

Organic & Biomolecular Chemistry

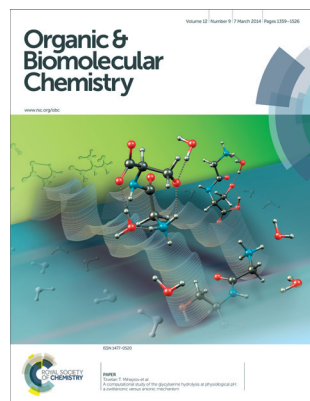
An international journal of synthetic, physical and biomolecular organic chemistry

www.rsc.org/obc

The Royal Society of Chemistry is the world's leading chemistry community. Through our high impact journals and publications we connect the world with the chemical sciences and invest the profits back into the chemistry community.

IN THIS ISSUE

ISSN 1477-0520 CODEN OBCRAK 12(9) 1359–1526 (2014)



Cover

See Tzvetan T. Mihaylov *et al.*,
pp. 1395–1404.

Image reproduced by
permission of Tzvetan
T. Mihaylov from *Org. Biomol.
Chem.*, 2014, **12**, 1395.

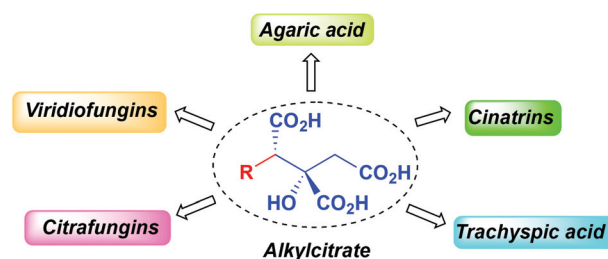
REVIEW

1367

Total synthesis of alkyl citrate natural products

Mark A. Rizzacasa* and Dayna Sturgess

This review highlights the synthesis of members of the alkyl citrate family of natural products. The focus is on the stereoselective construction of the alkyl citrate moiety common to these compounds.



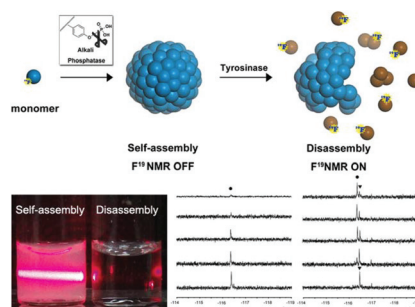
COMMUNICATIONS

1383

Enzyme-controllable F-NMR turn on through disassembly of peptide-based nanospheres for enzyme detection

Jie Gao, Yang Shi, Youzhi Wang, Yanbin Cai, Jie Shen,*
Deling Kong and Zhimou Yang*

The enzyme tyrosinase could trigger the disassembly of peptide-based nanospheres, resulting in F-NMR signal turning on.



Editorial staff

Editor

Richard Kelly

Deputy editor

Marie Cote

Senior publishing editor

Helen Saxton

Development editors

James Anson, Alessia Millemaggi

Publishing editors

Nicola Burton, Ruth Dilleen, Zoe Karthaus, Elisa Meschini, Roxane Owen, Simon Rankmore

Publishing assistants

Vicki Adams, Judy Allen

Publisher

Emma Wilson

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

For pre-submission queries please contact Richard Kelly, Editor. Email: 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

2014 Annual (print+electronic) subscription price: £4396; US\$8206. 2014 Annual (electronic) subscription price: £4176; US\$7795. 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.

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

For marketing opportunities relating to this journal, contact marketing@rsc.org



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. Broad in scope, it publishes research and reviews on topics across organic synthesis, physical organic chemistry, supramolecular chemistry and chemical biology.

Editorial board

Chair

Jeffrey Bode, ETH Zürich, Switzerland

Associate editors

Margaret Brimble, University of Auckland, New Zealand
Jin-Quan Yu, Scripps Research Institute, La Jolla, CA, USA
Andrei Yudin, University of Toronto, Canada

Editorial board members

Ashraf Brik, Ben-Gurion University of the Negev, Israel
Pauline Chiu, University of Hong Kong, China
Jonathan Clayden, University of Manchester, UK
Anthony Davis, University of Bristol, UK
Christian Hertweck, Leibniz-Institute Jena, Germany

Kenichiro Itami, Nagoya University, Japan
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
Luiz Carlos Dias, State University of Campinas, Brazil
Antonio Echavarren, Autonomous University of Madrid, Spain
Jonathan Ellman, Yale University, USA
Margaret Faul, Amgen, USA
Ben Feringa, University of Groningen, The 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
Krishna Kaliappan, IITB, India
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
Cristina Nevado, University of Zürich, Switzerland
Dhevalapally B. Ramachary, University of Hyderabad, India
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, University of Michigan, 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 2014. 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).

Registered Charity No. 207890.

