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IN THIS ISSUE

ISSN 1359-7345 CODEN CHCOFS (35) 4081-4220 (2008)



Cover

See Azusa Takai *et al.*, pp. 4171–4173. Mesoporous Pt nanotubes were realized by the volume control of lyotropic liquid crystals infiltrated inside the channels of porous anodic alumina membranes under a reduced pressure. Image reproduced by permission of Azusa Takai, Yusuke Yamauchi and Kazuyuki Kuroda from *Chem. Commun.*, 2008, 4171.



Inside cover

See Sarit S. Agasti *et al.*, pp. 4123–4125. Water-soluble flavin dendrons have been synthesised and the role dendron generation has on modulating the physical properties and reactivity of the flavin unit have been explored. Image reproduced by permission of Sarit S. Agasti, Stuart T. Caldwell, Graeme Cooke, Brian J. Jordan, Andrew Kennedy, Nadiya Kryvokhyzha, Gouher Rabani, Subinoy Rana, Amitav Sanyal and Vincent M. Rotello from *Chem. Commun.*, 2008, 4123.

CHEMICAL SCIENCE

C65

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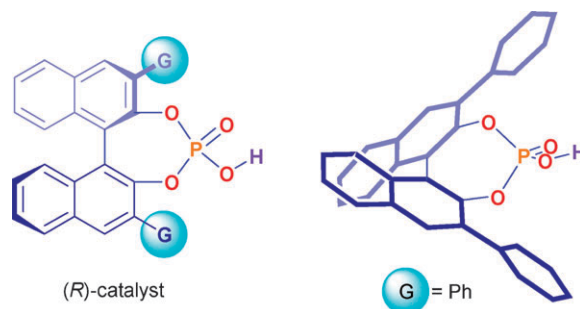
FEATURE ARTICLE

4097

Binaphthol-derived phosphoric acid as a versatile catalyst for enantioselective carbon–carbon bond forming reactions

Masahiro Terada*

Binaphthol-derived monoposphoric acids have been designed as novel chiral Brønsted-acid catalysts. The chiral phosphoric acids thus developed functioned as efficient enantioselective catalysts for a variety of carbon–carbon bond forming reactions affording enantioenriched products in excellent selectivities.



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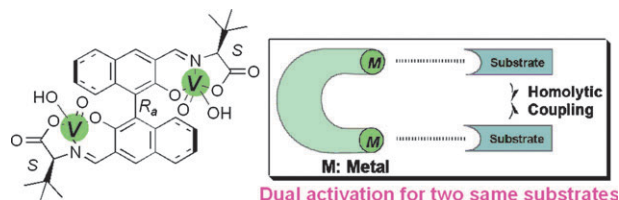
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4113

Dinuclear chiral vanadium catalysts for oxidative coupling of 2-naphthols *via* a dual activation mechanism

Shinobu Takizawa, Tomomi Katayama and Hiroaki Sasai*

Development of dual activation catalysts for an enantioselective oxidative coupling of 2-naphthols was described. To develop the most powerful catalyst, chiral dinuclear vanadium(v) complexes were prepared by using VOCl_3 .



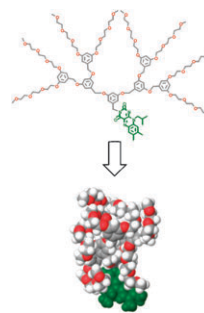
COMMUNICATIONS

4123

Dendron-based model systems for flavoenzyme activity: towards a new class of synthetic flavoenzyme

Sarit S. Agasti, Stuart T. Caldwell, Graeme Cooke,* Brian J. Jordan, Andrew Kennedy, Nadiya Kryvokhyzha, Gouher Rabani, Subinoy Rana, Amitav Sanyal and Vincent M. Rotello

Water-soluble flavin dendrons have been synthesized and the role dendrimer generation has on the physical and catalytic properties of the flavin moiety of these assemblies has been investigated.

