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IN THIS ISSUE

ISSN 1359-7345 CODEN CHCOFS (7) 641–764 (2007)



Cover

See D. Venkataraman *et al.*, page 692. The illustration shows the packing of naphthyl ether and naphthalimide moieties into segregated stacks due to mutually phobic side-chain interactions. Image reproduced by permission of Travis L. Benanti, Pranorm Saejueng and D. Venkataraman, from *Chem. Commun.*, 2007, 692.



Inside cover

See Nadia C. Mösch-Zanetti *et al.*, page 701. Molecular oxygen is activated by a molybdenum(IV) compound. The kinetics of its formation was monitored by UV/VIS spectrometry, while the Uhrnturm of Graz measured the time. Image reproduced by permission of Ganna Lyashenko, Gerald Saischek, Aritra Pal, Regine Herbst-Irmer and Nadia C. Mösch-Zanetti, from *Chem. Commun.*, 2007, 701.

CHEMICAL SCIENCE

C9

Drawing together the research highlights and news from all RSC publications, *Chemical Science* provides a 'snapshot' of the latest developments across the chemical sciences showcasing newsworthy articles, as well as the most significant scientific advances.

Chemical Science

February 2007/Volume 4/Issue 2

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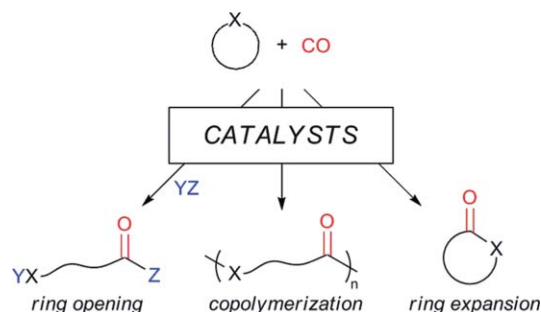
FEATURE ARTICLES

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Carbonylation of heterocycles by homogeneous catalysts

Tamara L. Church, Yutan D. Y. L. Getzler, Christopher M. Byrne and Geoffrey W. Coates*

This article summarizes the recent developments (particularly the uses of homogeneous organometallic catalysts) in ring-opening carbonylations, ring-opening carbonylative polymerizations and ring-expansion carbonylations of heterocycles, particularly epoxides, aziridines, lactones and oxazolines.



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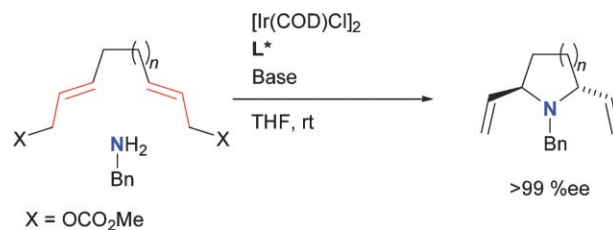
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Iridium-catalysed asymmetric allylic substitutions

Günter Helmchen,* Axel Dahnz, Pierre Dübon, Mathias Schelwies and Robert Weihofen

Ir-catalysed allylic substitutions can be carried out with a variety of C-, N- and O-nucleophiles to give branched substitution products with high degrees of regio- and enantioselectivity.



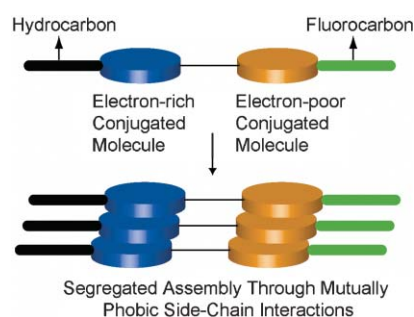
COMMUNICATIONS

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Segregated assemblies in bridged electron-rich and electron-poor π -conjugated moieties

Travis L. Benanti, Pranorm Saejueng and D. Venkataraman*

We report a general strategy for the spontaneous segregation of electron-rich and electron-poor π -conjugated molecules using mutually phobic aliphatic fluorocarbon–hydrocarbon interactions.

