

# Organic & Biomolecular Chemistry

An international journal of synthetic, physical and biomolecular organic chemistry

[www.rsc.org/obc](http://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](http://www.rsc.org)

## IN THIS ISSUE

ISSN 1477-0520 CODEN OBCRAK 11(1) 1–180 (2013)



### Cover

See Leung Sheh *et al.*, pp. 48–61.

Energetic basis for molecular recognition of peptides to DNA enlightens the universal EEC paradox.

Image reproduced by permission of Leung Sheh from *Org. Biomol. Chem.*, 2013, **11**, 48.



### Inside cover

See Alison Thompson *et al.*, pp. 62–68.

Image reproduced by permission of Alison Thompson from *Org. Biomol. Chem.*, 2013, **11**, 62.

## EDITORIAL

15

### Happy New Year from *Organic & Biomolecular Chemistry*

On behalf of the *OBC* Editorial Board and Editorial Team, we welcome you to this first issue of 2013.



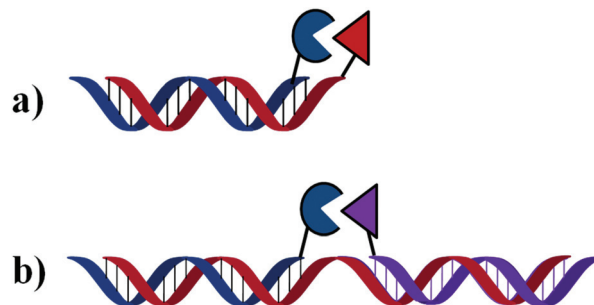
## EMERGING AREA

16

### Oligonucleotide-templated chemical reactions: pushing the boundaries of a nature-inspired process

Claudia Percivalle, Jean-François Bartolo and Sylvain Ladame\*

Widespread in nature, oligonucleotide-templated reactions of phosphodiester bond formation have inspired chemists who are now applying this elegant strategy to the catalysis of a broad range of otherwise inefficient reactions.



## EDITORIAL STAFF

**Editor**

Richard Kelly

**Deputy editor**

Marie Cote

**Development editor**

James Anson

**Senior publishing editor**

Helen Saxton

**Publishing editors**

Mark Archibald, Andrea Banham, Nicola Burton, Sarah Dixon, Sonja Hampel, Elisa Meschini, Roxane Owen

**Publishing assistants**

Aliya Anwar, Sarah Salter

**Publisher**

Emma Wilson

For queries about submitted papers, please contact Helen Saxton, Senior publishing editor in the first instance. E-mail: [obc@rsc.org](mailto:obc@rsc.org)

For pre-submission queries please contact Richard Kelly, Editor. Email: [obc-rsc@rsc.org](mailto:obc-rsc@rsc.org)

Organic & Biomolecular Chemistry (print: ISSN 1477-0520; electronic: ISSN 1477-0539) is published 48 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 Order Department, Royal Society of Chemistry, Thomas Graham House, Science Park, Milton Road, Cambridge, CB4 0WF, UK  
Tel +44 (0)1223 432398; E-mail [orders@rsc.org](mailto:orders@rsc.org)

2013 Annual (print+electronic) subscription price: £4147; US\$7741. 2013 Annual (electronic) subscription price: £3940; US\$7354. 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](http://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 Jamaica NY 11434.

US Postmaster: Send address changes to Organic & Biomolecular Chemistry (OBC), Air Business Ltd, c/o Worldnet Shipping Inc., 156-15, 146th Avenue, 2nd Floor, Jamaica, NY 11434, USA. All despatches outside the UK by Consolidated Airfreight.

The Royal Society of Chemistry takes reasonable care in the preparation of this publication but does not accept liability for the consequences of any errors or omissions. Inclusion of an item in this publication does not imply endorsement by The Royal Society of Chemistry of the content of the original documents to which that item refers.

Advertisement sales: Tel +44 (0) 1223 432246; Fax +44 (0) 1223 426017; E-mail [advertising@rsc.org](mailto:advertising@rsc.org)

For marketing opportunities relating to this journal, contact [marketing@rsc.org](mailto:marketing@rsc.org)



# Organic & Biomolecular Chemistry

An international journal of synthetic, physical and biomolecular organic chemistry

[www.rsc.org/obc](http://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.

