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Chemical Communications

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## IN THIS ISSUE

ISSN 1359-7345 CODEN CHCOFS (4) 397-512 (2008)



### Cover

See Jagadese J. Vittal *et al.*, pp. 441–443.

A cyclobutane ring formed from the solid-state photochemical cycloaddition of an hydrogen-bonded zwitter-ionic Pb(II) complex is found to isomerize in solution. Image reproduced by permission of Abdul Malik Puthan Peedikakkal, Lip L. Koh and Jagadese J. Vittal from *Chem. Commun.*, 2008, 441.



### Inside cover

See Jian Zhang and Xianhui Bu, pp. 444–446.

The charge separation in 3-D homochiral cadmium camphorates is favoured at room temperature and becomes progressively smaller with increasing reaction temperature.

Image reproduced by permission of Jian Zhang and Xianhui Bu from *Chem. Commun.*, 2008, 444.

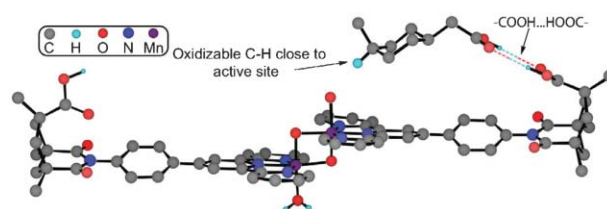
## FEATURE ARTICLES

413

### Molecular recognition in homogeneous transition metal catalysis: a biomimetic strategy for high selectivity

Siddhartha Das, Gary W. Brudvig and Robert H. Crabtree

A molecular recognition strategy involving hydrogen bonding and solvophobic forces has given almost completely regioselective functionalization of remote, unactivated C–H bonds of alkyl carboxylic acids.

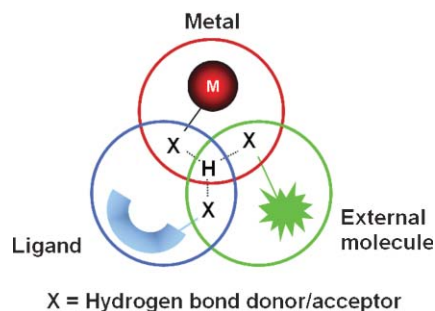


425

### The combination of transition metal ions and hydrogen-bonding interactions

Daniela Natale and Juan C. Mareque-Rivas\*

This feature article presents an overview of the types of hydrogen bonding interactions involving metal complexes and their functional effects. It shows with recent examples why hydrogen bonds have become a crucial functional and structural element in modern inorganic chemistry. The relevance of this combination in tackling current chemistry challenges such as energy production and the development of new materials and more effective catalysts, sensors and medicines is illustrated.



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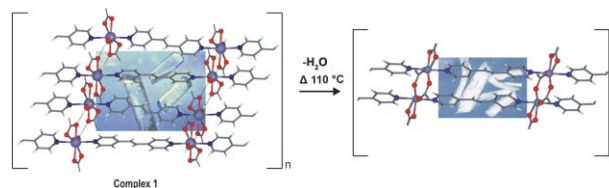
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438

### Photochemical [2 + 2] cycloaddition as a tool to study a solid-state structural transformation

Mangayarkarasi Nagarathinam and Jagadese J. Vittal\*

Schmidt's criteria for the photodimerization reaction are used as a tool to ascertain the solid-state structural transformation from a hydrogen-bonded 1D coordination polymer to a ladder-like polymer after removing the coordinated water.

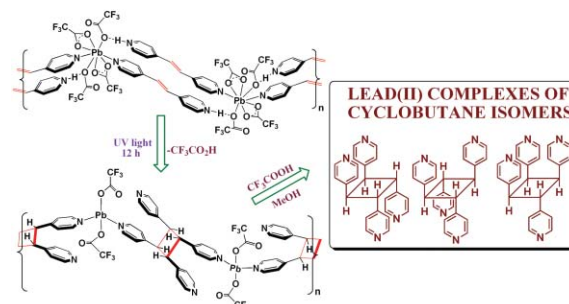


441

### Photodimerization of a 1D hydrogen-bonded zwitter-ionic lead(II) complex and its isomerization in solution

Abdul Malik Puthan Peedikakkal, Lip L. Koh and Jagadese J. Vittal\*

Photochemical cycloaddition of the hydrogen-bonded zwitter-ionic Pb complex involving  $\text{bpe-H}^{\oplus}$  is reported. The *rect*-tpcb ring formed isomerizes slowly in solution to *rcct*-tpcb and *rtct*-tpcb isomers, catalyzed by the  $\text{CF}_3\text{CO}_2\text{H}$  formed.

