

**Cover (far left)**

The background shows the optical texture of the liquid crystalline phase of a perylene bisimide dye. Remarkably, the luminescence of this liquid-crystalline dye is tunable from green through red by changing its concentration in toluene. At low concentration of 10^{-6} M this dye is green luminescent (nonaggregated monomer), while upon aggregation at 10^{-2} M it is red luminescent (pp. 1564–1579).

Inside cover (left)

In Heck reactions catalysed by ligand-free palladium, most of the palladium is stored in the form of nanoclusters (signified by the flock of sheep). Only a small amount of palladium (the single sheep) is active as catalyst going around in the catalytic cycle (pp. 1559–1563).



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

contents

OBITUARY

xiv



Ian P. Rothwell 1955–2004

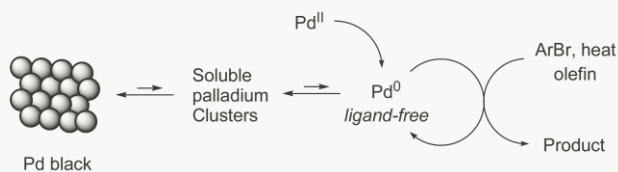
By Malcolm Chisholm

FOCUS ARTICLE

1559

Ligand-free Heck reactions using low Pd-loading

Manfred T. Reetz and Johannes G. de Vries



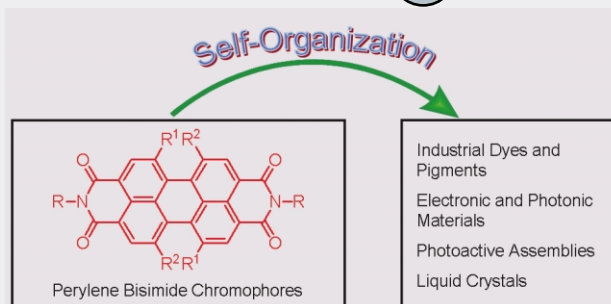
Ligand-free Heck reactions of aryl bromides are readily possible, provided that the palladium concentration is kept between 0.01–0.1 mol%; Pd-colloids serve as a reservoir for the active species.

FEATURE ARTICLE

1564

Perylene bisimide dyes as versatile building blocks for functional supramolecular architectures

Frank Würthner*



Perylene bisimide dyes and their organization into supramolecular architectures through hydrogen-bonding, metal ion coordination and π - π -stacking is discussed; further self-assembly leading to nano- and meso-scopic structures and liquid-crystalline compounds is also addressed.

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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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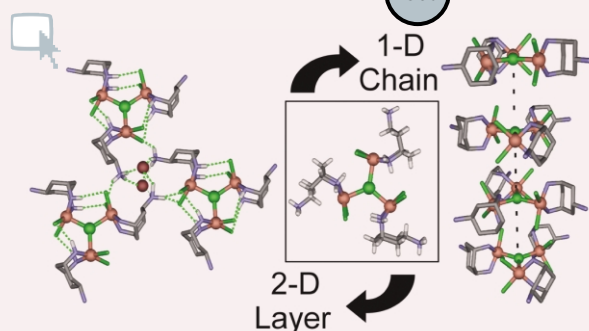
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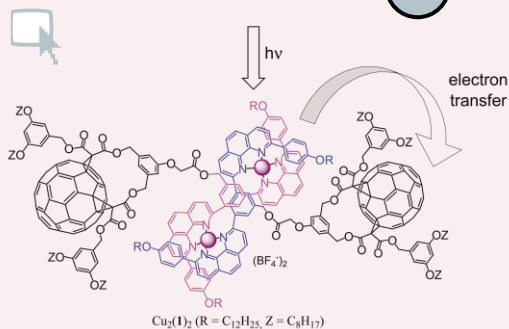


Supramolecular assembly of ligand-directed triangular $\{\text{Cu}^{\text{II}}_3\text{Cl}\}$ clusters with spin frustration and spin-chain behaviour

Georg Seeber, Paul Kögerler, Benson M. Kariuki and Leroy Cronin*

A 3-D array of triangular $\{\text{Cu}^{\text{II}}_3\text{Cl}\}$ clusters, the first example of a chloro ligand in a trigonal planar coordination mode, are assembled by the combination of coordinative and hydrogen-bonded interactions, and these combine to yield isolated 1-D chains that exhibit a combination of spin frustration and spin-chain behaviour.

1582

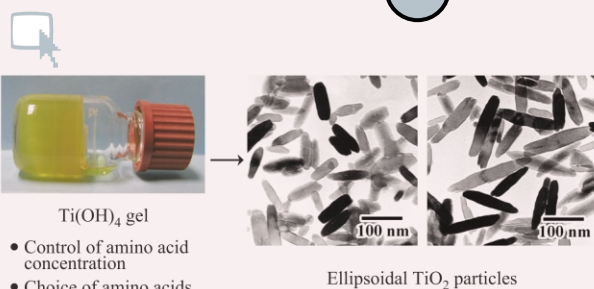


Fullerohelicates: a new class of fullerene-containing supermolecules

François Cardinali, Hind Mamlouk, Yannick Rio, Nicola Armaroli* and Jean-François Nierengarten*

A multicomponent array made of a bis-copper(I) helicate core and two peripheral fullerene subunits has been prepared and electron transfer from the photoexcited Cu(I)-complexed unit to C_{60} occurs.

1584

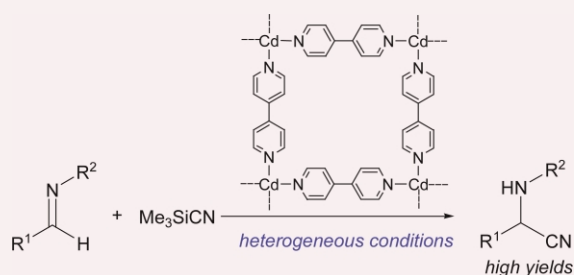


Shape control of anatase TiO_2 nanoparticles by amino acids in a gel-sol system

Kiyoshi Kanie and Tadao Sugimoto*

Ellipsoidal anatase TiO_2 nanoparticles of different aspect ratios were obtained by the gel-sol method in the presence of amino acids. The resulting particles were basically single crystals, but highly rough surfaces or partly polycrystalline structures were observed with a high concentration of glutamic or aspartic acid.