## EDITORIAL BOARD

**Chair**

Jeffrey Bode, ETH Zürich, Switzerland

**Associate Editors**

Jin-Quan Yu, Scripps Research

Institute, La Jolla, CA, USA

Andrei Yudin, University of Toronto,

Canada

Margaret Brimble, University of

Auckland, New Zealand

Ashraf Brik, Ben-Gurion University of the Negev, Israel

Pauline Chiu, University of Hong Kong, China

Anthony Davis, University of Bristol, UK

Christian Hertweck, Leibniz-Institute Jena, Germany

Kenichiro Itami, Nagoya University, Japan

Stephen Kent, University of Chicago, USA

Paolo Scrimin, University of Padova, Italy

Qi-Lin Zhou, Nankai University, China

## ADVISORY BOARD

Kyo Han Ahn, Pohang University of Science and Technology, Korea

Fredrik Almqvist, Umeå University, Sweden

Barry Carpenter, Cardiff University, UK

David Chen, Seoul National University, Korea

Shunsuke Chiba, Nanyang

Technological University, Singapore

Sheng-Hsien Chiu, National Taiwan University, Taiwan

Antonio Echavarren, Autonomous

University of Madrid, Spain

Jonathan Ellman, Yale University, USA

Margaret Faul, Amgen, USA

Ben Feringa, University of Groningen, Netherlands

Amar Flood, Indiana University

Bloomington, USA

Nobutaka Fujii, Kyoto University, Japan

Carmen Galan, University of Bristol, UK

Sam Gellman, University of Wisconsin, USA

Christian Hackenberger, Free University Berlin, Germany

Mimi Hii, Imperial College London, UK

Steven V Ley, University of Cambridge, UK

Shih-Yuan Liu, University of Oregon, USA

Stephen Loeb, University of Windsor, Canada

David Lupton, Monash University, Australia

Ilan Marek, Israel Institute of Technology, Israel

Keiji Maruoka, Kyoto University, Japan

Christina Nevado, University of Zurich, Switzerland

Viresh Rawal, University of Chicago, USA

Mark Rizzacasa, University of Melbourne, Australia

Richmond Sarpong, University of California, Berkeley, USA

Oliver Seitz, Humboldt University of Berlin, Germany

Jay Siegel, University of Zürich, Switzerland

Tibor Soos, Hungarian Academy of Sciences, Hungary

Corey Stephenson, Boston University, USA

Mark Taylor, University of Toronto, Canada

Dirk Trauner, Ludwig-Maximilian University Munich, Germany

Bruce Turnbull, University of Leeds, UK

Georgios Vassilikogiannakis, University of Crete, Greece

Helma Wennemers, University of Basel, Switzerland

Peter Wipf, University of Pittsburgh, USA

Shuli You, Shanghai Institute of Organic Chemistry, China

Li He Zhang, Peking University, China

Jian Zhou, East China Normal University, China

## 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>.

Authors may reproduce/republish portions of their published contribution without seeking permission from the RSC, provided that any such republication is accompanied by an acknowledgement in the form: (Original Citation)—Reproduced by permission of The Royal Society of Chemistry.

This journal is © The Royal Society of Chemistry 2013. Apart from fair dealing for the purposes of research or private study for non-commercial purposes, or criticism or review, as permitted under the Copyright, Designs and

Patents Act 1988 and the Copyright and Related Rights Regulation 2003, this publication may only be reproduced, stored or transmitted, in any form or by any means, with the prior permission in writing of the Publishers or in the case of reprographic reproduction in accordance with the terms of licences issued by the Copyright Licensing Agency in the UK. US copyright law is applicable to users in the USA.

The Royal Society of Chemistry takes reasonable care in the preparation of this publication but does not accept liability for the consequences of any errors or omissions.

Ⓢ The paper used in this publication meets the requirements of ANSI/NISO Z39.48-1992 (Permanence of Paper).

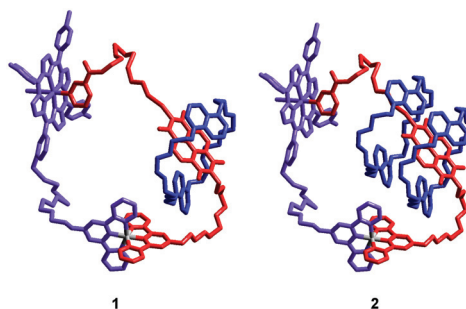
Royal Society of Chemistry: Registered Charity No. 207890.

27

### Synthesis of a four-component [3]catenane using three distinct noncovalent interactions

Miguel Á. Alemán García and Nick Bampos\*

A multicomponent assembly is described resulting in [2] and [3]catenanes using three flexible components and three distinct noncovalent interactions.

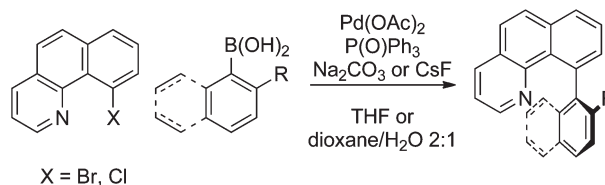


31

### Synthesis of sterically encumbered C10-arylated benzo[h]quinolines using *ortho*-substituted aryl boronic acids

Marko Weimar and Matthew J. Fuchter\*

The challenging coupling of 10-halobenzo[h]quinolines with *ortho*-substituted aryl boronic acids has been achieved using Pd(OAc)<sub>2</sub>/P(O)Ph<sub>3</sub> as the catalytic system.

