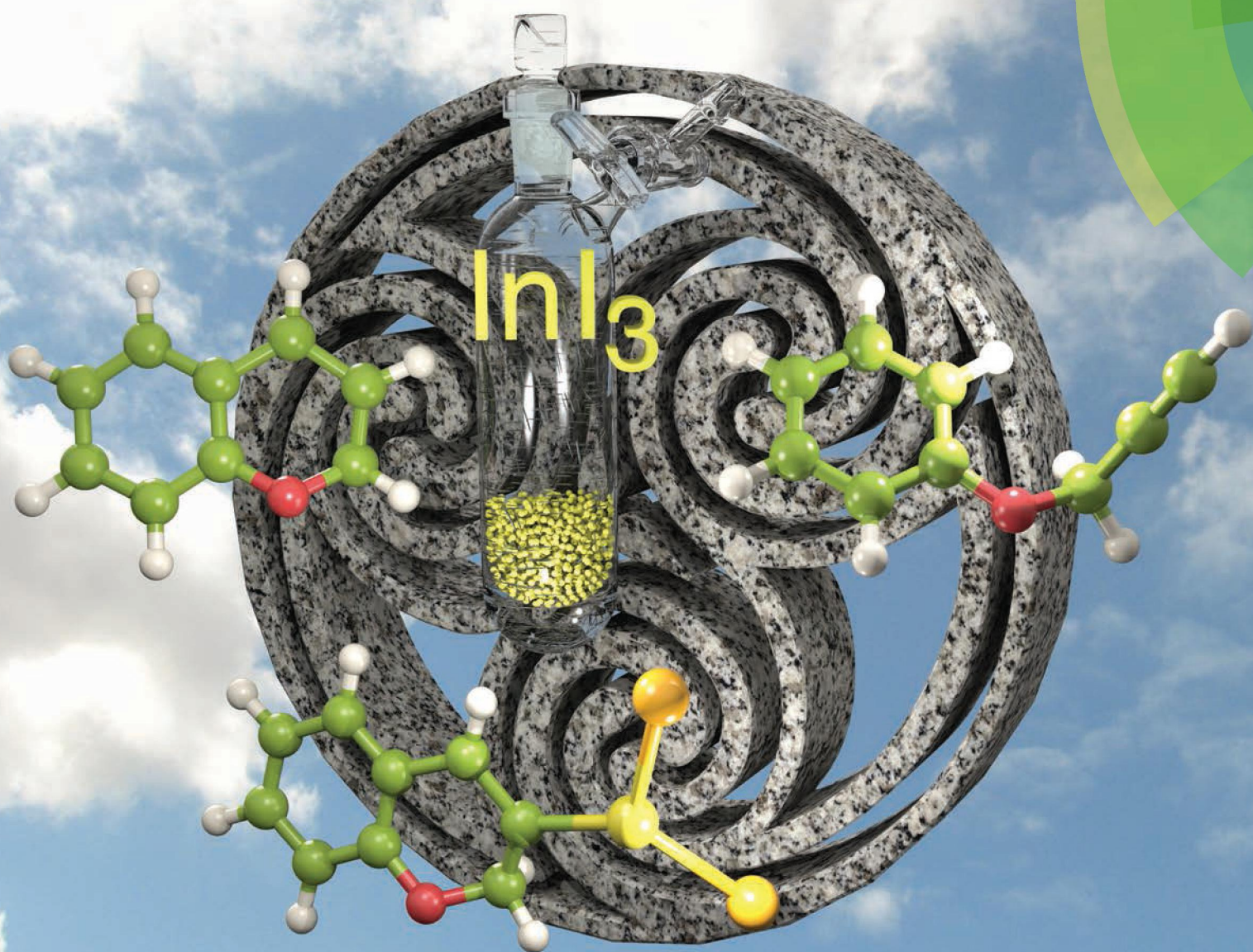


Organic & Biomolecular Chemistry

www.rsc.org/obc



ISSN 1477-0520



PAPER

José Pérez Sestelo *et al.*

Indium-catalyzed intramolecular hydroarylation of aryl propargyl ethers

Organic & Biomolecular Chemistry

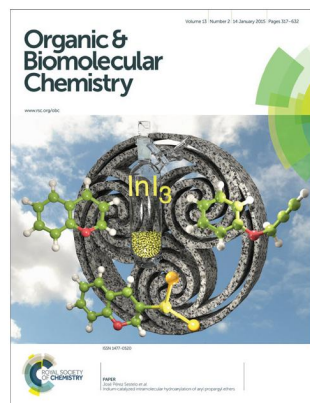
An international journal of synthetic, physical and biomolecular organic chemistry

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 13(2) 317–632 (2015)



Cover

See José Pérez Sestelo *et al.*, pp. 379–387.

