

**Cover**

A snapshot from a molecular dynamics simulation of the mechanical unfolding of barnase; the termini were pulled at a 'constant speed' of $0.01 \text{ \AA} \cdot \text{ps}^{-1}$ using a time-dependent harmonic potential on the terminal C^α carbon atoms. This frame was taken 5 ns into the simulation, at which point the protein is already substantially unfolded.

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contents

FEATURE ARTICLE

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What can atomic force microscopy tell us about protein folding?

Robert B. Best and Jane Clarke

Atomic force spectroscopy has emerged as a new tool for studying protein folding, which complements existing techniques. What can this new technique tell us about the unfolding energy surface and how can it be used to determine detailed unfolding pathways under force?



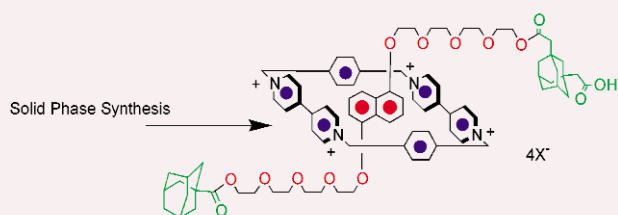
COMMUNICATIONS

194

Solid-phase template-directed synthesis of a [2]rotaxane using a solid-phase stopper

José A. Bravo, David Orain and Mark Bradley

The first synthesis of a rotaxane by solid phase chemistry has been achieved, using the resin bead as a 'Mega' stopper. One of the advantages of this methodology over traditional solution routes include the ability to use mass action to drive the chemistry, without complicating the purification process.

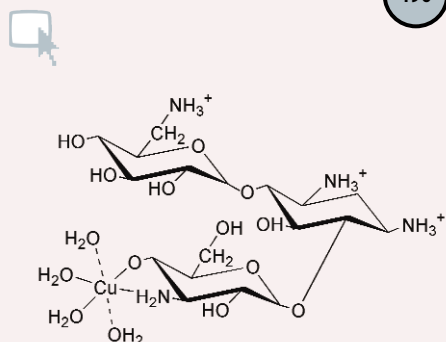


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***In vivo* cleavage of a target RNA by copper kanamycin A. Direct observation by a fluorescence assay**

Chun-An Chen and J. A. Cowan

A novel fluorescence assay to monitor *in vivo* cleavage chemistry of RNA target sequences has been established and used to demonstrate the activity of copper aminoglycoside mediated degradation of RNA in bacterial cells.

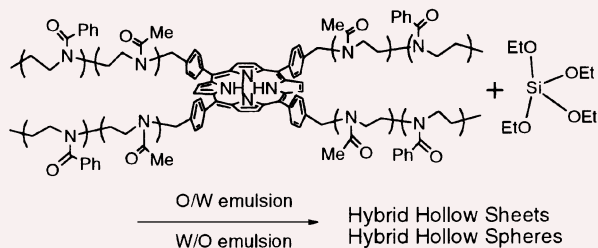


198

Silica–polyoxazoline hybrid with nanosized hollow enclosing porphyrin in hybrid walls

Ren-Hua Jin

A silica–polyoxazoline hybrid with nanosized hollows and porphyrin moieties was fabricated *via* an emulsion templated sol–gel route.

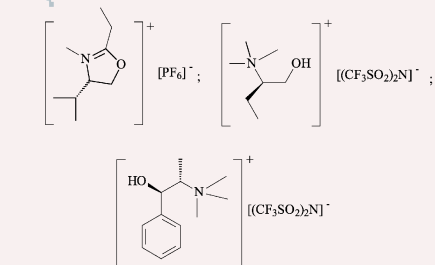


200

Synthesis and properties of ionic liquids derived from the ‘chiral pool’

Peter Wasserscheid, Andreas Bösmann and Carsten Bolm

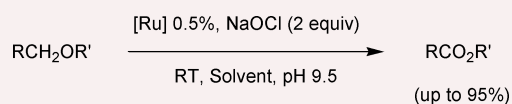
New chiral ionic liquids have been synthesised which are directly derived from the ‘chiral pool’ and therefore readily available in kg scale; NMR-measurements indicate that these liquids may be interesting solvents for enantioselective reactions and useful in chiral separation techniques.



202

Highly efficient use of NaOCl in the Ru-catalysed oxidation of aliphatic ethers to esters

Luca Gonsalvi, Isabel W. C. E. Arends and Roger A. Sheldon



R, R' = alkyl; Solvent = CH₂Cl₂, EtOAc

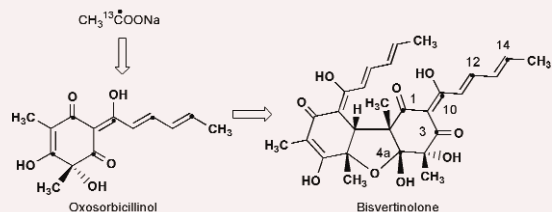
Selective oxidation of (cyclic) ethers using as little as 0.25% ruthenium catalyst together with the theoretical amount of NaOCl (2 equiv.) was achieved through careful pH control during the reaction.

