Organic & Biomolecular Chemistry

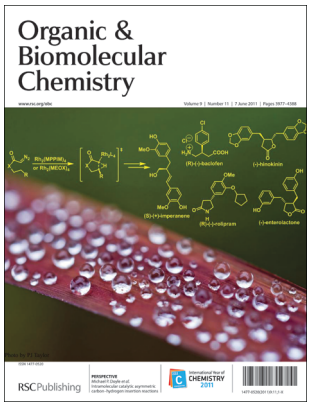
An international journal of synthetic, physical and biomolecular organic chemistry

www.rsc.org/obc

RSC Publishing is a not-for-profit publisher and a division of the Royal Society of Chemistry. Any surplus made is used to support charitable activities aimed at advancing the chemical sciences. Full details are available from www.rsc.org

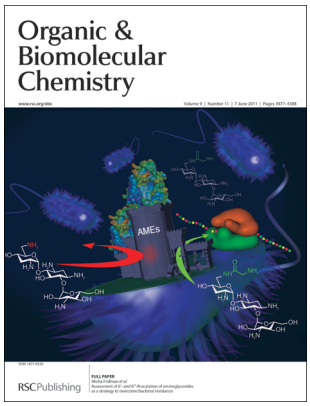
IN THIS ISSUE

ISSN 1477-0520 CODEN OBCRAK 9(11) 3977–4388 (2011)



Cover
See Michael P. Doyle *et al.*, pp. 4007–4016.

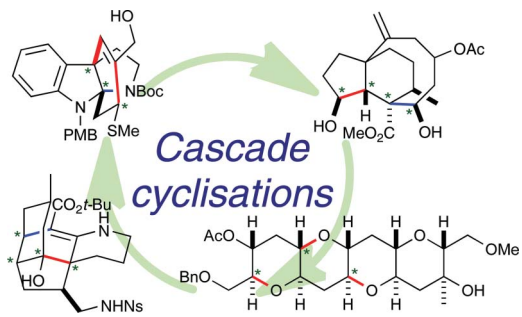
Image reproduced by permission of Michael P. Doyle from *Org. Biomol. Chem.*, 2011, **9**, 4007.



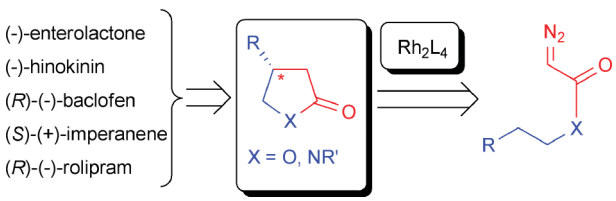
Inside cover
See Micha Fridman *et al.*, pp. 4057–4063.
Aminoglycoside acetyltransferase AAC(6′) belonging to the aminoglycoside-modifying enzymes (AMEs) does not deactivate the 6′-*N*-glycinyl tobramycin which attacks the bacterial ribosome (green arrow). In contrast, the parent aminoglycoside tobramycin cannot pass through bacterial resistance mechanism (red arrow).

Image reproduced by permission of Micha Fridman from *Org. Biomol. Chem.*, 2011, **9**, 4057.

EMERGING AREA



PERSPECTIVE



Organic & Biomolecular Chemistry (print: ISSN 1477-0520; electronic: ISSN 1477-0539) is published 24 times a year by the Royal Society of Chemistry, Thomas Graham House, Science Park, Milton Road, Cambridge, UK CB4 0WF. All orders, with cheques made payable to the Royal Society of Chemistry, should be sent to RSC Distribution Services, c/o Portland Customer Services, Commerce Way, Colchester, Essex, UK CO2 8HP. Tel +44 (0)1206 226050; E-mail sales@rscdistribution.org

2011 Annual (print+electronic) subscription price: £3726; US$6955. 2011 Annual (electronic) subscription price: £3353; US$6260 Customers in Canada will be subject to a surcharge to cover GST. Customers in the EU subscribing to the electronic version only will be charged VAT. If you take an institutional subscription to any RSC journal you are entitled to free, site-wide web access to that journal. You can arrange access via Internet Protocol (IP) address at www.rsc.org/ip. Customers should make payments by cheque in sterling payable on a UK clearing bank or in US dollars payable on a US clearing bank. Periodicals postage paid at Rahway, NJ, USA and at additional mailing offices. Airfreight and mailing in the USA by Mercury Airfreight International Ltd., 365 Blair Road, Avenel, NJ 07001, USA.

US Postmaster: send address changes to Organic & Biomolecular Chemistry (OBC) c/o Mercury Airfreight International Ltd., 365 Blair Road, Avenel, NJ 07001. All despatches outside the UK by Consolidated Airfreight.

