IN THIS ISSUE

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In this issue..

The feature article reviews recent achievements in the area of targeted synthesis of porphyrin-based framework solids by various non-covalent mechanisms of molecular recognition. See Israel Goldberg, pp. 1243–1254.



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Cover

Artistic interpretation of the selfassociation of tetra(thiafulvalene-crownether)-substituted phthalocyanine building blocks. See page 1255. Image reproduced by permission of Professor Roeland J. M. Nolte.

FEATURE ARTICLE

1243

Crystal engineering of porphyrin framework solids

Israel Goldberg

Crystal engineering of extended solids from porphyrin building blocks by self-assembly has led us to a variety of supramolecular framework materials with tunable pore structure, aiming mainly at the formulations of molecular sieves and zeolite analogs. This review surveys some practical strategies and effective cooperative mechanisms for the rational design of open porphyrin frameworks with diverse topologies, through systematic variation of the porphyrin platform and application of suitable templates.

COMMUNICATIONS

1255

Chiral molecular tapes from novel tetra(thiafulvalenecrown-ether)-substituted phthalocyanine building blocks

Joseph Sly, Peter Kasák, Elba Gomar-Nadal, Concepció Rovira, Lucía Górriz, Pall Thordarson, David B. Amabilino,* Alan E. Rowan* and Roeland J. M. Nolte*

A tetra(crown-ether) phthalocyanine (Pc) appended with four tetrathiafulvalene (TTF) units has been shown to self-assemble into helical tapes several micrometers in length.





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1258

Molecular chairs, zippers, zigzag and helical chains: chemical enumeration of supramolecular isomerism based on a predesigned metal-organic building-block

Jie-Peng Zhang, Yan-Yong Lin, Xiao-Chun Huang and Xiao-Ming Chen*

A predesigned metal–organic building-block $[Cu^{1}(2-pytz)]$ (2-Hpytz = 3,5-di-2-pyridyl-1,2,4-triazole) has been successfully used to synthesize four genuine supramolecular isomers.

1261

Supramolecular assemblies of DNA with neutral nucleoside amphiphiles

Philippe Barthelemy, Carla A. H. Prata, Shaun F. Filocamo, Chad E. Immoos, Benjamin W. Maynor, S. A. Nadeem Hashmi, Stephen J. Lee and Mark W. Grinstaff*

A neutral uridine-based amphiphile is described which condenses plasmid DNA. AFM studies show that the three distinct structural components of the amphiphile (*i.e.*, nucleobase, alkyl chains, and poly(ethylene glycol)) are required for the formation of DNA-amphiphile supramolecular assemblies on a mica surface.

1264

Fine-tuning the degree of organic functionalization of mesoporous silica nanosphere materials *via* an interfacially designed co-condensation method

Daniela R. Radu, Cheng-Yu Lai, Jianguo Huang, Xu Shu and Victor S.-Y. Lin*

A synthetic method that can fine tune the amount of chemically accessible organic functional groups of MCM-41 type mesoporous silica nanosphere (MSN) materials has been developed by electrostatically matching various anionic organoalkoxysilanes with the cationic CTAB micelles in a base-catalyzed condensation reaction of tetraethoxysilane.

1267

An unprecedented process involving normal and redox transmetallation reactions between Hg and Pt affording the unexpected K[Pt₂{CH₂C(O)Me}₆(μ -Cl)₃] complex: the key role of X-ray powder diffraction in unravelling its nature and structure

José Vicente,* Aurelia Arcas, Jesús M. Fernández-Hernández, Angelo Sironi and Norberto Masciocchi

By reacting Zeise's salt with $[Hg{CH_2C(O)Me}_2]$ a monoacetonyl platinum(II) or a tris(acetonyl) platinum(IV) species can be isolated.















Fluorescent

Nonfluorescent

Mutual structure-directing effects of a non-interpenetrated square grid coordination polymer and its complementary complex anion net

Leslie J. May and George K. H. Shimizu*

The cationic grid, $\{[Cu(1,2-bis(4-pyridylethane)_2)(H_2O)_2]^{2+}]_{\mu}$, and the complex anion, $Cu(4-pySO_3)_4(H_2O)_2]^{2-}$, neither of which have been previously observed, form a perfectly complementary supramolecular pair with respect to charge and H-bonding, to mutually stabilize each others formation.

