IN THIS ISSUE

ISSN 1359-7345 CODEN CHCOFS (48) 5933-6048 (2005)



Cover

See Takashi Uemura et al., page 5968. Radical polymerisation of styrene in a porous coordination polymer provides single polystyrene chains isolated in the nanochannels. Image reproduced by permission of Takashi Uemura, Kana Kitagawa, Satoshi Horike, Takashi Kawamura, Susumu Kitagawa, Motohiro Mizuno and Kazunaka Endo from Chem. Commun., 2005, 5968.



Inside cover See Radu Custelcean, Bruce A. Moyer and Benjamin P. Hay, page 5971. Coordination of sulfate via twelve complementary hydrogen bonds was achieved by encapsulation in a coordination polymer functionalized with urea groups. Image reproduced by permission of Radu Custelcean, Bruce A. Moyer and Benjamin P. Hay from Chem. Commun., 2005,

40TH ANNIVERSARY ARTICLE

5945

Sir Jack Baldwin, FRS: Biomimetic studies at Oxford

John E. Moses* and Robert M. Adlington*

Professor Sir Jack Baldwin, FRS, has recently stepped down as the Wayneflete Professor of Organic Chemistry at the University of Oxford. This article is intended to overview some aspects of Professor Baldwin's spectacular career, with an emphasis on his contributions towards the field of biomimetic synthesis. The picture shows Sir Jack giving a closing speech at his retirement dinner at Lady Margaret Hall, Oxford in August 2005.



5971.

FEATURE ARTICLE

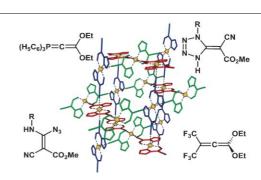
5953

Roots: From carbenes to allenes and coordination polymers

Ever present never twice the same

Rolf W. Saalfrank* and Harald Maid

Recognizing the similarities in different areas of chemistry allows the reaching of the new levels necessary for predicting potential results and progress in related fields. This Feature Article is designed to emphasise the achievements across different topics, together with a straightforward evaluation of developing new common aspects. It covers the chemistry of cumulated phosphoranes, tetrazolylidenes, of vinyl-azides and push–pull substituted allenes, as well as coordination polymers.



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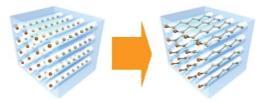
5968

Radical polymerisation of styrene in porous coordination polymers

Takashi Uemura, Kana Kitagawa, Satoshi Horike, Takashi Kawamura, Susumu Kitagawa,* Motohiro Mizuno and Kazunaka Endo

Radical polymerisation of styrene was performed in one-dimensional nanochannels of porous coordination polymers, showing stable propagating radicals in the nanochannels and a specific space effect of the host frameworks on the monomer reactivity.

Polymerisation in porous coordination polymer

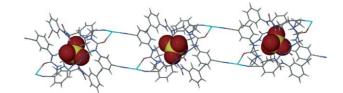


5971

A coordinatively saturated sulfate encapsulated in a metal-organic framework functionalized with urea hydrogen-bonding groups

Radu Custelcean,* Bruce A. Moyer and Benjamin P. Hay

Self-assembly of a tripodal urea-functionalized linker with Ag₂SO₄ generates a coordination polymer that encapsulates SO₄²⁻ via twelve complementary hydrogen bonds, which represents the highest coordination number observed for sulfate inside a host.

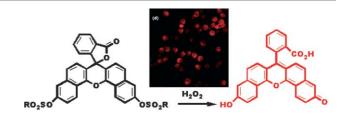


5974

Strong red fluorescent probes suitable for detecting hydrogen peroxide generated by mice peritoneal macrophages

Kehua Xu, Bo Tang,* Hui Huang, Guiwen Yang, Zhenzhen Chen, Ping Li and Liguo An

This paper reports the synthesis, fluorescence properties, and biological applications of naphthofluorescein disulfonate (NFDS-1), as a red fluorescence imaging probe to detect intracellular H₂O₂.

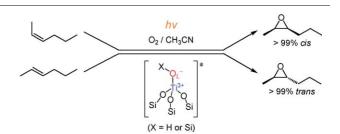


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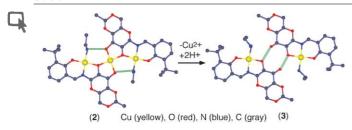
Acetonitrile-assisted highly selective photocatalytic epoxidation of olefins on Ti-containing silica with molecular oxygen

Yasuhiro Shiraishi,* Masatsugu Morishita and Takayuku Hirai

Highly selective photocatalytic epoxidation of olefins proceeds on Ti-containing silica with tetrahedrally coordinated Ti-oxide species with molecular oxygen and a simple addition of MeCN.