1586

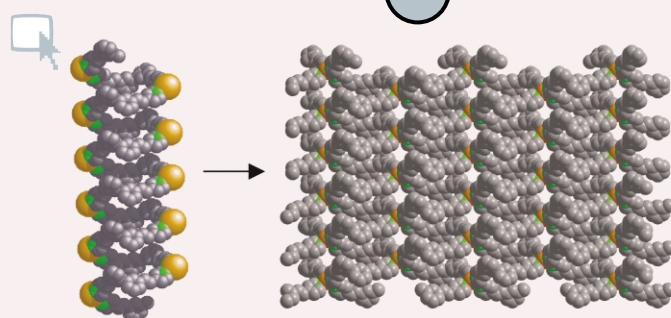


Heterogeneous catalysis of a coordination network: cyanosilylation of imines catalyzed by a Cd(II)-(4,4'-bipyridine) square grid complex

Osamu Ohmori and Makoto Fujita*

A coordination network prepared from $\text{Cd}(\text{NO}_3)_2$ and 4,4'-bipyridine (4,4'-bpy) catalyzed the cyanosilylation of imines under heterogeneous conditions.

1588

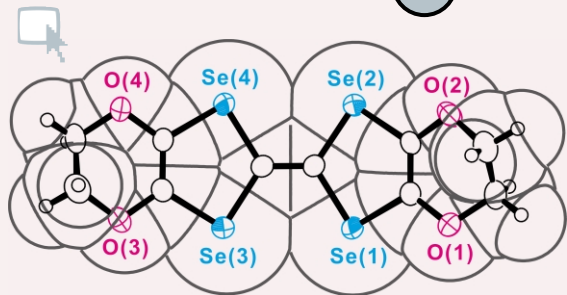


Luminescent homochiral silver(I) lamellar coordination networks built from helical chains

Chuan-De Wu, Helen L. Ngo and Wenbin Lin*

The reactions of 2,2'-dimethoxy-1,1'-binaphthyl-3,3'-bis(4-vinylpyridine) (L) with AgNO_3 or AgClO_4 afforded two luminescent homochiral lamellar coordination polymers, $(\text{AgL}_2)\text{X}$ ($\text{X} = \text{NO}_3^-$ or ClO_4^-), which are built from linking helical chains by Ag(I) atoms as hinges.

1590

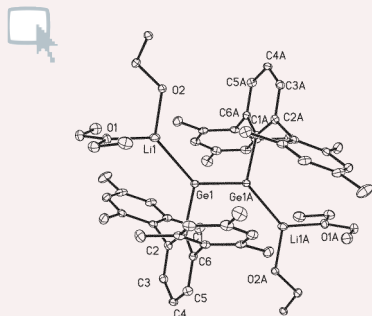


Synthesis, crystal structure and electrochemical properties of bis(ethylenedioxy)tetraselenafulvalene (BEDO-TSeF)

Tatsuro Imakubo,* Takashi Shirahata and Megumi Kibune

The first synthesis of BEDO-TSeF has been accomplished using elemental selenium as the only source of the selenium atoms, and its crystal structure and electrochemical properties are disclosed.

1592

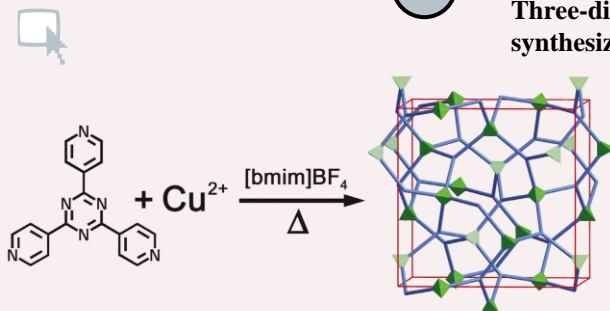


Effects of the alkali metal counter ions on the germanium–germanium double bond length in a heavier group 14 element ethenide salt

Anne F. Richards, Marcin Brynda and Philip P. Power*

The reduction of $\text{Ge}(\text{Cl})\text{C}_6\text{H}_3\text{-2,6-Mes}_2$ by lithium powder affords the *trans*-1,2-dilithiodigermene salt which is the first structurally characterized 1,2-dilithium salt of a group 14 element alkenide species and displays a much shorter Ge–Ge double bond distance than in related species with bridging alkali metal cations.

1594

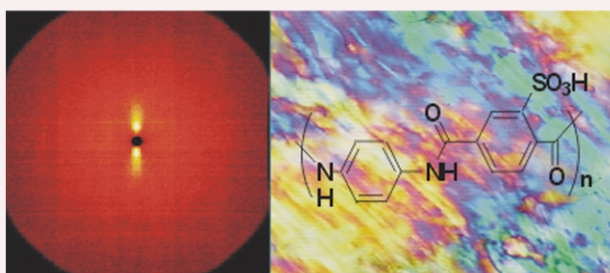


Three-dimensional metal–organic framework with (3,4)-connected net, synthesized from an ionic liquid medium

Danil N. Dybtsev, Hyunphil Chun and Kimoon Kim*

The synthesis of a new 3D metal–organic framework from an ionic liquid medium and its structure having a (3,4)-connected network which comprises 2-fold interpenetrating net with cubic- C_3N_4 topology are described.

1596

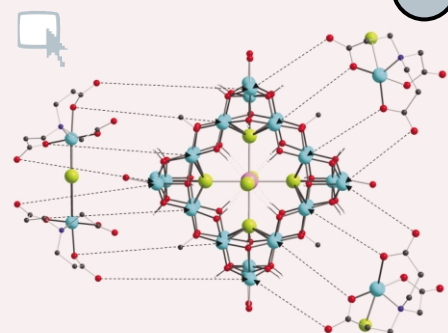


A supramolecular nematic phase in sulfonated polyaramides

Sebastien Viale, Adam S. Best, Eduardo Mendes, Wolter F. Jager and Stephen J. Picken*

A supramolecular assembly based on aggregates of the liquid crystalline polymer, “sulfo-invert-PPTA” that forms a nematic phase at very low polymer concentration in water, has been identified and characterized using WAXS and OPM.

1598



A new type of oxygen bridged $\text{Cu}_{36}^{\text{II}}$ aggregate formed around a central $\{\text{KCl}_6\}^{5-}$ unit

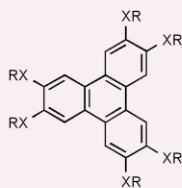
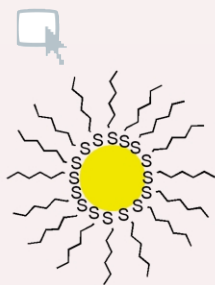
Muralee Murugesu, Rodolphe Clérac, Christopher E. Anson and Annie K. Powell*

A magnetically coupled $\text{Cu}_{36}^{\text{II}}$ aggregate forms around a potassium chloride unit.