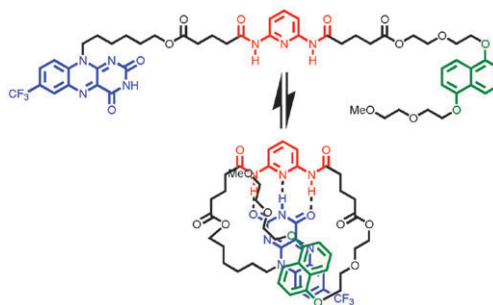


4126

Model systems for flavoenzyme activity: intramolecular self-assembly of a flavin derivative *via* hydrogen bonding and aromatic interactions

Stuart T. Caldwell, Graeme Cooke,* Shanika G. Hewage, Suhil Mabruk, Gouher Rabani, Vincent Rotello, Brian O. Smith, Chandramouleeswaran Subramani and Patrice Woisel

We describe the intramolecular self-assembly of a flavin derivative *via* hydrogen bonding and aromatic interactions.

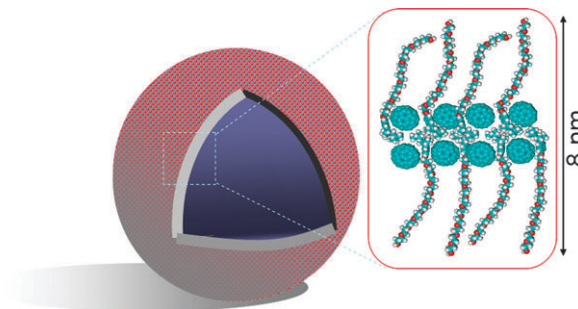


4129

Unilamellar composite vesicles and Y-junctions from pristine fullerene C_{60}

Illa Ramakanth, Balachandran Vijai Shankar and Archita Patnaik*

Aqueous phase formation of TX-100 vesicles with pristine C_{60} as an integral part of the bilayer architecture and Y-junctioned networks are reported through interactive non-covalent interactions.



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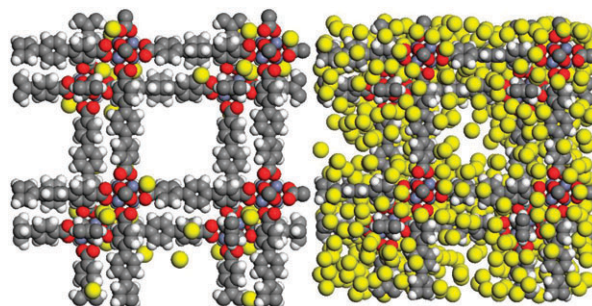
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4132

Is catenation beneficial for hydrogen storage in metal–organic frameworks?

Patrick Ryan, Linda J. Broadbelt and Randall Q. Snurr*

Grand canonical Monte Carlo (GCMC) simulations demonstrate that catenation can be beneficial for improving hydrogen storage in metal–organic frameworks at cryogenic temperatures and low pressures but not necessarily at room temperature.

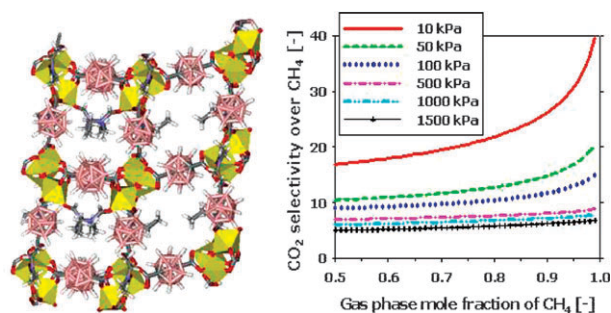


4135

Carborane-based metal–organic frameworks as highly selective sorbents for CO₂ over methane

Youn-Sang Bae, Omar K. Farha, Alexander M. Spokoyny, Chad A. Mirkin, Joseph T. Hupp and Randall Q. Snurr*

A carborane-based metal–organic framework with coordinatively unsaturated metal sites yields high selectivities for CO₂ over CH₄ (~17).