204

The biosynthesis of bisvertinolone: evidence for oxosorbicillinol as a direct precursor

Naoki Abe, Tadaharu Arakawa and Akira Hirota

Biosynthetic incorporation of labeled sodium acetate into oxosorbicillinol in *Trichoderma* sp. USF-2690 suggests that oxosorbicillinol is derived from six acetate units, and subsequent bioconversion of the labeled oxosorbicillinol to bisvertinolone in the fermentation of the strain suggests that bisvertinolone is biosynthesized from oxosorbicillinol and sorbicillinol in a Michael-type reaction.

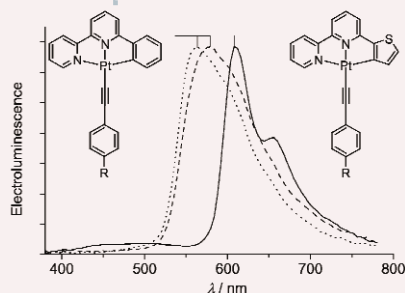


206

[(C[^]N[^]N[^])Pt(C≡C)_nR] (HC[^]N[^]N[^] = 6-aryl-2,2'-bipyridine, n = 1–4, R = aryl, SiMe₃) as a new class of light-emitting materials and their applications in electrophosphorescent devices

Wei Lu, Bao-Xiu Mi, Michael C. W. Chan, Zheng Hui, Nianyong Zhu, Shuit-Tong Lee and Chi-Ming Che

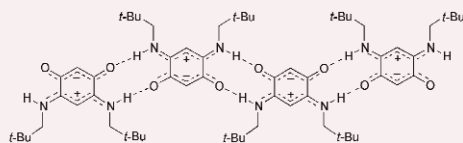
The σ-alkynyl ligand is the key to tunable, strong photophosphorescence and stability during vacuum deposition for these organometallic materials. Orange to red electrophosphorescence derived from the pictured emitters displays maximum luminance approaching 10000 cd m⁻².



208

Unprecedented zwitterion in quinonoid chemistry

Olivier Siri and Pierre Braunstein

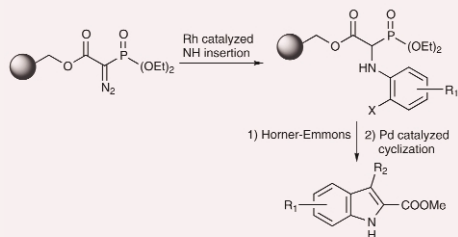


The first 12π -electron zwitterionic structure in quinonoid chemistry is described with the *N,N,O,O*-molecule shown in which the positive charge is π -delocalized between the nitrogen atoms and the negative charge between the oxygen atoms; depending on the crystallization solvent, a 1D-tape-like H-bonded network can be generated in the solid-state.

210

Immobilized α -diazophosphonoacetate as a versatile key precursor for palladium catalyzed indole synthesis on a polymer support

Kazuo Yamazaki and Yoshinori Kondo

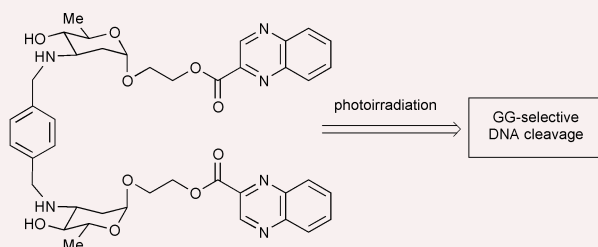


Rh(II)-catalyzed N–H insertion reaction of immobilized diazophosphonoacetate with 2-haloanilines followed by Horner–Emmons reaction gave immobilized enaminoesters, which were efficiently cyclized to indoles *via* intramolecular palladium catalyzed reaction on a polymer support.

212

Molecular design and evaluation of quinoxaline-carbohydrate hybrids as novel and efficient photo-induced GG-selective DNA cleaving agents

Kazunobu Toshima, Ryusuke Takano, Tomohiro Ozawa and Shuichi Matsumura

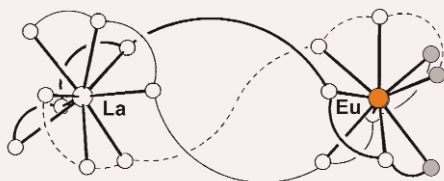


Quinoxaline, found in antitumor quinoxaline antibiotics, was found to cleave double stranded DNA at the 5' side guanine of 5'-GG-3' site on irradiation with long wavelength UV light without any additive; furthermore, a bis(quinoxaline-carbohydrate) hybrid system was very effective for DNA cleavage.