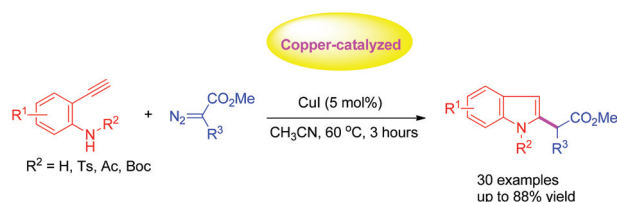
COMMUNICATIONS

1387

Copper-catalyzed annulation of α -substituted diazoacetates with 2-ethynylanilines: the direct synthesis of C2-functionalized indoles

Gang Liu, Guangyang Xu, Jian Li, Dong Ding and Jiangtao Sun*

Copper-catalyzed direct annulation of α -substituted diazoacetates with 2-ethynylanilines leading to C2-functionalized indoles was achieved under mild reaction conditions. The C2-(carboxylate methyl) substituted indoles were obtained in moderate to high yields.

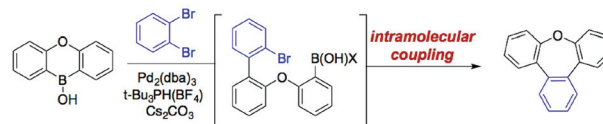


1391

Synthesis of benzannulated heterocycles by twofold Suzuki–Miyaura couplings of cyclic diarylborinic acids

Elena Dimitrijević, Madeline Cusimano and Mark S. Taylor*

Successive transmetalations of the two C–B bonds of cyclic borinic acids enable a novel synthesis of benzo-fused heterocycles from readily available precursors.



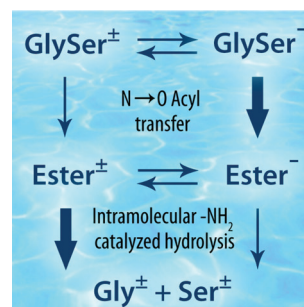
PAPERS

1395

A computational study of the glycyserine hydrolysis at physiological pH: a zwitterionic versus anionic mechanism

Tzvetan T. Mihaylov,* Tatjana N. Parac-Vogt and Kristine Pierloot

The hydrolysis of GlySer at physiological pH was investigated by modeling the most feasible reaction mechanisms in aqueous phase at the MP2/6-311+(2df,2p)//SMD-M06/6-311+(2df,2p) level of the theory.

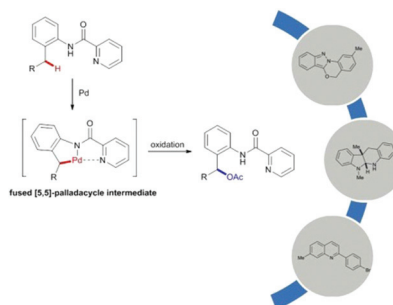


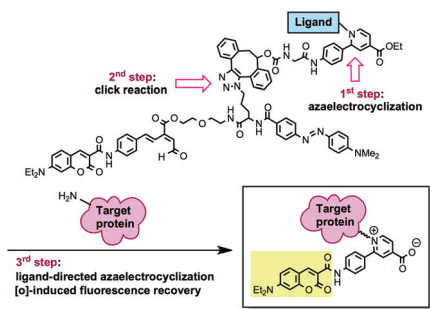
1405

Palladium catalyzed acetoxylation of benzylic C–H bonds using a bidentate picolinamide directing group

Tao Cheng, Weiyu Yin, Yi Zhang, Yingnan Zhang and Yong Huang*

Palladium catalysed oxygenation of inert benzylic C–H bonds offers a straightforward entry to heterocycle synthesis.

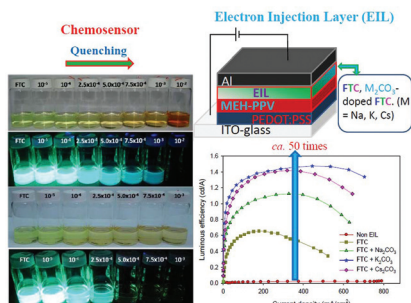




Katsunori Tanaka,* Masataka Kitadani, Ayumi Tsutsui,
Ambara R. Pradipta, Rie Imamaki, Shinobu Kitazume,
Naoyuki Taniguchi and Koichi Fukase*

A general probe designed to induce a cascading sequence of reactions on a target protein was efficiently synthesized.

1430

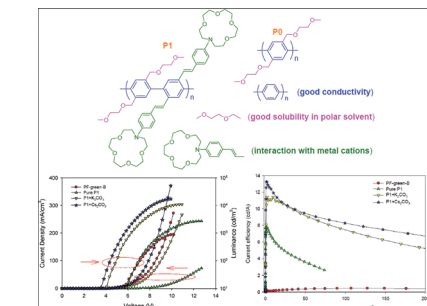


A fluorene-based material containing triple azacrown ether groups: synthesis, characterization and application in chemosensors and electroluminescent devices

Chia-Shing Wu, Ying-Ju Lin and Yun Chen*

A novel fluorene-based FTC containing triple azacrown ether groups is applied as a chemical sensor and as an electron injection layer simultaneously.