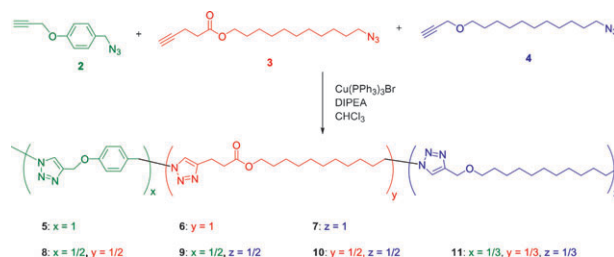


4138

Click chemistry step growth polymerization of novel α -azide- ω -alkyne monomers

Sandra Binauld, Denis Damiron, Thierry Hamaide, Jean-Pierre Pascault, Etienne Fleury and Eric Drockenmuller*

A novel A–B step growth polymerization strategy based on click chemistry polyaddition of tailor-made α -azide- ω -alkyne low molar mass monomers was developed, leading to polytriazole (co)polymers with tunable structures and properties.

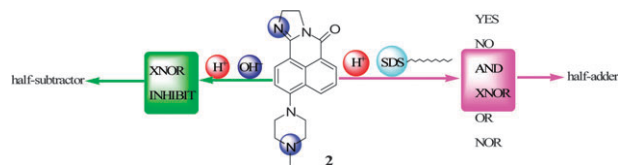


4141

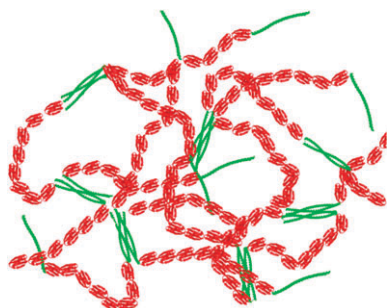
Multiple molecular logic functions and molecular calculations facilitated by surfactant's versatility

Junhong Qian, Xuhong Qian,* Yufang Xu and Shenyi Zhang

With the assistance of anionic surfactant SDS, compound **2** can perform ten logic functions (AND, OR, XNOR, INHIBIT, etc.) of two-input systems as well as half addition and half subtraction.



4144

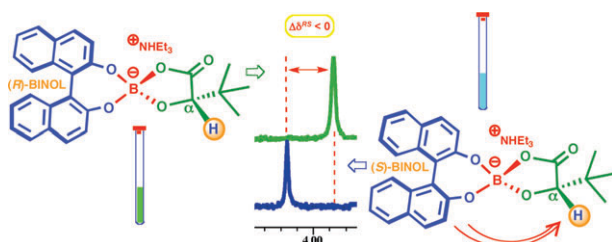


Engineering tandem modular protein based reversible hydrogels

Yi Cao and Hongbin Li*

The authors report the engineering of the first tandem modular protein based hydrogel that exhibits unique properties combining low erosion rate, fast and reversible sol-gel transition and antibody binding ability.

4147

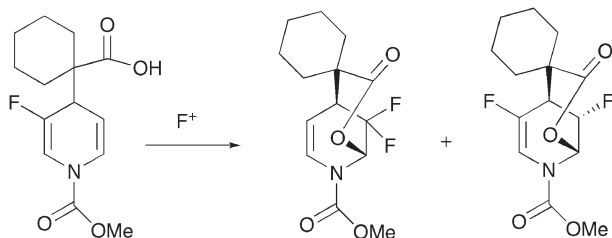


In tube determination of the absolute configuration of α - and β -hydroxy acids by NMR via chiral BINOL borates

Félix Freire, Emilio Quiñoá and Ricardo Riguera*

A NMR methodology through the formation of chiral BINOL borates in the NMR tube—that reunites the advantages of CDAs and CSAs^N—allows the assignment of the absolute configuration of α - and β -hydroxy acids.