1600

Inclusion of gold nanoparticles into a discotic liquid crystalline matrix

Sandeep Kumar* and V. Lakshminarayanan

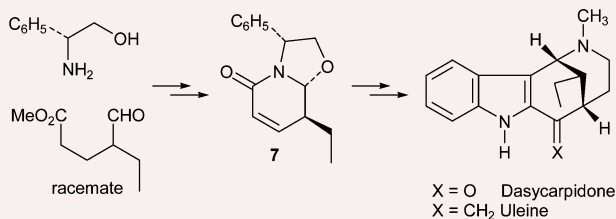


Polarizing optical microscopy, differential scanning calorimetry and DC conductivity experiments indicate inclusion of gold nanoparticles into a matrix of triphenylene based discotic liquid crystals.

1602

Enantioselective formal synthesis of uleine alkaloids from phenylglycinol-derived bicyclic lactams

Mercedes Amat,* Maria Pérez, Núria Llor, Marisa Martinelli, Elies Molins and Joan Bosch*

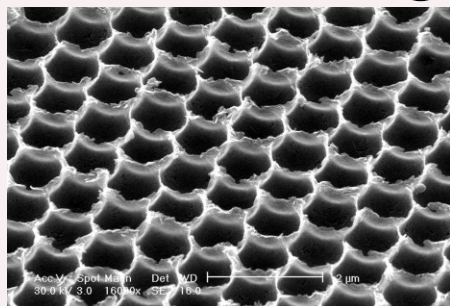


A short synthetic route for the enantioselective synthesis of the alkaloids of the uleine group is reported, the key step being the stereocontrolled conjugate addition of an indole-containing enolate to unsaturated lactam **7**.

1604

Fabrication of large-scale zinc oxide ordered pore arrays with controllable morphology

Bingqiang Cao, Weiping Cai,* Fengqiang Sun, Yue Li, Yong Lei and Lide Zhang

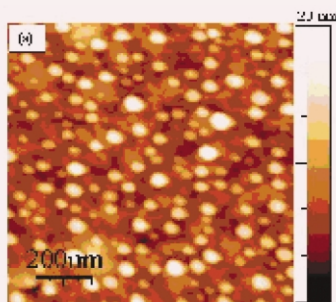


The fabrication of large-scale ZnO ordered pore arrays by the potentiostatic electrochemical deposition method based on a two-dimensional ordered colloidal monolayer template is reported. The pore morphology evolves from hemispherical to a well-like structure by control of the deposition potential.

1606

Fabrication and electrocatalytic properties of polyaniline/Pt nanoparticle composites

Anthony P. O'Mullane, Sara E. Dale, Julie V. Macpherson and Patrick R. Unwin*



Polyaniline (PANI)/Pt nanoparticle composites can be prepared by the spontaneous redox reaction of K₂PtCl₄ with PANI, to yield thin films that show electrocatalytic properties in both acidic and neutral aqueous media.

1608

Synthesis of novel dendrimer-like star block copolymers with definite numbers of arms by combination of ROP and ATRP

Youliang Zhao, Xintao Shuai, Chuanfu Chen and Fu Xi*

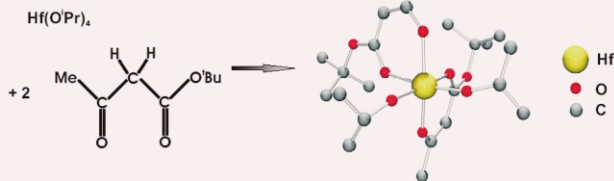


On the basis of hydroxy-terminated polyaryl ether dendrimers, a series of biodegradable dendrimer-like star block copolymers with precise number of arms were first synthesized by mechanism transformation.

1610

Mononuclear precursor for MOCVD of HfO₂ thin films

Arne Baunemann, Reji Thomas, Ralf Becker, Manuela Winter, Roland A. Fischer, Peter Ehrhart, Rainer Waser and Anjana Devi*

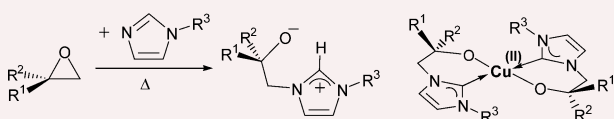


A novel mononuclear mixed alkoxide compound of Hf has been synthesised and structurally characterised. Application of this compound as a precursor in a production tool MOCVD reactor resulted in HfO₂ thin films exhibiting electrical properties which are promising for device applications.

1612

Asymmetric lithium(I) and copper(II) alkoxy-*N*-heterocyclic carbene complexes; crystallographic characterisation and Lewis acid catalysis

Polly L. Arnold,* Mark Rodden, Kate M. Davis, Andrew C. Scarisbrick, Alexander J. Blake and Claire Wilson



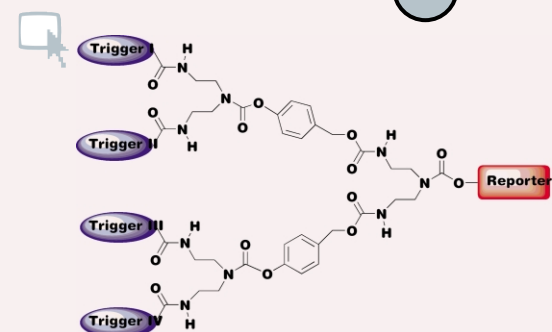
A one-pot synthesis of bidentate, alkoxy-*N*-heterocyclic carbene ligands provides new lithium alkoxy-carbenes and a range of covalently bound organometallic Cu(II) carbene complexes, which are catalytically active, in some cases enantioselectively, for conjugate addition reactions.

1614

Self-immolative dendrimer biodegradability by multi-enzymatic triggering

Roey J. Amir and Doron Shabat*

We synthesize self-immolative dendrimers that degrade readily on enzymatic triggering under physiological conditions.

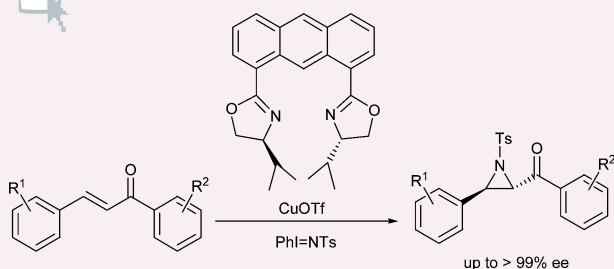


1616

Asymmetric aziridination of chalcones catalyzed by a novel backbone 1,8-bisoxazolinylanthracene (AnBOX)-copper complex

Jiaxi Xu,* Linge Ma and Peng Jiao

Highly enantioselective aziridination of chalcones catalyzed by a novel backbone 1,8-bisoxazolidinylanthracene (AnBOX) and CuOTf and the opposite enantioselectivity compared with the ligands of Evans are described.