5980

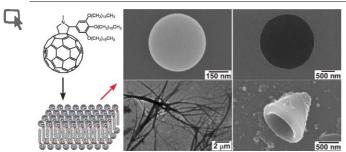


Crystal-to-crystal transformation from tri- to mononuclear Cu(II) complex with a sugar-derived ligand *via* proton transfer reaction and rearrangement of hydrogen bonding networks

Ajay K. Sah and Tomoaki Tanase*

The trinuclear Cu(II) complex of a glucopyranosylamine derived ligand has been converted into a mononuclear one *via* proton transfer reaction revealing the first instance of crystal-to-crystal transformation in the field of metal saccharide chemistry.

5982

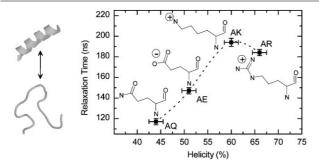


Hierarchical supramolecular fullerene architectures with controlled dimensionality

Takashi Nakanishi,* Wolfgang Schmitt, Tsuyoshi Michinobu, Dirk G. Kurth and Katsuhiko Ariga

Self-assembly of a long alkyl chain-substituted fullerene results in the formation of hierarchical supramolecular architectures such as vesicles, fibers, discs and cones.

5985

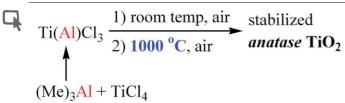


The effects of individual amino acids on the fast folding dynamics of α -helical peptides

Edward A. Gooding,* Angela Pozo Ramajo, JunWei Wang, Colin Palmer, Elizabeth Fouts and Martin Volk*

Nanosecond temperature jump experiments confirm for the first time that the substitution of individual amino acid residues in an α -helical peptide not only affects helical stability, but also the helix–coil kinetics.

5988



High-temperature stabilized anatase TiO_2 from an aluminum-doped $TiCl_3$ precursor

Sujith Perera and Edward G. Gillan*

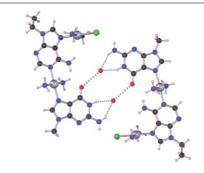
This study describes the synthesis of crystalline aluminum-doped anatase TiO₂ from an organometallic derived aluminum-doped TiCl₃ precursor. The anatase product is stable at 1000 °C for extended periods with average crystallite sizes close to 100 nm.

5991

Structural precursor of the hemideprotonated guanine pair

Michael Roitzsch and Bernhard Lippert*

The insertion of two water molecules between the Watson-Crick faces of two N(7) platinated guanine bases permits indirect base pairing. This pair is proposed to be the precursor of the corresponding hemideprotonated guanine pair, following removal of $H_5O_2^+$.

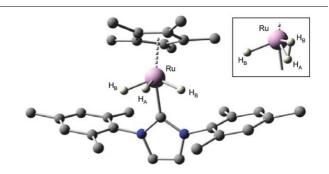


5994

Ruthenium trihydrides with N-heterocyclic carbene ligands: effects on quantum mechanical exchange coupling

Aled L. Jones, G. Sean McGrady,* Peter Sirsch and Jonathan W. Steed*

Ruthenium trihydrides containing N-heterocyclic carbene ligands display large quantum mechanical exchange couplings in their ¹H NMR spectra: DFT calculations are used to explore this phenomenon and to compare them to their phosphine congeners.



5997

Versatile new uranyl(VI) dihalide complexes supported by tunable organic amide ligands

Shanmugaperumal Kannan, Charles L. Barnes and Paul B. Duval*

A series of air-stable uranyl(VI) dihalide complexes supported by organic amide ligands offers facile benchtop syntheses, solubility in hydrocarbon solvents and tunable steric/electronic properties.

$$UO_2X_2 \bullet x(H_2O)$$

$$2L \qquad \qquad U$$

$$X \qquad \qquad U$$

$$X \qquad \qquad U$$

$$2: X = CI$$

$$3: X = Br \qquad R = PPr, PBu, s-Bu$$

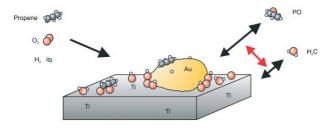
5999

Three-stage switching of surface wetting using phosphate-bearing polymer brushes

Feng Zhou and Wilhelm T. S. Huck*

Surface initiated polymer brushes containing phosphate side groups have been used to reversibly switch wettability and hydration between three different states depending on pH.