4150

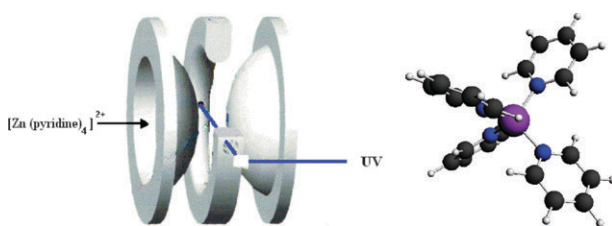


Lactonizations of carboxylic acid-substituted 3-fluorodihydropyridines with electrophiles: peculiar behaviour of F^+

Henri Rudler,* Andrée Parlier, Louis Hamon, Patrick Herson and Jean-Claude Daran

In contrast to other electrophiles, fluorine seems to like fluorine.

4153



State-resolved UV photofragmentation spectrum of the metal dication complex $[Zn(pyridine)_4]^{2+}$

Guohua Wu, Caroline Norris, Hamish Stewart, Hazel Cox* and Anthony J. Stace*

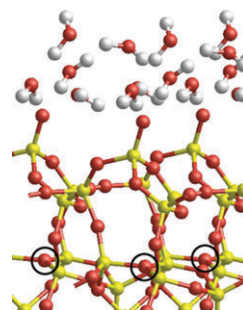
A combined theoretical and experimental study of electronic transitions in the complex $[Zn(pyridine)_4]^{2+}$ provides the first example of a state-resolved electronic spectrum to be recorded for a dication complex in the gas phase.

4156

Low reactivity of non-bridging oxygen defects on stoichiometric silica surfaces

Said Hamad and Stefan T. Bromley*

Using *ab initio* molecular dynamics simulations we demonstrate that surfaces of stoichiometric silica exhibiting non-bridging oxygen defects can be surprisingly resistant to attack by water.

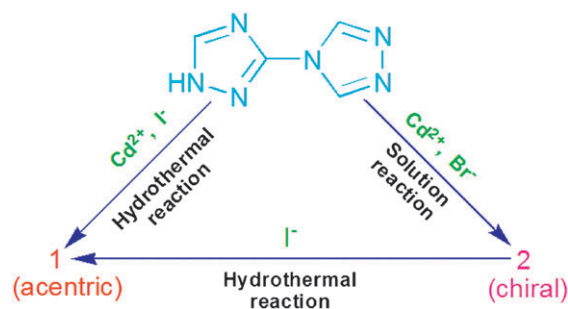


4159

Controlled generation of acentric and homochiral coordination compounds from a versatile asymmetric ligand 4-(1*H*-1,2,4-triazol-3-yl)-4*H*-1,2,4-triazole

Rui-Bo Zhang, Jian Zhang, Zhao-Ji Li, Ye-Yan Qin, Jian-Kai Cheng and Yuan-Gen Yao*

Luminescent acentric and homochiral coordination compounds, which possess new topological networks, multiple helical structures and NLO properties, have been generated through the control of a versatile asymmetric ligand as well as inorganic anions.

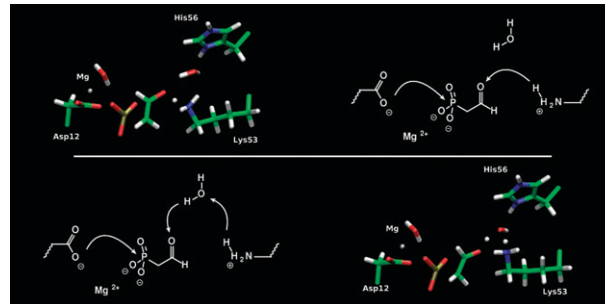


4162

Towards understanding phosphonoacetaldehyde hydrolase: an alternative mechanism involving proton transfer that triggers P–C bond cleavage

Borys Szeferczyk

The theoretical QM/MM study of the reaction catalysed by phosphonoacetaldehyde hydrolase indicates a possible alternative mechanism of the P–C bond cleavage that is facilitated by a proton transfer from catalytic residue to the substrate, instead of formation of a covalently bound intermediate (Schiff-base) postulated earlier.