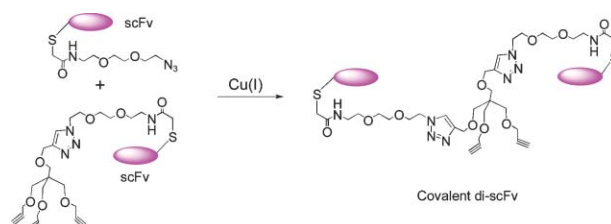


695

Construction of di-scFv through a trivalent alkyne–azide 1,3-dipolar cycloaddition

Arutselvan Natarajan, Wenjun Du, Cheng-Yi Xiong, Gerald L. DeNardo, Sally J. DeNardo* and Jacquelyn Gervay-Hague*

Site specific ligation of scFv was achieved using 1,3-dipolar cycloaddition. Incorporation of a trialkyne linker was critical for efficient conjugation.

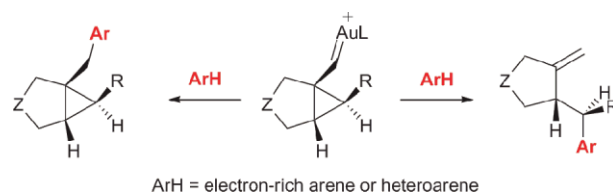


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Gold(I)-catalysed arylation of 1,6-enynes: different site reactivity of cyclopropyl gold carbenes

Cateljine H. M. Amijs, Catalina Ferrer and Antonio M. Echavarren*

Gold(I)-catalysed addition of electron-rich arenes and heteroarenes to 1,6-enynes gives two different types of products by reaction of the intermediate cyclopropyl gold carbenes at the cyclopropane or at the carbene.





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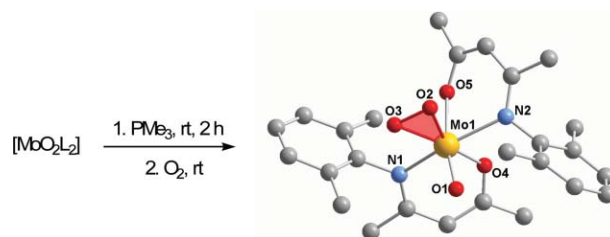
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Molecular oxygen activation by a molybdenum(IV) monooxo bis(β -ketiminato) complex

Ganna Lyashenko, Gerald Saischek, Aritra Pal, Regine Herbst-Irmer and Nadia C. Mösch-Zanetti*

Molybdenum(IV) monooxo compound that contains bis(β -ketiminato) ligands formed by oxygen atom transfer activates molecular oxygen forming a molybdenum(VI) monooxo peroxy complex. This represents a new entry into molybdenum peroxy derivatives.

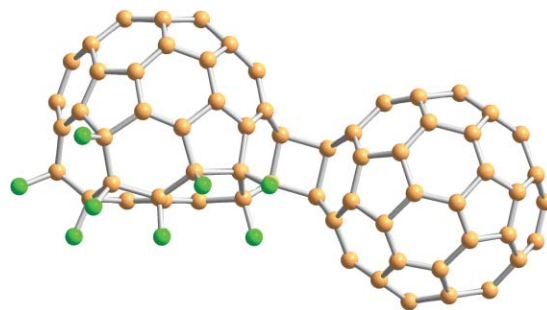


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The former “C₆₀F₁₆” is actually a double-caged adduct: (C₆₀F₁₆)(C₆₀)

Alexey A. Goryunkov, Ilya N. Ioffe, Pavel A. Khavrel, Stanislav M. Avdoshenko, Vitaly Yu. Markov, Z. Mazej, Lev N. Sidorov and Sergey I. Troyanov*

X-ray diffraction study of the substance originally believed to be C₆₀F₁₆ reveals a double-caged structure, (C₆₀F₁₆)(C₆₀), a finding supported by MALDI mass spectra and theoretical calculations.

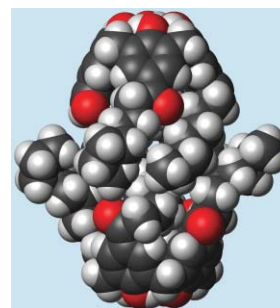


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A molecular turnstile in *para*-octanoyl calix[4]arene nanocapsules

Gennady S. Ananchenko,* Konstantin A. Udachin, Michaela Pojarova, Said Jebors, Anthony W. Coleman and John A. Ripmeester

The thermal treatment of different inclusion complexes of *para*-octanoyl calix[4]arene leads to the formation of a guest-free van der Waals nanocapsular framework possessing a remarkable stability caused by the high mobility of alkanoyl arms.