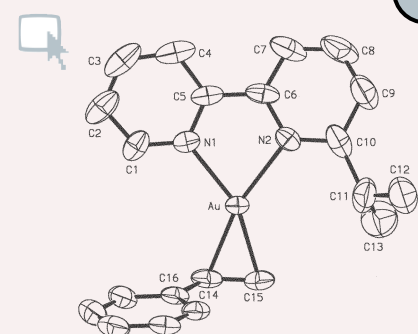


1618

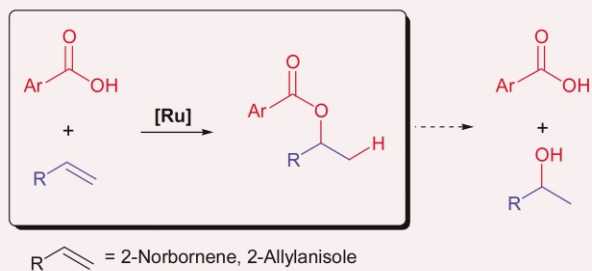
Reaction of gold(III) oxo complexes with alkenes. Synthesis of unprecedented gold alkene complexes, [Au(*N,N*-(alkene))][PF₆]. Crystal structure of [Au(bipy^{ip})(η^2 -CH₂=CHPh)][PF₆] (bipy^{ip} = 6-isopropyl-2,2'-bipyridine)

Maria A. Cinellu,* Giovanni Minghetti, Sergio Stoccoro, Antonio Zucca and Mario Manassero

The first gold alkene complexes supported by nitrogen ligands have been obtained by reaction of gold(III) oxo complexes with alkenes. Both spectroscopic and structural data suggest a non negligible π contribution in the olefin–gold bond.



1620

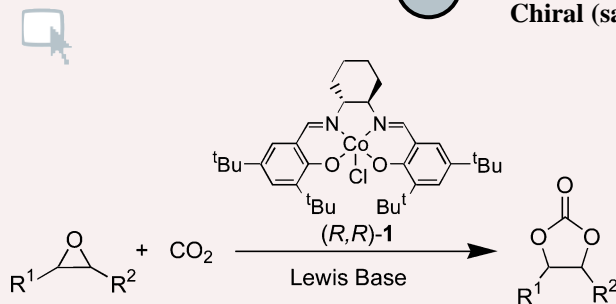


Ruthenium catalyzed addition reaction of carboxylic acid across olefins without β -hydride elimination

Yohei Oe,* Tetsuo Ohta and Yoshihiko Ito

The catalytic system $(Cp^*RuCl_2)_2/6AgOTf/2Ligand$ promotes the addition reaction of carboxylic acid derivatives with olefins, giving the corresponding saturated esters which are used as precursors of alcohols and starting carboxylic acids.

1622

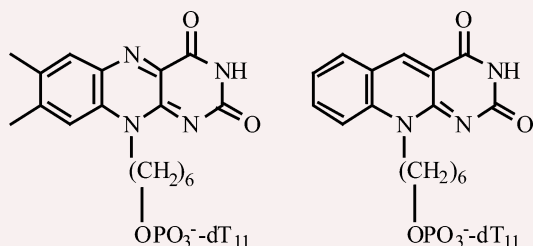


Chiral (salen)Co^{III} catalyst for the synthesis of cyclic carbonates

Robert L. Paddock and SonBinh T. Nguyen*

A catalyst system comprised of a (salen)Co^{III} complex and a Lewis base is investigated for the reaction of CO₂ and a variety of epoxides to form cyclic carbonates. Application of this catalyst system in the kinetic resolution of propylene oxide is also discussed.

1624

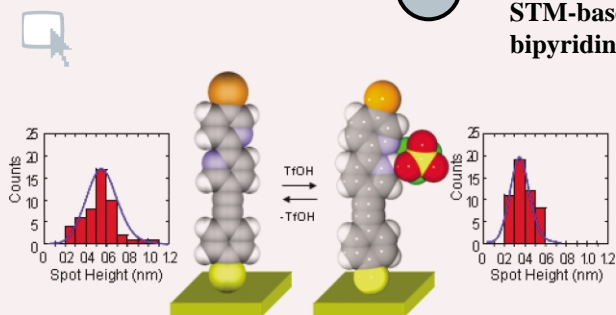


New flavin and deazaflavin oligonucleotide conjugates for the amperometric detection of DNA hybridization

Serge Cosnier,* Chantal Gondran, Cécile Dueymes, Philippe Simon, Marc Fontecave and Jean-Luc Décout

A new amperometric detection of DNA hybridization has been designed via the functionalization of an oligonucleotide by flavin and deazaflavin derivatives exhibiting both electroactive and fluorescent properties.

1626

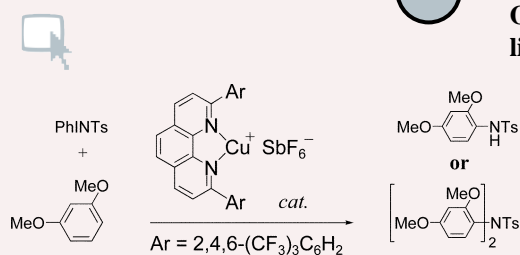


STM-based molecular detection of "catch-and-release" of protons for bipyridine bound to phenylene-ethynylene thiol

Emiko Koyama,* Takao Ishida, Hideo Tokuhisa, Abdelhak Belaissaoui, Yoshinobu Nagawa and Masatoshi Kanosato

The protonation/deprotonation response of a novel thiol adsorbed to a Au surface was investigated with scanning tunneling microscopy (STM), showing reversible changes in the average heights and the height distribution arising from protonation/deprotonation.

1628

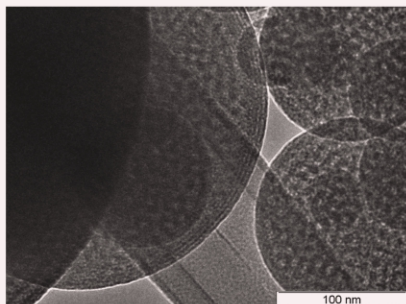


Oxidation-resistant, sterically demanding phenanthrolines as supporting ligands for copper(I) nitrene transfer catalysts

Charles W. Hamilton, David S. Laitar and Joseph P. Sadighi*

A cationic copper(I) complex of a new 1,10-phenanthroline ligand substituted with heavily fluorinated aryl rings catalyses nitrene transfer to the C-H bonds of electron-rich arenes.