214

Discriminating between lanthanide ions: self-assembly of heterodimetallic triple-stranded helicates

Nicolas André, Rosario Scopelliti, Gérard Hopfgartner, Claude Piguet and Jean-Claude G. Bünzli

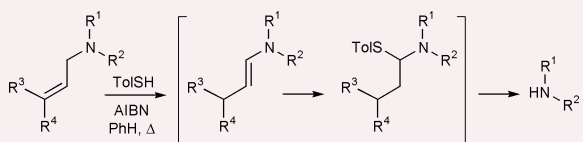


Discrimination between lanthanide ions with ionic radii differing by about 0.1 Å is achieved by means of a heteroditopic ligand which self-assembles with these ions to yield f–f heterodimetallic helicates.

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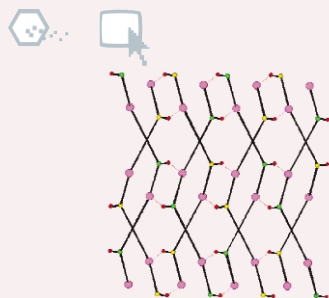
Tandem radical and non-radical reactions mediated with thiols—a new method of cleavage of allylic amines

Michèle P. Bertrand, Stéphanie Escoubet, Stéphane Gastaldi and Vitaliy I. Timokhin



ArS^\bullet promotes the isomerisation of allylic amines to enamines. Subsequent cleavage occurs *via* a thioaminal intermediate.

218



Design of 3-D coordination networks: topology and metrics

Sylvie Ferlay, Stéphanie Koenig, Mir Wais Hosseini, Jérôme Pansanel, André De Cian and Nathalie Kyritsakas

A doubly interpenetrated pseudo-diamondoid 3-D network was obtained using Ag^+ cation and a hetero tetradentate tecton with C_{3v} symmetry bearing three CN and one OH group.

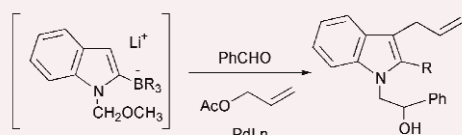
220



A novel C–C bond formation reaction with 1-methoxymethylindolylborate

Minoru Ishikura, Hiromi Kato and Nobuyuki Ohnuki

A novel cascade of intramolecular alkyl–boryl migration in indolylborate was applied for a ‘one-pot’ construction of tri-substituted indoles.



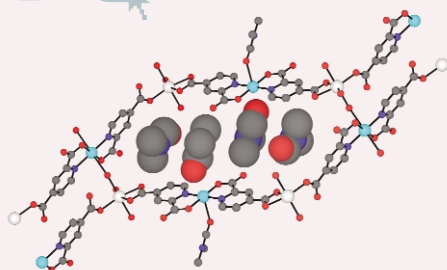
222



New microporous coordination polymer affording guest-coordination sites at channel walls

Shin-ichiro Noro, Susumu Kitagawa, Masahiro Yamashita and Tatsuo Wada

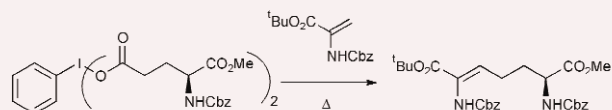
By utilizing a novel synthetic scenario (two-step self-assembly), a microporous coordination polymer with guest-coordination sites at channel walls was rationally synthesized.



224

Conjugate addition of radicals generated from diacyloxyiodobenzenes to dehydroamino acid derivatives; a synthesis of diaminopimelic acid analogues

Andrew Sutherland and John C. Vederas



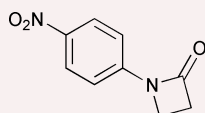
Radical decomposition of bis((2*S*)-*N*-benzyloxycarbonyl-2-aminopentan-5-carboxy-1-methyl ester)iodobenzene followed by decarboxylation and subsequent conjugate addition with a series of selectively protected dehydroamino acids leads to new analogues of diaminopimelic acid.

226

Polyclonal antibody-catalysed hydrolysis of a β -lactam

Elizabeth L. Ostler, Marina Resmini, Guillaume Boucher, Nickolas Romanov, Keith Brocklehurst and Gerard Gallacher

We report the first example of antibody-catalysed hydrolysis of a β -lactam where the antibodies were generated by a simple transition-state analogue; in this example the antibodies are polyclonal.



