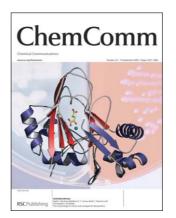
IN THIS ISSUE

ISSN 1359-7345 CODEN CHCOFS (34) 4237-4368 (2005)



Cover

See Takashi Jin, Fumihiko Fujii, Hiroshi Sakata, Mamoru Tamura and Masataka Kinjo, page 4300. New functional quantum dots can be used for the detection of the neurotransmitter acetylcholine. Image reproduced by permission of Takashi Jin *et al.*, from *Chem. Commun.*, 2005, 4300.



Inside cover See Nadia J. Kershaw, Matthew E. C. Caines, Mark C. Sleeman and Christopher J. Schofield, page 4251. The enzymology of clavam and carbapenem biosynthesis. Image reproduced by permission of Christopher J. Schofield *et al.*, from *Chem. Commun.*, 2005, 4251.

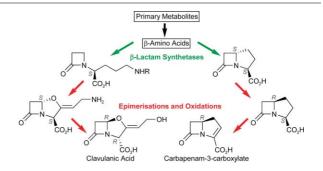
FEATURE ARTICLE

4251

The enzymology of clavam and carbapenem biosynthesis

Nadia J. Kershaw, Matthew E. C. Caines, Mark C. Sleeman and Christopher J. Schofield*

The enzymology of clavam and carbapenem biosynthesis is reviewed. The common role of unusual oxidation and epimerisation reactions is highlighted.



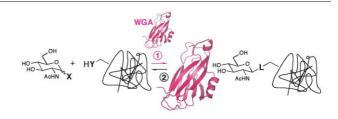
COMMUNICATIONS

4264

Ligand amplification in a dynamic combinatorial glycopeptide library

Tom Hotchkiss, Holger B. Kramer, Katie J. Doores, David P. Gamblin, Neil J. Oldham and Benjamin G. Davis*

N-acetyl glucosamine binding protein amplifies the concentration of one member in a dynamic combinatorial glycopeptide library based on exchanging disulfides.



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4267

Aptamer biosensor for label-free impedance spectroscopy detection of proteins based on recognition-induced switching of the surface charge

Marcela C. Rodriguez, Abdel-Nasser Kawde and Joseph Wang*

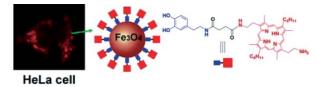
The aptamer-protein binding event reverses the electrode charge from negative to positive, leading to attraction of the previously repelled negatively-charged marker towards the electrode surface decreasing the interfacial electron transfer.

4270

Synthesis and cellular uptake of porphyrin decorated iron oxide nanoparticles—a potential candidate for bimodal anticancer therapy

Hongwei Gu, Keming Xu, Zhimou Yang, Chi K. Chang and Bing Xu*

Using a dopamine-based anchor to covalently attach porphyrin derivatives to iron oxide nanoparticles may lead to a simple and general pathway to produce a bimodal anticancer agent for both photodynamic therapy and hyperthermia treatment.



4273

Gold nanoparticle-based competitive colorimetric assay for detection of protein-protein interactions

Charng-Sheng Tsai, Ting-Bin Yu and Chao-Tsen Chen*

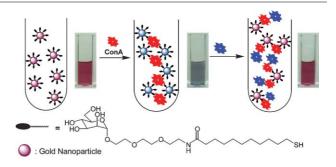
A gold nanoparticle-based competitive colorimetric assay uses the ensemble of Concanavalin (ConA) and mannopyranosideencapsulated gold nanoparticles (Man-GNPs) to identify the binding partners for ConA and the binding constants are determined based on the wavelength shifts.

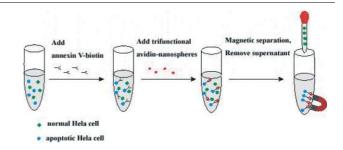
4276

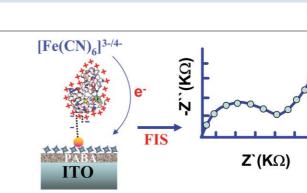
Biofunctionalization of fluorescent-magnetic-bifunctional nanospheres and their applications

Guo-Ping Wang, Er-Qun Song, Hai-Yan Xie, Zhi-Ling Zhang, Zhi-Quan Tian, Chao Zuo, Dai-Wen Pang,* Dao-Cheng Wu and Yun-Bo Shi

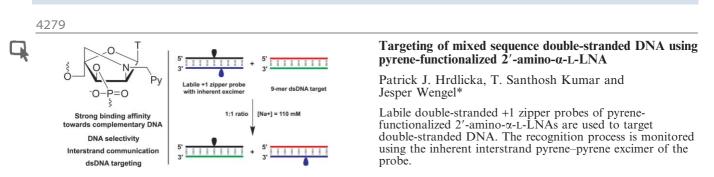
Novel fluorescent-magnetic-biotargeting trifunctional nanospheres with surface-confined IgG, avidin, and biotin were constructed, which can be used in a number of biomedical applications, including visual sorting and manipulation of apoptotic cells as demonstrated here.