1630

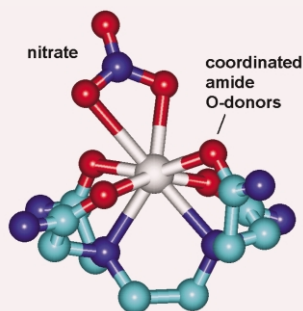


One-pot aerosol synthesis of ordered hierarchical mesoporous core-shell silica nanoparticles

S. Areva, C. Boissière, D. Grosso, T. Asakawa, C. Sanchez and M. Lindén*

A spray-drying procedure for the direct synthesis of mesoporous core-shell structured silica using mixed surfactants as the structure-directing agent is reported. The core-shell structure was induced through the difference in surface activity of one fluorocarbon and one hydrocarbon based surfactant.

1632



Possible insights into metal ion recognition in calcium-binding proteins provided by complexing properties of ligands containing amide oxygen donors

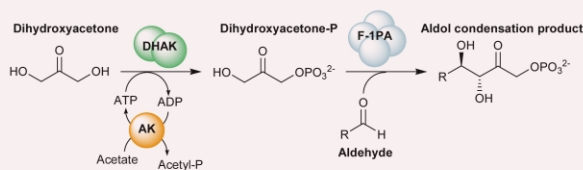
Chynthia J. Siddons and Robert D. Hancock*

The metal ion complexing properties of an amide-containing ligand suggest that at least part of the selectivity for Ca^{2+} over Mg^{2+} in calcium binding proteins comes from intrinsic coordinating properties of the amide oxygen-donor.

1634

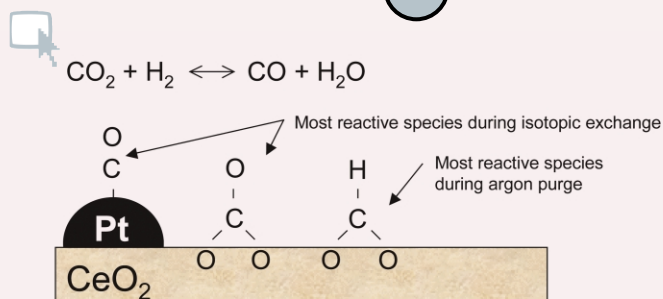
Multienzyme system for dihydroxyacetone phosphate-dependent aldolase catalyzed C–C bond formation from dihydroxyacetone

Israel Sánchez-Moreno, Juan Francisco García-García, Agatha Bastida and Eduardo García-Junceda*



A multienzyme system composed of recombinant dihydroxyacetone kinase, fucose-1-phosphate aldolase and acetate kinase, allows a practical one-pot C–C bond formation from dihydroxyacetone and an aldehyde.

1636



On the importance of steady-state isotopic techniques for the investigation of the mechanism of the reverse water-gas-shift reaction

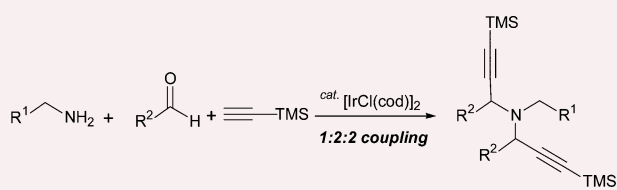
Daniele Tibiletti, Alexandre Goguet, Frederic C. Meunier,* John P. Breen and Robbie Burch

The reactivity of surface intermediates in the reverse water-gas-shift reaction on a Pt/CeO₂ catalyst is critically dependent on the reaction conditions.

1638

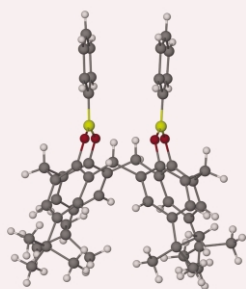
Iridium-catalyzed coupling of simple primary or secondary amines, aldehydes and trimethylsilylacetylene: preparation of propargylic amines

Satoshi Sakaguchi, Tomoya Mizuta, Masuyuki Furuwan, Takashi Kubo and Yasutaka Ishii*



The 1 : 2 : 2 coupling product of amine, aldehyde and alkyne was obtained by allowing primary amine to react with aldehyde and trimethylsilylacetylene in the presence of a catalytic amount of $[\text{IrCl}(\text{cod})]_2$.

1640



3

Synthesis and structural characterisation of the first bis(bora)calixarene: a selective, bidentate, fluorescent fluoride sensor

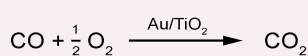
Susumu Arimori, Matthew G. Davidson,* Thomas M. Fyles, Thomas G. Hibbert, Tony D. James and Gabriele I. Kociok-Köhn

A bis(bora)calixarene, **3**, the first lower-rim boron derivatised calixarene to be structurally characterised, is synthesised by the reaction of PhBCl₂ with 4-*tert*-butylcalix[4]arene, and is demonstrated to be a sensitive and selective fluorescent fluoride sensor.

1642

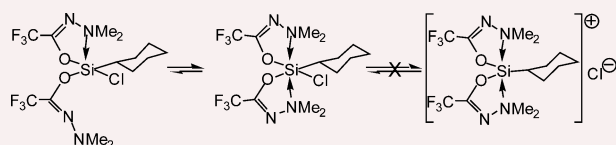
The influence of metal loading and pH during preparation on the CO oxidation activity of Au/TiO₂ catalysts

François Moreau, Geoffrey C. Bond and Adrian O. Taylor*



Au/TiO₂ catalysts with gold loadings in the range 0.06–1.9 wt.%, prepared by a repeatable ‘deposition–precipitation’ method at pH 9, have a constant and high specific activity for the oxidation of CO: $3.9(\pm 0.4) \times 10^{-4} \text{ mol}_{\text{CO}} \text{ s}^{-1} \text{ g}_{\text{Au}}^{-1}$ at 243 K.

1644

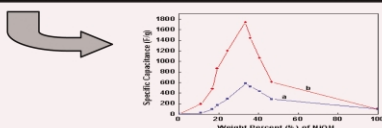
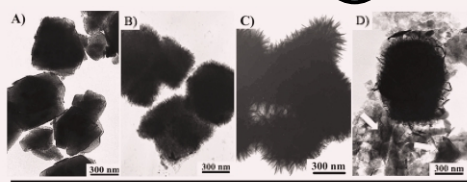


Neutral and ionic dissociation patterns in hexacoordinate silicon chelates: a model nucleophilic substitution at pentacoordinate silicon

Boris Gostevskii, Keren Adear, Akella Sivaramakrishna, Gilad Silbert, Dietmar Stalke, Nikolaus Kocher, Inna Kalikhman* and Daniel Kost*

A model nucleophilic-displacement reaction coordinate at pentacoordinate silicon is demonstrated by neutral and ionic dissociation equilibria through a stable hexacoordinate complex.