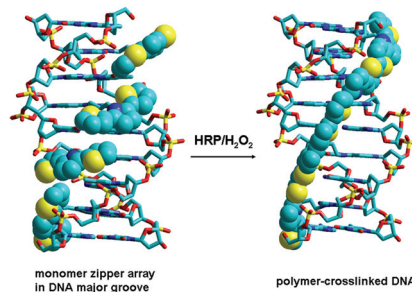


35

### Structural stabilization of DNA-templated nanostructures: crosslinking with 2,5-bis(2-thienyl)pyrrole monomers

Wen Chen and Gary B. Schuster\*

Ordered 2,5-bis(2-thienyl)pyrrole (SNS) zipper arrays are formed by hybridization of complementary DNA oligomers each containing covalently bound SNS monomers.

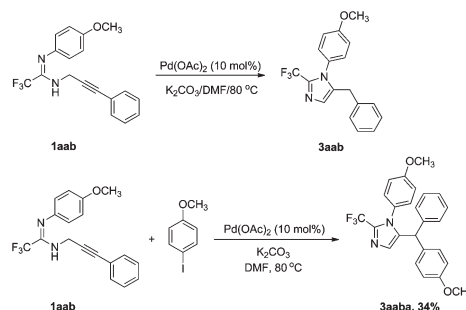


41

### Pd-catalyzed coupling reaction of fluorinated propargyl amidines with aryl iodides

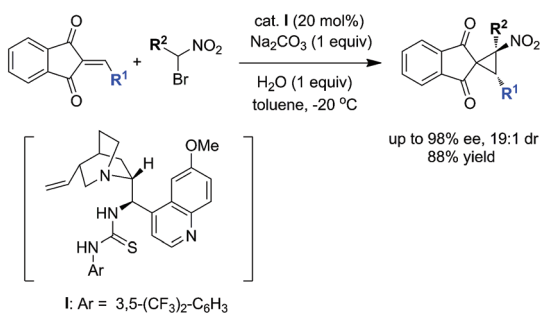
Shan Li, Yafen Yuan, Yajun Li, Zhengke Li, Lisi Zhang and Yongming Wu\*

Catalyzed by ligand free Pd(OAc)<sub>2</sub>, 2,5-disubstituted imidazole was prepared in good yield by the reaction of fluorinated propargyl amidines with iodoarene.



## COMMUNICATIONS

44



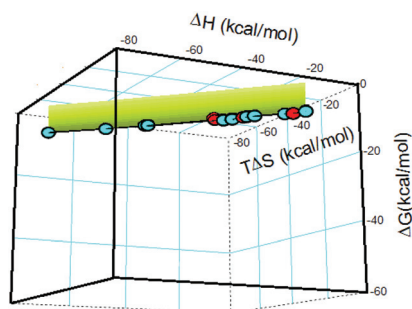
### An efficient organocatalytic enantioselective synthesis of spironitrocyclopropanes

Utpal Das, Yi-Ling Tsai and Wenwei Lin\*

An organocatalytic construction of substituted spironitrocyclopropanes starting from 2-arylidene-1,3-indandiones and bromonitroalkanes has been described.

## PAPERS

48

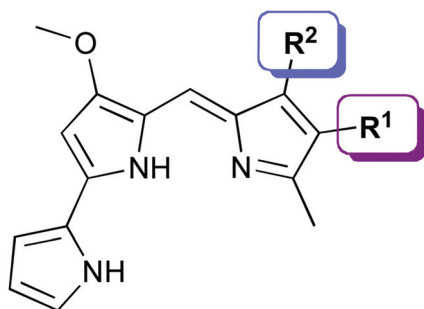


### Energetic studies on DNA–peptide interaction in relation to the enthalpy–entropy compensation paradox

Robin C. K. Yang, Jonathan T. B. Huang, Shih-Chuan Chien, Roy Huang, Kee-Ching G. Jeng, Yen-Chung Chen, Mokai Liao, Jia-Rong Wu, Wei-Kang Hung, Chia-Chun Hung, Yu-Ling Chen, Michael J. Waring and Leung Sheh\*

The energetic basis for molecular recognition of peptides to DNA enlightens the universal EEC paradox.

62

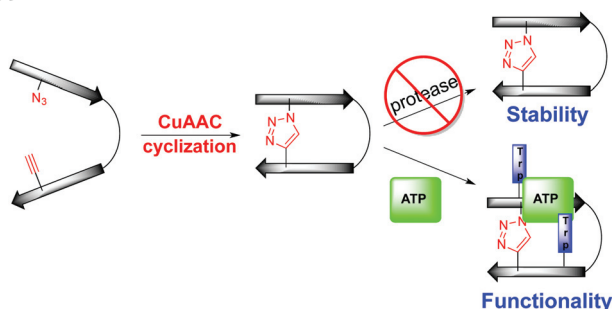


### Investigations regarding the utility of prodigiosenes to treat leukemia

Deborah A. Smithen, A. Michael Forrester, Dale P. Corkery, Graham Dellaire, Julie Colpitts, Sherri A. McFarland, Jason N. Berman and Alison Thompson\*

Structural modification of the C-ring resulted in a series of prodigiosenes that displayed interesting activity *in vitro* and *in vivo*.