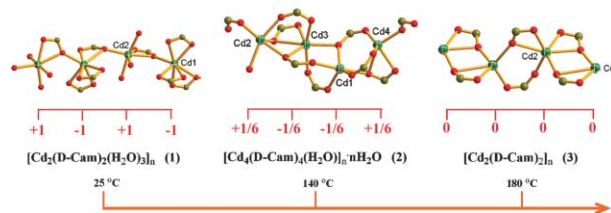


444

### Temperature dependent charge distribution in three-dimensional homochiral cadmium camphorates

Jian Zhang and Xianhui Bu\*

While the charge distribution plays a key role in the formation of microporous inorganic materials such as zeolites, its role in metal-organic frameworks is less clear. Reported here are three homochiral camphorates that exhibit various degrees of charge separation controllable by temperature.

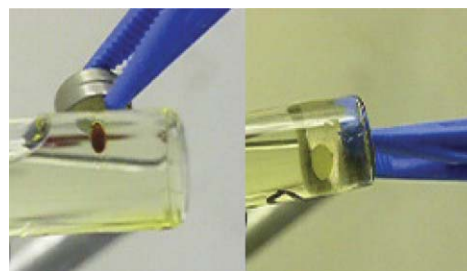


447

### Structure and magnetic behavior of transition metal based ionic liquids

Rico E. Del Sesto,\* T. Mark McCleskey, Anthony K. Burrell, Gary A. Baker, Joe D. Thompson, Brian L. Scott, John S. Wilkes and Peg Williams

Ionic liquids (ILs) containing transition metal based anions were studied. The larger tetraalkylphosphonium cations produce ILs that are immiscible with and stable in water, allowing a droplet of the IL to be manipulated in an aqueous milieu using an external magnetic field.



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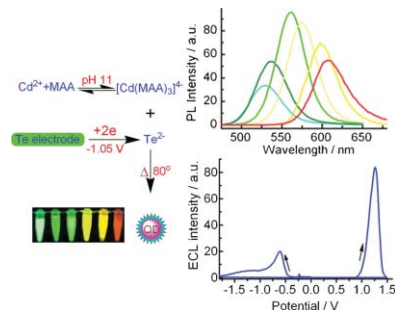
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450

### Facile synthesis and application of highly luminescent CdTe quantum dots with an electrogenerated precursor

Cunwang Ge, Min Xu, Jing Liu, Jianping Lei and Huangxian Ju\*

An electrogenerated precursor has been developed for green synthesis of highly luminescent aqueous CdTe quantum dots with unique quantum yield and strong electrogenerated luminescence, which have potential application as biolabels in highly sensitive biosensing and cell imaging.

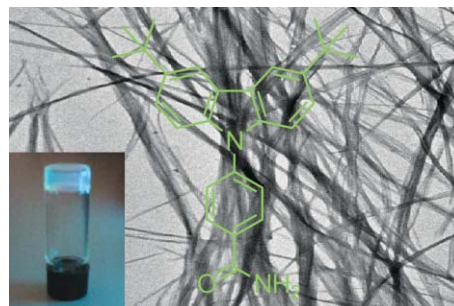


453

### Novel carbazole-based organogels modulated by *tert*-butyl moieties

Xinchun Yang, Ran Lu,\* Tinghua Xu, Pengchong Xue, Xingliang Liu and Yingying Zhao

*tert*-Butyl groups can modulate the self-assembling properties of carbazole derivatives; organogel fibers with a bright blue emission are generated, directed by the cooperation of hydrogen bonding as well as  $\pi$ - $\pi$  interactions.

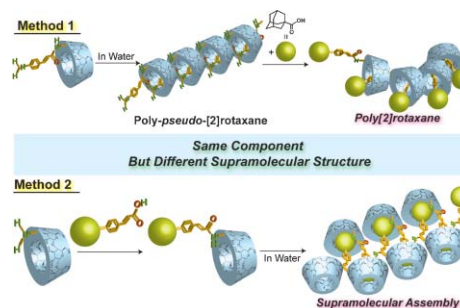


456

### Formation of supramolecular isomers; poly[2]rotaxane and supramolecular assembly

Atsuhisa Miyawaki, Masahiko Miyauchi, Yoshinori Takashima, Hiroyasu Yamaguchi and Akira Harada\*

Poly[2]rotaxane and supramolecular assembly have been prepared by modified cyclodextrins bearing an adamantyl group in an aqueous medium. Although each unit of poly[2]rotaxane and supramolecular assembly is the same building block, each structure of supramolecular complexes revealed to be quite different.