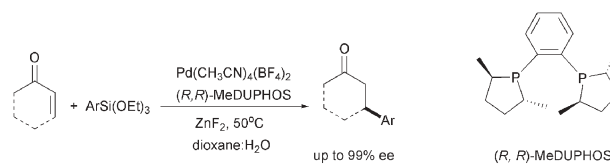


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Enantioselective palladium-catalysed conjugate addition of arylsiloxanes

Francesca Gini, Bart Hessen, Ben L. Feringa and Adriaan J. Minnaard*

The complex formed from Pd(CH₃CN)₄(BF₄)₂ and (*R,R*)-MeDUPHOS is a highly enantioselective catalyst for the asymmetric conjugate addition of aryltriethylsiloxanes to a variety of unsaturated ketones, lactones and lactams.



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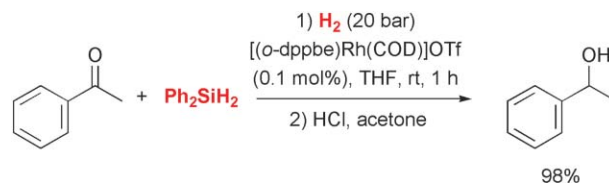
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From inconsistent results to high speed hydrosilylation

Virginie Comte,* Cédric Balan, Pierre Le Gendre* and Claude Moise

The hydrosilylation of various ketones in the presence of cationic rhodium catalysts containing diphosphine ligands proceeds much faster under dihydrogen pressure than under an inert atmosphere.

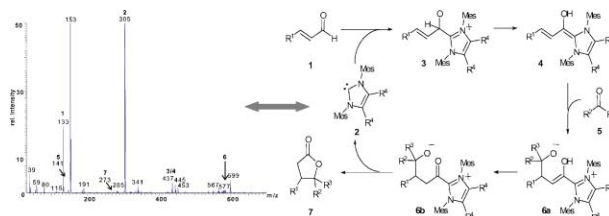


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Investigating organocatalytic reactions: mass spectrometric studies of a conjugate *umpolung* reaction

Wolfgang Schrader,* Peni Purwa Handayani, Christian Burstein and Frank Glorius

An organocatalyzed conjugate *umpolung* reaction has been studied in detail using electrospray mass spectrometry, which allows support of the catalytic cycle by intercepting intermediates.

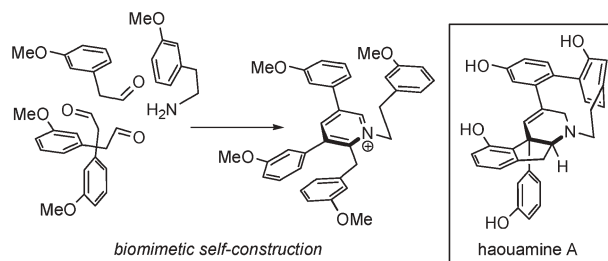


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Biogenetic hypothesis and first steps towards a biomimetic synthesis of haouamines

Edmond Gravel, Erwan Poupon* and Reynald Hocquemiller

Following a plausible biogenetic pathway, advanced intermediates for the total synthesis of haouamines have been prepared. This is the first biomimetic approach reported for this class of complex alkaloids.