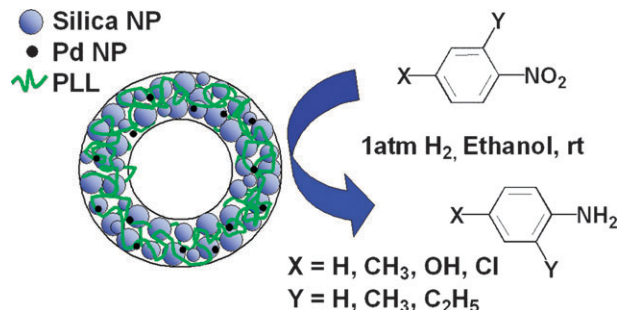


4165

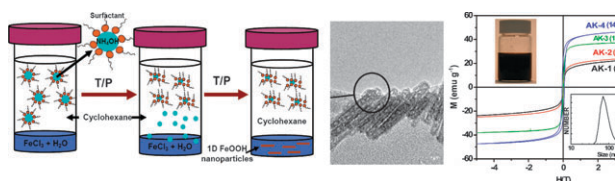
Trapping Pd(0) in nanoparticle-assembled microcapsules: an efficient and reusable catalyst

Arlin Jose Amali and Rohit Kumar Rana*

Pd nanoparticles dually encased by soft (polymer) and hard (inorganic) materials in a microcapsule structure, obtained *via* a nanoparticle self-assembly method, exhibit excellent catalytic activity, with efficient catalyst recovery and reusability.



4168

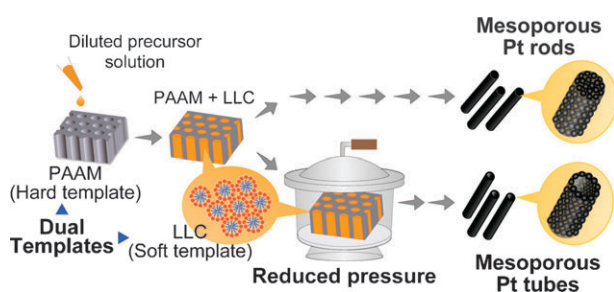


Controlled formation of porous magnetic nanorods *via* a liquid/liquid solvothermal method

Oscar Bomati-Miguel, Aldo F. Rebolledo and Pedro Tartaj*

A liquid/liquid solvothermal method is reported for the preparation of porous magnetic nanorods with sizes readily modulated to at least within an order of magnitude difference.

4171

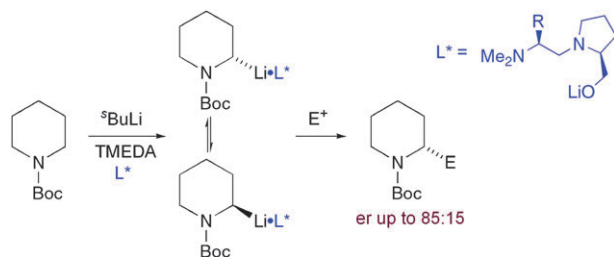


Fabrication of mesoporous Pt nanotubes utilizing dual templates under a reduced pressure condition

Azusa Takai, Yusuke Yamauchi* and Kazuyuki Kuroda*

Pt nanotubes with mesoporous walls were realized through the expansion of lyotropic liquid crystals infiltrated inside the channels of porous anodic alumina membranes under a reduced pressure condition.

4174

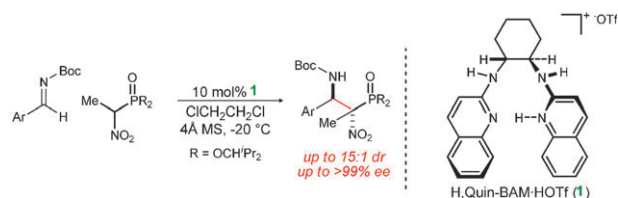


Dynamic resolution of *N*-Boc-2-lithiopiperidine

Iain Coldham,* Sophie Raimbault, Praful T. Chovatia, Jignesh J. Patel, Daniele Leonori, Nadeem S. Sheikh and David T. E. Whittaker

The *2S* and *2R* organolithium complexes can be resolved in the presence of a chiral ligand.