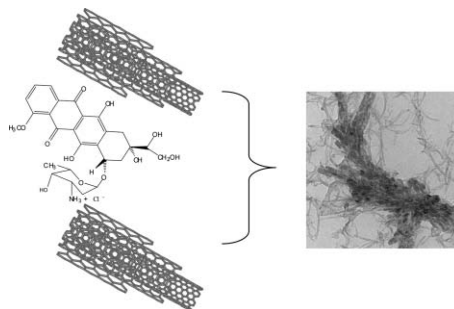


459

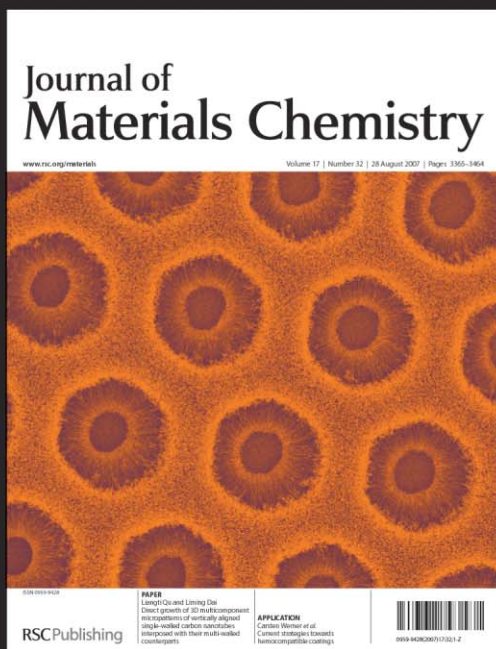
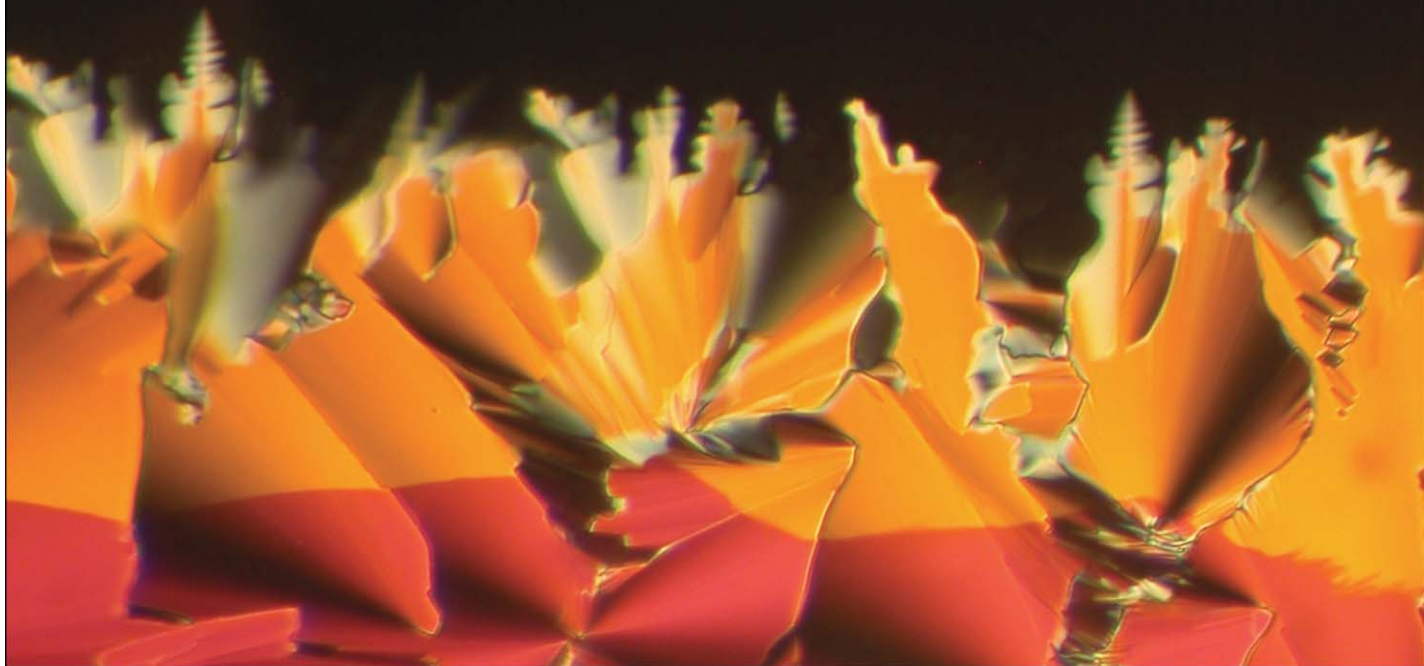
### Multiwalled carbon nanotube–doxorubicin supramolecular complexes for cancer therapeutics

Hanene Ali-Boucetta, Khuloud T. Al-Jamal, David McCarthy, Maurizio Prato, Alberto Bianco and Kostas Kostarelos\*

Multiwalled carbon nanotube aqueous dispersions using block copolymers are able to form supramolecular complexes with the aromatic chromophore and anticancer agent doxorubicin *via*  $\pi$ - $\pi$  stacking and enhance its cytotoxic activity.