Mannose-substituted PPEs detect lectins: A model for Ricin sensing

Ik-Bum Kim, James N. Wilson and Uwe H. F. Bunz*

A novel mannose-substituted and water soluble poly(*para* phenyleneethynylene) has been prepared as a highly effective fluorescent sensor that detects the sugar binding protein (lectin) Concanavalin A by a quenching assay.

Inclusion of C_{60} into an adjustable porphyrin dimer generated by dynamic disulfide chemistry

Amy L. Kieran, Sofia I. Pascu, Thibaut Jarrosson and Jeremy K. M. Sanders*

A new, highly flexible porphyrin dimer was isolated from a dynamic disulfide library; this receptor adjusts to fit guests with a wide range of steric requirements

1279

1276





Supramolecular porphyrin–fullerene *via* 'two-point' binding strategy: Axial-coordination and cation–crown ether complexation

Francis D'Souza,* Raghu Chitta, Suresh Gadde, Melvin E. Zandler, Atula S. D. Sandanayaka, Yasuyuki Araki and Osamu Ito*

A highly stable porphyrin–fullerene conjugate with defined distance and orientation, was formed using a newly developed 'two-point' binding strategy involving axial-coordination and cation–crown ether complexation.

1282

9 Fast and mild palladium(II)-catalyzed 1,4-oxidation of 1,3-dienes via activation of molecular oxygen with a designed cobalt(II) porphyrin

Renzo C. Verboom, Vincent F. Slagt and Jan-E. Bäckvall*

The use of Co(porphyrin)-amide ligand 2 in the palladium(II)-catalyzed 1,4-diacetoxylation of conjugated dienes under O2 results in aerobic oxidation. The reaction can also be performed under air.

1285

A switchable macrocycle-clip complex that functions as a NOR logic gate

Pin-Nan Cheng, Pinn-Tsong Chiang and Sheng-Hsien Chiu*

A new molecular switch based on a macrocycle-clip complex can be operated as a two-input NOR functioning molecular logic gate.

1288

Switchable rewritability of Ag–TiO₂ nanocomposite films with multicolor photochromism

Kenji Naoi, Yoshihisa Ohko and Tetsu Tatsuma*

The photochromism and rewritability of Ag-TiO2 films were deactivated by modification with thiols to make it possible to retain color images displayed on the films, while the deactivated properties were fully reactivated by UV-irradiation.

1291

One teflon[®]-like channelled nanoporous polymer with a chiral and new uninodal 4-connected net: sorption and catalytic properties

Angeles Monge,* Natalia Snejko, Enrique Gutiérrez-Puebla, Manuela Medina, Concepción Cascales, Caridad Ruiz-Valero, Marta Iglesias and Berta Gómez-Lor

 $Zn(C_{17}H_8F_6O_4)$ is the first example of a fluoro-lined nanotube organo-inorganic 3D polymeric chiral compound, which possesses two types of isolated channels. The structure is a new uninodal 4-connected net and the compound exhibits selective sorption and catalytic chiral recognition properties.









1294

1300







R-OMe



Structure controlled self-assembly of Cu(II) salicylic aldehyde and aldimine derivative complexes

Philipp Zell, Florian Mögele, Ulrich Ziener and Bernhard Rieger*

The orientation of metal complexes in 2D surface structures and hence the relative distances between the individual Cu(II) ions have been controlled by ligand design as a tool for the "fine-tuning" of intermolecular interactions.

Silyl methallylsulfinates: efficient and powerful agents for the chemoselective silylation of alcohols, polyols, phenols and carboxylic acids

Xiaogen Huang, Cotinica Craita, Loay Awad and Pierre Vogel*

Alcohols, phenols and carboxylic acids are silylated with very good yield in the presence of silyl methallylsulfinates under non-basic conditions and with the formation of volatile co-products.