Organic & Biomolecular Chemistry

An international journal of synthetic, physical and biomolecular organic chemistry

www.rsc.org/obc

Organic & Biomolecular Chemistry brings together molecular design, synthesis, structure, function and reactivity in one journal. It publishes fundamental work on synthetic, physical and biomolecular organic chemistry as well as all organic aspects of: chemical biology, medicinal chemistry, natural product chemistry, supramolecular chemistry, macromolecular chemistry, theoretical chemistry, and catalysis.

INFORMATION FOR AUTHORS

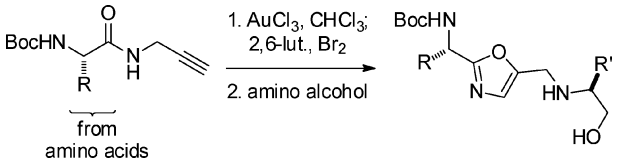
Full details on how to submit material for publication in Organic & Biomolecular Chemistry are given in the Instructions for Authors (available from http://www.rsc.org/authors). Submissions should be made *via* the journal's homepage: http://www.rsc.org/obc.

4017

Gold-catalysed synthesis of amino acid-derived 2,5-disubstituted oxazoles

Christopher L. Paradise, Pooja R. Sarkar, Mina Razzak and Jef K. De Brabander*

We describe a tandem gold(III)-catalysed cycloisomerisation–bromination of propargylic amides, followed by further processing into valuable *bis*-heterocyclic compounds including novel dipeptide mimetics.

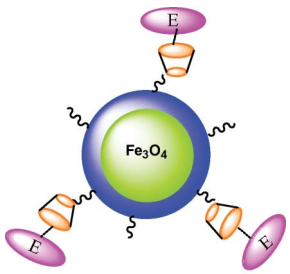


4021

Improvement of catalytic properties of *Candida Rugosa* lipase by sol–gel encapsulation in the presence of magnetic calix[4]arene nanoparticles

Serkan Sayin, Elif Yilmaz and Mustafa Yilmaz

Candida rugosa lipase (CRL) was encapsulated within a chemically inert sol-gel support prepared by polycondensation with tetraethoxysilane and octyltriethoxysilane in the presence of *N*-methylglucamine based calix[4]arene magnetic nanoparticles. The results indicate that the magnetic calix[4]arene based encapsulated lipase showed particularly high conversion and enantioselectivity.



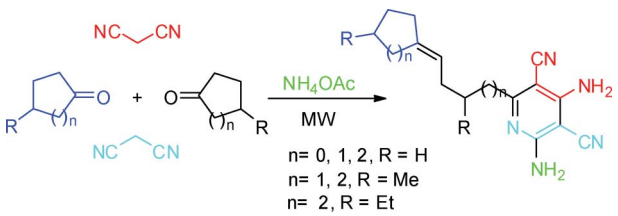
(Calix-MN-SE-Lipase)

4025

New multicomponent cyclization: domino synthesis of pentasubstituted pyridines under solvent-free conditions

Bo Jiang, Xiang Wang, Feng Shi, Shu-Jiang Tu* and Guigen Li*

New five-component domino reaction for rapid synthesis of highly functionalized pentasubstituted pyridines was established giving up to 87% yield.

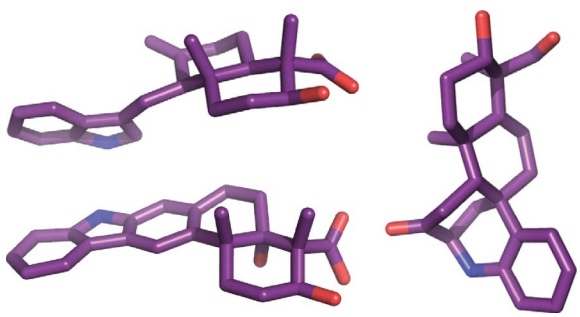


4029

A family of multicyclic indolosesquiterpenes from a bacterial endophyte

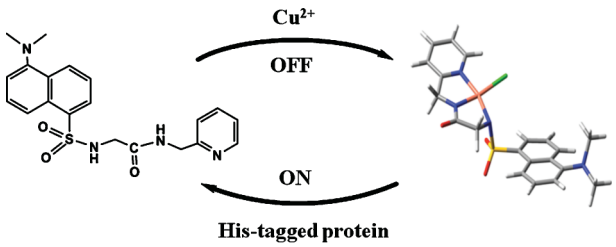
Ling Ding, Armin Maier, Heinz-Herbert Fiebig, Wen-Han Lin and Christian Hertweck*

Three multicyclic indolosesquiterpenes, xiamycin B, indosespene, and sespenine, along with the known xiamycin A, share an unusual terpene alkaloid pathway in *Streptomyces* sp. HKI0595, an endophyte of the widespread mangrove tree *Kandelia candel*.



