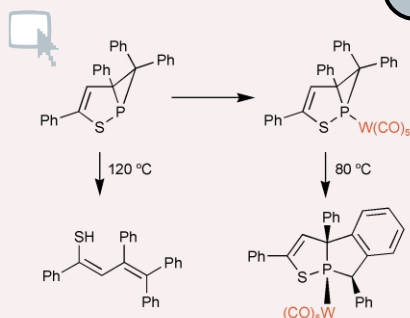


2794

Different thermal reactivity of a 1,2-thiaphospholo[a]phosphirane in free and metal carbonyl complexed form

Tamaki Jikyo and Gerhard Maas*

The thermal reactivity of 3,5,6-tetraphenyl-1-phospha-2-thiabicyclo[3.1.0]hex-3-ene depends on whether or not it is complexed with a tungsten or iron carbonyl fragment.

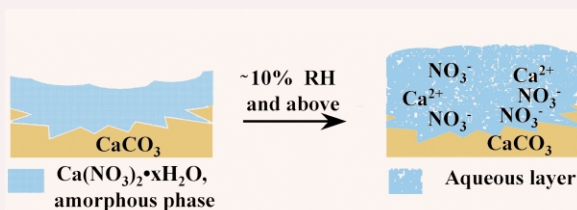


2796

Phase transitions in calcium nitrate thin films

Hind A. Al-Abadleh, B. J. Krueger, J. L. Ross and V. H. Grassian*

In calcium nitrate thin films, deliquescence occurs at lower relative humidity than expected from bulk phase thermodynamics and lower than the recommended humidity for the preservation of artifacts and antiques.

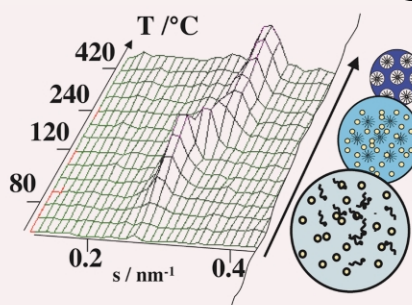


2798

First *in-situ* SAXS studies of the mesostructure of spherical silica and titania particles during spray-drying process

C. Boissiere, D. Grosso, H. Amenitsch, A. Gibaud, A. Coupé, N. Baccile and C. Sanchez*

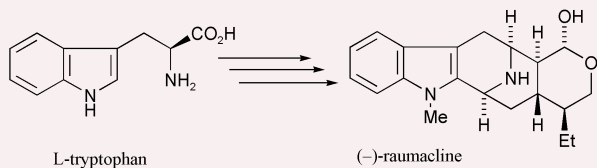
Mesoorganisation of SiO₂ and TiO₂ particles prepared by spray drying have been analysed for the first time through *in-situ* SAXS.



2800

Total synthesis of (–)-raumacline

Patrick D. Bailey, Paul D. Clingan, Timothy J. Mills, Richard A. Price and Robin G. Pritchard

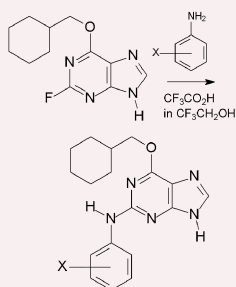


The total synthesis of (–)-raumacline from L-tryptophan is achieved, featuring a *cis*-specific Pictet–Spengler reaction, a stereoselective Dieckmann cyclization, and an epimerization step that allows complete stereocontrol of five chiral centres.

2802

Facilitation of addition–elimination reactions in pyrimidines and purines using trifluoroacetic acid in trifluoroethanol

Hayley J. Whitfield, Roger J. Griffin, Ian R. Hardcastle, Andrew Henderson, Jerome Meneyrol, Veronique Mesguiche, Kerry L. Sayle and Bernard T. Golding*

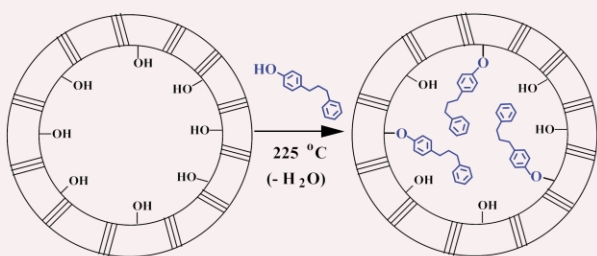


S_NAr displacement reactions of 6-cyclohexylmethoxy-2-fluoropurine, 6-amino-2-butylsulfonyl-4-cyclohexylmethoxypyrimidine and 2-amino-6-chloropurine with substituted anilines (*e.g.* the weakly nucleophilic 4-aminobenzenesulfonamide) are dramatically accelerated in the presence of trifluoroacetic acid and occur especially efficiently in 2,2,2-trifluoroethanol solvent.