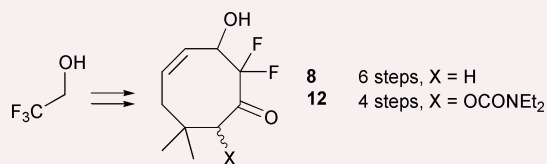
228



Rapid assembly of highly-functionalised difluorinated cyclooctenones *via* ring-closing metathesis

Benson M. Kariuki, W. Martin Owton, Jonathan M. Percy, Stéphane Pintat, Clive A. Smith, Neil S. Spencer, Andrew C. Thomas and Martin Watson

Metallated difluoroenol acetal and carbamate chemistry allowed the rapid assembly of highly-functionalised difluorinated dienes which underwent ring-closing metathesis to novel difluorinated cyclooctenones.



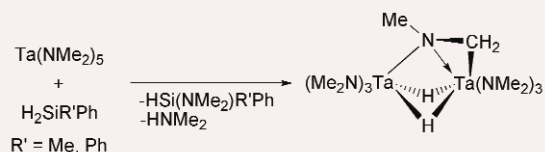
230



Direct observation of η^2 -imine formation through β -H abstraction between amide ligands. Neutron and X-ray diffraction structure of a dihydride imine ditantalum complex

Hu Cai, Tianniu Chen, Xiaoping Wang, Arthur J. Schultz, Thomas F. Koetzle and Ziling Xue*

Reactions of Ta(NMe₂)₅ with D₂SiR'Ph were found to give a dideuteride η^2 -imine complex (Me₂N)₃Ta(μ -D)₂(μ -N- η^2 -N,C-CH₂NMe)Ta(NMe₂)₃ through C–H activation of an amide ligand *via* β -H abstraction.



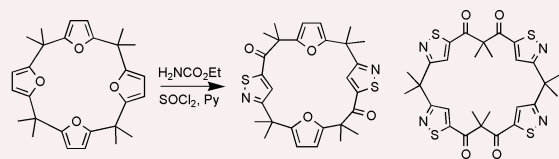
232



Direct conversion of macrocyclic furans into macrocyclic isothiazoles

Jérôme Guillard, Otto Meth-Cohn, Charles W. Rees, Andrew J. P. White and David J. Williams

The first calixhetarenes with more than one heteroatom in the constituent rings, namely macrocyclic isothiazoles, have been prepared by direct one-pot conversion of preformed calixfurans.



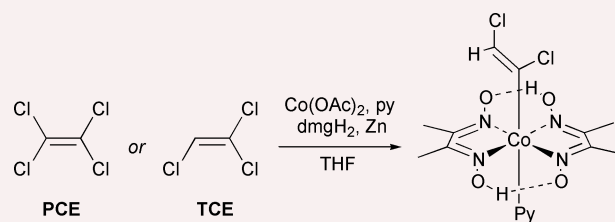
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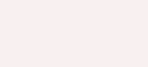
Synthesis of (chlorovinyl)cobaloxime complexes, model complexes of proposed intermediates in the B₁₂-catalyzed dehalogenation of chlorinated ethylenes

Autumn E. Rich, Angela D. DeGreeff and Kristopher McNeill

The synthesis and preliminary reactivity studies of (chlorovinyl)cobalt complexes is reported. The method of synthesis and the reactions of these complexes with reducing agents support the intermediacy of such species in the dehalogenation of chlorinated ethylenes by B₁₂.



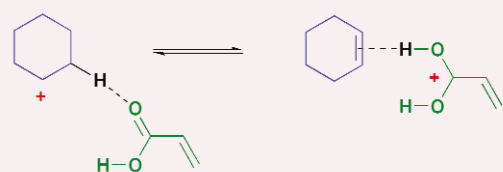
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Ion-neutral complexes formation and 1,3-proton transfer in the chemical ionization of alkylcyclohexyl benzoates

Chagit Denekamp and Amnon Stanger

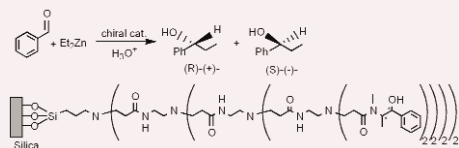
CI, CID, labelling experiments and DFT calculations are used for the elucidation of the mechanism for the decomposition of cyclohexyl benzoates, which proceeds through 1,3-H shift and two equilibrating ion-neutral complexes.