COMMUNICATIONS

4032

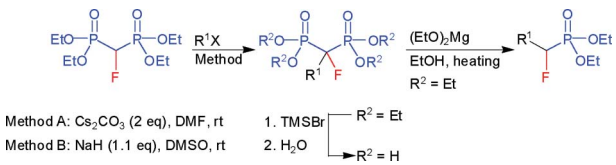


The switch-on luminescence sensing of histidine-rich proteins in solution: a further application of a Cu^{2+} ligand

Bin Wang, Yang Gao, Hong-Wei Li, Zhi-Peng Hu and Yuqing Wu*

A new **1**/Cu^{2+} complex for the detection of His-tagged protein has been developed, based on an improved probe by closely mimicing the structure of a peptide, ATCUN. In aqueous solution, **1**/Cu^{2+} has good selectivity to histidine and cysteine, and further can detect histidine-rich protein by releasing the quenched fluorescence.

4035

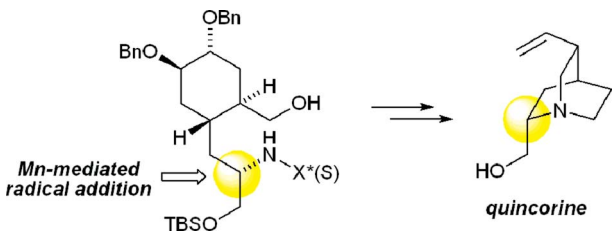


A new route to α-alkyl-α-fluoromethylenebisphosphonates

Petr Beier,* Stanislav Opekar, Mikhail Zibinsky, Inessa Bychinskaya and G. K. Surya Prakash*

Synthesis of α-alkyl-α-fluoromethylenebisphosphonates and corresponding bisphosphonic acids is described by alkylation of tetraethyl fluoromethylenebisphosphonate. Alkoxide–induced carbon–phosphorus bond cleavage of title compounds gave α-fluorophosphonates.

4039



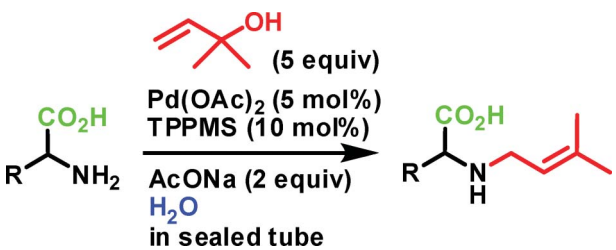
Intermolecular radical addition to *N*-acylhydrazones as a stereocontrol strategy for alkaloid synthesis: formal synthesis of quinine

Gregory K. Friestad,* An Ji, Chandra Sekhar Korapala and Jun Qin

Synthesis of quinine demonstrates the utility of intermolecular radical addition to imino compounds as a stereocontrol strategy in synthesis of complex multifunctional targets.

PAPERS

4044



Palladium-catalyzed mono-*N*-allylation of unprotected amino acids with 1,1-dimethylallyl alcohol in water

Hidemasa Hikawa* and Yuusaku Yokoyama*

Palladium-catalyzed *N*-allylation of unprotected amino acids with 1,1-dimethylallyl alcohol gave mono-*N*-allylated amino acids in good yield.

International Symposia on Advancing the Chemical Sciences (ISACS)

Challenges in Chemical Biology (ISACS5)

26-29 July 2011 ● University of Manchester, UK

Topical

Gain a state-of-the-art review of current research developments and identify future challenges in a comprehensive plenary programme.

Interactive

Network with the editors of *Chemical Science* and international colleagues and present your research in the poster sessions.

Dynamic

Hear from a new generation of exceptional, internationally renowned researchers.

New for 2011

We have a limited number of oral presentation slots available at ISACS5. If you are interested in presenting your work at the meeting, visit the website to submit your abstract for consideration by the conference committee.

About ISACS

The International Symposia on Advancing the Chemical Sciences (ISACS) is a significant new global symposia series, supporting the launch of the new RSC flagship journal *Chemical Science*.
Related events include:

Challenges in Renewable Energy (ISACS4)

5-8 July 2011 ● Boston, USA ● www.rsc.org/isacs4

Challenges in Organic Materials & Supramolecular Chemistry (ISACS6)

2-5 September 2011 ● Beijing, China ● www.rsc.org/isacs6

Speakers include

Thomas Steitz, *USA (Nobel Laureate)*
Venki Ramakrishnan, *UK (Nobel Laureate)*
Donna Blackmond, *USA*
Ben Davis, *UK*
Andrew Ellington, *USA*
Hermann Gaub, *Germany*
Reza Ghadiri, *USA*
Nathaneal Gray, *USA*
Andrew Griffiths, *France*
Bartosz A Grzybowski, *USA*
Johan Hofkens, *Belgium*
Philip Holliger, *UK*
Clyde Hutchison, *USA*
Stefan Knapp, *UK*
Stephen Mann, *UK*
David Rees, *UK*
Hugh Rosen, *USA*
William Shih, *USA*
Hiroaki Suga, *Japan*
Erik Winfree, *USA*