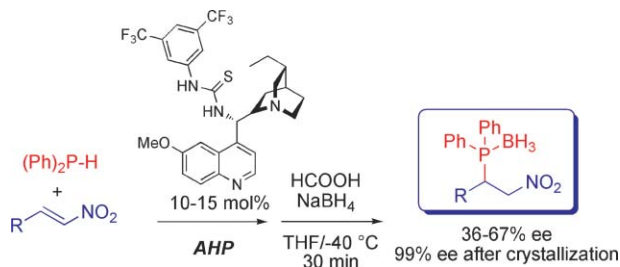


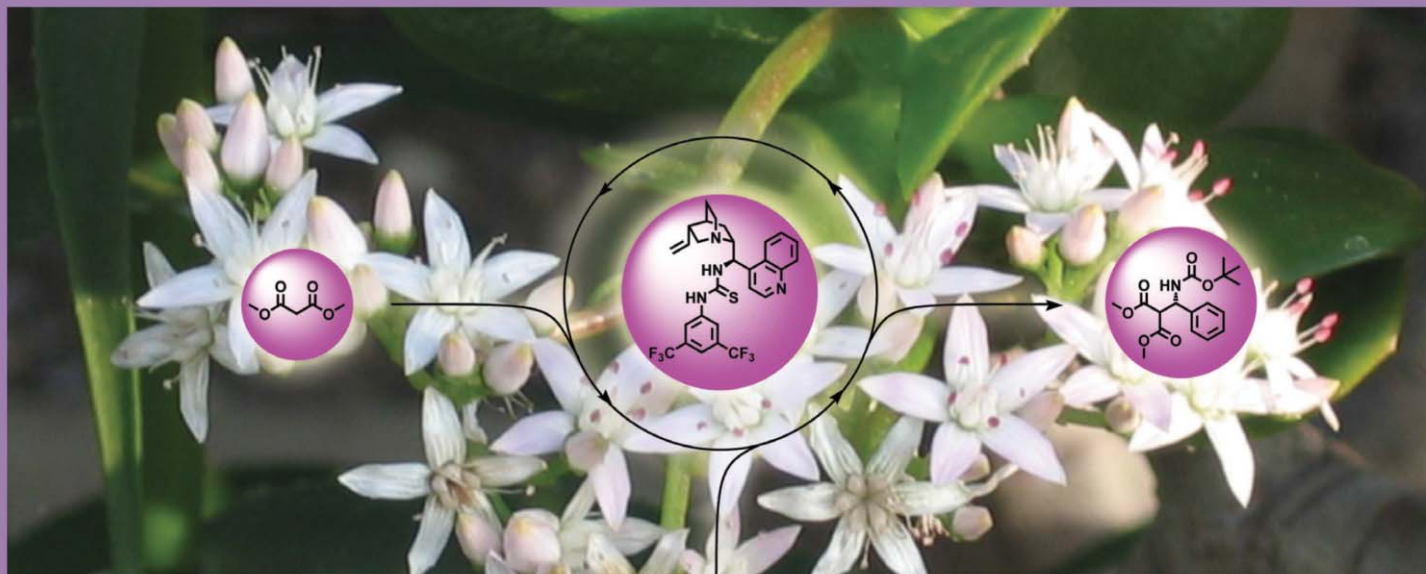
722

Organocatalytic asymmetric hydrophosphination of nitroalkenes

Giuseppe Bartoli, Marcella Bosco, Armando Carlone, Manuela Locatelli, Andrea Mazzanti, Letizia Sambri and Paolo Melchiorre*

The first direct asymmetric hydrophosphination (AHP) of nitroalkenes, catalyzed by a bifunctional *Cinchona* alkaloid derivative, has been accomplished.





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Professor Andrew Evans US Associate Editor for organic chemistry

Professor Evans currently holds the Heath Harrison Chair of Organic Chemistry at the University of Liverpool, having recently moved from Indiana University. His research group is engaged in the development of new diastereo- and enantioselective metal-catalyzed reactions that facilitate the expeditious total synthesis of complex pharmacologically important natural products.

Happy to receive papers on important developments in organic chemistry, Professor Evans can be contacted via chemcomm@indiana.edu



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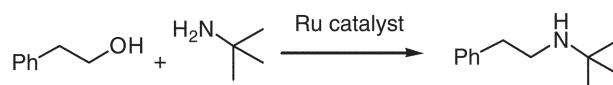
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Ruthenium catalysed *N*-alkylation of amines with alcohols

Malai Haniti S. A. Hamid and Jonathan M. J. Williams*

Borrowing hydrogen in the *N*-alkylation of amines has been achieved using a ruthenium based catalyst.

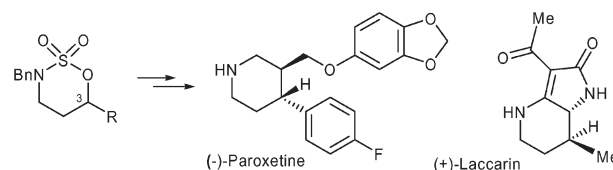


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Stereospecific construction of substituted piperidines. Synthesis of (–)-paroxetine and (+)-laccarin

John F. Bower, Thomas Riis-Johannessen, Peter Szeto, Andrew J. Whitehead and Timothy Gallagher*

Short and efficient enantioselective syntheses of (–)-paroxetine and (+)-laccarin are described based on the highly stereospecific cleavage of C(3)-substituted 1,3-cyclic sulfamidates.