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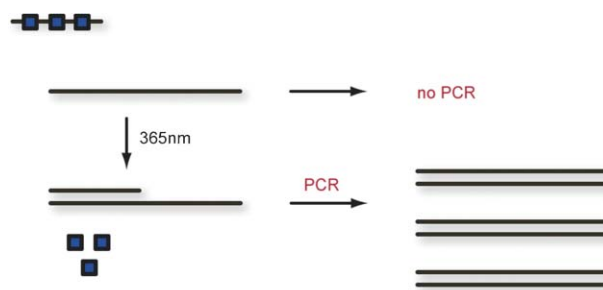
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462

**Light-triggered polymerase chain reaction**

Douglas D. Young, Wessleigh F. Edwards, Hrvoje Lusic, Mark O. Lively and Alexander Deiters\*

Photochemical activation and deactivation of DNA amplification *via* PCR was achieved through the site-specific incorporation of caged nucleotides into PCR primers.

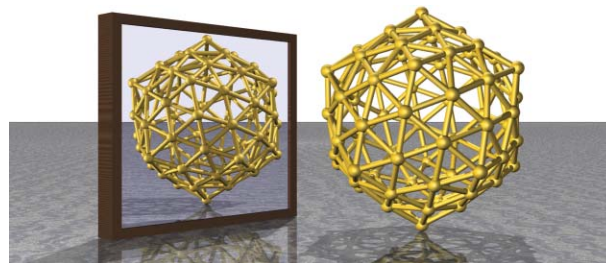


465

**Icosahedral Au<sub>72</sub>: a predicted chiral and spherically aromatic golden fullerene**

Antti J. Karttunen, Mikko Linnolahti,\* Tapani A. Pakkanen and Pekka Pyykkö

Quantum chemical calculations demonstrate the spherical aromaticity and high thermodynamic stability of Au<sub>72</sub>, a predicted *I*-symmetric golden fullerene.

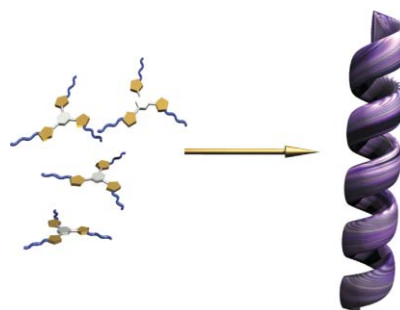


468

**Self-assembly of tris(phenylisoxazoly)benzene and its asymmetric induction of supramolecular chirality**

Takeharu Haino,\* Masahiro Tanaka and Yoshimasa Fukazawa

Tris(phenylisoxazoly)benzene derivatives stack in a columnar fashion to form helical fibers that act as an organogelator. The columnar assembly forms in solution *via* solvophobic  $\pi$ - $\pi$  stacking and dipole-dipole interactions. The assembly adopts a helical structure, which is biased upon the addition of a tiny proportion of a chiral source, giving rise to the induced CD.

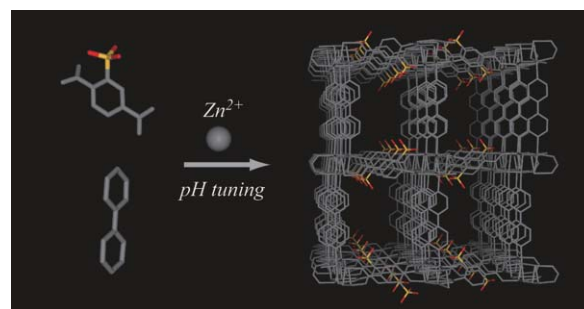


471

**Coordination pillared-layer type compounds having pore surface functionalization by anionic sulfonate groups**

Satoshi Horike, Sareeya Bureekaew and Susumu Kitagawa\*

Pillared-layer type 3D porous coordination polymers with 2-sulfonylterephthalate, 4,4'-bipyridine and Zn<sup>2+</sup> have metal-free sulfonate groups on the pore walls, providing Lewis basic property for acid guest sorption.