Conference Committee

Hagan Bayley, *UK (Conference Chair)*
Benjamin Cravatt, *USA*
Thomas Carell, *Germany*
Nigel Scrutton, *UK*
Sabine Flitsch, *UK*

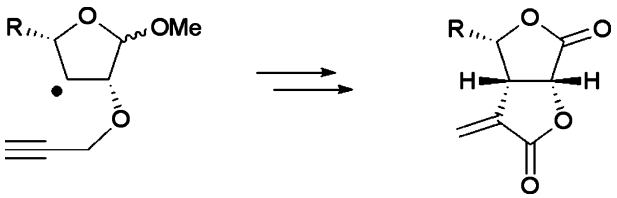
International Year of CHEMISTRY 2011

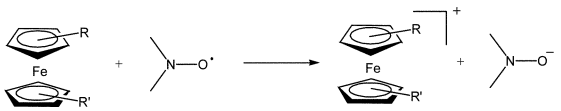
Co-sponsored by:
RSC Chemical Biology Interface Division

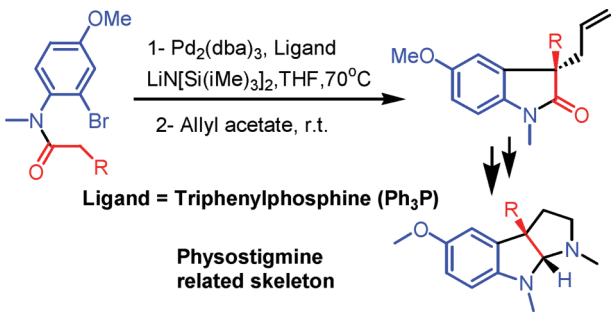
Sign up for news updates and register at:

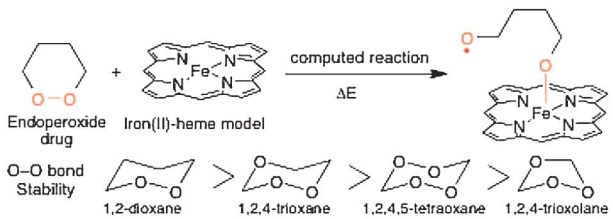
RSC | Advancing the Chemical Sciences

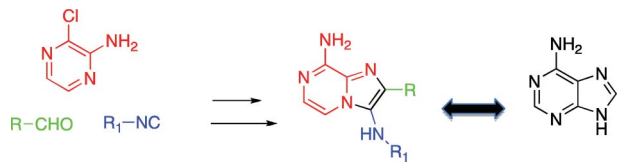
www.rsc.org/isacs5







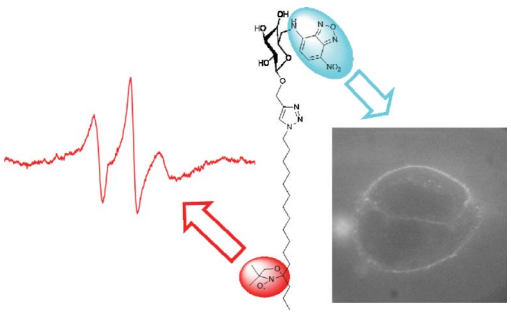




Groebke multicomponent reaction and subsequent nucleophilic aromatic substitution for a convenient synthesis of 3,8-diaminoimidazo[1,2-*a*]pyrazines as potential kinase inhibitors

Margherita Guasconi, Xiaoyun Lu, Alberto Massarotti, Antonio Caldarelli, Elisa Ciraolo, Gian Cesare Tron, Emilio Hirsch, Giovanni Sorba and Tracey Pirali*

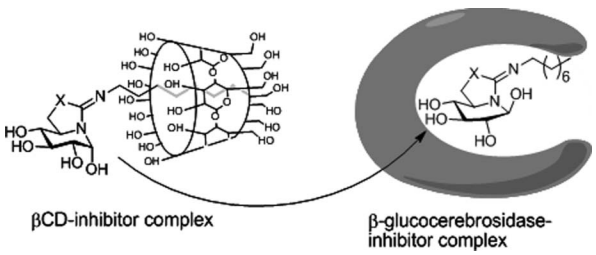
In a program aimed at discovering novel protein kinase inhibitors, a convenient synthesis of 3,8-diaminoimidazo[1,2-*a*]pyrazines has been developed exploiting the isocyanide-based multicomponent Groebke reaction.