238

Dendritic chiral auxiliaries on silica: a new heterogeneous catalyst for enantioselective addition of diethylzinc to benzaldehyde

Young-Min Chung and Hyun-Ku Rhee

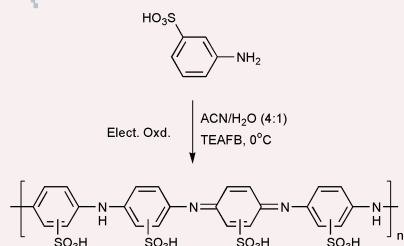


Silica supported dendritic chiral auxiliaries are effective for the enantioselective addition of diethylzinc to benzaldehyde.

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Electrochemical synthesis of fully sulfonated *n*-dopable polyaniline: poly(metanillic acid)

K. Krishnamoorthy, A. Q. Contractor and Anil Kumar

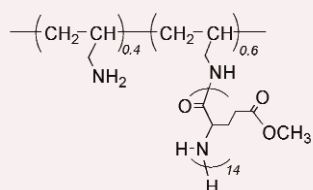


Electrochemical homopolymerization of metanillic acid has been achieved for the first time using a 4:1 acetonitrile–water mixture to get 100% sulfonated polyaniline which is soluble in both organic and aqueous solvents, electrically conducting, and is *n*-dopable.

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pH-Regulated formation of amyloid-like β -sheet assemblies from polyglutamate grafted polyallylamine

Tomoyuki Koga, Kazuhiro Taguchi, Takatoshi Kinoshita and Masahiro Higuchi

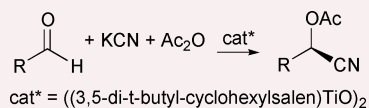


A novel artificial protein with simple primary structure, poly(γ -methyl-L-glutamate)-grafted polyallylamine, has been prepared and the resultant peptide has shown a unique property of pH-regulated conformation and morphology.

244

Catalytic asymmetric synthesis of *O*-acetyl cyanohydrins from KCN, Ac₂O and aldehydes

Yuri N. Belokon, Andrey V. Gutnov, Margarita A. Moskalenko, Lidia V. Yashkina, Denis E. Lesovoy, Nicolai S. Ikonnikov, Vladimir S. Larichev and Michael North

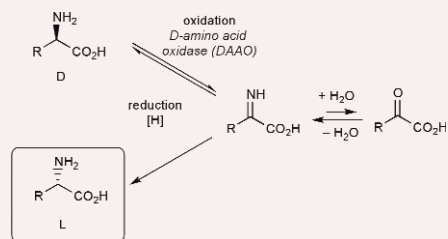


The first asymmetric, catalytic synthesis of cyanohydrin derivatives using potassium cyanide as an inexpensive and non-volatile cyanide source is reported. Reactions are carried out in the presence of acetic anhydride to generate cyanohydrin acetates with up to 93% enantiomeric excess.

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Deracemisation and stereoinversion of α -amino acids using D-amino acid oxidase and hydride reducing agents

Timothy M. Beard and Nicholas J. Turner

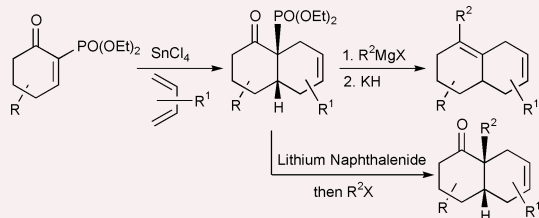


The deracemisation and stereoinversion of both cyclic and acyclic DL- α -amino acids, using porcine kidney D-amino acid oxidase (DAAO) and a hydride reducing agent (NaCNBH₃–NaBH₄), has been investigated.

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Diels–Alder chemistry of 2-diethoxyphosphinylcyclohex-2-enones. A new approach to complex phosphonates and synthetic applications of the β -keto phosphonate system

Chiung-Fang Chien, Jen-Dar Wu, Tai Wei Ly, Kak-Shan Shia and Hsing-Jang Liu

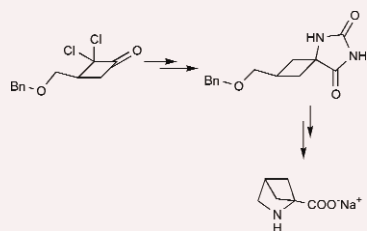


A Diels–Alder reaction provides a direct access to some phosphonate-containing polycyclic compounds. The phosphonate group can also act as a handle for further reductive alkylation or intramolecular Wadsworth–Horner–Emmons process.

250

A new and short method for the synthesis of 2,4-methanoproline

Thomas Rammeloo and Christian V. Stevens

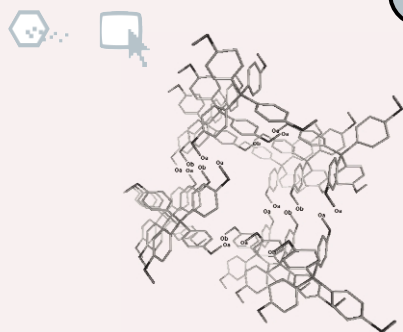


2,4-Methanoproline, a supposed non-proteinogenic anti-feedant, was synthesised in 5 steps starting from allyl benzyl ether in 10% overall yield with an intramolecular nucleophilic substitution as the key step for the formation of the bicyclic skeleton.