69



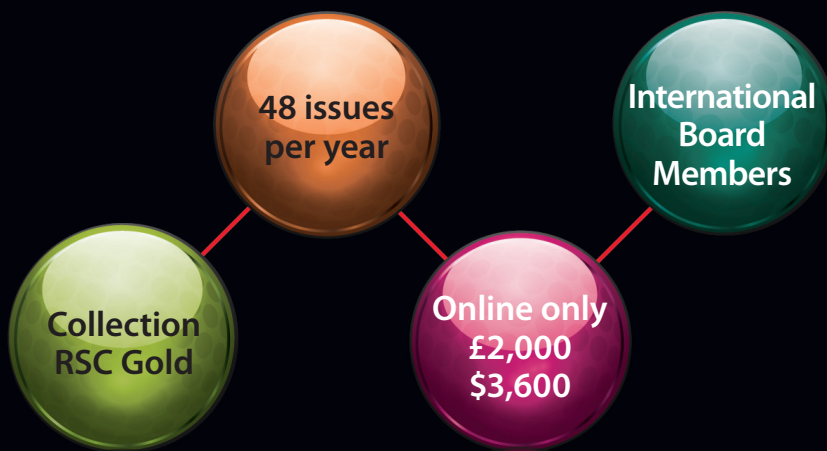
### Positional effects of click cyclization on $\beta$ -hairpin structure, stability, and function

Jessica H. Park and Marcey L. Waters\*

“Clicking” of  $\beta$ -hairpin peptides at several different positions was explored as a method to influence folding, protease resistance, and function.

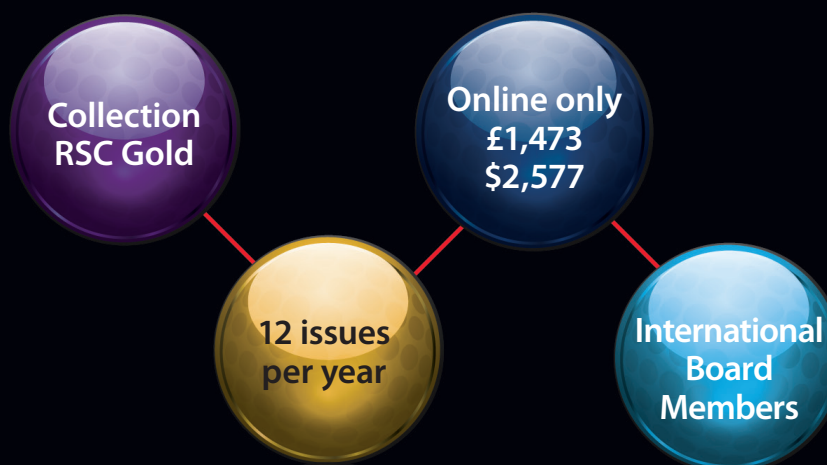
# Affordable, high impact science

FREE ACCESS to these peer-reviewed journals is ending. You and your colleagues can continue to access high quality, high impact science through discounted packages or affordable institutional subscriptions.



**An international online journal covering all of the chemical sciences, including multidisciplinary and emerging areas**

- Over 260,000 downloads to date\*
- Highest quality communications, papers and reviews
- Weekly issues providing a wealth of cutting-edge content across the chemical sciences



**A multidisciplinary journal focusing on all fundamental science and technological aspects of catalysis**

- Over 150,000 downloads to date\*
- Original articles, communications and reviews
- Balanced mix of applied, fundamental, experimental and computational research

\*correct at end of September 2012

“ I need to know  
the structure of  
this compound ”

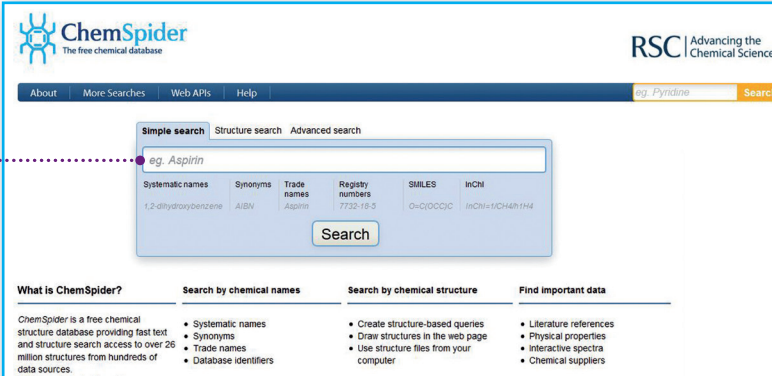
## ChemSpider can help you!

We know that chemical naming is hard and that trivial names hide complex structures.

We want to make it easy for you to find this information wherever you are:

- In the lab
- At home
- At a conference


A simple and intuitive  
text search.