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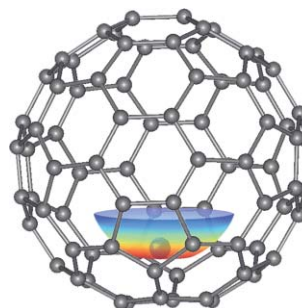


474

### The structural determination of endohedral metallofullerene Gd@C<sub>82</sub> by XANES

Lei Liu, Bin Gao, Wangsheng Chu, Dongliang Chen, Tiandou Hu, Chunru Wang, Lothar Dunsch, Augusto Marcelli, Yi Luo and Ziyu Wu\*

Although the Gd ion in Gd@C<sub>82</sub> has been shown to lie above the C–C bond on the C<sub>2</sub> axis as an anomalous structure from the MEM/Rietveld analysis, the present XANES study reveals that it lies above the hexagon on the C<sub>2</sub> axis as a normal structure, and Gd oscillates around its equilibrium position.

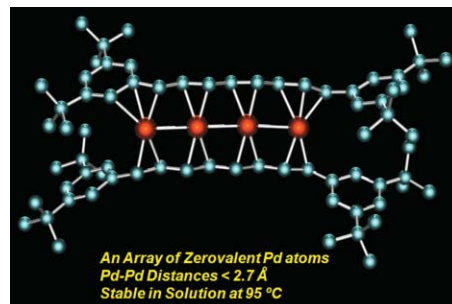


477

### A stable zerovalent palladium chain enveloped by a π-electron sheath of conjugated polyene ligands

Yasuki Tatsumi, Tetsuro Murahashi,\* Mitsue Okada, Sensuke Ogoshi and Hideo Kurosawa

A surprisingly stable homoleptic Pd(0)<sub>4</sub> chain complex of non-activated olefins was isolated and structurally characterized by X-ray crystallographic analysis. The unique structure and bonding are compared to those of the corresponding dicationic [Pd<sub>4</sub>]<sup>2+</sup> chain sandwich complex.

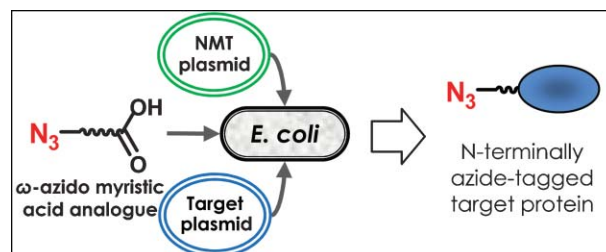


480

### Site-specific N-terminal labelling of proteins *in vitro* and *in vivo* using N-myristoyl transferase and bioorthogonal ligation chemistry

William P. Heal, Sasala R. Wickramasinghe, Paul W. Bowyer, Anthony A. Holder, Deborah F. Smith, Robin J. Leatherbarrow\* and Edward W. Tate\*

N-Myristoyl transferase-mediated protein labelling with azide-bearing substrates: a highly selective and practical method for *in vitro* and *in vivo* N-terminal labelling of proteins using bioorthogonal ligation chemistry.

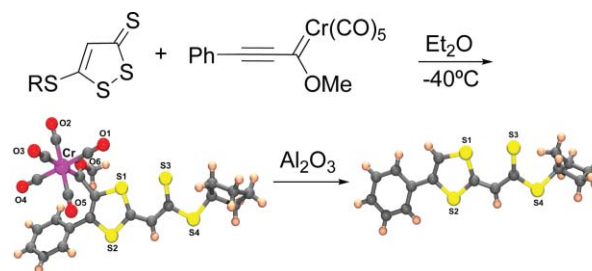


483

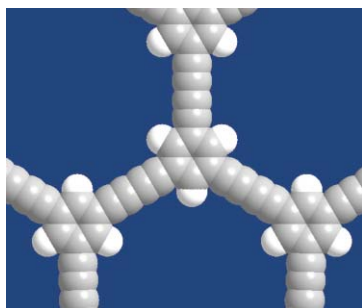
### Alkynyl Fischer carbene complex as a traceless directing group for the regioselective cycloaddition of dithiolethiones to arylacetylene: synthesis of E-dithiafulvene thione and dithioesters

Alejandro M. Granados,\* Alejandro M. Fracaroli, Rita H. de Rossi, Pedro Fuertes and Tomás Torroba\*

A phenylethynyl Fischer carbene complex was used as a traceless directing group for the regioselective cycloaddition of dithiolethiones to arylacetylene, which constitutes the first synthesis of E-dithiafulvene thione or dithioesters.