1646

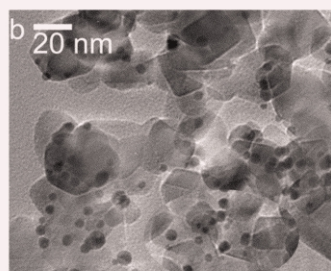


Preparation of novel nano-composite Ni(OH)₂/USY material and its application for electrochemical capacitance storage

Lin Cao, Ling-Bin Kong, Yan-Yu Liang and Hu-Lin Li*

A novel nano-composite material of Ni(OH)₂/USY was prepared in our lab. This nanostructure creates electrochemical accessibility of electrolyte OH⁻ ions to Ni(OH)₂ thin layers and a fast diffusion rate within the redox phase.

1648

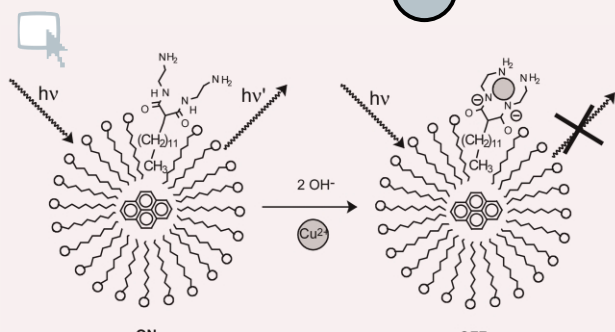


Direct propylene epoxidation on chemically reduced Au nanoparticles supported on titania

Ju Chou and Eric W. McFarland*

The activity and selectivity of supported gold nanoclusters are highly dependent upon the preparation route. By chemical reduction in suspension followed by impregnation on titania (P25), Au nanoclusters (4.6 nm) selective for propylene epoxidation were synthesized and characterized by TEM and XPS.

1650



Using micelles for a new approach to fluorescent sensors for metal cations

Yuri Diaz Fernandez,* Aurora Pérez Gramatges, Valeria Amendola, Francesco Foti, Carlo Mangano, Piersandro Pallavicini* and Stefano Patroni

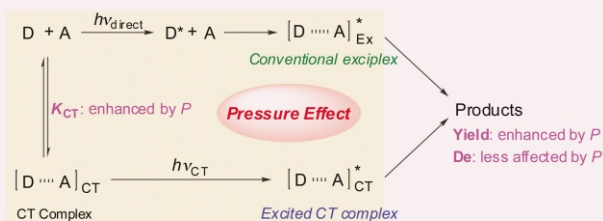
Self-assembly inside micelles allows for the easy building of a water soluble multicomponent fluorescent sensor for Cu^{2+} and Ni^{2+} .

1652

Pressure control of diastereodifferentiating [2 + 2] photocycloaddition of (*E*)-stilbene to chiral fumarate upon direct and charge-transfer excitation

Hideaki Saito, Tadashi Mori,* Takehiko Wada and Yoshihisa Inoue*

Significant enhancements in complexation (K_{CT}) and product yield (particularly for CT excitation) were achieved on increasing the pressure without any substantial change in diastereoselectivities in both excitation modes.

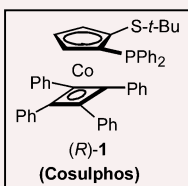
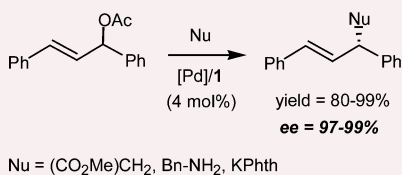


1654

First planar chiral bidentate ligand based on a (η^5 -cyclopentadienyl)(η^4 -cyclobutadiene) cobalt backbone: high efficiency in enantioselective palladium-catalyzed allylic substitutions

Ramón Gómez Arrayás, Olga García Mancheño and Juan C. Carretero*

The planar chiral (η^5 -cyclopentadienyl)(η^4 -tetraphenylcyclobutadiene)cobalt complex (*R*)-**1** has been developed. This P,S-bidentate ligand provides very high enantioselectivities in Pd-catalyzed allylic substitutions with carbon and nitrogen nucleophiles.

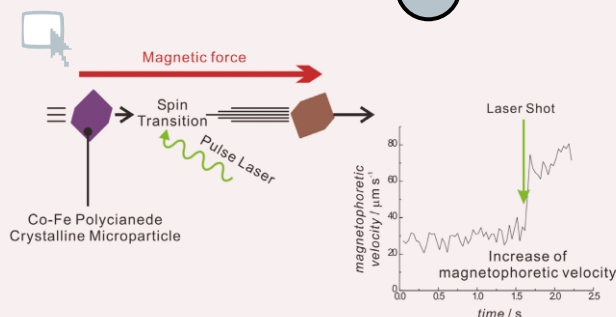


1656

Magnetophoretic detection of photo-induced spin transition

Masayori Suwa and Hitoshi Watarai*

Magnetophoretic velocimetry detected the spin transition of a single Co-Fe Prussian Blue analogous micro-crystal in water induced by a single-shot pulse laser.

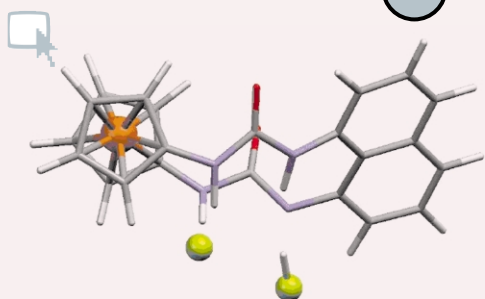


1658

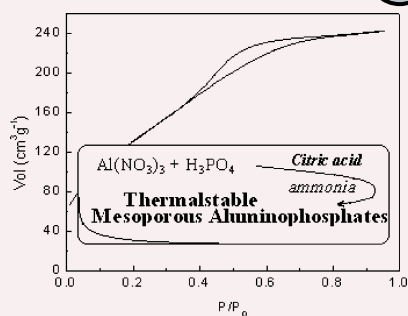
A new fluoride selective electrochemical and fluorescent chemosensor based on a ferrocene-naphthalene dyad

Francisco Otón, Alberto Tárraga,* María D. Velasco, Arturo Espinosa and Pedro Molina*

A new supramolecular sensor for fluoride ions, based on a bis(urea) binding site and ferrocene and naphthalene reporting groups, is described.