The image includes a Celtic symbol, which bears a resemblance to a catalytic cycle, and was designed with assistance from Prof. Eugenio Vázquez, University of Santiago de Compostela.

Image reproduced by permission of José Pérez Sestelo from *Org. Biomol. Chem.*, 2015, **13**, 379.

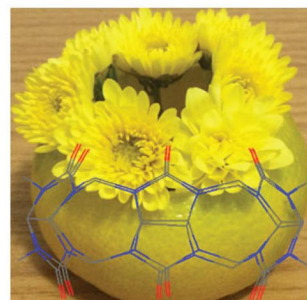
REVIEW

330

Cucurbituril-based supramolecular engineered nanostructured materials

Sinem Gürbüz, Muazzam Idris and Dönüs Tuncel*

Nanostructured materials, including nanoparticles, nanocomposites, vesicles, and rods, have been prepared by taking advantage of the interesting features of cucurbituril homologues.



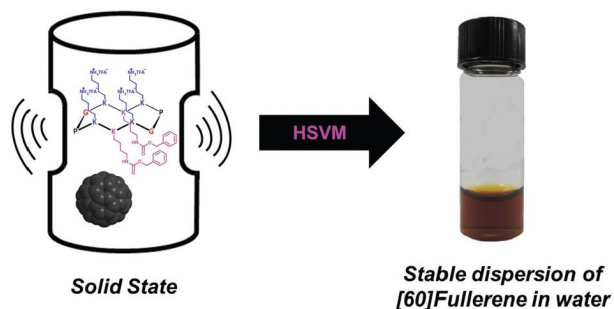
COMMUNICATIONS

348

A peptide topological template for the dispersion of [60]fullerene in water

S. Bartocci, D. Mazzier, A. Moretto and M. Mba*

Stable dispersions of [60]fullerene in water with concentrations of up to 1.3 mg mL^{-1} have been obtained using a peptide topological template as the carrier.



Editorial staff

Editor

Richard Kelly

Deputy editor

Marie Cote

Editorial production manager

Helen Saxton

Development editor

James Anson

Publishing editors

Nicola Burton, Zoe Karthäuser, Elisa Meschini, Roxane Owen, Simon Rankmore, Donna Smith

Publishing assistants

Emily Finney, Rosalind Searle

Publisher

Emma Wilson

For queries about submitted papers, please contact Helen Saxton, Editorial production manager 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

2015 Annual (print+electronic) subscription price: £4572; US\$8534. 2015 Annual (electronic) subscription price: £4343; US\$8107. 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

Andrei Yudin, University of Toronto, Canada

Associate editors

Margaret Brimble, University of Auckland, New Zealand
Jin-Guan Yu, Scripps Research Institute, La Jolla, CA, USA

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
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, Chinese Taipei

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 2015. 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.

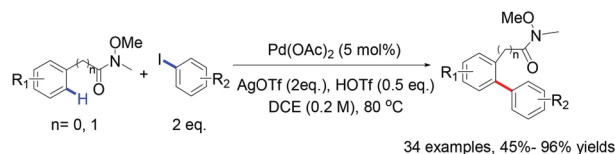
COMMUNICATIONS

353

Pd(II)-catalyzed C–H arylation of aryl and benzyl Weinreb amides

Yan Wang, Kai Zhou, Quan Lan and Xi-Sheng Wang*

The first example of palladium-catalyzed *ortho*-C–H arylation of aryl and benzyl Weinreb amides was developed, in which HOTf was used as a key promoter.

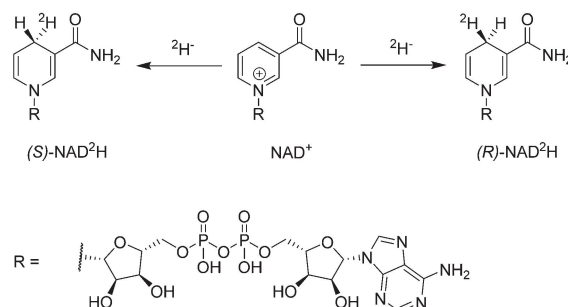


357

Artificial metalloenzymes for the diastereoselective reduction of NAD⁺ to NAD^{2H}

Tommaso Quinto, Daniel Häussinger, Valentin Köhler and Thomas R. Ward*

Stereoselectively labelled isotopomers of NAD(P)H are highly relevant for mechanistic studies of enzymes which utilize them as redox equivalents.