4177



A diastereo- and enantioselective synthesis of α -substituted *anti*- α,β -diaminophosphonic acid derivatives

Jeremy C. Wilt, Maren Pink and Jeffrey N. Johnston*

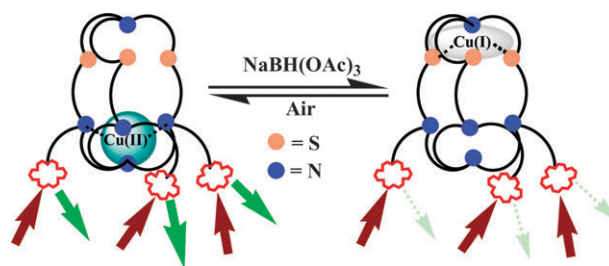
Highly diastereo- and enantioselective additions of α -nitrophosphonates to imines are described. High diastereoselection relies on the finding that chiral proton catalyst **1** and branched phosphonate esters combine to raise diastereoselection from 1 : 1 (substrate-controlled) to as high as 15 : 1 in favor of the *anti*-diastereomer.

4180

Translocation of copper within the cavity of cryptands: reversible fluorescence signaling

Kalyan K. Sadhu and Parimal K. Bharadwaj*

A copper ion is shown to reversibly translocate within the cavity of two new cryptands, which can be monitored by fluorescence ON–OFF signaling for potential use as a molecular photonic device.

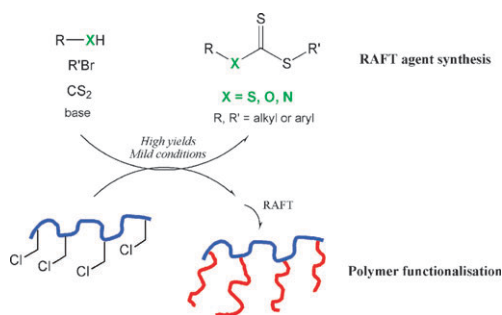


4183

Facile one pot synthesis of a range of reversible addition–fragmentation chain transfer (RAFT) agents

Jared Skey and Rachel K. O'Reilly*

A universal strategy for the synthesis of trithiocarbonates, xanthates and dithiocarbamates for application as chain transfer agents in RAFT/MADIX polymerisation is presented. This mild synthetic strategy has also been explored as an efficient post-polymerisation polymer functionalisation strategy and in the synthesis of multifunctional CRP initiators.

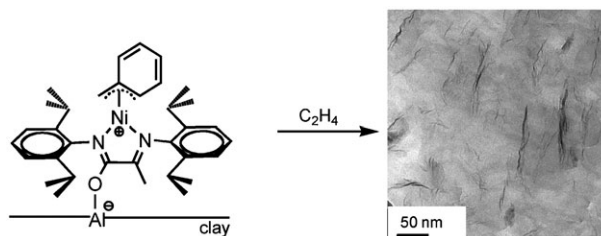


4186

Highly dispersed clay–polyolefin nanocomposites free of compatibilizers, via the *in situ* polymerization of α -olefins by clay-supported catalysts

Susannah L. Scott,* Brian C. Peoples, Cathleen Yung, René S. Rojas, Vikram Khanna, Hironari Sano, Toru Suzuki and Fumihiko Shimizu

In situ polymerization by catalysts supported on acid-treated montmorillonite produces well-dispersed clay–polyolefin nanocomposites directly, without the use of surfactants.