1660

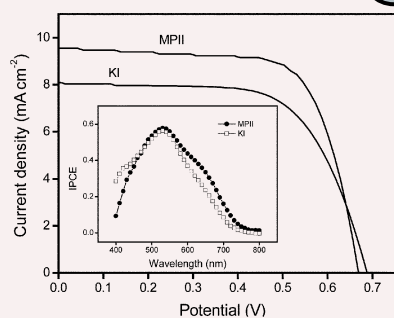


Synthesis of amorphous mesoporous aluminophosphate materials with high thermal stability using a citric acid route

Gang Liu, Mingjun Jia,* Zhou Zhou, Wenxiang Zhang,* Tonghao Wu and Dazhen Jiang

Amorphous mesoporous aluminophosphates synthesized by using a citric acid route, exhibit high specific surface areas, narrow pore size distributions and excellent thermal stabilities.

1662



Dye-sensitized nanocrystalline solar cells based on composite polymer electrolytes containing fumed silica nanoparticles

Jong Hak Kim, Moon-Sung Kang, Young Jin Kim, Jongok Won, Nam-Gyu Park and Yong Soo Kang*

We report remarkably high energy conversion efficiency (4.5% at 100 mW cm⁻²) of a dye-sensitized solar cell in the solid state, using composite polymer electrolytes containing fumed silica nanoparticles.

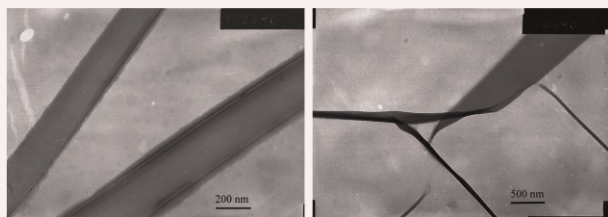
1664



Nanotubes of poly(phenylene vinylene) derivative at the air/water interface

Lin Guo,* Zhongkui Wu and Yingqiu Liang*

Different sizes of poly(2-methoxy-5-(*n*-hexadecyloxy)-*p*-phenylene vinylene) nanotubes have been fabricated at the air/water interface by LB techniques.

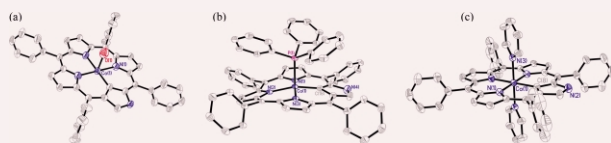


1666



Dianionic and trianionic macrocycles in cobalt N-confused porphyrin complexes

John D. Harvey and Christopher J. Ziegler*



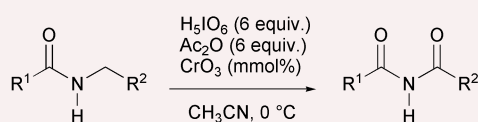
We report the syntheses of cobalt N-confused porphyrins; this work completes the series of the late first-row transition metals that have been incorporated into the core of N-confused porphyrin, and in these compounds the macrocycles can act as either a -2 or -3 anion.

1668



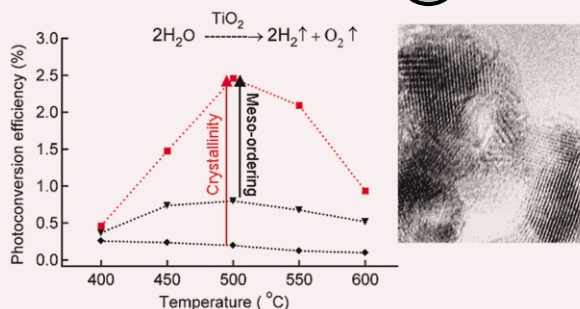
Novel chromium(VI) catalyzed oxidation of *N*-alkylamides to imides with periodic acid

Liang Xu, Suhong Zhang and Mark L. Trudell*



A novel and practical procedure for preparation of imides is described using chromium(VI) oxide to catalyze the oxidation of *N*-alkylamides with periodic acid in the presence of acetic anhydride in acetonitrile.

1670



Synthesis and photocatalytic properties of highly crystalline and ordered mesoporous TiO₂ thin films

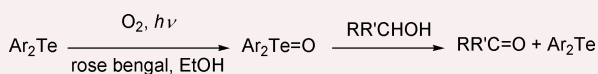
Jing Tang, Yiying Wu, Eric W. McFarland and Galen D. Stucky*

Highly crystalline and ordered mesoporous TiO₂ thin films have been synthesized by stabilization of the mesostructure with confined carbon; the films exhibit 2.5% photoconversion efficiency for the water photolysis at zero-bias and Xe lamp illumination of 40 mW cm⁻².

1672

Photosensitized oxygenation of diaryl tellurides to telluroxides and their oxidizing properties

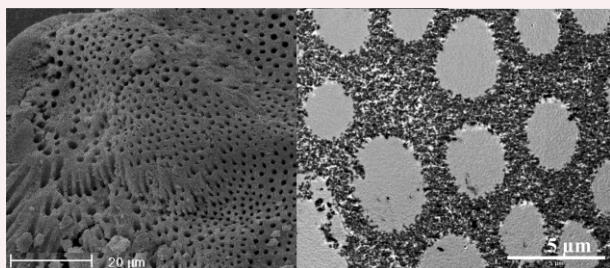
Makoto Oba, Masaki Endo, Kozaburo Nishiyama,* Akihiko Ouchi and Wataru Ando*



Ar = 2,4,6-trimethylphenyl (Mes) or 2,4,6-triisopropylphenyl (Tip)

Photosensitized oxygenation of bulky diaryl tellurides afforded the corresponding telluroxides which were found to be mild oxidizing agents towards simple alcohols.

1674



A novel and template-free method for the spontaneous formation of aluminosilicate macro-channels with mesoporous walls

Alexandre Léonard and Bao-Lian Su*

Hierarchical meso-macroporous aluminosilicates made of straight tubular macrochannels separated by disordered mesoporous walls were prepared by a simple and template-free synthesis pathway.