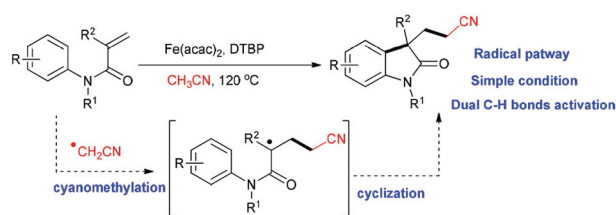


361

Fe-promoted radical cyanomethylation/arylation of arylacrylamides to access oxindoles via cleavage of the sp³ C–H of acetonitrile and the sp² C–H of the phenyl group

Changduo Pan, Honglin Zhang and Chengjian Zhu*

Radical cyanomethylation/arylation of arylacrylamides to access oxindoles with acetonitrile as a radical precursor is described, involving dual C–H bond functionalization.

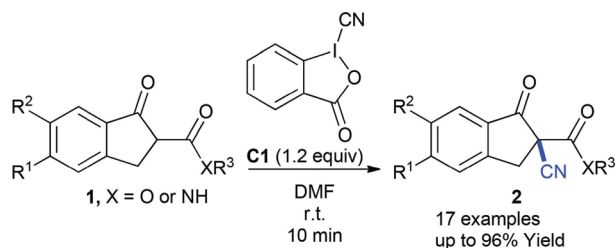


365

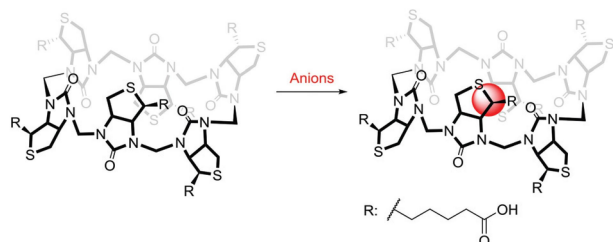
The direct electrophilic cyanation of β-keto esters and amides with cyano benziodoxole

Yao-Feng Wang, Jiashen Qiu, Dejie Kong, Yongtao Gao, Feipeng Lu, Pran Gopal Karmaker and Fu-Xue Chen*

Electrophilic cyanating agent **C1** accomplishes the direct cyanation of β-keto esters and amides without any catalyst and base.



369

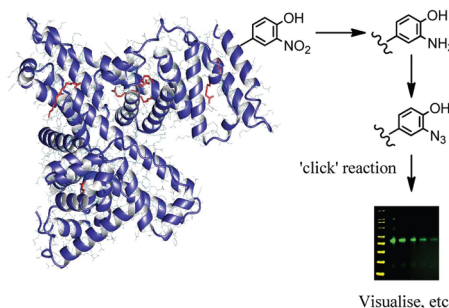


Anion binding by biotin[6]uril in water

Micke Lisbjerg, Bjarne E. Nielsen, Birgitte O. Milhøj, Stephan P. A. Sauer and Michael Pittelkow*

We show that the newly discovered 6 + 6 biotin-formaldehyde macrocycle Biotin[6]uril binds a variety of anionic guest molecules in water. We discuss how and why the anions are bound based on data obtained using NMR spectroscopy, mass spectrometry, isothermal titrations calorimetry (ITC), computational calculations and single crystal X-ray crystallography.

374



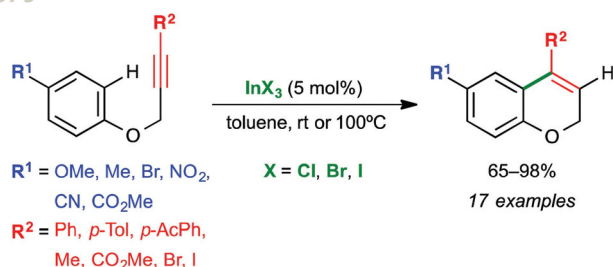
Bioorthogonal labelling of 3-nitrotyrosine in peptides and proteins through diazotisation mediated azidation

John Y. Ng and Jason W. H. Wong*

A bioorthogonal method of transforming 3-nitrotyrosine to 3-azidoxytyrosine is described, providing new opportunities to study 3-nitrotyrosine in biological samples.

