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IN THIS ISSUE

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Cover



See Alexander Rothenberger et al., page 1499. Coordination chemistry with novel anions that are generated in situ. Similar to an example of a randomly-generated tiling, the coinage metal cluster and the alkali metal-containing coordination polymer represent two of many new oligomeric and polymeric arrangements of group 15/16 anions. Image reproduced by permission of Alexander Rothenberger, Maryam Shafaei-Fallah and Weifeng Shi, from Chem. Commun., 2007, 1499.

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C25

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Chemical Science

April 2007/Volume 4/Issue 4 www.rsc.org/chemicalscience

FEATURE ARTICLE

1487

Design of chiral organocatalysts for practical asymmetric synthesis of amino acid derivatives

Keiji Maruoka,* Takashi Ooi and Taichi Kano

A series of new C_2 -symmetric chiral phase-transfer catalysts and chiral bifunctional amino-catalysts derived from commercially available (*R*)- or (*S*)-binaphthol have been designed and successfully applied to the highly practical asymmetric synthesis of various amino acid derivatives.



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1496

Organometallic molecular materials: self-assembly through hydrogen bonding of an organoplatinum network structure with zeolite-like topology

Fenbao Zhang, Michael C. Jennings and Richard J. Puddephatt*

The complex $[Pt(OH)_2Me_2(dpa)]$, dpa = di-2-pyridylamine, undergoes a remarkable form of self-assembly through NH···O and OH···O hydrogen bonding to give an organometallic network with a zeolitic topology.

1499

A recipe for new organometallic polymers and oligomers? Synthesis and structure of an oligo- and a polymeric arrangement of P–S anions

Alexander Rothenberger,* Maryam Shafaei-Fallah and Weifeng Shi

The potential of neutral molecules to undergo fragmentation and subsequent organisation into clusters or polymers is demonstrated. The results feature new P/S anions and the first example of a $[P(\mu_3-S)_4]^{3-}$ ion embedded in a cluster cation.

1502

Highly efficient P–N nickel(II) complexes for the dimerisation of ethylene

Antoine Buchard, Audrey Auffrant, Christian Klemps, Laurence Vu-Do, Leïla Boubekeur, Xavier F. Le Goff and Pascal Le Floch*

New P–N ligands featuring a phosphino group and an iminophosphorane moiety were successfully employed in the nickel-catalysed dimerisation of ethylene.

1505

Straightforward construction of diarylmethane skeletons via aryne insertion into carbon–carbon σ -bonds

Hiroto Yoshida,* Masahiko Watanabe, Takami Morishita, Joji Ohshita and Atsutaka Kunai*

Two mol equiv. of arynes are found to couple with nitriles at the C–C and C–H σ -bonds, assembling diverse diarylmethane skeletons in a straightforward manner. Overall, the transformation enables three C–C and one C–H bond forming processes to take place all in one pot.













Highlighting Heme

Natural Product Reports has gathered together a series of forwardlooking articles from leading international experts in the heme enzyme area. Co-edited by Emma Raven and Paul Ortiz, it is essential reading for anyone working on heme enzymes. It will also be of wider interest to those requiring an overview of the current understanding of the bioinorganic chemistry of heme iron.

The Janus Nature of Heme Thomas L. Poulos

Spectroscopic characterization of heme iron-nitrosyl species and their role in NO reductase mechanisms in diiron proteins Pierre Moënne-Loccoz

Heme and Virulence: How bacterial pathogens regulate, transport and utilize heme Angela Wilks and Kimberly A. Burkhard

Structure and Catalytic Mechanism of Heme Oxygenase Masaki Unno, Toshitaka Matsui and Masao Ikeda-Saito

Heme to protein linkages in mammalian peroxidases: Impact on spectroscopic, redox and catalytic properties. *M. Zederbauer, P. G. Furtmüller, S. Brogioni, C. Jakopitsch, G. Smulevich and C. Obinger*

Properties of an Unusual Heme Cofactor in PLP-dependent Cystathionine b-Synthase *Sangita Singh, Peter Madzelan and Ruma Banerjee*

Structural Modelling of Metal Ion Binding to Human Hæmopexin *Marcia R. Mauk, Federico I. Rosell and A. Grant Mauk*

Diversity and Conservation of Interactions for Binding Heme in b-type Heme Proteins *Sabine Schneider, Jon Marles-Wright, Katherine H. Sharp and Max Paoli*