486

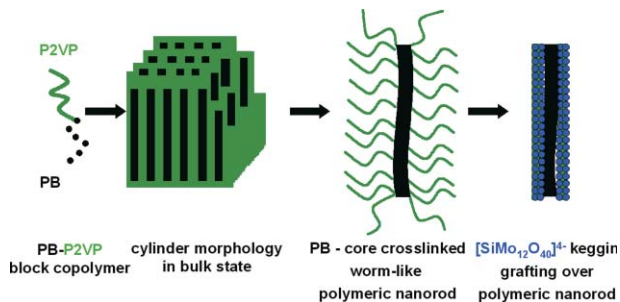


### Conjugated microporous poly(phenylene butadiynylene)s

Jia-Xing Jiang, Fabing Su, Hongjun Niu, Colin D. Wood, Neil L. Campbell, Yaroslav Z. Khimyak and Andrew I. Cooper\*

The direct synthesis of homocoupled microporous polymers (HCMPs) is demonstrated, producing conjugated poly(phenylene butadiynylene)s with surface areas up to  $842 \text{ m}^2 \text{ g}^{-1}$ .

489

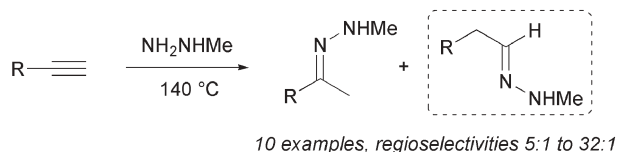


### Core-crosslinked block copolymer nanorods as templates for grafting $[\text{SiMo}_{12}\text{O}_{40}]^{4-}$ Keggin ions

Ram Sai Yelamanchili, Andreas Walther, Axel H. E. Müller\* and Josef Breu\*

Core-crosslinked PB-P2VP block copolymer nanorods are used as templates for the synthesis of Keggin-type heteropolyoxometalate (POM) nanostructures by grafting  $[\text{SiMo}_{12}\text{O}_{40}]^{4-}$  Keggin ions on the template.

492

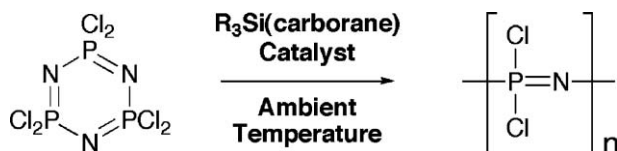


### Intermolecular Cope-type hydroamination of alkynes using hydrazines

Pamela H. Cebrowski, Jean-Grégoire Roveda, Joseph Moran, Serge I. Gorelsky and André M. Beauchemin\*

Metal-free, intermolecular hydroaminations are performed upon heating aryl acetylenes and  $\text{MeNHNH}_2$  at  $140\text{ }^\circ\text{C}$ , with preferential formation of the linear, “anti-Markovnikov” hydrazones. A concerted mechanism is proposed and supported by DFT calculations.

494



### Ambient temperature ring-opening polymerisation (ROP) of cyclic chlorophosphazene trimer $[\text{N}_3\text{P}_3\text{Cl}_6]$ catalyzed by silylium ions

Yun Zhang, Keith Huynh, Ian Manners\* and Christopher A. Reed\*

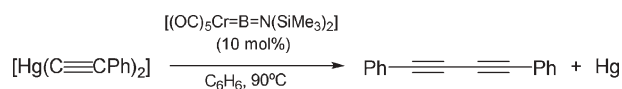
Ambient temperature ring-opening polymerisation of  $[\text{N}_3\text{P}_3\text{Cl}_6]$  is achieved with trialkylsilyl carborane catalysts such as  $\text{Et}_3\text{Si}(\text{CHB}_{11}\text{H}_5\text{Br}_6)$ , offering the possibility of better control over phosphazene polymer properties.

497

### Group VI metal aminoborylene complex-catalyzed demercuration reactions of bis(alkynyl)mercurials

Leonardo Apostolico, Holger Braunschweig,\*  
Andrew G. Crawford, Thomas Herbst and Daniela Rais

The aminoborylene complex-catalyzed demercuration of  $[\text{Hg}(\text{C}\equiv\text{CR})_2]$  represents the very first catalytic application of borylene complexes, and the first instance in which a transition metal complex with a direct metal–boron bond participates in a catalytic cycle without transfer of the boron-based ligand to a substrate.