1440

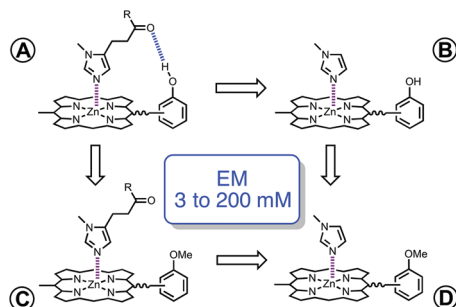


Water/alcohol soluble electron injection material containing azacrown ether groups: synthesis, characterization and application to enhancement of electroluminescence

Chia-Shing Wu, Huai-An Lu, Chiao-Pei Chen,
Tzung-Fang Guo and Yun Chen*

A copoly(*p*-phenylene) containing pendant azacrown ether and ethylene glycol ether groups was prepared as a highly efficient electron injection layer for PLEDs.

1440



Measurement of supramolecular effective molarities for intramolecular H-bonds in zinc porphyrin–imidazole complexes

Michael A. Jinks, Hongmei Sun and
Christopher A. Hunter*

Effective molarities measured for the formation of H-bonds in a variety of zinc porphyrin-imidazole complexes are all between 3 mM and 200 mM, and the values measured for flexible and rigid ligand systems are comparable.

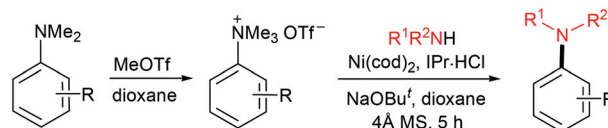
PAPERS

1448

Nickel-catalyzed cross-coupling of aryltrimethylammonium triflates and amines

Xue-Qi Zhang and Zhong-Xia Wang*

Ni(cod)₂-IPr effectively catalyzes cross-coupling of aryltrimethylammonium triflates and amines in the presence of NaOBu^t and 4 Å molecular sieves.

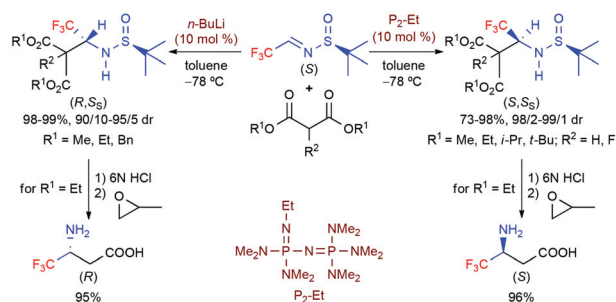


1454

Asymmetric Mannich reaction between (*S*)-*N*-(*tert*-butanesulfinyl)-3,3,3-trifluoroacetaldimine and malonic acid derivatives. Stereodivergent synthesis of (*R*)- and (*S*)-3-amino-4,4,4-trifluorobutanoic acids

Norio Shibata,* Takayuki Nishimine, Naoyuki Shibata, Etsuko Tokunaga, Kosuke Kawada, Takumi Kagawa, José Luis Aceña, Alexander E. Sorochinsky and Vadim A. Soloshonok*

Mannich additions of malonates to the title CF₃-sulfinylimine are described.

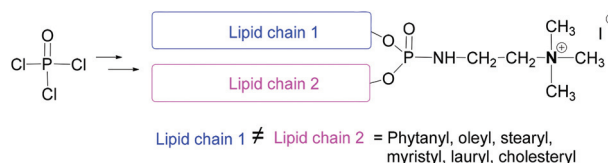


1463

Cationic lipophosphoramidates with two different lipid chains: synthesis and evaluation as gene carriers

Stéphanie S. Le Corre, Mathieu Berchel, Nawal Belmadi, Caroline Denis, Jean-Pierre Haelters, Tony Le Gall, Pierre Lehn, Tristan Montier and Paul-Alain Jaffrès*

The synthesis of a series of new cationic lipids possessing two different lipid chains is detailed. The transfection efficacies have shown the interest to associate a phytanyl chain with either, a lauryl or oleyl chain.