ChemSpider ID: 9041149  
Molecular Formula:  $C_{50}H_{70}O_{14}$   
Average mass: 895.08197 Da  
Monoisotopic mass: 894.47699 Da

Systematic name  
2-[(1S,3R,4aR,5aS,6aR,7Z,9aS,10aR,11aS,12aR,13R,14aS,15aR,16aS,20aR,21aS,22aR,23aS,25aR,26aS,27aR,28aS,29aR,30aS)-1-Hydroxy-9a,13,20,21a,25a,26a,30a-heptamethyl-18-oxo-2,3,4a,5,5a,6a,9,9a,10a,11,11a,12a,13,14,14a,15a,16,16a,18,20a,21a,22,22a,23a,24,25,25a,26a,27,27a,28a,29,29a,30a-tetrahydro-1H-pyrido[2',3':5',6']pyrano[2'',3'':5'',6'']pyrano[3,2-b]pyrano[2''''',3''''',5''''',6''''']pyrano[2''''',3''''',5''''',6''''']pyrano[2''''',3''''',6''''',7''''']oxepino[2''',3''',6''',7''']oxepino[2'',3'',5'',6'']pyrano[2',3',5',6']pyrano[2,3-g]oxocin-3-yl)methyl]acrylaldehyde

Once you've found a structure,  
save it in a format that can be  
opened in any chemical  
drawing program; use it again  
and again.



Search term: **brevetoxin B** (Found by approved synonym)

**Brevetoxin B**

ChemSpider ID: 9041149  
Molecular Formula:  $C_{50}H_{70}O_{14}$   
Average mass: 895.08197 Da  
Monoisotopic mass: 894.47699 Da

Systematic name  
2-[(1S,3R,4aR,5aS,6aR,7Z,9aS,10aR,11aS,12aR,13R,14aS,15aR,16aS,20aR,21aS,22aR,23aS,25aR,26aS,27aR,28aS,29aR,30aS)-1-Hydroxy-9a,13,20,21a,25a,26a,30a-heptamethyl-18-oxo-2,3,4a,5,5a,6a,9,9a,10a,11,11a,12a,13,14,14a,15a,16,16a,18,20a,21a,22,22a,23a,24,25,25a,26a,27,27a,28a,29,29a,30a-tetrahydro-1H-pyrido[2',3':5',6']pyrano[2'',3'':5'',6'']pyrano[3,2-b]pyrano[2''''',3''''',5''''',6''''']pyrano[2''''',3''''',5''''',6''''']pyrano[2''''',3''''',6''''',7''''']oxepino[2''',3''',6''',7''']oxepino[2'',3'',5'',6'']pyrano[2',3',5',6']pyrano[2,3-g]oxocin-3-yl)methyl]acrylaldehyde

View the image in 3D



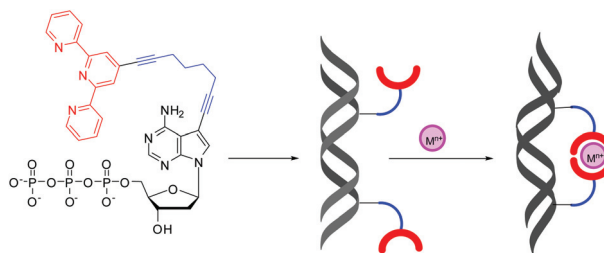
And remember, **ChemSpider** gives you access to a database containing 28 million chemical structures and all of this information: **FREE**, for **Anyone**, **Anytime**, **Anywhere**

78

### Synthesis of nucleosides and dNTPs bearing oligopyridine ligands linked through an octadiyne tether, their incorporation into DNA and complexation with transition metal cations

Lubica Kalachova, Radek Pohl, Lucie Bednářová, Jindřich Fanfrlík and Michal Hocek\*

Nucleotides bearing terpyridine ligands attached *via* a flexible octadiynyl tether were prepared and enzymatically incorporated into DNA where formation of intra-strand complexes with  $\text{Fe}^{2+}$  and  $\text{Ni}^{2+}$  was observed.

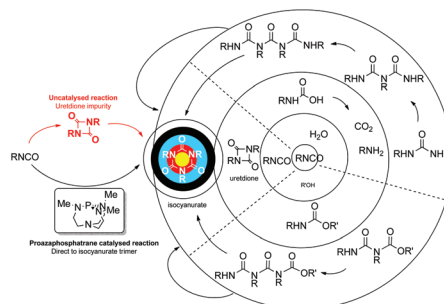


90

### The formation of high-purity isocyanurate through proazaphosphatrane-catalysed isocyanate cyclo-trimerisation: computational insights

Jack N. Gibb and Jonathan M. Goodman\*

Polyurethane foams are widely used materials and control of their physical properties is a significant challenge.

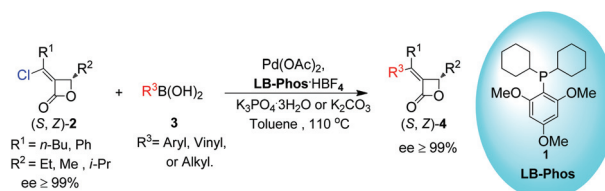


98

### Pd-catalyzed Suzuki coupling reaction of chloroalkylidene- $\beta$ -lactones with LB-Phos as the ligand

Pengbin Li, Bo Lü, Chunling Fu and Shengming Ma\*

The LB-Phos- $\text{HBF}_4$  salt was applied for the Pd-catalysed Suzuki coupling reactions of optically active (*Z*)- $\alpha$ -chloroalkylidene- $\beta$ -lactones.