π -Indenyl tin(II) and lead(II) compounds

Jamie N. Jones and Alan H. Cowley

The first π -indenyl tin(II) complexes are reported; the bis(π -indenyl) tin(II) derivative has an essentially parallel arrangement of indenyl rings.



ca. 82

Microflow electroorganic synthesis without supporting electrolyte

Roberto Horcajada, Masayuki Okajima, Seiji Suga and Jun-ichi Yoshida*

Anodic methoxylation of several organic compounds has been successfully achieved in the absence of intentionally added supporting electrolyte using an electrochemical microflow system.

1306



Erhong Hao and M. Graça H. Vicente*

The high yield synthesis of two new porphyrincobaltacarborane conjugates is described. These conjugates of high boron content display spectroscopic properties characteristic of porphyrin macrocycles and may have application in the BNCT treatment of tumors.

1309

A facile and versatile preparation of bilindiones and biladienones from tetraarylporphyrins

Takae Yamauchi, Tadashi Mizutani,* Kenji Wada, Shoji Horii, Hirotaka Furukawa, Shigeyuki Masaoka, Ho-Chol Chang and Susumu Kitagawa

Coupled oxidation of iron tetraarylporphyrins with dioxygen and ascorbic acid afforded meso-arylbilindiones and meso-arylbiladienones in 20-63% yield. The crystal structure of meso-triphenylbilindione showed that it has larger helix pitch than β -octaethylbilindione.

1312

An unexpected bonding interaction between d_{xy} and a_{1y} orbitals mediated by porphyrin deformation

Ru-Jen Cheng,* Yen-Ku Wang, Ping-Yu Chen, Ya-Ping Han and Chih-Ching Chang

The extraordinary bonding interaction between d_{xy} and a_{1u} orbitals in the saddle-shaped [Fe(OETPP)(THF)₂]⁺ complex offers a novel symmetry-controlled mechanism for the formation of the unusual intermediate-spin electronic structure $(d_{xz}d_{yz})^3(d_{xy})^1(d_z2)^1$ that is consistent with the reported NMR data.

1315

An efficient synthesis of propargylamines via C-H activation catalyzed by copper(I) in ionic liquids

Soon Bong Park and Howard Alper*

An efficient three-component coupling of aldehydes with amines and alkynes to form propargylamines has been developed via Č-H activation catalyzed by a copper(I) compound in an ionic liquid; the catalysts were recycled five times without any significant loss of catalytic activity.





R₂R₃NH $R_2 R_3$ [bmim]PF₆ R1CHO Cu cat. -R4 R_4







groups.

1330

Electrodeposition of ferrocenoyl peptide disulfides

Grzegorz A. Orlowski, Somenath Chowdhury, Yi-Tao Long, Todd C. Sutherland and Heinz-Bernhard Kraatz*

Using electrodeposition of cyclic and acyclic Fc–peptide disulfides tightly-packed Fc–peptide monolayers were conveniently formed, which exhibit significant differences in their electron transfer kinetics.

1333

Anisotropic ion conduction in a unique smectic phase of self-assembled amphiphilic ionic liquids

Tomohiro Mukai, Masafumi Yoshio, Takashi Kato, Masahiro Yoshizawa and Hiroyuki Ohno*

Anisotropic ion conduction was observed in a supercooled smectic phase composed of crystallized alkyl chains and fused ionic layer.





1336

Asymmetric alkylation of dimethoxyphosphoryl-N-[1-(S)- α -methylbenzyl]acetamide enolates. Synthesis of both stereoisomers from the same source of chirality by changing the equivalents of LDA

Mario Ordóñez,* Eugenio Hernández-Fernández, Janet Xahuentitla and Carlos Cativiela

A new methodology has been developed for the synthesis of both stereoisomers from a single chiral source.

1339



Robert J. Baker, Cameron Jones* and Damien M. Murphy

The first gallyl–Group 4 complex has been prepared *via* an unusual reaction involving an unprecedented oxidative insertion of a transition metal centre into a digallane(4).