Nitroxide–fluorophore double probes: a potential tool for studying membrane heterogeneity by ESR and fluorescence

Stane Pajk,* Maja Garvas, Janez Štrancar and Slavko Pečar

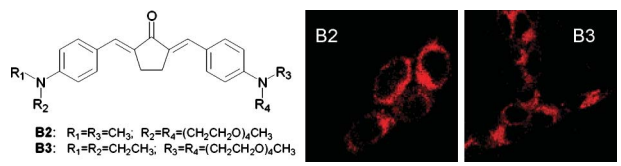
New nitroxide–fluorophore double probes that enable investigation of lipid dynamics and visualization by fluorescence microscopy.



Cyclodextrin-mediated crystallization of acid β-glucosidase in complex with amphiphilic bicyclic nojirimycin analogues

Boris Brumshtein, Matilde Aguilar-Moncayo, Juan M. Benito, José M. García Fernandez, Israel Silman, Yoseph Shaaltiel, David Aviezer, Joel L. Sussman, Anthony H. Futerman and Carmen Ortiz Mellet*

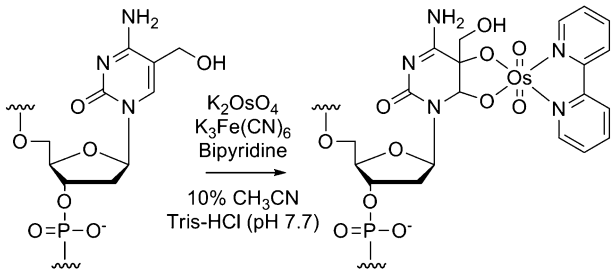
Cyclodextrin-based host–guest chemistry has been exploited to facilitate co-crystallization of recombinant human acid β-glucosidase with amphiphilic bicyclic nojirimycin analogues of the sp^2-iminosugar type.

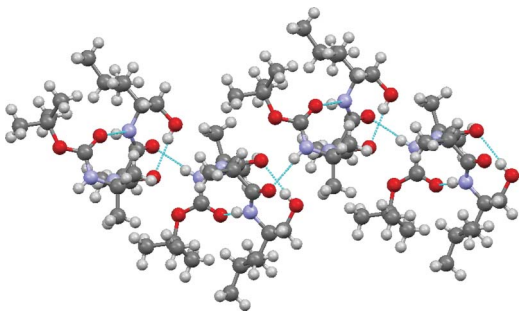


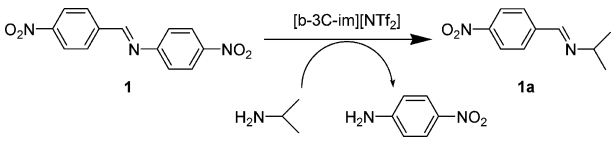
Polyethylene glycol-functionalized benzylidene cyclopentanone dyes for two-photon excited photodynamic therapy

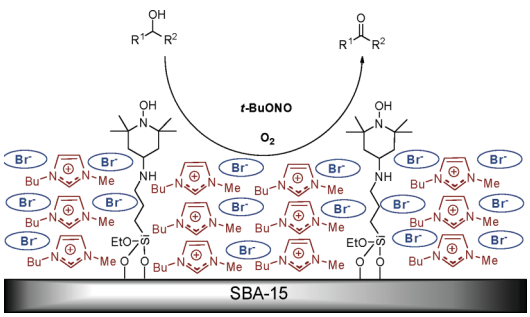
Yuxia Zhao, Weijia Wang, Feipeng Wu,* Yang Zhou, Naiyan Huang, Ying Gu, Qianli Zou and Wei Yang

Water-soluble benzylidene cyclopentanone dyes show strong photodynamic activity under one- and two-photon excitation in *in vitro* cell experiments.

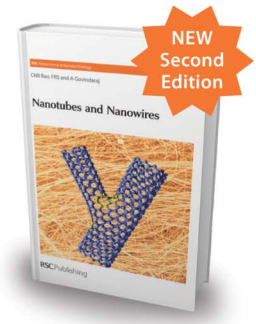








The RSC Nanoscience & Nanotechnology Series



Nanotubes and Nanowires

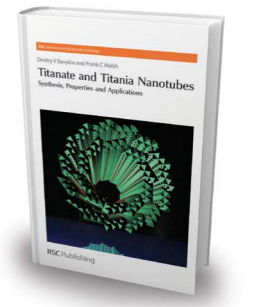
C N Ram Rao and A Govindaraj, JNCASR, India
ISBN: 9781849730587
Price: £139.99

This fully revised new edition is an up-to-date survey of the research areas of carbon nanotubes, inorganic nanotubes and nanowires including synthesis, characterisation, properties and applications. An ideal resource both for graduates needing an introduction to the field of nanomaterials as well as for professionals and researchers in academia and industry.

Phage Nanobiotechnology

Edited by Valery Petrenko, Auburn University, USA | George Smith, University of Missouri, USA
ISBN: 9780854041848
Price: £121.99

This comprehensive book, of value to researchers in all fields, brings the phage display technique closer to specialists in areas of medicine, science and technology, where phage-derived nanomaterials can be most beneficial.