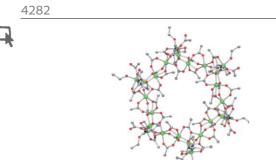












çO Me

 \hat{R}_3

Ŕ3

R

R

Br

 R_1

Me OAc

tsE

tsE

(3S,5aR)

Me

(S,Z)

Br

Rì

tsZ

tsZ

(3R,5aR)

High-nuclearity homometallic iron and nickel clusters: Fe₂₂ and Ni₂₄ complexes from the use of *N*-methyldiethanolamine

Dolos Foguet-Albiol, Khalil A. Abboud and George Christou*

The use of *N*-methyldiethanolamine (mdaH₂) in reactions with Fe(III) and Ni(II) sources has led to very large Fe₂₂ and Ni₂₄ products, possessing S = 0 and S = 6 ground states, respectively.

Mechanistic subtleties in the cyclopentannelation of allenolate allyl carbamates: the origin of the center-to-center chirality transfer

Olalla Nieto Faza, Carlos Silva López, Rosana Álvarez and Ángel R. de Lera*

A theoretical study of the stereospecificity in the cyclization of allenolate allyl carbamates shows that this process, displaying features of pericyclic and ionic mechanisms, is concerted and benefits from $\pi_{allenolate} - \sigma^*_{leaving group}$ charge donation.

4288

4285

B

R

(R,E)

R1 = Me, Ph

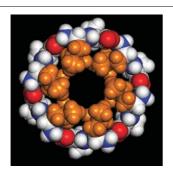
 $\mathbf{R_2} = O(-), OLi$ $\mathbf{R_3} = Me, tBu$

Me

 $\int R_2$

 R_3

q



Microporous organic crystals: an unusual case for L-leucyl–L-serine

Carl Henrik Görbitz,* Mette Nilsen, Kai Szeto and Linda Wibecke Tangen

Cocrystallized acetonitrile solvent molecules located inside 5.2 Å channels in the crystal structure of L-leucyl–L-serine have been replaced by I_2 molecules with full retention of the peptide scaffold.

4291

Dynamic helicity inversion in an octahedral cobalt(II) complex system *via* solvato-diastereomerism

Hiroyuki Miyake,* Hideki Sugimoto, Hitoshi Tamiaki and Hiroshi Tsukube

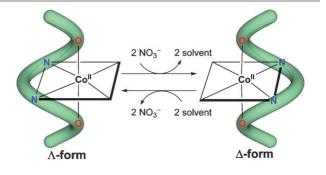
The helical sense of a mononuclear Co(II) complex with an amino acid-based chiral tetradentate ligand was dynamically inverted by changing the solvent component.

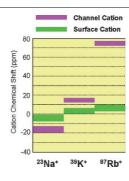


Solid-state ⁸⁷Rb NMR signatures for rubidium cations bound to a G-quadruplex

Ramsey Ida and Gang Wu*

First solid-state ⁸⁷Rb NMR characterization for Rb⁺ cations bound to a G-quadruplex structure.



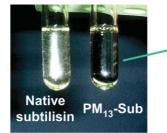


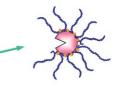
4297

Comb-shaped poly(ethylene glycol)-modified subtilisin Carlsberg is soluble and highly active in ionic liquids

Kazunori Nakashima, Tatsuo Maruyama, Noriho Kamiya and Masahiro Goto*

Subtilisin Carlsberg conjugated with comb-shaped poly(ethylene glycol) exhibits higher transesterification activity in $[\text{Emim}][\text{Tf}_2\text{N}]$ than in organic solvents commonly used for enzymatic biotransformation.