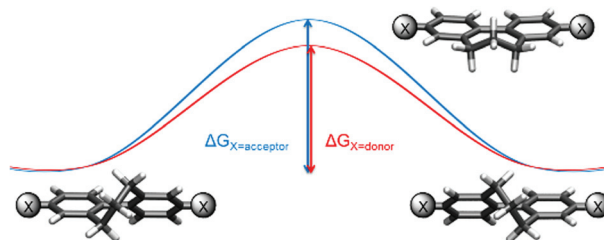


110

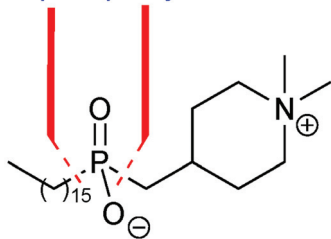
### Atropisomerization of di-*para*-substituted propyl-bridged biphenyl cyclophanes

Jürgen Rotzler, Heiko Gsellinger, Angela Bihlmeier, Markus Gantenbein, David Vonlanthen, Daniel Häussinger,\* Wim Klopper\* and Marcel Mayor\*

The influence of electron donors and electron acceptors of variable strength in the 4 and 4' position of 2 and 2' propyl-bridged axial chiral biphenyl cyclophanes on their atropisomerization process was studied.



119

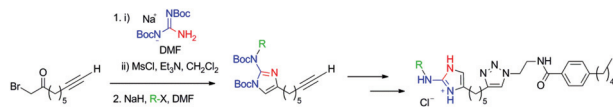
Radical  
hydrophosphinylation

## Synthesis of a phosphinate analogue of the anti-tumour phosphate di-ester perifosine via sequential radical processes

Marios S. Markoulides and Andrew C. Regan\*

A phosphinate analogue of the anti-tumour phosphate di-ester perifosine has been prepared using two sequential radical hydrophosphinylation reactions.

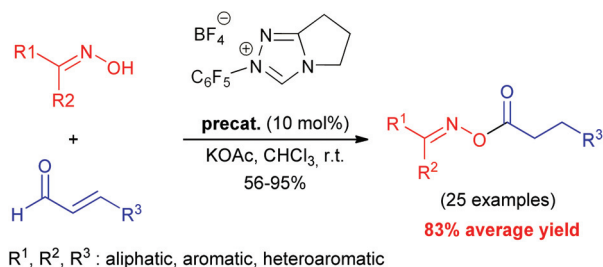
130

*N*-Substituted 2-aminoimidazole inhibitors of MRSA biofilm formation accessed through direct 1,3-bis(*tert*-butoxycarbonyl)guanidine cyclization

Andrew A. Yeagley, Zhaoming Su, Kára D. McCullough, Roberta J. Worthington and Christian Melander\*

A method to access 2-substituted derivatives of a known 2-aminoimidazole/triazole biofilm modulator was developed. Analogues possessing aliphatic substituents exhibited increased biofilm inhibition activity against MRSA compared to the parent compound. Several analogues also displayed synergy with oxacillin against MRSA.

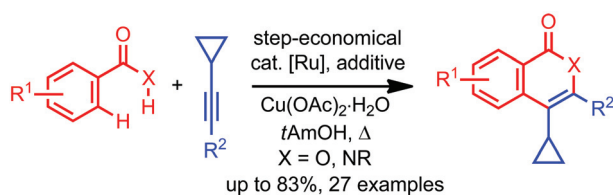
138

*N*-Heterocyclic carbene catalyzed synthesis of oxime esters

Dieter Enders,\* André Grossmann and David Van Craen

A selective, redox- and atom economical synthesis of oxime esters via *N*-heterocyclic carbene catalyzed redox esterification has been developed. The new protocol opens a convenient and direct entry to the title compounds in very high yields without any significant limitations in the substrate scope.

142



## Ruthenium-catalyzed C–H/O–H and C–H/N–H bond functionalizations: oxidative annulations of cyclopropyl-substituted alkynes

Monica Deponti, Sergei I. Kozhushkov, Dmitry S. Yufit and Lutz Ackermann\*

Ruthenium-catalyzed syntheses of cyclopropyl-substituted isocoumarins and isoquinolones were accomplished through highly chemo-, site- and regioselective C–H/O–H and C–H/N–H bond functionalizations with cyclopropylacetylenes.

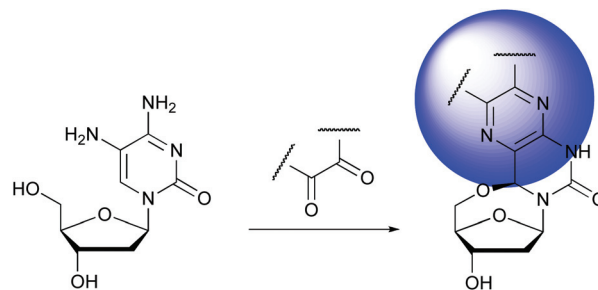


149

### Syntheses and photophysical properties of 5'-6-locked fluorescent nucleosides

Kristmann Gislason, Dnyaneshwar B. Gophane and Snorri Th. Sigurdsson\*

The condensation reaction between 5-amino-2'-deoxycytidine and 1,2-diketones was utilized to obtain nine 5'-6-locked fluorescent nucleosides with diverse photophysical properties. One nucleoside was also studied in model DNA structures, including a nicked duplex.