Titanate and Titania Nanotubes Synthesis, Properties and Applications

Dmitry Bavykin and Frank Walsh, University of Southampton, UK
ISBN: 9781847559104
Price: £87.99

A unique compilation of data from a wide range of chemical and spectroscopic instrumentation and their integration into nanostructure characterisation and a valuable reference point aimed at professionals, postgraduates and industrial research workers in nanomaterials.

Polymer Nanocomposites by Emulsion and Suspension Polymerization

Edited by Vikas Mittal, BASF, Germany
ISBN: 9781847552259
Price: £132.99

Brings together all the research and technologies in the field, provides comprehensive information about one of the most important facets of polymer nanocomposites technology - synthesis in emulsion and suspension. Ideal for the experienced researcher as well as a valuable point of reference for newcomer to the field.

This book series covers the wide ranging areas of Nanoscience and Nanotechnology. In particular, the series provides a comprehensive source of information on research associated with nanostructured materials and miniaturised lab on a chip technologies. Titles are intended to provide an accessible reference for professionals and researchers in academia and industry and all are fully referenced to primary literature.

New for 2011

Fullerenes: Principles and Applications
NEW Second Edition
Price £144.99 | ISBN 9781849731362

Fully revised and up-dated, written by leading experts in the field the second edition summarises the basic principles of fullerene chemistry but also highlights some of the most remarkable advances that have occurred in recent years. This book will appeal to researchers in both academia and industry.

Biological Interactions with Surface Charge Biomaterials
Price £121.99 | ISBN 9781849731850

This book is the first to comprehensively address the complex phenomenon of biological interactions with the surface charge of biomaterials. Aimed at academics, practitioners and graduate students this book makes a significant contribution to the newly emerging discipline of 'nanomedicine'.

Nanostructured Catalysts
Price £144.99 | ISBN 9780854041862

A comprehensive up-to-date summary of the existing information on the structural/electronic properties, chemistry and catalytic properties of vanadium and molybdenum containing catalysts.

RSC Nanoscience and Nanotechnology Series Package

- Includes 10 leading series titles
- Comprehensive source of information
- Cutting-edge developments in research

SAVE 35% on the individual purchase price of these leading series titles – purchase as one complete package!

To order any of these titles please email books@rsc.org or visit the website!

RSC Publishing

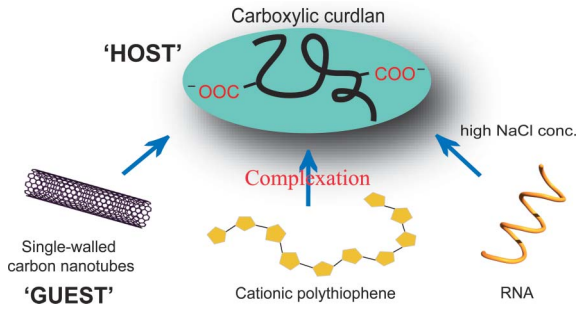
www.rsc.org/nanoscience

4266

A pH-responsive carboxylic β-1,3-glucan polysaccharide for complexation with polymeric guests

Le Thi Ngoc Lien, Tomohiro Shiraki, Arnab Dawn, Youichi Tsuchiya, Daisuke Tokunaga, Shun-ichi Tamaru,* Naoya Enomoto, Junichi Hojo and Seiji Shinkai*

A β-1,3-glucan polysaccharide bearing dissociable COOH groups behaves as a host for SWNTs, RNA and conjugated polymers, resulting in supramolecular complexes.

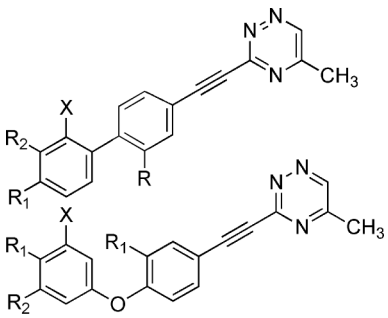


4276

Synthesis and evaluation of 1,2,4-methyltriazines as mGluR5 antagonists

Jeremy P. Olson, Moses G. Gichinga, Elizabeth Butala, Hernan A. Navarro, Brian P. Gilmour and F. Ivy Carroll*

We report the synthesis of 11 new mGluR5 antagonists based on 1,2,4-methyltriazine. The most potent analogue **5f** has an IC50 of 28.2 nM at mGluR5.