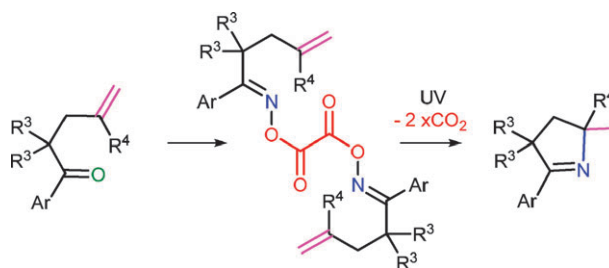


4189

From dioxime oxalates to dihydropyrroles and phenanthridines via iminyl radicals

Fernando Portela-Cubillo, Eoin M. Scanlan, Jackie S. Scott and John C. Walton*

Dioxime oxalates cleanly release iminyl radicals on UV irradiation. The method is useful for spectroscopic work and can be adapted for preparations of N-heterocycles. Iminyl radicals with pent-4-enyl substituents yield 3,4-dihydro-2H-pyrroles and 2-iminylbiphenyls ring close to give phenanthridines.



4192

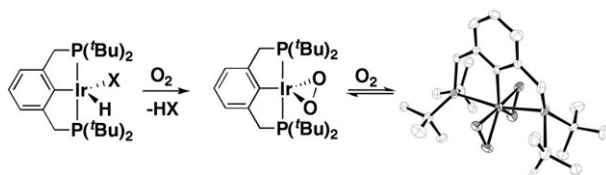


Catalytic properties of MIL-101

Antje Henschel, Kristina Gedrich, Ralph Kraehnert and Stefan Kaskel*

MIL-101, a chromium based metal–organic framework ($\text{Cr}_3\text{X}(\text{H}_2\text{O})_2\text{O}(\text{bdc})_3$; X = F, OH; bdc = benzene-1,4-dicarboxylate) has a very high activity in Lewis acid catalyzed reactions and is an excellent Pd support for the hydrogenation of alkenes.

4195

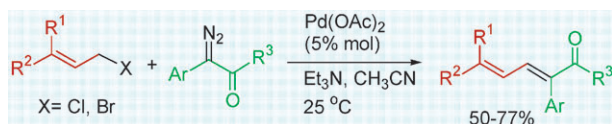


Reactions of iridium hydride pincer complexes with dioxygen: new dioxygen complexes and reversible O₂ binding

D. Bridget Williams, Werner Kaminsky, James M. Mayer* and Karen I. Goldberg*

Dioxygen promotes reductive elimination from pincer-iridium hydrocarbyl-hydride and dihydride complexes, leading to unusual mono- and bis-O₂ adducts.

4198

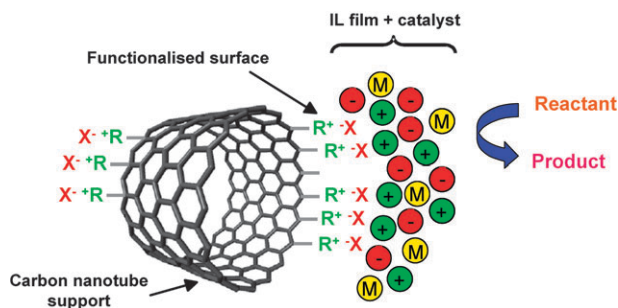


Palladium-catalyzed reaction of allyl halides with α -diazocarbonyl compounds

Shufeng Chen and Jianbo Wang*

The $\text{Pd}(\text{OAc})_2$ -catalyzed reaction between α -diazocarbonyl compounds and allyl bromides or chlorides leads to the formation of 1,3-diene derivatives.

4201



Supported ionic liquid phase catalysis on functionalized carbon nanotubes

Laura Rodríguez-Pérez, Emmanuelle Teuma, Andrea Falqui, Montserrat Gómez and Philippe Serp*

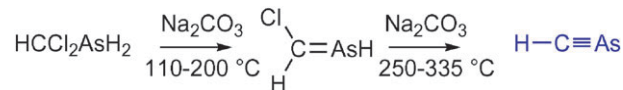
Highly active rhodium catalysts have been prepared by immobilization of an ionic liquid film on carbon nanotubes functionalized with imidazolium-based ionic moieties.