Reactivity patterns of cytochrome P450 enzymes: Multifunctionality of the active species and the two states – two oxidants conundrum Sason Shaik, Hajime Hirao, and Devesh Kumar

Variations on a (T)Heme – Novel Mechanisms, Redox Partners and Catalytic Functions in the Cytochrome P450 Superfamily Andrew W. Munro, Hazel M. Girvan and Kirsty J. McLean



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1508



The total synthesis of siphonazole, a structurally unusual bis-oxazole natural product

Jörg Linder and Christopher J. Moody*

The first synthesis of the unusual bis-oxazole natural product siphonazole is reported.

1510

A bimetallic N-heterocyclic carbene complex featuring a short Cr–Cr distance

Kevin A. Kreisel, Glenn P. A. Yap and Klaus H. Theopold*

The reaction of a chelating bis(carbene) complex of CrCl₂ with MeMgCl in THF yields an unusual bimetallic complex featuring a short Cr–Cr distance, novel ligand coordination, and CH₃ ligand exchange.



1512

The hexamethylpentalene dianion and other reagents for organometallic pentalene chemistry

Andrew E. Ashley, Andrew R. Cowley and Dermot O'Hare*

A novel trialkylborohydride-mediated conjugate reduction of an isomer of hexamethylpentalene permits the isolation of the lithium salt of the hydropermethylpentalene anion. Subsequent metallation provides the hexamethylpentalene dianion, which has been structurally characterised. These represent new synthons for future progress in metal-pentalene chemistry.

1515

Bis(permethylpentalene)cerium – another ambiguity in lanthanide oxidation state

Andrew Ashley, Gabor Balazs, Andrew Cowley, Jennifer Green, Corwin H. Booth and Dermot O'Hare*

 $Ce(\eta^8-C_8Me_6)_2$ has been studied using a variety of techniques including XANES spectroscopy and DFT calculations; the former gives strong evidence for a formal valency close to Ce^{III} in this molecule and provides an example of the self-contained Kondo effect.







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1518



Activity-based fingerprinting and inhibitor discovery of cysteine proteases in a microarray

Mahesh Uttamchandani, Kai Liu, Resmi C. Panicker and Shao Q. Yao*

A panel of 20 peptide vinyl sulfone probes has been synthesized and used to generate activity-based fingerprinting profiles of cysteine proteases in both gel- and microarraybased formats. The inhibitor fingerprints of representative small molecule inhibitors targeted against 4 cysteine proteases were also obtained, in high-throughput, using the same protein microarray platform.



Sorption of nitrogen oxides in a nonporous crystal

Praveen K. Thallapally,* B. Peter McGrail* and Jerry L. Atwood*

The uptake of various nitrogen oxides was studied with the well known nonporous *p-tert*-butylcalix[4]arene at 1 atm pressure and room temperature.



1524

A 'metallic tape' stabilized by an unprecedented $(\mu_5 - \kappa^2, \kappa^2, \kappa^2, \kappa^1, \kappa^1 -)$ scorpionate binding mode

James R. Gardinier,* Rosalice M. Silva, Chengeto Gwengo and Sergey V. Lindeman

A new coordination mode for a 'soft' scorpionate has been discovered that may portend the use of such ligands in materials chemistry.

1527

A pcu-type metal–organic framework with spindle $[Zn_7(OH)_8]^{6+}$ cluster as secondary building units

Jian-Rong Li, Ying Tao, Qun Yu and Xian-He Bu*

A novel **pcu**-type metal–organic framework complex, $\{[Zn_7(OH)_8(DTA)_3] \cdot H_2O\}_n (DTA^{2^-} = 9,10$ -ditetrazolateanthracene) was obtained by the *in situ* solvothermal reaction of 9,10-dicyanoanthracene and NaN₃/ZnCl₂, in which tetrazolate ligands DTA^{2^-} as linkers bridge unprecedented heptanuclear spindle $[Zn_7(OH)_8]^{6^+}$ cluster SBUs as nodes. The optical and electric properties of this complex were also investigated.





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1530

Enantioselective oxidation of *O*-methyl-*N*-hydroxylamines using monoamine oxidase N as catalyst

Tom S. C. Eve, Andrew Wells and Nicholas J. Turner*

E. coli cells expressing a monoamine oxidase N (MAO-N) variant have been used for the efficient and highly stereoselective oxidation of (\pm) -3 yielding (*R*)-3 (e.e. = 99%) and (*E*)-8.