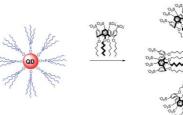
Subtilisin modified with comb-shaped PEG (PM₁₃-Sub)

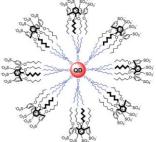
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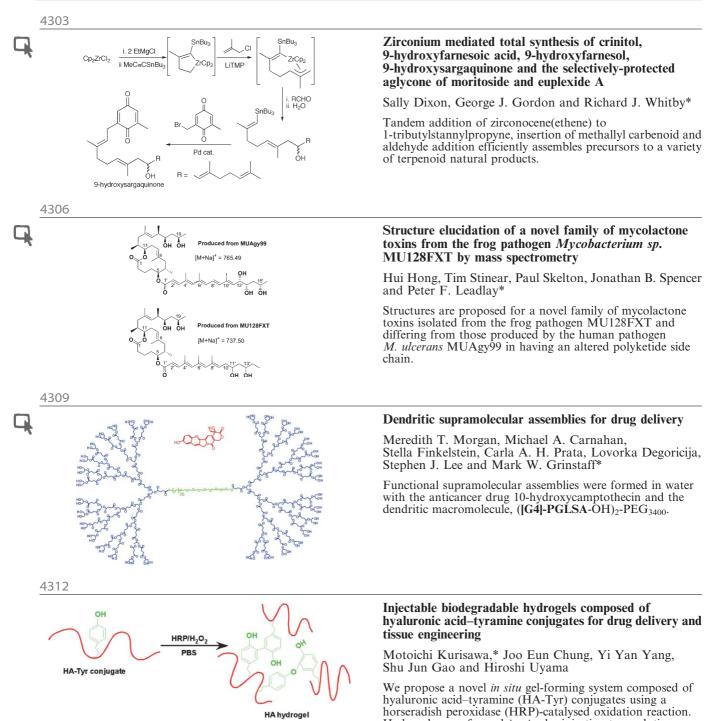
Amphiphilic *p*-sulfonatocalix[4]arene-coated CdSe/ZnS quantum dots for the optical detection of the neurotransmitter acetylcholine

Takashi Jin,* Fumihiko Fujii, Hiroshi Sakata, Mamoru Tamura and Masataka Kinjo

Water-soluble CdSe/ZnS (core-shell) semiconductor quantum dots surface-modified with amphiphilic *p*-sulfonatocalix[4]arene derivatives were synthesized for the optical detection of the neurotransmitter acetylcholine.







hyaluronic acid-tyramine (HA-Tyr) conjugates using a horseradish peroxidase (HRP)-catalysed oxidation reaction. Hydrogels were formed *in vivo* by injecting two solutions through syringes: (i) HA-Tyr solution containing H_2O_2 and (ii) HRP as a catalyst which induces the oxidative coupling of the phenol moiety.

4315

Virus-glycopolymer conjugates by copper(I) catalysis of atom transfer radical polymerization and azide-alkyne cycloaddition

Sayam Sen Gupta, Krishnaswami S. Raja, Eiton Kaltgrad, Erica Strable and M. G. Finn*

The construction of polymer-covered surfaces is made convenient by Cu^{I} catalysis of polymerization, end-labeling, and attachment steps. The example described here is fluorophore-labeled glycopolymer chains on a virus particle scaffold.

4318

Electrochemical detection of *Arachis hypogaea* (peanut) agglutinin binding to monovalent and clustered lactosyl motifs immobilized on a polypyrrole film

Marie-Pierre Dubois, Chantal Gondran, Olivier Renaudet, Pascal Dumy, Hugues Driguez, Sébastien Fort* and Serge Cosnier*

Direct detection of peanut agglutinin/lactose interactions was realized by an electrochemical approach based on a polypyrrole coated electrode displaying pendant carbohydrates.

4321

Simple and quick chemical aminoacylation of tRNA in cationic micellar solution under ultrasonic agitation

Naoto Hashimoto, Keiko Ninomiya, Takamasa Endo and Masahiko Sisido*

Aminoacylation of a tRNA with a non-natural amino acid was achieved by using an *N*-protected amino acid cyanomethyl ester as a substrate solubilized in CTACl micelle under ultrasonic agitation.