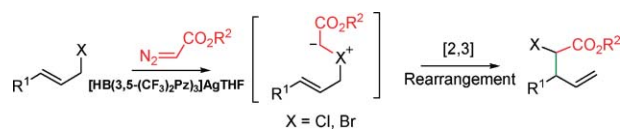


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Silver-catalyzed [2,3]-rearrangement of halonium ylides derived from allyl and propargyl halides and alkyl diazoacetates

Pasupathy Krishnamoorthy, R. Greg Browning, Shreeyukta Singh, Rasapalli Sivappa, Carl J. Lovely* and H. V. Rasika Dias*

A silver(I) complex derived from a polyfluorinated tris(pyrazolyl)borate effectively catalyzes carbene transfer to allylic and propargylic halides, leading to the formation of α -haloacetate derivatives.

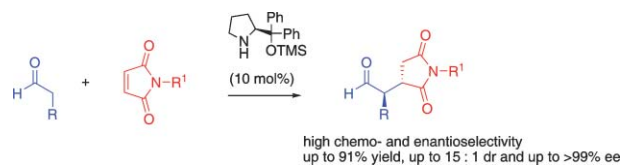


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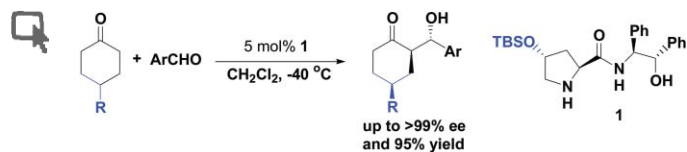
Organocatalytic enantioselective conjugate addition of aldehydes to maleimides

Gui-Ling Zhao, Yongmei Xu, Henrik Sundén, Lars Eriksson, Mahmoud Sayah and Armando Córdoba*

The first highly enantioselective organocatalytic conjugate addition of unmodified aldehydes to maleimides is presented. The novel reaction gives access to α -substituted succinimides with up to 15 : 1 dr and generally 97 \rightarrow 99% ee.



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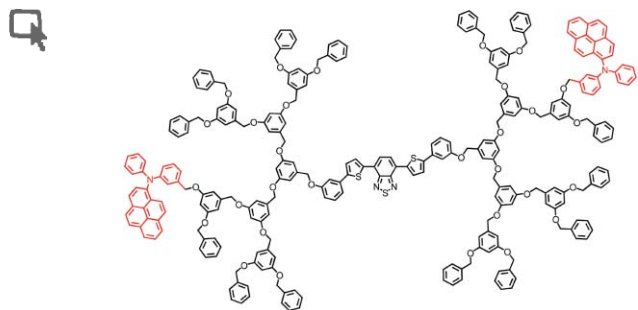


Enantioselective desymmetrization of prochiral cyclohexanone derivatives *via* the organocatalytic direct aldol reaction

Jun Jiang, Long He, Shi-Wei Luo, Lin-Feng Cun and Liu-Zhu Gong*

Asymmetric desymmetrization of 4-substituted cyclohexanones using proline amide-catalyzed direct aldol reaction afforded β -hydroxyketones with three stereogenic centers in high enantioselectivities of up to >99% ee.

739

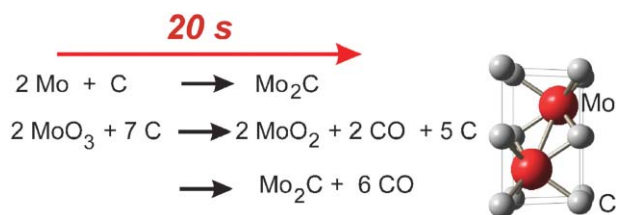


Probing the periphery of dendrimers by heterogeneous electron transfer

K. Krishnamoorthy, Raghunath Reddy Dasari, Arpornrat Nantalaksakul and S. Thayumanavan*

Generation-dependence in the accessibility to the peripheral electroactive functionalities in dendrimers has been studied by comparing two different classes of dendrimers with linear molecules.

742

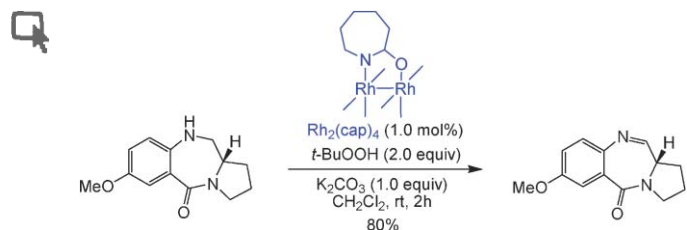


Ultra-rapid processing of refractory carbides; 20 s synthesis of molybdenum carbide, Mo₂C

Simon R. Vallance, Sam Kingman* and Duncan H. Gregory*

Ultra-rapid microwave reactions yield refractory carbides such as superconducting Mo₂C in 20 s; dielectric properties of the components mediate the reaction process.