252

Structure and exchange in silicon-linked tetradicals

Yi Liao, Martha Baskett, Paul M. Lahti and Fernando Palacio

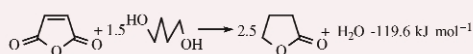


The EPR spectroscopy, crystallography, and magnetic susceptibility of tetrakis(*N*-oxyl-2,2,6,6-tetramethylpiperidin-4-yl)silane and tetrakis(4-*N*-*tert*-butyl-*N*-aminoxylphenyl)silane show that silicon acts as a weak intramolecular exchange linker for polynitroxides, although both tetradicals show onset of inter-spin exchange at reduced temperatures.

254

A novel route for synthesis of γ -butyrolactone through the coupling of hydrogenation and dehydrogenation

Yu-Lei Zhu, Hong-Wei Xiang, Gui-Sheng Wu, Liang Bai and Yong-Wang Li

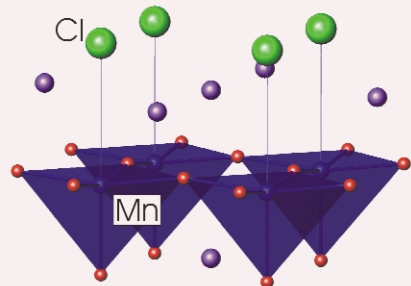


A coupling process of the hydrogenation of maleic anhydride and the dehydrogenation of 1,4-butanediol has been invented for the synthesis of γ -butyrolactone over a Cu–Zn catalyst, realizing optimal hydrogen utilization and better energy efficiency.

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New layered manganese oxide halides

Christopher S. Knee and Mark T. Weller



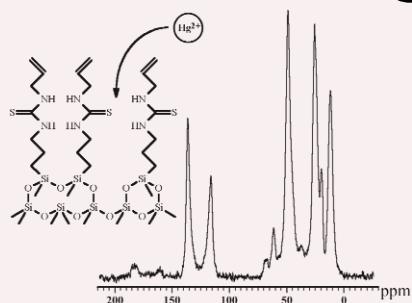
The synthesis and structures of two novel manganese based oxide chlorides are reported. These materials belong to a large family of manganese(III/IV) oxide halides and have potential interest with respect to magnetoresistance effects.

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1-Allyl-3-propylthiourea modified mesoporous silica for mercury removal

Valentyn Antochshuk and Mietek Jaroniec

1-Allyl-3-propylthiourea modified mesoporous silica has high adsorption capacity for mercury ions and its regeneration can be accomplished by washing with 10% thiourea in aqueous 0.05 M HCl.

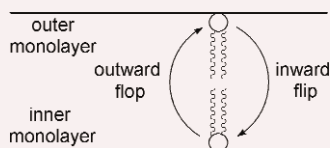


260

Selective phosphatidylethanolamine translocation across vesicle membranes using synthetic translocases

J. Middleton Boon, Rameshwer Shukla, Bradley D. Smith, Giulia Licini and Paolo Scrimin

Two sulfonamide derivatives of tris(aminoethyl)amine selectively facilitate the translocation of a fluorescent phospholipid probe containing the phosphoethanolamine head-group across vesicle membranes.

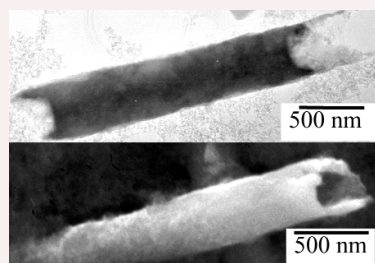


262

A simple route towards tubular ZnO

Jun Zhang, Lingdong Sun, Chunsheng Liao and Chunhua Yan

A simple route through thermal treatment of $\text{Zn}(\text{NH}_3)_4^{2+}$ precursor in ethanol is presented to fabricate tubular ZnO, which exhibits strong UV emission and has potential applications.