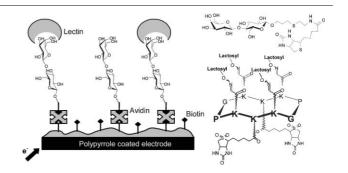
4324

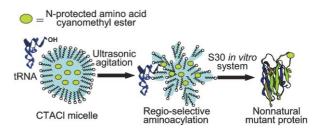
The vicinal $\rm F-C-C-F$ moiety as a tool for influencing peptide conformation

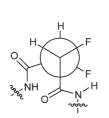
Martin Schüler, David O'Hagan* and Alexandra M. Z. Slawin

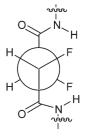
The *erythro-* and *threo-*isomers of bis(amino acid) 2,3difluorosuccinamides display very different conformations. In particular, the *gauche-*preference for the vicinal C–F bonds influences the relative positioning (*gauche vs. anti*) of the amide groups, a property which could find utility in influencing the conformation of peptide mimetics.







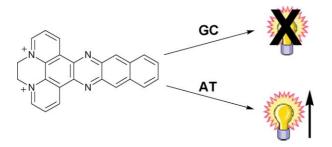




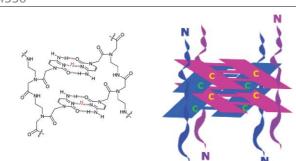
erythro-isomer

threo-isomers

4327







$\mathbf{Q} \xrightarrow{4333} \mathbf{Click}_{\mathsf{N_3}} \xrightarrow{\mathsf{Me}}_{\mathsf{N_2}} \xrightarrow{\mathsf{Click}}_{\mathsf{Chemistry}} \xrightarrow{\mathsf{New}}_{\mathsf{N}} \xrightarrow{\mathsf{Me}}_{\mathsf{N}} \xrightarrow{\mathsf{New}}_{\mathsf{N}} \xrightarrow{\mathsf{New}}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}}_{\mathsf{N}} \xrightarrow{\mathsf{New}}_{\mathsf{N}} \xrightarrow{\mathsf{New}}_{\mathsf{N}} \xrightarrow{\mathsf{New}}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}} \xrightarrow{\mathsf{New}$

Water-soluble organic dppz analogues—tuning DNA binding affinities, luminescence, and photo-redox properties

Tim Phillips, Chatna Rajput, Lance Twyman, Ihtshamul Haq and Jim A. Thomas*

A water-soluble organic anion related to the dppz fragment binds to duplex DNA with an affinity comparable to [Ru^{II}(dppz)] complexes showing a two orders of magnitude preference for GC over AT sequences. Binding to GC sequences is accompanied by luminescent quenching, while binding to AT sequences results in luminescent enhancement.

PNA C–C⁺ *i*-motif: superior stability of PNA TC₈ tetraplexes compared to DNA TC₈ tetraplexes at low pH

Nagendra K. Sharma and Krishna N. Ganesh*

Self-assembly of PNA TC₈ leads to formation of a hitherto unknown C–C⁺ tetraplex (*i*-motif) in acidic pH, with higher stability than the analogous dTC_8 .

Click-chemistry as an efficient synthetic tool for the preparation of novel conjugated polymers

Dirk Jan V. C. van Steenis, Olivier R. P. David, Gino P. F. van Strijdonck, Jan H. van Maarseveen* and Joost N. H. Reek*

We have developed a general efficient route towards conjugated polymers utilizing click chemistry and in the current example fluorene based polymers have been prepared. The pyridyl-triazole units within the polymer can be used for further post-functionalization.

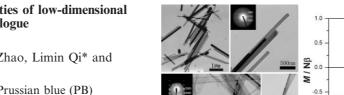
(Z)-Selective cross-dimerization of arylacetylenes with silylacetylenes catalyzed by vinylideneruthenium complexes

Hiroyuki Katayama,* Hiroshi Yari, Masaki Tanaka and Fumiyuki Ozawa*

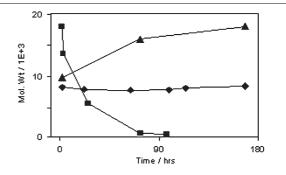
The vinylideneruthenium(II) complex bearing bulky and basic triisopropylphosphine ligands, $RuCl_2(=C=CHPh)(PPr'_3)_2$, serves as a good catalyst precursor for (*Z*)-selective cross-dimerization between arylacetylenes and silylacetylenes in the presence of *N*-methylpyrrolidine.