PAPERS

379

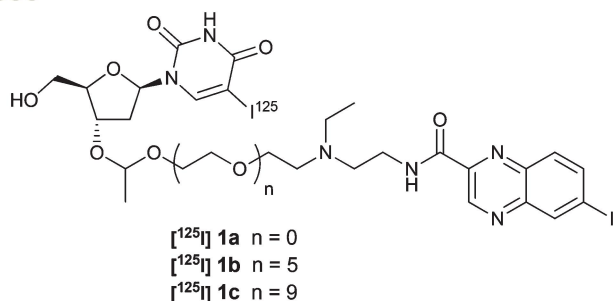


Indium-catalyzed intramolecular hydroarylation of aryl propargyl ethers

Lorena Alonso-Marañón, M. Montserrat Martínez, Luis A. Sarandeses and José Pérez Sestelo*

Indium(III) halides are efficient and selective catalysts for the intramolecular hydroarylation of aryl propargyl ethers.

388



PEGylation enhances the tumor selectivity of melanoma-targeted conjugates

Mathieu André, Sophie Besse, Jean-Michel Chezal and Emmanuelle Mounetou*

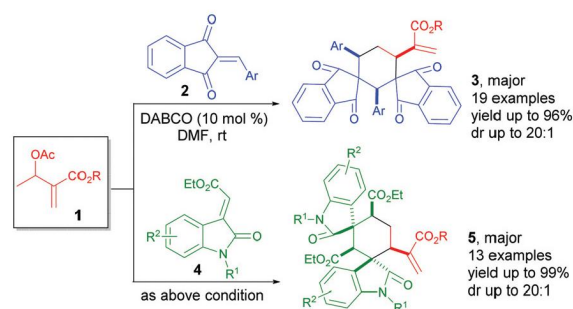
Three preselected conjugates of 5-iodo-2'-deoxyuridine (IUdR) to the ICF01012 melanoma-carrier, PEGylated and non-PEGylated, were radiolabelled with iodine-125, and their *in vivo* distribution profile was evaluated for potential intratumoural selective delivery.

398

Construction of dispirocyclohexanes *via* amine-catalyzed [2 + 2 + 2] annulations of Morita–Baylis–Hillman acetates with exocyclic alkenes

Rongshun Chen, Silong Xu,* Xia Fan, Hanyuan Li, Yuhai Tang and Zhengjie He*

Divergent amine-catalyzed [2 + 2 + 2] annulations of Morita–Baylis–Hillman acetates with exocyclic alkenes were developed for the convergent construction of multistereogenic dispirocyclohexanes.

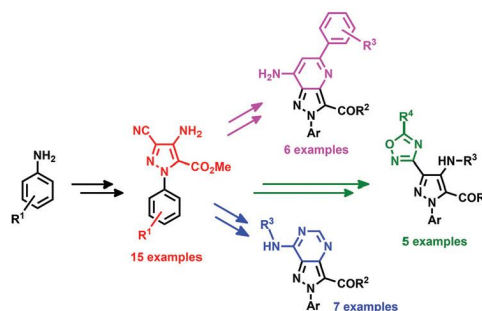


409

Microwave-assisted preparation of 4-amino-3-cyano-5-methoxycarbonyl-*N*-arylpyrazoles as building blocks for the diversity-oriented synthesis of pyrazole-based polycyclic scaffolds

Laurent Le Corre, Lotfi Tak-Tak, Arthur Guillard, Guillaume Prestat, Christine Gravier-Pelletier and Patricia Busca*

The obtention of 4-amino-3-cyano-*N*-arylpyrazoles *via* a μ W mediated cyclization allowed the diversity oriented syntheses of various heterocyclic scaffolds.

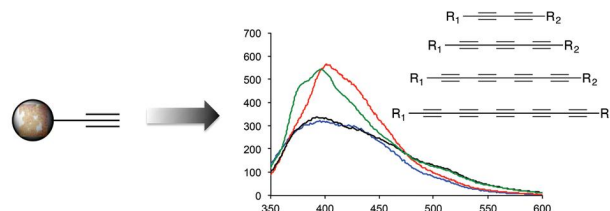


424

Preparation of asymmetrical polyynes by a solid-supported Glaser–Hay reaction

Jessica S. Lampkowski, Corinn E. Durham, Marshall S. Padilla and Douglas D. Young*

A novel approach to the synthesis of asymmetrical polyynes utilizing solid-supported chemistry.