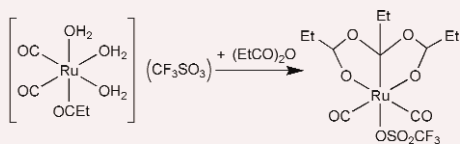


264

The acylation of an acyl complex resulting in a labile OCO tridentate ligand

Tiziana Funaioli, Giulia Falchi, Fabio Marchetti and Giuseppe Fachinetti

The reaction of propionic anhydride with $[\text{fac-Ru}(\text{C}(\text{O})\text{Et})(\text{CO})_2(\text{H}_2\text{O})_3][\text{CF}_3\text{SO}_3]$ produces a new propylidyn dipropionato group, which behaves as a tridentate ligand giving the neutral complex $\text{Ru}\{\text{C}(\text{Et})(\text{OC}(\text{O})\text{Et})_2\}(\text{CO})_2(\text{CF}_3\text{SO}_3)$.

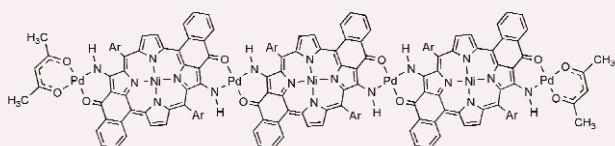


266

A modular approach to porphyrin oligomers using metal ions as connectors

S. Richeter, C. Jeandon, R. Ruppert and H. J. Callot

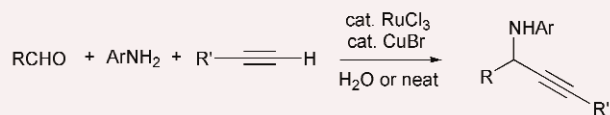
Porphyrins bearing two external coordination sites allowed the stepwise preparation of polymeric oligomers connected by metal centers.



268

Highly efficient Grignard-type imine additions *via* C–H activation in water and under solvent-free conditions

Chao-Jun Li and Chunmei Wei

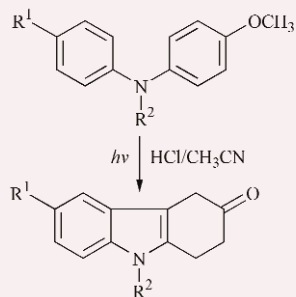


A highly effective Cu–Ru catalyzed addition of terminal alkynes to imines *via* C–H activation has been achieved in water or under solvent-free conditions.

270

Novel photoreaction of *N*-alkyl(*p*-methoxyphenyl)arylamines assisted by protic acids

Jinn-Hsuan Ho and Tong-Ing Ho

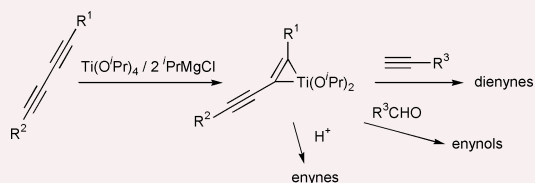


A novel photochemical transformation from *N*-alkyl(*p*-methoxyphenyl)arylamines to 1,2,4-trihydro(4*aH*)-carbazol-3-ones is reported with the assistance of protonation at the dihydrocarbazole intermediate followed by sequential formal [1,5]hydrogen, [1,3]hydrogen shifts and proton assisted hydrolysis.

272

Site-selective mono-titanation of conjugated diynes with a Ti(II) alkoxide reagent. Concise preparation of stereo-defined enynes and dienynes

Christophe Delas, Hirokazu Urabe and Fumie Sato

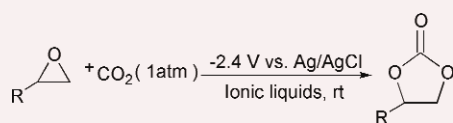


Conjugated diynes underwent selective mono-titanation with a Ti(II) reagent to give 1 : 1 diyne–titanium alkoxide complexes, which reacted with proton, aldehyde, and another acetylene to give stereo-defined enynes, enynols, and dienynes.

274

Electrochemical activation of carbon dioxide in ionic liquid: synthesis of cyclic carbonates at mild reaction conditions

Hongzhou Yang, Yanlong Gu, Youquan Deng and Feng Shi

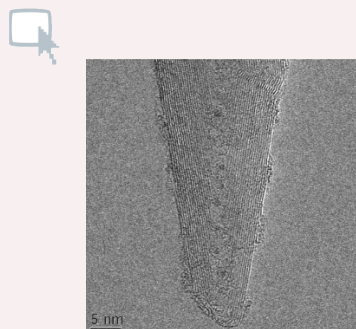


Electrocatalytic cycloaddition of carbon dioxide to epoxides in room temperature ionic liquids as reaction media without any additional supporting electrolyte and catalyst could be conducted with high to excellent performances under mild conditions.