1532

Ligand flexibility and framework rearrangement in a new family of porous metal–organic frameworks

Samuel M. Hawxwell, Guillermo Mínguez Espallargas, Darren Bradshaw, Matthew J. Rosseinsky, Timothy J. Prior, Alastair J. Florence, Jacco van de Streek and Lee Brammer*

Ligand flexibility permits framework rearrangement upon evacuation and gas uptake in a new family of porous MOFs.

1535

Electropolymeric materials incorporating subsite structures related to iron-only hydrogenase: active ester functionalised poly(pyrroles) for covalent binding of {2Fe3S}-carbonyl/cyanide assemblies

Saad K. Ibrahim, Xiaoming Liu, Cédric Tard and Christopher J. Pickett*

The authors report the assembly of the first electropolymeric materials incorporating catalytic diiron subsites related to those of the iron-only hydrogenases.

1538

Heteronuclear Mn–Ca/Sr complexes, and Ca/Sr EXAFS spectral comparisons with the Oxygen-Evolving Complex of Photosystem II

Abhudaya Mishra, Junko Yano, Yulia Pushkar, Vittal K. Yachandra, Khalil A. Abboud and George Christou*

The first mixed-metal Mn–Sr complex has been prepared, and this and a previously reported Mn–Ca complex have allowed Sr and Ca EXAFS comparisons with the Oxygen-Evolving Complex (OEC) of Photosystem II.













Sulfate separation by selective crystallization of a urea-functionalized metal–organic framework

Radu Custelcean,* Vincent Sellin and Bruce A. Moyer

Competitive crystallization of a Ni coordination framework functionalized with urea hydrogen-bonding groups results in exclusive separation of sulfate from aqueous anionic mixtures.



A multi-ion particle sensor

Maria Jose Ruedas-Rama, Xiaojuan Wang and Elizabeth A. H. Hall*

The first sub-micron polyacrylic sensor containing two independent ion-sensing systems is shown, that uses a single excitation wavelength and separates signals by using quantum dot donors to form FRET pairs with other fluorophores.

1547



1550



Study on high-efficiency fluorescent microcapsules doped with europium β -diketone complex by LbL self-assembly

Ren-Jie Zhang,* Ji-wei Cui, De-Ming Lu and Wan-Guo Hou

Microcapsules are fabricated by the layer-by-layer selfassembly of polyelectrolytes and a europium β -diketone complex emitting red fluorescence. Luminescence of the microcapsules with only one layer of europium complex can be observed by the naked eye.

Preparation of transparent oxyapatite ceramics by combined use of freeze-drying and spark-plasma sintering

A. Chesnaud, C. Bogicevic, F. Karolak, C. Estournès and G. Dezanneau*

Lanthanum silicate oxyapatites with formula La_{9.33}Si₆O₂₆, an ion-conducting material for solid oxide fuel cell electrolytes, have been obtained in form of dense transparent ceramics, by combining freeze-drying and spark plasma sintering methods.

1553



/. 10 HPPh2 , 60 % Pd(OAc) NEt3, CH3CN, 60°C, 3 days

2/. Methyl amine/NH₄OH, 60 3/. FPLC, eluant H₂O/TEAA

Self-organization of porphyrin units induced by magnetic field during sol-gel polymerization

Frédéric Lerouge, Geneviève Cerveau, Robert J. P. Corriu,* Christine Stern and Roger Guilard*

The use of a magnetic field during the hydrolytic sol–gel polycondensation of porphyrins tetrasubstituted by $Si(OR)_3$ groups induces the stacking of the macrocycle units in the resulting hybrid materials.

1556

Phosphine containing oligonucleotides for the development of metallodeoxyribozymes

Loïc Ropartz, Nico J. Meeuwenoord, Gijsbert A. van der Marel, Piet W. N. M. van Leeuwen, Alexandra M. Z. Slawin and Paul C. J. Kamer*

Novel transition metal catalysts based on oligonucleotides can be easily obtained by functionalization of 5-iodouridine with phosphine ligands, resulting in good asymmetric induction in palladium catalyzed allylic nucleophilic substitution.