6002

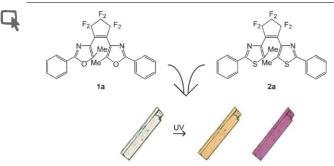


The role of water in the epoxidation over gold-titania catalysts

T. Alexander Nijhuis* and Bert M. Weckhuysen

In the epoxidation of propene over gold–titania catalysts, water and water formation strongly influence the epoxidation and *vice versa*. Competitive adsorption of water reduces catalyst activity but increases catalyst stability.

6005

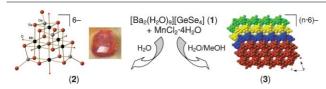


Photochromism of single crystals composed of dioxazolylethene and dithiazolylethene

Lumi Kuroki, Shizuka Takami, Katsunori Shibata and Masahiro Irie*

Dioxazolylethene 1a, which is photochemically inactive in the homo-crystal, becomes photoactive in the mixed crystal containing both 1a and dithiazolylethene 2a. The mixed crystal shows two-color photochromism.

6008



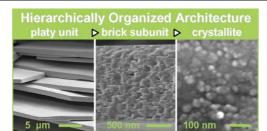
Ternary Mn/Ge/Se anions from reactions of [Ba₂(H₂O)₉][GeSe₄]: Synthesis and characterization of compounds containing discrete or polymeric [Mn₆Ge₄Se₁₇]⁶⁻ units

Maike Melullis, Rodolphe Clérac and Stefanie Dehnen*

Reactions of $[Ba_2(H_2O)_9][GeSe_4]$ with $MnCl_2\cdot 4H_2O$ in aqueous or H_2O /methanol solutions yielded compounds $[Ba_3(H_2O)_{15}][Mn_6(H_2O)_6(\mu_6\text{-Se})(GeSe_4)_4]\cdot 9H_2O$ (2) or $[Ba_3(H_2O)_{16}][Mn_6(H_2O)_3(\mu_6\text{-Se})(GeSe_4)_4]$ (3) containing novel M/14/16 anions.

6011





Hierarchically organized architecture of potassium hydrogen phthalate and poly(acrylic acid): toward a general strategy for biomimetic crystal design

Yuya Oaki and Hiroaki Imai*

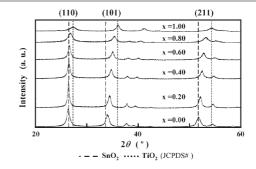
A hierarchically organized architecture in multiple scales emerged from potassium hydrogen phthalate crystals and poly(acrylic acid). The exquisite interaction between crystal and polymer led to the hierarchical crystal growth.

6014

Crystal growth of metastable rutile-type $Ti_rSn_{1-r}O_2$ solid solutions in an aqueous system

Hiroaki Uchiyama and Hiroaki Imai*

Nanoscale grains of rutile-type $Ti_xSn_{1-x}O_2$ were directly grown over the whole range of [Ti]/([Ti] + [Sn]) ratio in aqueous solutions. The structures and properties of the solid solutions depended on the composition.



6017



Ir-catalyzed almost perfect enantioselective synthesis of helical polyaryls based on an axially-chiral sequence

Takanori Shibata* and Kyoji Tsuchikama

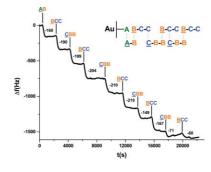
Ir-catalyzed [2 + 2 + 2] cycloaddition of tetraynes or an octayene with monoalkynes provides helically-chiral polyaryls in an almost enantiomerically pure form.

6020

45- and 70-Base DNA supramolecular polymerizations on quartz crystal microbalance biosensor

Mathieu Lazerges, Hubert Perrot,* Niriniony Rabehagasoa, Elisabeth Antoine and Chantal Compere

Supramolecular polymerizations of 45- and 70-base DNAs on the surface of an *in-situ* time-resolved 7 MHz quartz crystal microbalance biosensor.



6023



Isolation and structure of a silicocationic species with 1,3-aryl bridging between silicon atoms: a bis-silylated benzenium ion or a bridging Ph group?