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Oxidation of secondary amines catalyzed by dirhodium caprolactamate

Hojae Choi and Michael P. Doyle*

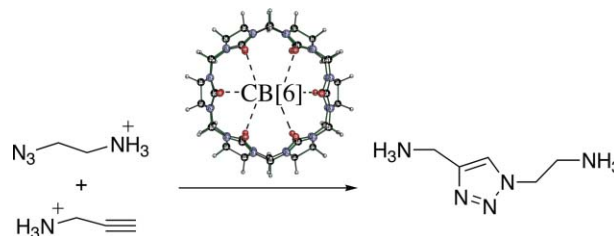
Secondary amines are converted to their corresponding imines with high chemo- and regioselectivity in oxidation reactions of *tert*-butyl hydroperoxide catalyzed by dirhodium caprolactamate [Rh₂(cap)₄].

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A theoretical analysis of a classic example of supramolecular catalysis

Peter Carlqvist and Feliu Maseras*

Computational chemistry is applied to the study of the 1,3-dipolar cycloaddition between an azide and an alkyne inside the macrocycle cucurbit[6]uril.

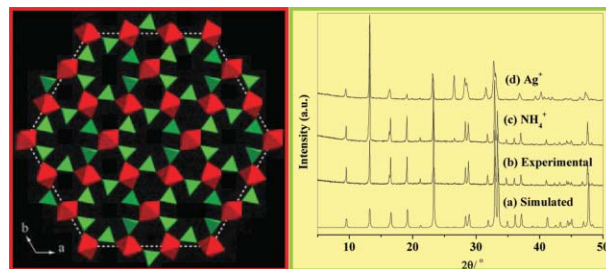


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A (4,2)-connected niobium phosphate $\text{Na}_{13}\text{Nb}_3\text{P}_6\text{O}_{28}\text{F}_2$ with a unique 6.12-net layer: Synthesis, structure and ion exchange properties

Guang-Zhen Liu, Shou-Tian Zheng and Guo-Yu Yang*

A two-step hydrothermal process was employed to make a very open (4,2)-connected layered niobium phosphate, $\text{Na}_{13}\text{Nb}_3\text{P}_6\text{O}_{28}\text{F}_2$. The sheet consists of alternating 4-connected niobium octahedra and 2-connected phosphorus tetrahedra, forming a unique 6.12-net that has never previously been found in metal phosphates.

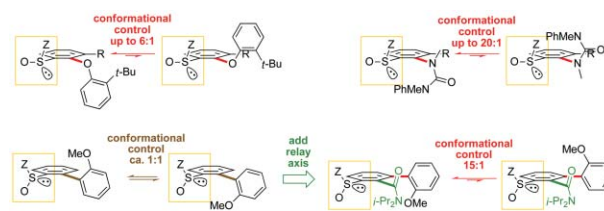


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Achieving conformational control over C–C, C–N and C–O bonds in biaryls, N,N' -diarylureas and diaryl ethers: advantages of a relay axis

Mark S. Betson, Ann Bracegirdle, Jonathan Clayden,* Madeleine Helliwell, Andrew Lund, Mark Pickworth, Timothy J. Snape and Christopher P. Worrall

A sulfoxide substituent can control the orientation of a C–C, C–N or C–O bond.

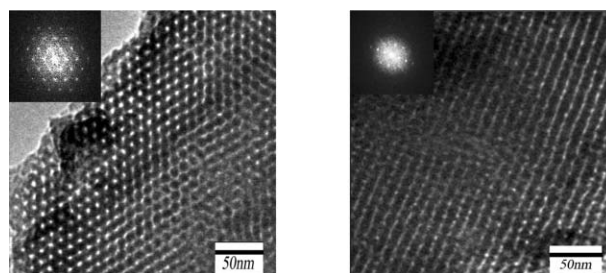


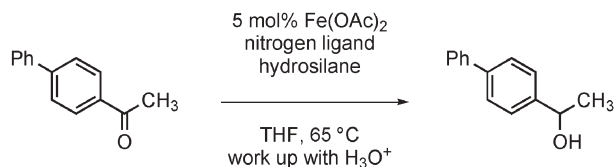
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Facile synthesis of ordered mesoporous carbons from F108/resorcinol–formaldehyde composites obtained in basic media