1348

1351

F₂C

Q





R3 R2

Fac SO

H₃C(0)C

A pyrimidine-like nickel(II) DNA base pair

Christopher Switzer* and Dongwon Shin

We report an improbable naturally inspired DNA self-pair based on a pyrimidine scaffold. The $Pyr^{p} \cdot Ni^{2+} \cdot Pyr^{p}$ metallo base-pair ($Pyr^{p} = 4-(2'-pyridyl)$ -pyrimidinone) has mismatch discrimination and stability on a par with natural Watson– Crick base-pairs despite assuming a base–base distance predicted to be half the corresponding natural dimension.

Electrosynthesis of hydrogen peroxide in room temperature ionic liquids and *in situ* epoxidation of alkenes

Michael Chi-Yung Tang, Kwok-Yin Wong and Tak Hang Chan*

Hydrogen peroxide can be electrosynthesized from oxygen in [bmim][BF₄]-water or 0.04M NaOH and used *in situ* for the epoxidation of alkenes.

The first 4,1,10-MC₂B₁₀ supraicosahedral metallacarboranes and a route to previously inaccessible 4,1,12-ruthenium arene species

David Ellis, Maria Elena Lopez, Ruaraidh McIntosh, Georgina M. Rosair, Alan J. Welch* and Romain Quenardelle

Reduction and subsequent metallation of 1,12-*closo*- $C_2B_{10}H_{12}$ affords the first examples of 4,1,10- MC_2B_{10} supraicosahedral metallacarboranes: these undergo quantitative isomerisation to the corresponding 4,1,12 isomers on heating.

1-Triflylpyrroles as efficient dienophiles in normal electron demand [4+2] cycloaddition reactions under pressure

Antony Chrétien, Isabelle Chataigner and Serge R. Piettre*

1-Triflylpyrroles bearing acetyl group(s) on position 3, or 2 and 4, are efficient dienophiles in normal electron demand Diels–Alder reactions activated by high pressures and Lewis acids.

1354



Nakia Maulucci, Francesco De Riccardis,* Cinzia Barbara Botta, Agostino Casapullo, Elena Cressina, Massimo Fregonese, Paolo Tecilla* and Irene Izzo*

The synthesis of a new class of amphiphilic calix[4]arene-based ionophores, relying on direct reductive amination as a key step, and the evaluation of their H^+ and Na^+ transporting properties is described.





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AUTHOR INDEX

Alper, Howard, 1315 Amabilino, David B., 1255 Amat, Mercedes, 1327 Araki, Yasuyuki, 1279 Arcas, Aurelia, 1267 Awad, Loay, 1297 Bäckvall, Jan-E., 1282 Baker, Robert J., 1339 Barthelemy, Philippe, 1261 Bassas, Oriol, 1327 Bernardi, Luca, 1324 Bosch, Joan, 1327 Botta, Cinzia Barbara, 1354 Bunz, Uwe H. F., 1273 Casapullo, Agostino, 1354 Cascales, Concepción, 1291 Cativiela, Carlos, 1336 Chan, Tak Hang, 1345 Chang, Chih-Ching, 1312 Chang, Ho-Chol, 1309 Chataigner, Isabelle, 1351 Chen, Ping-Yu, 1312 Chen, Xiao-Ming, 1258 Cheng, Pin-Nan, 1285 Cheng, Ru-Jen, 1312 Chiang, Pinn-Tsong, 1285 Chitta, Raghu, 1279 Chiu, Sheng-Hsien, 1285 Chowdhury, Somenath, 1330 Chrétien, Antony, 1351 Chung, Young Keun, 1321 Cowley, Alan H., 1300 Craita, Cotinica, 1297 Cressina, Elena, 1354 De Riccardis, Francesco, 1354 D'Souza, Francis, 1279 Ellis, David, 1348 Fernández-Hernández, Jesús M., 1267