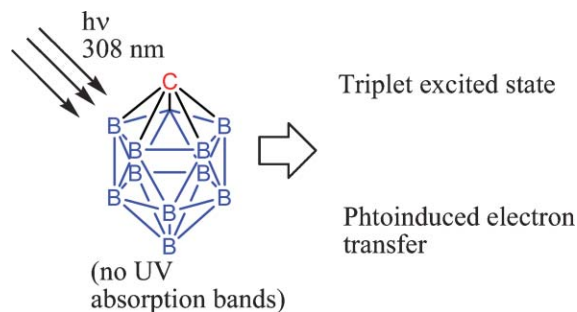


499

### Unexpected photochemistry and charge-transfer complexes of $[\text{CB}_{11}\text{H}_{12}]^-$ carborane

Francesc X. Llabrés i Xamena, Laura Teruel,  
María S. Galletero, Avelino Corma\* and  
Hermenegildo García\*

The triplet excited state of  $\text{Cs}[\text{CB}_{11}\text{H}_{12}]$  has been characterized. Also the charge transfer complex of carborane and methylviologen gives rise to photoinduced electron transfer.

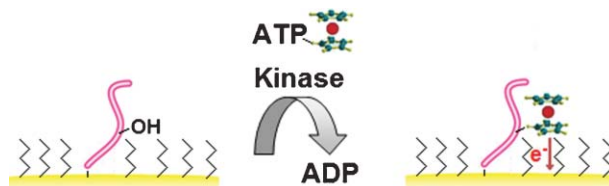


502

### Electrochemical detection of kinase-catalyzed phosphorylation using ferrocene-conjugated ATP

Haifeng Song, Kagan Kerman and  
Heinz-Bernhard Kraatz\*

Adenosine-5'-[ $\gamma$ -ferrocene] triphosphate is exploited as a co-substrate for the kinase-catalyzed phosphorylation of a surface-immobilized substrate peptide, in which the  $\gamma$ -ferrocene phosphate is transferred to the peptide and then detected using electrochemical techniques.

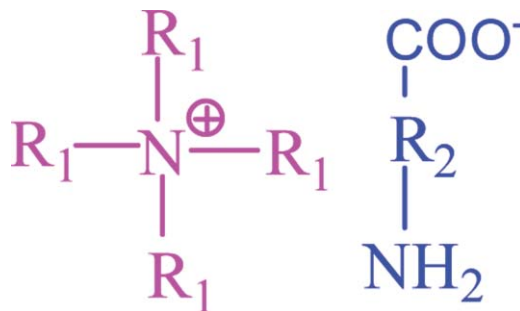


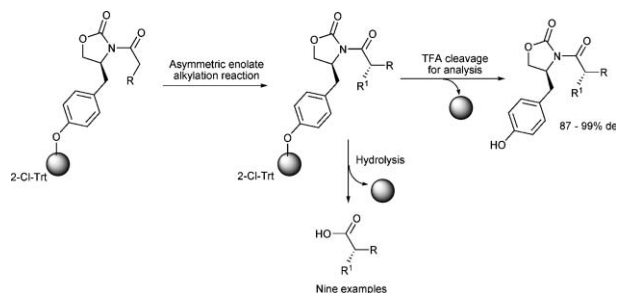
505

### Tetraalkylammonium amino acids as functionalized ionic liquids of low viscosity

Ying-Ying Jiang, Guan-Nan Wang, Zheng Zhou,  
You-Ting Wu,\* Jiao Geng and Zhi-Bing Zhang

Four of nine tetraalkylammonium-based amino-acid ionic liquids (AAILs) prepared in this work show lower viscosities than all previously reported AAILs and their reversible  $\text{CO}_2$  absorption approaches 0.5 mol per mol ionic liquid.





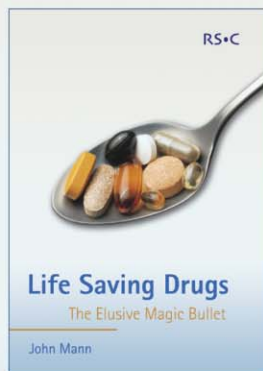
### A cleavable linker strategy for optimising enolate alkylation reactions of a polymer-supported Evans' oxazolidin-2-one

Rachel Green, Andrew T. Merritt and Steven D. Bull\*

A cleavable linker strategy has been used to optimise the enolate alkylation reactions of a recyclable L-tyrosine derived polymer-supported oxazolidin-2-one for the asymmetric synthesis of a series of chiral  $\alpha$ -alkyl acids.