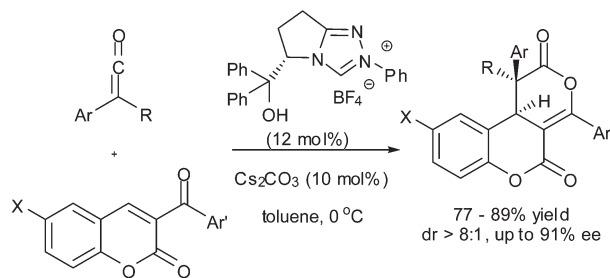


158

### N-heterocyclic carbene-catalyzed [4 + 2] cycloaddition of ketenes and 3-arylcoumarins: highly enantioselective synthesis of dihydrocoumarin-fused dihydropyranones

Teng-Yue Jian, Xiang-Yu Chen, Li-Hui Sun and Song Ye\*

Dihydrocoumarin-fused dihydropyranones were synthesized in high yield with good diastereo- and enantioselectivity *via* NHC-catalyzed [4 + 2] cyclization of ketenes and 3-arylcoumarins.

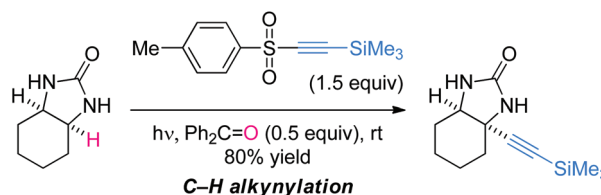


164

### Photochemically induced radical alkylation of C(sp<sup>3</sup>)-H bonds

Tamaki Hoshikawa, Shin Kamijo and Masayuki Inoue\*

The present direct alkylation of C(sp<sup>3</sup>)-H bonds enables construction of various tri- and tetra-substituted carbons from heteroatom-substituted methylenes, methines and alkanes in a highly chemoselective fashion.

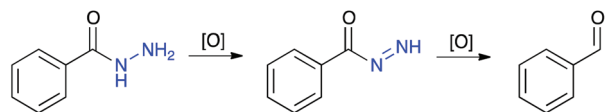


170

### Mechanistic investigation of the oxidation of hydrazides: implications for the activation of the TB drug isoniazid

Ruth I. J. Amos, Brendon S. Gourlay, Brian F. Yates, Carl H. Schiesser, Trevor W. Lewis and Jason A. Smith\*

In the oxidation of arylhydrazides the diimide intermediate is the source for nucleophilic acyl substitution products and the precursor for acyl radical formation, there is no evidence for the formation of acyl cations.



# High Impact Organic Chemistry

Covering all organic areas, including synthesis, supramolecular, natural products and organic materials



Scan the QR code, search and view the latest articles online

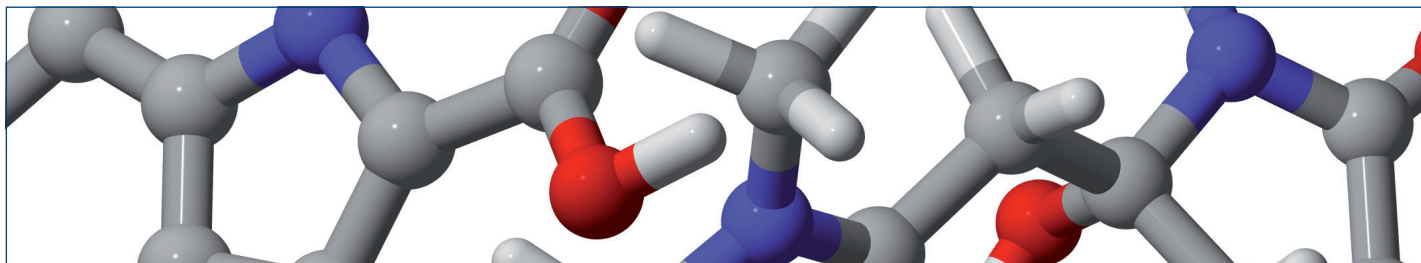


Organic Materials Physical Inorganic Nanoscience  
 Food Biological Environmental Analytical Energy  
 Catalysis Chemical Biology & Medicinal General Chemistry

RSC Publishing

[www.rsc.org/OrganicPort](http://www.rsc.org/OrganicPort)

Registered Charity Number 207890



## 23rd International Symposium

# Synthesis in Organic Chemistry

22-25 July 2013

St Catherine's College, University of Oxford, UK

Register  
early to  
guarantee  
your place

Synthesis in Organic Chemistry provides an international showcase for the core area of organic chemistry - synthesis - covering all aspects of contemporary organic synthesis and providing a forum for the ever more exciting methodologies and strategies that continue to emerge.