Filocamo, Shaun F., 1261 Fregonese, Massimo, 1354 Furukawa, Hirotaka, 1309 Gadde, Suresh, 1279 Goldberg, Israel, 1243 Gomar-Nadal, Elba, 1255 Gómez-Lor, Berta, 1291 Górriz, Lucía, 1255 Grinstaff, Mark W., 1261 Gutiérrez-Puebla, Enrique, 1291 Han, Ya-Ping, 1312 Hao, Erhong, 1306 Hashmi, S. A. Nadeem, 1261 Hernández-Fernández, Eugenio, 1336 Horcajada, Roberto, 1303 Horii, Shoji, 1309 Huang, Jianguo, 1264 Huang, Xiao-Chun, 1258 Huang, Xiaogen, 1297 Iglesias, Marta, 1291 Immoos, Chad E., 1261 Ito, Osamu, 1279 Izzo, Irene, 1354 Jarrosson, Thibaut, 1276 Jones, Cameron, 1339 Jones, Jamie N., 1300 Jørgensen, Karl Anker, 1324 Kasák, Peter, 1255 Kato, Takashi, 1333 Kieran, Amy L., 1276 Kim, Ik-Bum, 1273 Kim, So Yeon, 1321 Kitagawa, Susumu, 1309 Kraatz, Heinz-Bernhard, 1330 Lai, Cheng-Yu, 1264 Lee, Stephen J., 1261

Lin, Victor S.-Y., 1264 Lin, Yan-Yong, 1258 Loh, Teck-Peng, 1318 Long, Yi-Tao, 1330 Lopez, Maria Elena, 1348 Masaoka, Shigeyuki, 1309 Masciocchi, Norberto, 1267 Maulucci, Nakia, 1354 May, Leslie J., 1270 Maynor, Benjamin W., 1261 McIntosh, Ruaraidh, 1348 Medina, Manuela, 1291 Mizutani, Tadashi, 1309 Mögele, Florian, 1294 Monge, Angeles, 1291 Mukai, Tomohiro, 1333 Murphy, Damien M., 1339 Naoi, Kenji, 1288 Nolte, Roeland J. M., 1255 Ohko, Yoshihisa, 1288 Ohno, Hiroyuki, 1333 Okajima, Masayuki, 1303 Ordóñez, Mario, 1336 Orlowski, Grzegorz A., 1330 Park, Kang Hyun, 1321 Park, Soon Bong, 1315 Pascu, Sofia I., 1276 Pastó, Mireia, 1327 Pericàs, Miquel A., 1327 Piettre, Serge R., 1351 Prata, Carla A. H., 1261 Quenardelle, Romain, 1348 Radu, Daniela R., 1264 Rieger, Bernhard, 1294 Rosair, Georgina M., 1348 Rovira, Concepció, 1255 Rowan, Alan E., 1255 Ruiz-Valero, Caridad, 1291 Sandanayaka, Atula S. D., 1279 Sanders, Jeremy K. M., 1276 Shimizu, George K. H., 1270 Shin, Dongwon, 1342 Shu, Xu, 1264 Sironi, Angelo, 1267 Slagt, Vincent F., 1282 Sly, Joseph, 1255 Snejko, Natalia, 1291 Suga, Seiji, 1303 Sutherland, Todd C., 1330 Switzer, Christopher, 1342 Tan, Kui-Thong, 1318 Tang, Michael Chi-Yung, 1345 Tatsuma, Tetsu, 1288 Tecilla, Paolo, 1354 Teo, Yong-Chua, 1318 Thordarson, Pall, 1255 Verboom, Renzo C., 1282 Vicente, José, 1267 Vicente, M. Graça H., 1306 Vogel, Pierre, 1297 Wada, Kenji, 1309 Wang, Yen-Ku, 1312 Welch, Alan J., 1348 Wilson, James N., 1273 Wong, Kwok-Yin, 1345 Xahuentitla, Janet, 1336 Yamauchi, Takae, 1309 Yoshida, Jun-ichi, 1303 Yoshio, Masafumi, 1333 Yoshizawa, Masahiro, 1333 Zandler, Melvin E., 1279 Zell, Philipp, 1294 Zhang, Jie-Peng, 1258 Ziener, Ulrich, 1294

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