1559

Partially-methylated amyloses as effective hosts for inclusion complex formation with polymeric guests

Toshiyuki Kida, Takashi Minabe, Shogo Okabe and Mitsuru Akashi*

Partially-methylated amyloses efficiently form inclusion complexes with polytetrahydrofuran and poly(ε -caprolactone) by simply mixing them in DMSO-H₂O (1 : 9) solution.

polymeric guest

inclusion complex

partially-methylated amylose

ADDITION AND CORRECTION

1562

Preparation of transparent oxyapatite ceramics by combined use of freeze-drying and spark plasma sintering

A. Chesnaud, C. Bogicevic, F. Karolak, C. Estournès and G. Dezanneau

AUTHOR INDEX

Abboud, Khalil A., 1538 Akashi, Mitsuru, 1559 Ashley, Andrew E., 1512, 1515 Atwood, Jerry L., 1521 Auffrant, Audrey, 1502 Balazs, Gabor, 1515 Bogicevic, C., 1550 Booth, Corwin H., 1515 Boubekeur, Leïla, 1502 Bradshaw, Darren, 1532 Brammer, Lee, 1532 Bu, Xian-He, 1527 Buchard, Antoine, 1502 Cerveau, Geneviève, 1553 Chesnaud, A., 1550 Christou, George, 1538 Corriu, Robert J. P., 1553 Cowley, Andrew R., 1512, 1515 Cui, Ji-wei, 1547 Custelcean, Radu, 1541 Dezanneau, G., 1550 Estournès, C., 1550 Eve, Tom S. C., 1530 Florence, Alastair J., 1532 Gardinier, James R., 1524

Green, Jennifer, 1515 Guilard, Roger, 1553 Gwengo, Chengeto, 1524 Hall, Elizabeth A. H., 1544 Hawxwell, Samuel M., 1532 Hou, Wan-Guo, 1547 Ibrahim, Saad K., 1535 Jennings, Michael C., 1496 Kamer, Paul C. J., 1556 Kano, Taichi, 1487 Karolak, F., 1550 Kida, Toshiyuki, 1559 Klemps, Christian, 1502 Kreisel, Kevin A., 1510 Kunai, Atsutaka, 1505 Le Floch, Pascal, 1502 Le Goff, Xavier F., 1502 Lerouge, Frédéric, 1553 Li, Jian-Rong, 1527 Lindeman, Sergey V., 1524 Linder, Jörg, 1508 Liu, Kai, 1518 Liu, Xiaoming, 1535 Lu, De-Ming, 1547 Maruoka, Keiji, 1487

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McGrail, B. Peter, 1521 Meeuwenoord, Nico J., 1556 Minabe, Takashi, 1559 Mínguez Espallargas, Guillermo, 1532 Mishra, Abhudaya, 1538 Moody, Christopher J., 1508 Morishita, Takami, 1505 Moyer, Bruce A., 1541 O'Hare, Dermot, 1512, 1515 Ohshita, Joji, 1505 Okabe, Shogo, 1559 Ooi, Takashi, 1487 Panicker, Resmi C., 1518 Pickett, Christopher J., 1535 Prior, Timothy J., 1532 Puddephatt, Richard J., 1496 Pushkar, Yulia, 1538 Ropartz, Loïc, 1556 Rosseinsky, Matthew J., 1532 Rothenberger, Alexander, 1499 Ruedas-Rama, Maria Jose, 1544 Sellin, Vincent, 1541 Shafaei-Fallah, Maryam, 1499 Shi, Weifeng, 1499

Silva, Rosalice M., 1524 Slawin, Alexandra M. Z., 1556 Stern, Christine, 1553 Tao, Ying, 1527 Tard, Cédric, 1535 Thallapally, Praveen K., 1521 Theopold, Klaus H., 1510 Turner, Nicholas J., 1530 Uttamchandani, Mahesh, 1518 van der Marel, Gijsbert A., 1556 van de Streek, Jacco, 1532 van Leeuwen, Piet W. N. M., 1556 Vu-Do, Laurence, 1502 Wang, Xiaojuan, 1544 Watanabe, Masahiko, 1505 Wells, Andrew, 1530 Yachandra, Vittal K., 1538 Yano, Junko, 1538 Yao, Shao Q., 1518 Yap, Glenn P. A., 1510 Yoshida, Hiroto, 1505 Yu, Qun, 1527 Zhang, Fenbao, 1496 Zhang, Ren-Jie, 1547

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