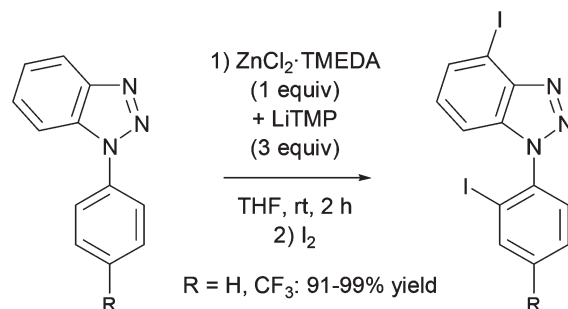


1475

Deproto-metallation using a mixed lithium–zinc base and computed CH acidity of 1-aryl 1*H*-benzotriazoles and 1-aryl 1*H*-indazoles

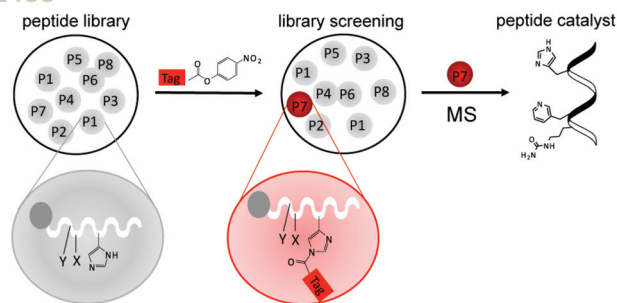
Elisabeth Nagaradja, Floris Chevallier,* Thierry Roisnel, Vincent Dorcet, Yury S. Halauko,* Oleg A. Ivashkevich, Vadim E. Matulis and Florence Mongin*

1-Aryl-1*H*-benzotriazoles have been deproto-metallated using a 2,2,6,6-tetramethylpiperidino-based Li–Zn combination. The regioselectivity has been discussed in the light of DFT-calculated CH acidities (THF solution).



PAPERS

1488

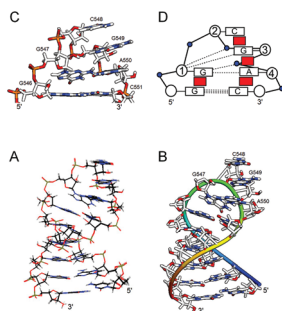


Identification and optimization of short helical peptides with novel reactive functionality as catalysts for acyl transfer by reactive tagging

Silvia Bezer, Masaomi Matsumoto, Michael W. Lodewyk, Stephen J. Lee, Dean J. Tantillo, Michel R. Gagné* and Marcey L. Waters*

A high-throughput screening method is used for identification of new catalytic functionalities for transesterification reaction in organic solvent.

1495

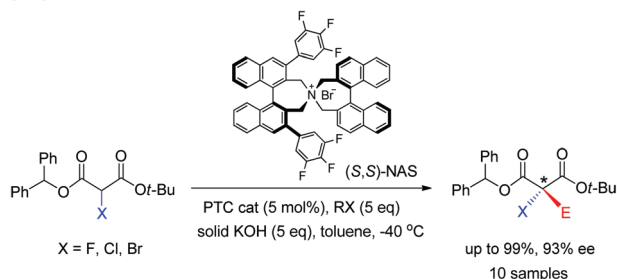


NMR elucidation of the role of Mg^{2+} in the structure and stability of the conserved RNA motifs of the EMCV IRES element

Sadia Mohammed, Marie M. Phelan, Usman Rasul and Vasudevan Ramesh*

NMR structure of the highly conserved and functionally significant 16mer RNA motif endowed with a classic "GNRA" tetraloop of the novel EMCV IRES element.

1510

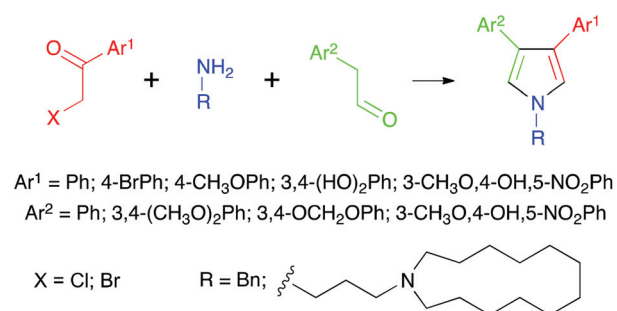


Enantioselective synthesis of α -halo- α -alkylmalonates *via* phase-transfer catalytic α -alkylation

Suckchang Hong, Minsik Kim, Myunggi Jung, Min Woo Ha, Myungmo Lee, Yohan Park, Mi-hyun Kim, Taek-Soo Kim, Jihoon Lee and Hyeung-geun Park*

A new enantioselective synthetic method for α -halo- α -alkylmalonates is reported.

1518



A concise formation of *N*-substituted 3,4-diarylpyrroles – synthesis and cytotoxic activity

Maxim Egorov, Bernard Delpech,* Geneviève Aubert, Thierry Cresteil, Maria Concepcion Garcia-Alvarez, Pascal Collin and Christian Marazano

Sequential condensation of a phenacyl halide with a primary amine and a phenylacetaldehyde led to *N*-substituted 3,4-diarylpyrroles. Synthesis of analogs of the marine alkaloid halitulin and cytotoxic studies are reported.