2804

Pore size effects in the pyrolysis of 1,3-diphenylpropane confined in mesoporous silicas

Michelle K. Kidder, Phillip F. Britt, Zongtao Zhang, Sheng Dai and A. C. Buchanan, III*

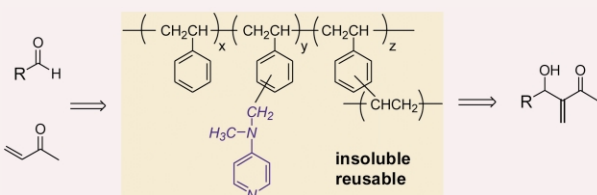


A new method for derivatizing the silanols of mesoporous silicas, SBA-15 and MCM-41, with a substituted phenol is described, and pore confinement and surface curvature are shown to impact the reaction rate and product selectivity for the pyrolysis of surface-immobilized 1,3-diphenylpropane.

2806

Heterogeneous Baylis–Hillman using a polystyrene-bound 4-(*N*-benzyl-*N*-methylamino)pyridine as reusable catalyst

Avelino Corma,* Hermenegildo García* and Antonio Leyva

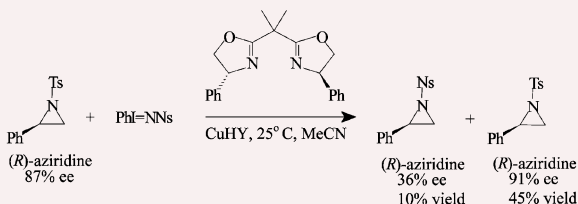


An insoluble Merrifield type resin having 4-aminopyridine units is a suitable and reusable heterogeneous catalyst for the Baylis–Hillman coupling of aromatic aldehydes and α,β -unsaturated ketones.

2808

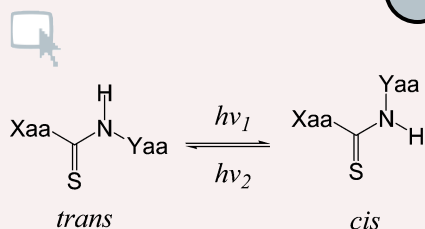
Observation of the enhancement in enantioselectivity with conversion for the aziridination of styrene using copper bis(oxazoline) complexes

John Gullick, Sophia Taylor, Darragh Ryan, Paul McMorn, Mike Coogan, Donald Bethell, Philip C. Bulman Page, Frederick E. Hancock, Frank King and Graham J. Hutchings*



During the aziridination of styrene using copper bis(oxazoline) complexes, the ee increases with conversion due to further reactions of the aziridine with the nitrene donor and sulfonamide by-product.

2810

Concentration of *cis* conformers:

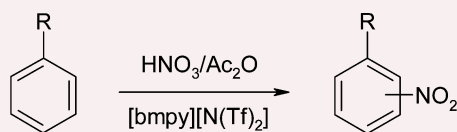
Steady state: <1%; Photostationary state: 20%

Direct photomodulation of peptide backbone conformations

Jianzhang Zhao,* Dirk Wildemann, Mario Jakob, Carolyn Vargas and Cordelia Schiene-Fischer*

A new method for photomodulation of the backbone conformation of peptides by using the photoresponsive unit of secondary thiopeptide bonds is described. With this method, a peptide bond at any specific site within the peptide chain can be photoswitched to be in either *cis* or *trans* conformations.

2812



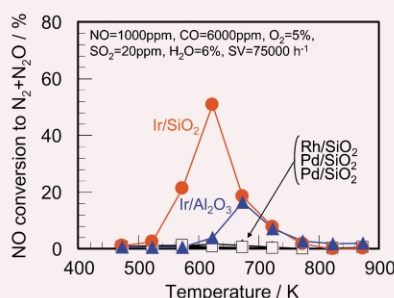
R = Me, OMe, Br, Cl

Aromatic nitrations in ionic liquids: the importance of cation choice

N. Llewellyn Lancaster* and Verónica Llopis-Mestre

The reactivity of the $\text{HNO}_3\text{-Ac}_2\text{O}$ system is enhanced in the ionic liquid [bmpy][N(Tf)₂], such that even deactivated aromatics can be nitrated. The regioselectivities are comparable to those achieved in dichloromethane.