### Supporting Speakers

**Dr Edward A Anderson**, *UK*

**Dr Chris Braddock**, *UK*

**Dr Paul Davies**, *UK*

**Professor Ian Fairlamb**, *UK*

**Professor Rachel O'Reilly**, *UK*

### Speakers

**Professor Harry Anderson**, *UK*

**Professor Phil S Baran**, *USA*

**Professor Scott E Denmark**, *USA*

**Professor Darren J Dixon**, *UK*

**Professor Ben L Feringa**, *the Netherlands*

**Professor Alison Frontier**, *USA*

**Professor Frank Glorius**, *Germany*

**Professor Timothy F Jamison**, *USA*

**Professor Karl Anker Jørgensen**, *Denmark*

**Professor Mohammad Movassaghi**, *USA*

**Professor Kenso Soai**, *Japan*

**Professor Jim Thomas**, *UK*

**Professor Shu-Li You**, *China*

### Key Deadlines

Poster abstract submission – 11 March 2013

Early bird registration – 11 March 2013

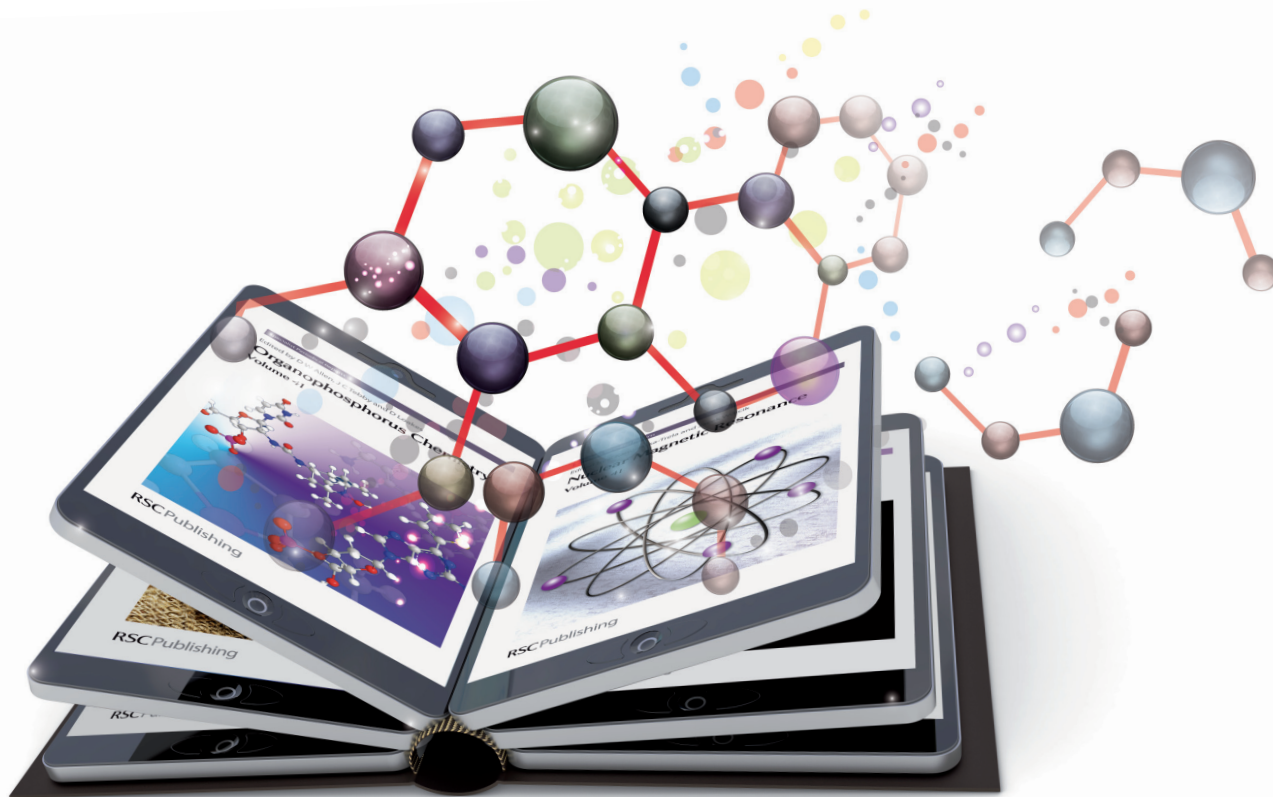
Standard registration – 15 May 2013

071217

# RSC eBooks: The Complete Collection 1968 - 2013

The RSC eBook Collection is a definitive point of reference for anyone working in the chemical sciences, featuring over 1,000 fully-searchable titles and spanning 45 years of groundbreaking research. Available for purchase as a complete collection, it can also be divided to suit your budget needs by year range and subject area.

- Unlimited access and unlimited usage
- All 25,000 chapters can be read and downloaded individually
- MARC records and comprehensive usage statistics free of charge
- All titles DOI indexed to chapter level
- Powerful search functionality
- Perpetual ownership guaranteed



Recommend to your Librarian today or visit us online to find out more!