4204

Methyldiynearsine (HC≡As): synthesis and direct characterization by UV-photoelectron spectroscopy and mass spectrometry

Jean-Claude Guillemin,* Anna Chrostowska,*
Alain Dargelos, Thi Xuan Mai Nguyen, Alain Graciaa
and Pierre Guenot

Methyldiynearsine, the simplest arsaalkyne, has been synthesized and characterized in the gas phase by UV photoelectron spectroscopy and mass spectrometry.

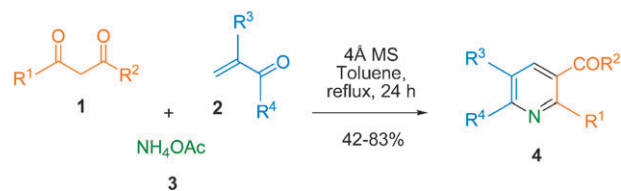


4207

Metal-free Michael addition initiated multicomponent oxidative cyclodehydration route to polysubstituted pyridines from 1,3-dicarbonyls

Frédéric Liéby-Muller, Christophe Allais,
Thierry Constantieux* and Jean Rodriguez*

A simple metal-free, step-economic and selective access to mono- and polycyclic pyridines is reported, *via* a 4 Å molecular sieves promoted domino three-component reaction from readily available 1,3-dicarbonyl systems.

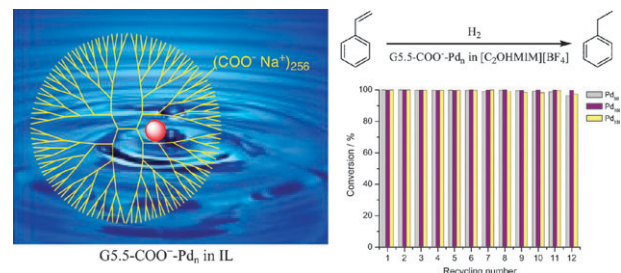


4210

Enhanced stability of charged dendrimer-encapsulated Pd nanoparticles in ionic liquids

Guangnan Ou, Li Xu, Biyan He and Youzhu Yuan*

Highly stable dendrimer-encapsulated Pd nanoparticles in ionic liquids were prepared for the first time by using charged PAMAM dendrimers as templates, which could maintain hydrogenation efficiency for up to at least 12 recycles.

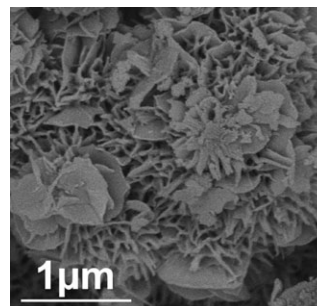


4213

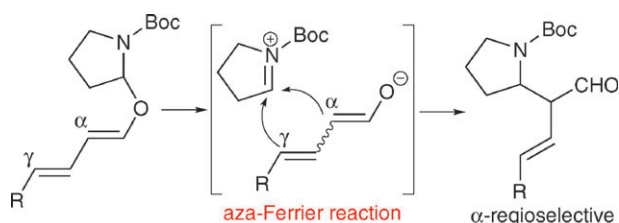
Facile approach to prepare loose-packed NiO nano-flakes materials for supercapacitors

Jun-Wei Lang, Ling-Bin Kong,* Wei-Jin Wu,
Yong-Chun Luo and Long Kang

The nickel oxide nano-flakes materials prepared by a facile approach have high specific capacitance and excellent cycle life for supercapacitors.



4216



Brønsted acid catalyzed regioselective aza-Ferrier reaction: a novel synthetic method for α -(*N*-Boc-2-pyrrolidinyl) aldehydes

Eiji Tayama,* Seijun Otoyama and Wataru Isaka

The Brønsted acid catalyzed aza-Ferrier reaction of *N*-Boc-2-(1,3-dienyloxy)pyrrolidines affords the corresponding α -(*N*-Boc-2-pyrrolidinyl) aldehydes in excellent yields with high α -regioselectivities.

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