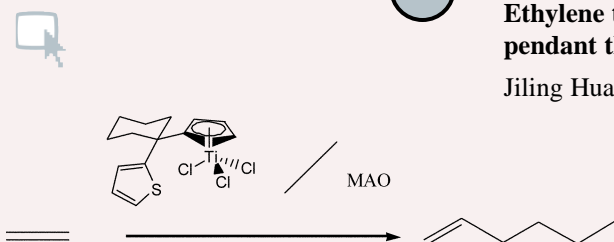
2814

**Ir/SiO₂ as a highly active catalyst for the selective reduction of NO with CO in the presence of O₂ and SO₂**

Masaaki Haneda,* Tomohiro Yoshinari, Kazuhito Sato, Yoshiaki Kintaichi and Hideaki Hamada

Coexisting SO₂ drastically promotes the selective reduction of NO with CO in the presence of O₂ over Ir/SiO₂.

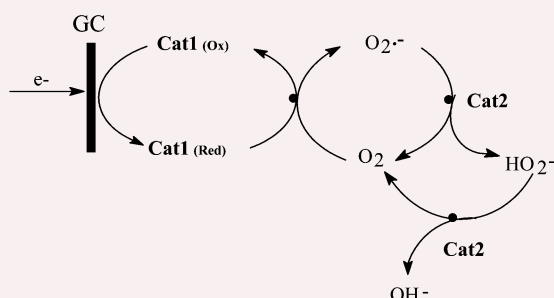
2816

**Ethylene trimerization with a half-sandwich titanium complex bearing a pendant thienyl group**

Jiling Huang,* Tianzhi Wu and Yanlong Qian

A half-sandwich titanium complex with a pendant thienyl group, activated by methylalumoxane (MAO), can trimerize ethylene to 1-hexene with considerable activity and high selectivity.

2818

**A novel electrochemical strategy for developing alkaline air electrodes by a combined use of dual functional catalysts**

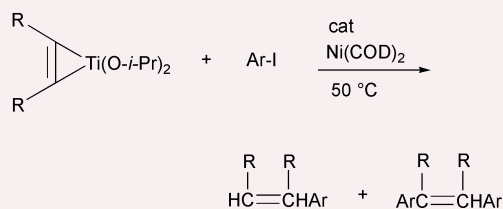
Lanqun Mao, Kazuki Arihara, Tadashi Sotomura and Takeo Ohsaka*

A novel electrochemical strategy for the development of alkaline air electrodes is proposed based on a combined use of dual catalysts for redox-mediating O₂ reduction and disproportionating superoxide and peroxide.

2820

Cross-coupling reaction of thermally stable titanium(II)-alkyne complexes with aryl halides catalysed by a nickel complex

Yasushi Obora, Hiroyuki Moriya, Makoto Tokunaga and Yasushi Tsuji*

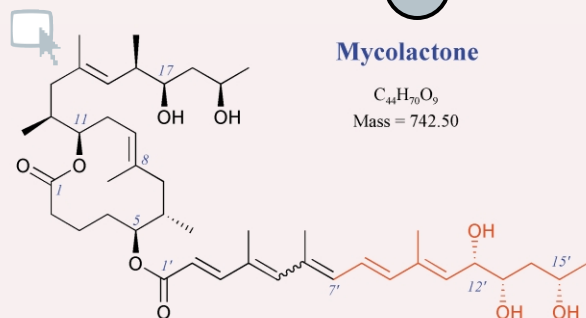


The first cross-coupling reaction of thermally stable titanium(II)-alkyne complexes with aryl iodides in the presence of a catalytic amount of Ni(cod)₂.