276

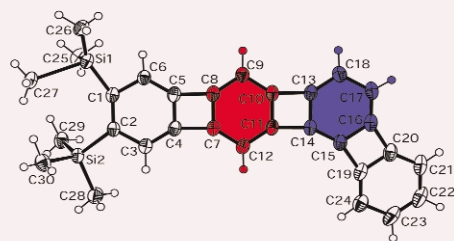
Bimetallic nanoparticles aligned at the tips of carbon nanotubes

Sophie Hermans, Jeremy Sloan, Douglas S. Shephard, Brian F. G. Johnson and Malcolm L. H. Green



Cluster-derived bimetallic nanoparticles have been deposited onto multi-wall carbon nanotubes and shown to be generally homogeneously dispersed, of uniform small sizes, of the same composition as the starting mixed-metal clusters, and to have a tendency to align at the tips of the tubules.

278

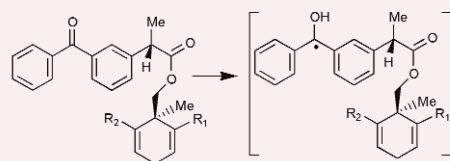


Synthesis of bent [4]phenylene (cyclobuta[1,2-*a*:3,4-*b'*]bisbiphenylene) and structure of a bis(trimethylsilyl) derivative: the last [4]phenylene isomer

Dennis T.-Y. Bong, Lionel Gentric, Daniel Holmes, Adam J. Matzger, Frank Scherhag and K. Peter C. Vollhardt

The effect of benzocyclobutadienofusion of linear and angular [3]phenylene, respectively, as manifested in the title compound, results in increased diatropism and reduced reactivity of the central ring of the former (red), but decreased diatropism and increased reactivity of that of the latter (blue).

280



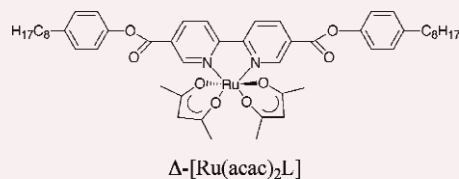
- 1 $R_1 = \text{CH}_3$; $R_2 = \text{H}$
2 $R_1 = \text{H}$; $R_2 = \text{CH}_3$

Stereoselective intramolecular hydrogen abstraction by a chiral benzophenone derivative

Miguel A. Miranda, Luis A. Martínez, Abdelouahid Samadi, Francisco Boscá and Isabel M. Morera

Using specifically designed bichromophores containing both a chiral benzophenone and a chiral hydrogen source it is shown here that photochemical hydrogen abstraction can be a stereoselective process.

282

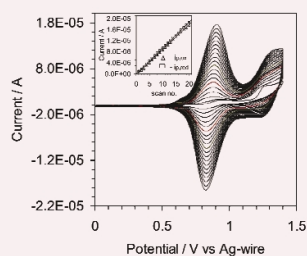


The effect of $\Delta\Lambda$ chirality on molecular organization in two-dimensional films of a Ru(II) complex with a mesogenic ligand

Kentaro Okamoto, Yuki Matsuoka, Noboru Wakabayashi, Akihiko Yamagishi and Naomi Hoshino

An amphiphilic, 6-coordinate, and Δ,Λ -isomeric Ru(II) complex has been synthesized with a mesogenic ligand. A remarkable difference has been noticed for the monolayer state between a racemic mixture and the Δ -isomer.

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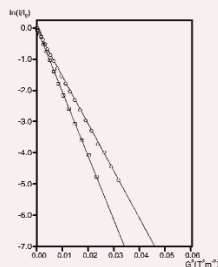


A rod-like polymer containing {Ru(terpy)}₂ units prepared by electrochemical coupling of pendant thienyl moieties

Johan Hjelm, Edwin C. Constable, Egbert Figgemeier, Anders Hagfeldt, Robyn Handel, Catherine E. Housecroft, Emad Mukhtar and Emma Schofield

Electropolymerisation of a ruthenium complex bearing a pendant thienyl functionality leads to films or a high nuclearity ruthenapolymer

286



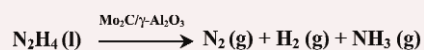
Selective anion effects in chiral complexes of iridium *via* diffusion and HOESY data: relevance to catalysis

Daniela Drago, Paul S. Pregosin and Andreas Pfaltz

PGSE diffusion measurements help to define how anions interact with cations in catalytically relevant Ir-complexes. A combined PGSE/HOESY approach is recommended.

A novel catalyst for hydrazine decomposition: molybdenum carbide supported on γ -Al₂O₃

Xiaowei Chen, Tao Zhang, Pinliang Ying, Mingyuan Zheng, Weicheng Wu, Liangen Xia, Tao Li, Xiaodong Wang and Can Li



An alumina-supported Mo₂C catalyst is found to be as active as a conventionally used Ir/ γ -Al₂O₃ catalyst for catalytic decomposition of hydrazine tested in a monopropellant thruster for the first time.

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