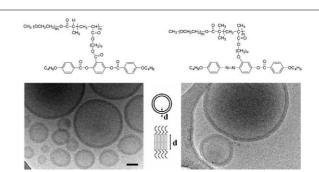
4336 $Ar \longrightarrow + R_3Si \longrightarrow \begin{bmatrix} CI & PPr_3^{i_3} \\ ...,CI \\ PPr_3^{i_3} (5 \text{ mol}\%) \\ ...,CI \\ ...$

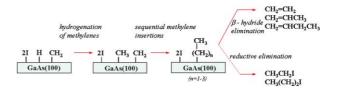
4339



LIE 200m 2







Shape-dependent magnetic properties of low-dimensional nanoscale Prussian blue (PB) analogue $SmFe(CN)_6$ ·4H₂O

Hao-Ling Sun, Hongtao Shi, Fei Zhao, Limin Qi* and Song Gao*

Unique nanorods and nanobelts of Prussian blue (PB) analogue $SmFe(CN)_6$ · $4H_2O$ have been successfully synthesized by using reverse micelles as colloidal soft templates; magnetic studies show that the shape of the material is a dominating factor for its coercivity.

4342

Self-repairing polymers: poly(dioxaborolane)s containing trigonal planar boron

Weijun Niu, Caroline O'Sullivan, Brett M. Rambo, Mark D. Smith and John J. Lavigne*

Poly(dioxaborolane)s containing trigonal planar boron are stable, self-repairing polymers which can be shortened or lengthened post-polymerization. Degraded material is repaired without the need for added catalyst or reintroduction to synthetic conditions.

4345

Polymer vesicles formed by amphiphilic diblock copolymers containing a thermotropic liquid crystalline polymer block

Jing Yang, Daniel Lévy, Wei Deng, Patrick Keller and Min-Hui Li*

Novel amphiphilic diblock copolymers composed of PEG and a thermotropic liquid crystalline polymer have been synthesized using atom transfer radical polymerization. Cryo-electron microscopy studies revealed that these amphiphiles self-assemble in water to form unilamellar vesicles.

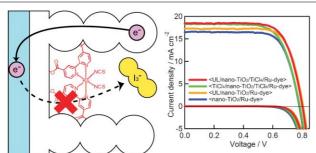
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Evidence of carbon-carbon bond formation on GaAs(100) *via* Fischer-Tropsch methylene insertion reaction mechanism

Neil T. Kemp and Nagindar K. Singh*

Sequential multiple methylene (CH₂) insertions into adsorbed methyl species on clean gallium-rich GaAs(100)-(4 \times 1) occur to form higher alkenes (ethene, propene, butene) and two higher alkyl iodides (iodoethane, iodopropane), not reported for a semiconductor surface previously.



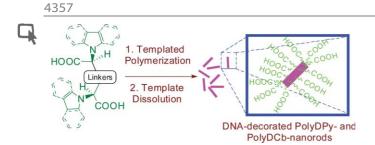


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trans-[Pt(pyr)2(9-EtGua)Cl]

Pt migration

Control of dark current in photoelectrochemical $(TiO_2/I^--I_3^-)$ and dye-sensitized solar cells

Seigo Ito,* Paul Liska, Pascal Comte, Raphaël Charvet, Peter Péchy, Udo Bach, Lukas Schmidt-Mende, Shaik Mohammed Zakeeruddin, Andreas Kay, Mohammad K. Nazeeruddin and Michael Grätzel

The charge-recombination blocking effects of Ru dye, a TiCl₄ treatment and compact TiO₂ underlayer have been clarified in dye-sensitized solar cells. Controlling the dark current achieved impressive efficiency (10.8%).

Photostability of a highly luminescent europium β-diketonate complex in imidazolium ionic liquids

Peter Nockemann, Eva Beurer, Kris Driesen, Rik Van Deun, Kristof Van Hecke, Luc Van Meervelt and Koen Binnemans*

Solutions of a luminescent europium(III) tetrakis(β -diketonate) complex in imidazolium ionic liquids have a higher photostability than solutions of the same complex in classic organic solvents.

Polydipyrrole- and polydicarbazole-nanorods as new nanosized supports for DNA hybridization

Jean-Paul Lellouche,* Senthil Govindaraji, Augustine Joseph, Jyongsik Jang* and Kyung Jin Lee

AAO template-synthesized novel functional COOH polydipyrrole- and polydicarbazole nanorods were tested for covalent DNA attachment and hybridization.

Thiolate-bridged heterodinuclear platinum-zinc chelates as models for ternary platinum-DNA-protein complexes and zinc ejection from zinc fingers. Evidence from studies using ESI-mass spectrometry

Qin Liu, Melissa Golden, Marcetta Y. Darensbourg and Nicholas Farrell*

Structures for models of ternary platinum–DNA–protein complexes and zinc ejection from zinc fingers by platinum were deduced from the ESI-MS spectra of the interaction of model Zn and Pt complexes.

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