Nicholas Choi, Paul D. Lickiss,* Mary McPartlin, Phindile C. Masangane and Guilaine L. Veneziani

Hydride abstraction from $(Me_3Si)_3CSiMePhH$ affords the cation $[(Me_3Si)_2CSiMe_2-Ph-SiMe_2]^+$, which is shown by X-ray crystallography to contain the first structurally characterised example of a Ph group bridging between two silicon atoms.

$$(Me_3Si)_2C$$
 Si
 Me_2
 Si
 Me_2

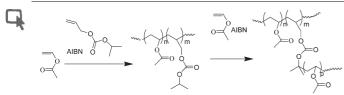
6026

Enantioselective hydrogenation of polar substrates in inverted supercritical CO₂/aqueous biphasic media

Katja Burgemeister, Giancarlo Franciò, Herbert Hugl and Walter Leitner*

An inverted supercritical CO₂/aqueous biphasic catalytic system allows highly enantioselective hydrogenation of polar water-soluble substrates and efficient recycling of the CO₂-philic catalysts.

6029

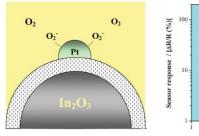


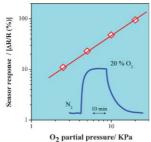
Preparation of highly branched poly(vinyl acetate) by transfer to allylic carbonate comonomers

Stephen Rimmer,* Stephen Collins and Prodip Sarker

The copolymerization of allyl isopropyl carbonates with vinyl acetate produces branched polymers. The branching is controlled by transfer to the isopropyl group of the carbonate comonomer.

6032



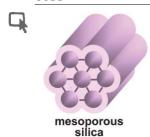


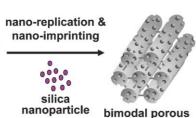
A highly sensitive oxygen sensor operating at room temperature based on platinum-doped In₂O₃ nanocrystals

Giovanni Neri, Anna Bonavita, Giuseppe Micali,* Giuseppe Rizzo, Signorino Galvagno, Markus Niederberger and Nicola Pinna

Semiconducting n-type Pt-doped $\rm In_2O_3$ nanocrystals, synthesised by a nonaqueous sol-gel method, have been used to develop an oxygen sensor showing unique high sensitivity towards oxygen at room temperature.

6035





carbon

Rational design of ordered mesoporous carbon with controlled bimodal porosity *via* dual silica templating route

Hyung Ik Lee, Chanho Pak, Chae-Ho Shin, Hyuk Chang, Doyoung Seung, Jae Eui Yie and Ji Man Kim*

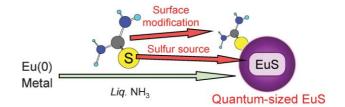
Combining both nano-replication and nano-imprinting techniques using dual silica templates provides a simple way to synthesize ordered mesoporous carbons with bimodal pore size distributions (\sim 1.5 nm and \sim 3.5 nm).

6038

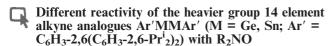
Size-controlled synthesis of quantum-sized EuS nanoparticles and tuning of their Faraday rotation peak

Tomoharu Kataoka, Yasunori Tsukahara, Yasuchika Hasegawa and Yuji Wada*

Size-controlled EuS nanoparticles were synthesized by reaction of europium metal with thiourea, giving Faraday rotation peaks adjustable by control of the EuS particle size.



6041



Geoffrey H. Spikes, Yang Peng, James C. Fettinger, Jochen Steiner and Philip P. Power*

The reactions of the digermanium and ditin alkyne analogues Ar'MMAr' (M = Ge or Sn) with R_2NO ,

 $(R_2NO = Me_2\dot{C}(CH_2)_3CMe_2\dot{N}O \text{ or } N_2O)$, result in complete MM bond cleavage to afford the germylene :Ge(Ar')ONR2 or the germanium(II) or tin(II) hydroxides $\{M(Ar')(\mu-OH)\}_2$.

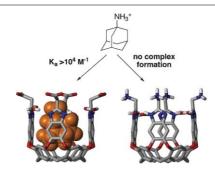
Ar'GeGeAr'

6044

Binding properties of cavitands in aqueous solution—the influence of charge on guest selectivity

Clemens H. Haas, Shannon M. Biros and Julius Rebek, Jr.*

The charge of the upper rim has influences on the binding properties of cavitands in aqueous media.



ADDITIONS AND CORRECTIONS

6046

Synthesis of single-phase anatase nanocrystallites at near room temperatures

Walid A. Daoud and John H. Xin

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