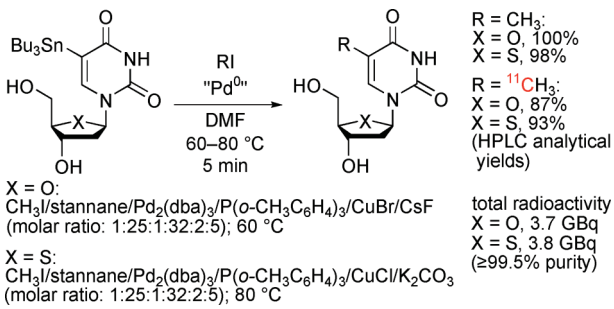


4287

Highly efficient syntheses of [methyl-^{11}C]thymidine and its analogue 4′-[methyl-^{11}C]thiothymidine as nucleoside PET probes for cancer cell proliferation by Pd^0-mediated rapid *C*-[^{11}C]methylation

Hiroko Koyama, Siqin, Zhouen Zhang, Kengo Sumi, Yuma Hatta, Hiroko Nagata, Hisashi Doi and Masaaki Suzuki*

[Methyl-^{11}C]thymidine and 4′-[methyl-^{11}C]thiothymidine as nucleoside PET probes for cancer cell proliferation have been synthesized efficiently based on the Pd^0-mediated rapid cross-coupling using [^{11}C]CH_3I and tributylstannyl precursors.

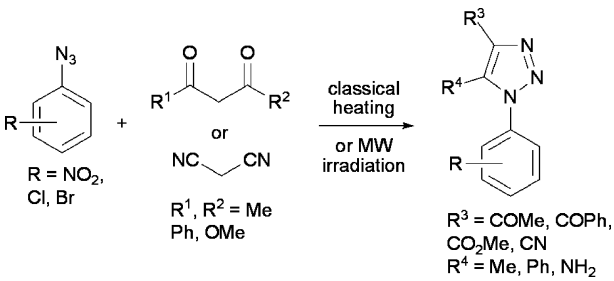


4295

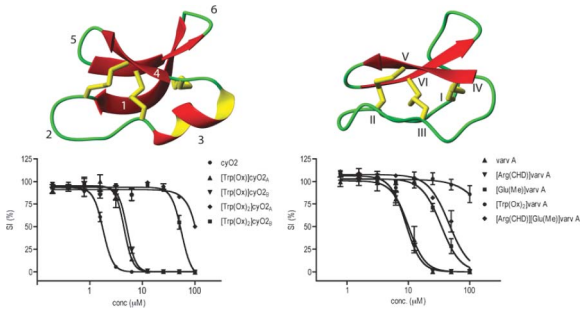
A combined experimental and theoretical study of the thermal cycloaddition of aryl azides with activated alkenes

Sarah Zeghada, Ghenia Bentabed-Ababsa,* Aïcha Derdour, Safer Abdelmounim, Luis R. Domingo,* José A. Sáez, Thierry Roisnel, Ekhlass Nassar and Florence Mongin*

Cycloadditions between aryl azides and activated alkenes *e.g.* coming from β-dicarbonyl compounds were performed; their mechanism and regioselectivity were theoretically studied using DFT methods at the B3LYP/6-31G* level.



4306

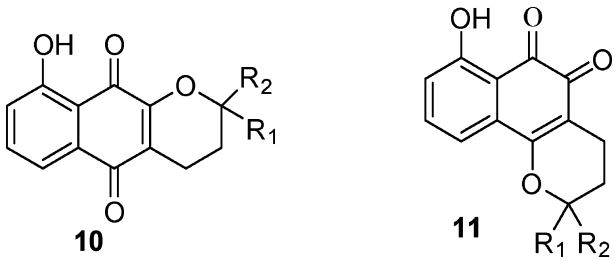


Cytotoxic potency of small macrocyclic knot proteins: Structure–activity and mechanistic studies of native and chemically modified cyclotides

Robert Burman, Anders Herrmann, Rossetti Tran, Jan-Erik Kivelä, Andrei Lomize, Joachim Gullbo and Ulf Göransson*

Investigations on the cytotoxic effects of cyclic disulfide rich mini-proteins—cyclotides—show the importance of a correct distribution of charged and hydrophobic residues for maintaining the ultimate degree of potency.

4315

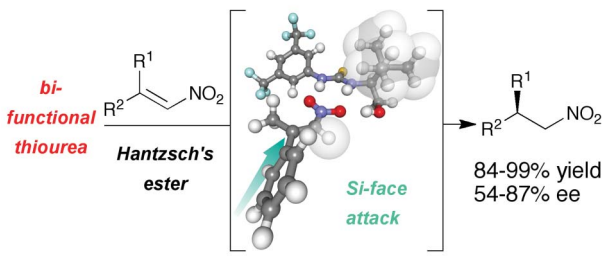


Synthesis of new 9-hydroxy-α- and 7-hydroxy-β-pyran naphthoquinones and cytotoxicity against cancer cell lines

David R. da Rocha,* Ana C. G. de Souza, Jackson A. L. C. Resende, Wilson C. Santos, Evelyne A. dos Santos, Cláudia Pessoa, Manoel O. de Moraes, Letícia V. Costa-Lotufo, Raquel C. Montenegro and Vitor F. Ferreira*

A synthetic route to 9-hydroxy-α- and 7-hydroxy-β-pyran naphthoquinones and its cytotoxicity against cancer lines were studied.