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 Amat, Mercedes, 1602
 Amendola, Valeria, 1650
 Amir, Roey J., 1614
 Ando, Wataru, 1672
 Anson, Christopher E., 1598
 Areva, S., 1630
 Arimori, Susumu, 1640
 Armaroli, Nicola, 1582
 Arnold, Polly L., 1612
 Arrayás, Ramón Gómez, 1654
 Asakawa, T., 1630
 Bastida, Agatha, 1634
 Baunemann, Arne, 1610
 Becker, Ralf, 1610
 Belaïssaoui, Abdelhak, 1626
 Best, Adam S., 1596
 Blake, Alexander J., 1612
 Boissière, C., 1630
 Bond, Geoffrey C., 1642
 Bosch, Joan, 1602
 Breen, John P., 1636
 Brynda, Marcin, 1592
 Burch, Robbie, 1636
 Cai, Weiping, 1604
 Cao, Bingqiang, 1604
 Cao, Lin, 1646
 Cardinali, François, 1582
 Carretero, Juan C., 1654
 Chen, Chuanfu, 1608
 Chou, Ju, 1648
 Chun, Hyungphil, 1594
 Cinellu, Maria A., 1618
 Clérac, Rodolphe, 1598
 Cosnier, Serge, 1624
 Cronin, Leroy, 1580
 Dale, Sara E., 1606
 Davidson, Matthew G., 1640
 Davis, Kate M., 1612
 de Vries, Johannes G., 1559
 Décout, Jean-Luc, 1624
 Devi, Anjana, 1610
 Dueymes, Cécile, 1624
 Dybtsev, Danil N., 1594
 Ehrhart, Peter, 1610
 Endo, Masaki, 1672
 Espinosa, Arturo, 1658
 Fernandez, Yuri Diaz, 1650
 Fischer, Roland A., 1610
 Fontecave, Marc, 1624
 Foti, Francesco, 1650
 Fujita, Makoto, 1586
 Furuwan, Masuyuki, 1638
 Fyles, Thomas M., 1640
 García-García, Juan Francisco, 1634
 García-Junceda, Eduardo, 1634
 Goguet, Alexandre, 1636
 Gondran, Chantal, 1624
 Gostevskii, Boris, 1644
 Gramatges, Aurora Pérez, 1650
 Grosso, D., 1630
 Guo, Lin, 1664
 Hamilton, Charles W., 1628
 Hancock, Robert D., 1632
 Harvey, John D., 1666
 Hibbert, Thomas G., 1640
 Imakubo, Tatsuro, 1590
 Inoue, Yoshihisa, 1652
 Ishida, Takao, 1626
 Ishii, Yasutaka, 1638
 Ito, Yoshihiko, 1620
 Jager, Wolter F., 1596
 James, Tony D., 1640
 Jia, Mingjun, 1660
 Jiang, Dazhen, 1660
 Jiao, Peng, 1616
 Kalikhman, Inna, 1644
 Kanesato, Masatoshi, 1626
 Kang, Moon-Sung, 1662
 Kang, Yong Soo, 1662
 Kanie, Kiyoshi, 1584
 Kariuki, Benson M., 1580
 Kibune, Megumi, 1590
 Kim, Jong Hak, 1662
 Kim, Kimoon, 1594
 Kim, Young Jin, 1662
 Kocher, Nikolaus, 1644
 Kociok-Köhn, Gabriele I., 1640
 Kögerler, Paul, 1580
 Kong, Ling-Bin, 1646
 Kost, Daniel, 1644
 Koyama, Emiko, 1626
 Kubo, Takashi, 1638
 Kumar, Sandeep, 1600
 Laitar, David S., 1628
 Lakshminarayanan, V., 1600
 Lei, Yong, 1604
 Léonard, Alexandre, 1674
 Li, Hu-Lin, 1646
 Li, Yue, 1604
 Liang, Yan-Yu, 1646
 Liang, Yingqiu, 1664
 Lin, Wenbin, 1588
 Lindén, M., 1630
 Liu, Gang, 1660
 Llor, Núria, 1602
 Ma, Linge, 1616
 McFarland, Eric W., 1648, 1670
 Macpherson, Julie V., 1606
 Mamlouk, Hind, 1582
 Manassero, Mario, 1618
 Mancheño, Olga García, 1654
 Mangano, Carlo, 1650
 Martinelli, Marisa, 1602
 Mendes, Eduardo, 1596
 Meunier, Frederic C., 1636
 Minghetti, Giovanni, 1618
 Mizuta, Tomoya, 1638
 Molina, Pedro, 1658
 Molins, Elies, 1602
 Moreau, François, 1642
 Mori, Tadashi, 1652
 Murugesu, Muralee, 1598
 Nagawa, Yoshinobu, 1626
 Ngo, Helen L., 1588
 Nguyen, SonBinh T., 1622
 Nierengarten, Jean-François, 1582
 Nishiyama, Kozaburo, 1672
 Oba, Makoto, 1672
 Oe, Yohei, 1620
 Ohmori, Osamu, 1586
 Ohta, Tetsuo, 1620
 O'Mullane, Anthony P., 1606
 Otón, Francisco, 1658
 Ouchi, Akihiko, 1672
 Paddock, Robert L., 1622
 Pallavicini, Piersandro, 1650
 Park, Nam-Gyu, 1662
 Patroni, Stefano, 1650
 Pérez, Maria, 1602
 Picken, Stephen J., 1596
 Powell, Annie K., 1598
 Power, Philip P., 1592
 Reetz, Manfred T., 1559
 Richards, Anne F., 1592
 Rio, Yannick, 1582
 Rodden, Mark, 1612
 Sadighi, Joseph P., 1628
 Saito, Hideaki, 1652
 Sakaguchi, Satoshi, 1638
 Sanchez, C., 1630
 Sánchez-Moreno, Israel, 1634
 Scarisbrick, Andrew C., 1612
 Seeber, Georg, 1580
 Shabat, Doron, 1614
 Shirahata, Takashi, 1590
 Shuai, Xintao, 1608
 Siddons, Chynthia J., 1632
 Silbert, Gilad, 1644
 Simon, Philippe, 1624
 Sivaramakrishna, Akella, 1644
 Stalke, Dietmar, 1644
 Stoccoro, Sergio, 1618
 Stucky, Galen D., 1670
 Su, Bao-Lian, 1674
 Sugimoto, Tadao, 1584
 Sun, Fengqiang, 1604
 Suwa, Masayori, 1656
 Tang, Jing, 1670
 Tárraga, Alberto, 1658
 Taylor, Adrian O., 1642
 Thomas, Reji, 1610
 Tibiletti, Daniele, 1636
 Tokuhisa, Hideo, 1626
 Trudell, Mark L., 1668
 Unwin, Patrick R., 1606
 Velasco, María D., 1658
 Viale, Sebastien, 1596
 Wada, Takehiko, 1652
 Waser, Rainer, 1610
 Watarai, Hitoshi, 1656
 Wilson, Claire, 1612
 Winter, Manuela, 1610
 Won, Jongok, 1662
 Wu, Chuan-De, 1588
 Wu, Tonghao, 1660
 Wu, Yiyang, 1670
 Wu, Zhongkui, 1664
 Würthner, Frank, 1564
 Xi, Fu, 1608
 Xu, Jiayi, 1616
 Xu, Liang, 1668
 Zhang, Lide, 1604
 Zhang, Suhong, 1668
 Zhang, Wenxiang, 1660
 Zhao, Youliang, 1608
 Zhou, Zhou, 1660
 Ziegler, Christopher J., 1666
 Zucca, Antonio, 1618

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