2822

Mycolactone

C₄₄H₇₀O₉
Mass = 742.50

**Identification using LC-MSⁿ of co-metabolites in the biosynthesis of the polyketide toxin mycolactone by a clinical isolate of *Mycobacterium ulcerans***

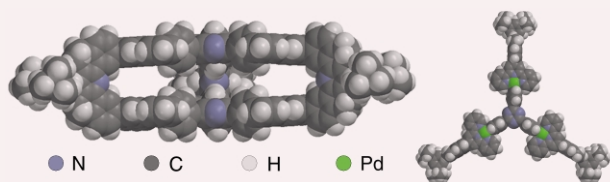
Hui Hong, Paul J. Gates, James Staunton, Tim Stinear, Stewart T. Cole, Peter F. Leadlay and Jonathan B. Spencer*

LC-MSⁿ analyses of mycolactone and its co-metabolites from a cell extract show that the structural variation is restricted to C7'–C15' of the molecule (shown in red). The implications for the biosynthesis of mycolactone are considered.

2824

Molecular recognition. Self-assembly of molecular trigonal prisms and their host–guest adducts

James D. Crowley, Andrew J. Goshe and Brice Bosnich*

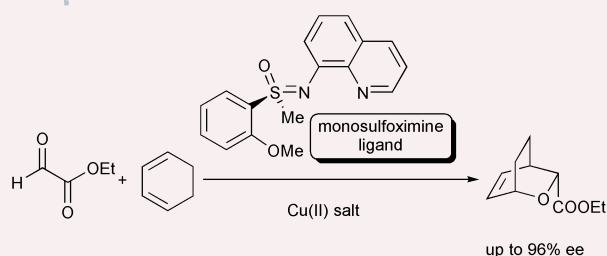


Two supramolecular trigonal prisms, each bearing three molecular clefts are shown to form 1:6 and 1:7 host–guest complexes with 9-methylanthracene and one of the prisms forms a 1:2 host–guest complex with a tritopic tri-anthracene guest that registers with the recognition sites of the host.

2826

A new class of C₁-symmetric monosulfoximine ligands for enantioselective hetero Diels–Alder reactions

Carsten Bolm,* Marinella Verrucci, Oliver Simic, Pier Giorgio Cozzi, Gerhard Raabe and Hiroaki Okamura

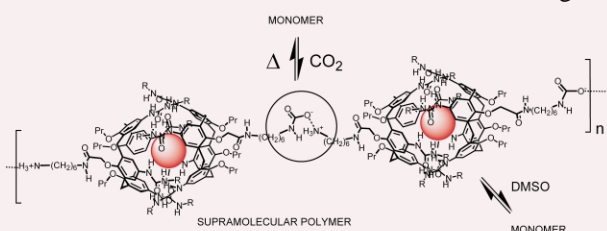


The first example of a copper-catalyzed hetero Diels–Alder reaction in the presence of a C₁-symmetric monosulfoximine ligand is reported. Cycloadducts with up to 96% ee were obtained.

2828

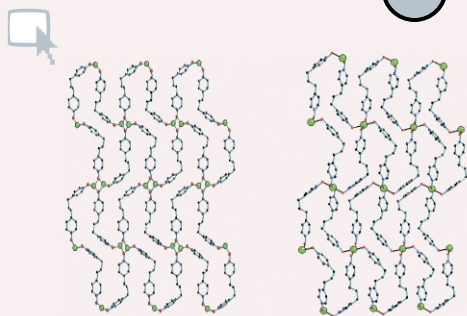
Applying reversible chemistry of CO₂ to supramolecular polymers

Heng Xu, Erin M. Hampe and Dmitry M. Rudkevich*



Novel supramolecular, reversibly formed polymers have been constructed through chemical fixation of CO₂ and hydrogen bonding. In addition to reversible carbamate bonds, these polymers also possess multiple self-assembling capsules.

2830

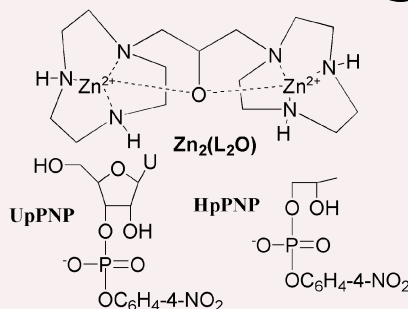


Controlled crystallization of mixed-ligand complexes of 1,3-bis(4-pyridyl)propane-*N,N'*-dioxide with metal(II) thiocyanates: isostructurality in coordination networks bearing different mono- and di-nuclear nodes

Lin-Ping Zhang, Wen-Jie Lu and Thomas C. W. Mak*

1,3-Bis(4-pyridyl)propane-*N,N'*-dioxide (L) acts in the novel μ_3 -*O,O,O'* and common μ -*O,O'* modes in $[\text{CoL}(\text{H}_2\text{O})(\text{SCN})_2]_\infty$ and $[\text{CoL}_2(\text{SCN})_2]_\infty$, respectively, to generate coordination networks having the same (4, 4) topology but different di- and mono-nuclear nodes.