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## AUTHOR INDEX

- Ali-Boucetta, Hanene, 459  
 Al-Jamal, Khuloud T., 459  
 Apostolico, Leonardo, 497  
 Baker, Gary A., 447  
 Beauchemin, André M., 492  
 Bianco, Alberto, 459  
 Bowyer, Paul W., 480  
 Braunschweig, Holger, 497  
 Breu, Josef, 489  
 Brudvig, Gary W., 413  
 Bu, Xianhui, 444  
 Bull, Steven D., 508  
 Bureekaew, Sareeya, 471  
 Burrell, Anthony K., 447  
 Campbell, Neil L., 486  
 Cebrowski, Pamela H., 492  
 Chen, Dongliang, 474  
 Chu, Wangsheng, 474  
 Cooper, Andrew I., 486  
 Corma, Avelino, 499  
 Crabtree, Robert H., 413  
 Crawford, Andrew G., 497  
 Das, Siddhartha, 413  
 Deiters, Alexander, 462  
 Del Sesto, Rico E., 447  
 de Rossi, Rita H., 483  
 Dunsch, Lothar, 474  
 Edwards, Wesley F., 462  
 Fracaroli, Alejandro M., 483  
 Fuertes, Pedro, 483  
 Fukazawa, Yoshimasa, 468  
 Galletero, María S., 499  
 Gao, Bin, 474  
 García, Hermenegildo, 499  
 Ge, Cunwang, 450  
 Geng, Jiao, 505  
 Gorelsky, Serge I., 492  
 Granados, Alejandro M., 483  
 Green, Rachel, 508  
 Haino, Takeharu, 468  
 Harada, Akira, 456  
 Heal, William P., 480  
 Herbst, Thomas, 497  
 Holder, Anthony A., 480  
 Horike, Satoshi, 471  
 Hu, Tiandou, 474  
 Huynh, Keith, 494  
 Jiang, Jia-Xing, 486  
 Jiang, Ying-Ying, 505  
 Ju, Huangxian, 450  
 Karttunen, Antti J., 465  
 Kerman, Kagan, 502  
 Khimyak, Yaroslav Z., 486  
 Kitagawa, Susumu, 471  
 Koh, Lip L., 441  
 Kostarelos, Kostas, 459  
 Kraatz, Heinz-Bernhard, 502  
 Kurosawa, Hideo, 477  
 Leatherbarrow, Robin J., 480  
 Lei, Jianping, 450  
 Linnolahti, Mikko, 465  
 Liu, Jing, 450  
 Liu, Lei, 474  
 Liu, Xingliang, 453  
 Lively, Mark O., 462  
 Llabrés i Xamena, Francesc X., 499  
 Lu, Ran, 453  
 Luo, Yi, 474  
 Lusic, Hrvoje, 462  
 Manners, Ian, 494  
 Marcelli, Augusto, 474  
 Mareque-Rivas, Juan C., 425  
 McCarthy, David, 459  
 McCleskey, T. Mark, 447  
 Merritt, Andrew T., 508  
 Miyauchi, Masahiko, 456  
 Miyawaki, Atsuhisa, 456  
 Moran, Joseph, 492  
 Müller, Axel H. E., 489  
 Murahashi, Tetsuro, 477  
 Nagarathinam, Mangayarkarasi, 438  
 Natale, Daniela, 425  
 Niu, Hongjun, 486  
 Ogoshi, Sensuke, 477  
 Okada, Mitsue, 477  
 Pakkanen, Tapani A., 465  
 Peedikakkal, Abdul Malik Puthan, 441  
 Prato, Maurizio, 459  
 Pyykkö, Pekka, 465  
 Rais, Daniela, 497  
 Reed, Christopher A., 494  
 Roveda, Jean-Grégoire, 492  
 Scott, Brian L., 447  
 Smith, Deborah F., 480  
 Song, Haifeng, 502  
 Su, Fabing, 486  
 Takashima, Yoshinori, 456  
 Tanaka, Masahiro, 468  
 Tate, Edward W., 480  
 Tatsumi, Yasuki, 477  
 Teruel, Laura, 499  
 Thompson, Joe D., 447  
 Torroba, Tomás, 483  
 Vittal, Jagadeś J., 438, 441  
 Walther, Andreas, 489  
 Wang, Chunru, 474  
 Wang, Guan-Nan, 505  
 Wickramasinghe, Sasala R., 480  
 Wilkes, John S., 447  
 Williams, Peg, 447  
 Wood, Colin D., 486  
 Wu, You-Ting, 505  
 Wu, Ziyu, 474  
 Xu, Min, 450  
 Xu, Tinghua, 453  
 Xue, Pengchong, 453  
 Yamaguchi, Hiroyasu, 456  
 Yang, Xinchun, 453  
 Yelamanchili, Ram Sai, 489  
 Young, Douglas D., 462  
 Zhang, Jian, 444  
 Zhang, Yun, 494  
 Zhang, Zhi-Bing, 505  
 Zhao, Yingying, 453  
 Zhou, Zheng, 505

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