Changyi Liu, Lixia Li, Huaihe Song* and Xiaohong Chen

Highly ordered mesoporous carbon with cubic $Im\bar{3}m$ symmetry has been synthesized successfully *via* a direct carbonization of self-assembled F108 ($\text{EO}_{132}\text{PO}_{50}\text{EO}_{132}$) and resorcinol–formaldehyde composites obtained in a basic medium of nonaqueous solution.





An iron-catalysed hydrosilylation of ketones

Hisao Nishiyama* and Akihiro Furuta

The combination of Fe(OAc)₂ and multi-nitrogen-based ligands can efficiently catalyse hydrosilylation of ketones to give the corresponding alcohols in high yields including asymmetric catalysis.

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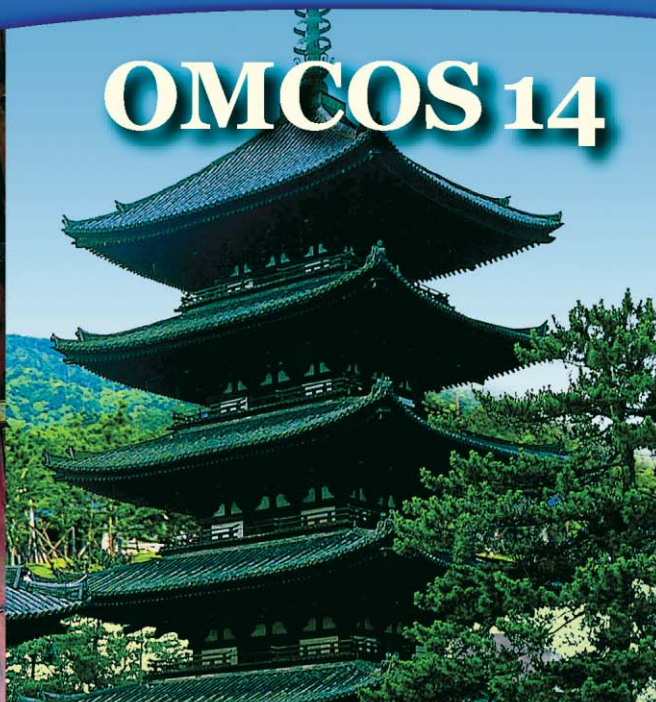
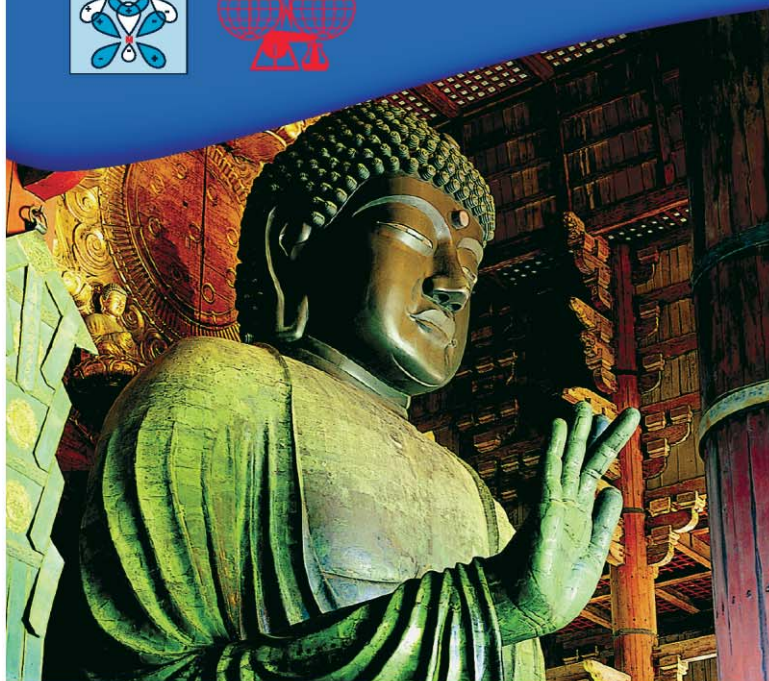
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