2832

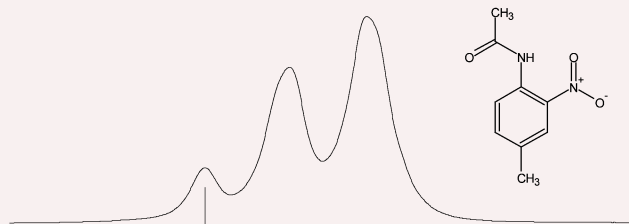


Substrate specificity for catalysis of phosphodiester cleavage by a dinuclear Zn(II) complex

Meng-Yin Yang, John P. Richard* and Janet R. Morrow*

The 2.4 kcal mol⁻¹ greater stabilization of the transition state for cleavage of the minimal substrate **HpPNP** compared to the nucleoside substrate **UpPNP** by **Zn₂(L₂O)** provides evidence that access to the core of **Zn₂(L₂O)** is blocked for the bulkier nucleoside substrate.

2834

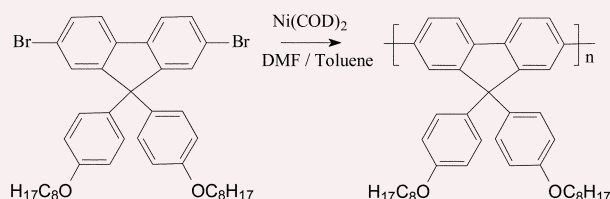


Refinement of hydrogen atomic position in a hydrogen bond using a combination of solid-state NMR and computation

Robin K. Harris,* Phuong Y. Ghi, Robert B. Hammond, Cai-Yun Ma and Kevin J. Roberts

The hydrogen-bonded protons in solid methyl nitroacetanilide and L-histidine hydrochloride have been located by high-speed magic-angle spinning ¹H NMR and density functional theory shielding computations.

2836

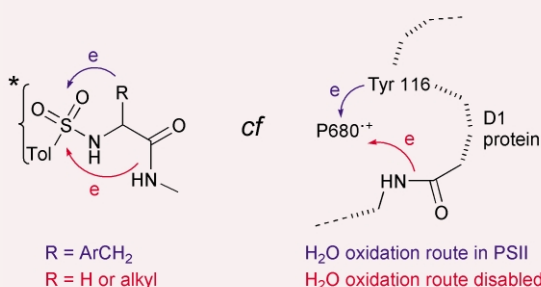


Alkoxyphenyl-substituted polyfluorene: a stable blue-light-emitting polymer with good solution processability

Ji-Hoon Lee* and Do-Hoon Hwang*

A new alkoxyphenyl-substituted poly(fluorene) emitted pure blue light, the characteristics of which did not change upon thermal annealing or normal operation of the EL device.

2838



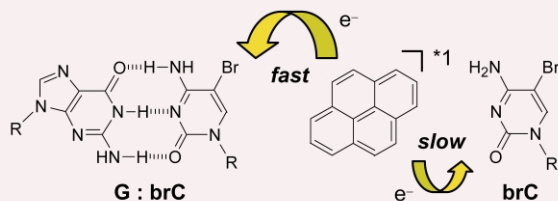
Competitive electron transfers from a tyrosyl side-chain and peptide bond in the photodegradation of *N*-tosyl α -aminomethylamides: an insight into photosynthesis and photodamage in the biological oxidation of water?

Roger R. Hill,* Sharon A. Moore and David R. Roberts

Differences in the electron transfer processes undergone by photo-excited *N*-tosyl-aminomethylamides with an aryl side-chain compared with aliphatic analogues provides a possible model for redox options in PSII.

Effects of base pairing on the one-electron reduction rate of cytosine

Kiyohiko Kawai,* Aya Yokoojji, Sachiko Tojo and Tetsuro Majima*



Experiments in dichloromethane using nucleoside derivatives demonstrate that base pairing with guanine accelerates the one-electron reduction rate of cytosine.

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