4323

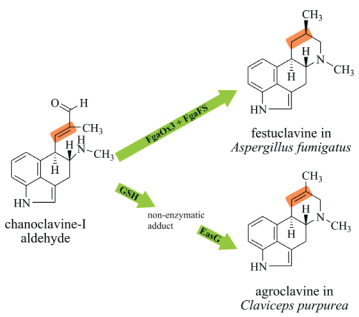


Readily available hydrogen bond catalysts for the asymmetric transfer hydrogenation of nitroolefins

Jakob F. Schneider, Markus B. Lauber, Vanessa Muhr, Domenic Kratzer and Jan Paradies*

Easily available thiourea catalysts for the asymmetric transfer hydrogenation have been developed.

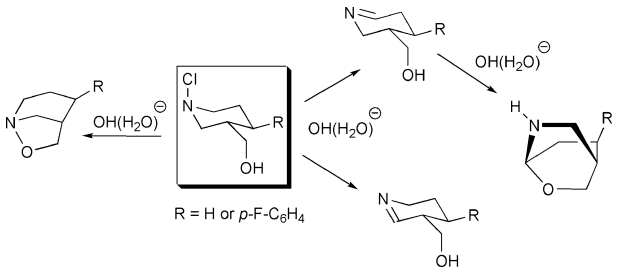
4328

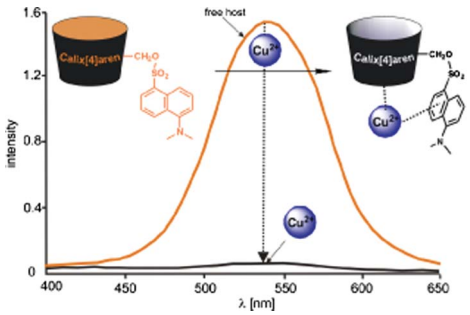


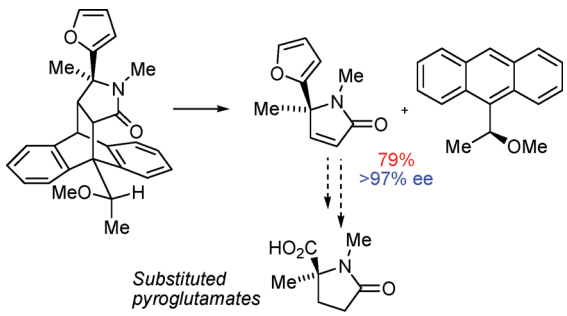
New insights into ergot alkaloid biosynthesis in *Claviceps purpurea*: An agroclavine synthase EasG catalyses, *via* a non-enzymatic adduct with reduced glutathione, the conversion of chanoclavine-I aldehyde to agroclavine

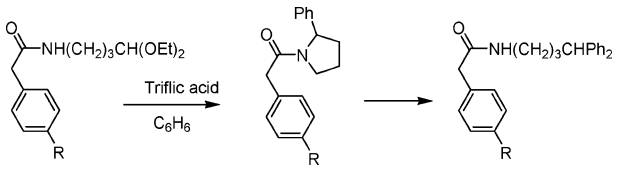
Marco Matuschek, Christiane Wallwey, Xiulan Xie and Shu-Ming Li*

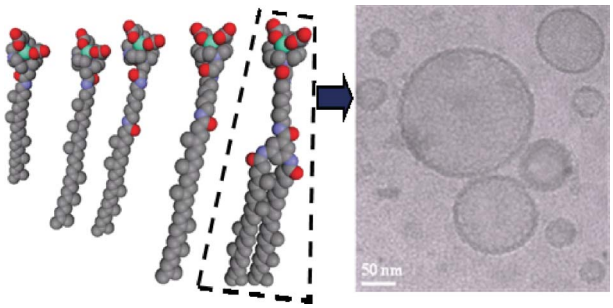
The conversion of chanoclavine-I aldehyde to agroclavine was catalysed by EasG of *C. purpurea* in the presence of NADPH, *via* a non-enzymatic adduct with reduced glutathione (GSH).











Synthesis and physicochemical characterization of new squalenoyl amphiphilic gadolinium complexes as nanoparticle contrast agents

Mohammad Othman, Didier Desmaële, Patrick Couvreur, Luce Vander Elst, Sophie Laurent, Robert N. Muller, Claudie Bourgaux, Estelle Morvan, Thierry Pouget, Sinda Lepêtre-Mouelhi, Philippe Durand and Ruxandra Gref*

Novel amphiphilic squalene–Gd^{3+} complexes spontaneously self-organise as micelles or liposomes and have potential as contrast agents.