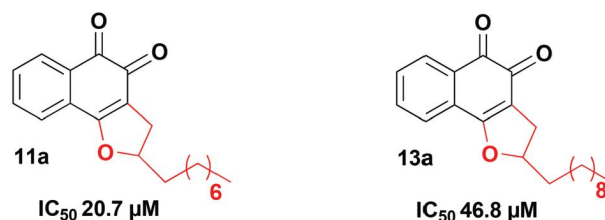


428

Novel naphthoquinone derivatives and evaluation of their trypanocidal and leishmanicidal activities

Aline Alves dos Santos Naujorks, Adriano Olímpio da Silva, Rosangela da Silva Lopes, Sérgio de Albuquerque, Adilson Beatriz, Maria Rita Marques and Dênis Pires de Lima*

Novel naphthoquinone derivatives were synthesized and tested for trypanocidal and leishmanicidal activities. Compound **11a** was the most active against *T. cruzi*.



438

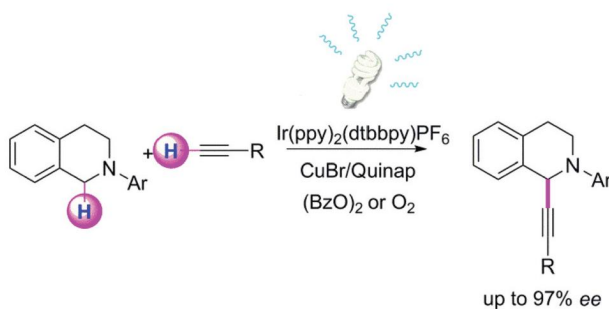


Bidirectional macrocyclization of peptides by double multicomponent reactions

Manuel G. Ricardo, Fidel E. Morales, Hilda Garay, Osvaldo Reyes, Dimitar Vasilev, Ludger A. Wessjohann* and Daniel G. Rivera*

Peptide macrocyclization by multicomponent reactions.

447

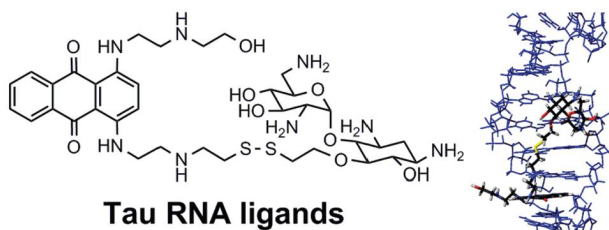


Efficient merging of copper and photoredox catalysis for the asymmetric cross-dehydrogenative-coupling of alkynes and tetrahydroisoquinolines

Inna Perepichka, Soumen Kundu, Zoë Hearne and Chao-Jun Li*

A highly efficient catalytic asymmetric alkylation of prochiral CH₂ groups in tetrahydroisoquinoline was developed using copper catalyzed cross-dehydrogenative-coupling of sp³ and sp C–H bonds with the assistance of a photocatalyst and visible light.

452



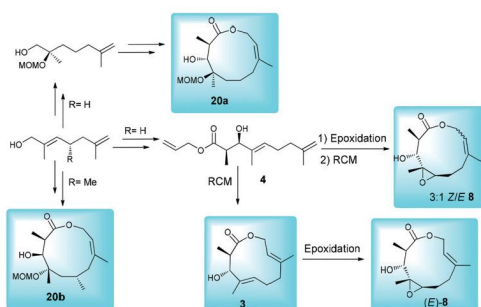
Tau RNA ligands

Ametantrone-based compounds as potential regulators of Tau pre-mRNA alternative splicing

Gerard Artigas, Paula López-Senín, Carlos González, Núria Escaja and Vicente Marchán*

Ligands with a preferred binding site in the bulged region of the stem–loop structure of Tau pre-mRNA have been identified by dynamic combinatorial chemistry.

465



The synthesis of 3-hydroxy-2,4,8-trimethyldec-8-enolides and an approach to 3,4-dihydroxy-2,4,6,8-tetramethyldec-8-enolide

J. M. Botubol-Ares, M. J. Durán-Peña, A. J. Macías-Sánchez, J. R. Hanson, I. G. Collado and R. Hernández-Galán*

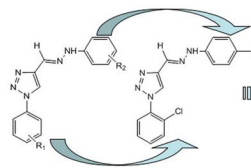
The enantioselective synthesis of **3**, (*E*)-**8**, (*Z*)-**8**, **20a** and **20b** is reported using RCM as key step. Based in spectroscopic differences, the structure assigned to the natural product has been revised to that of a γ -butyrolactone.

477

Synthesis and antifungal activity of 1,2,3-triazole phenylhydrazone derivatives

Zhi-Cheng Dai, Yong-Fei Chen, Mao Zhang, Sheng-Kun Li, Ting-Ting Yang, Li Shen, Jian-Xin Wang, Shao-Song Qian, Hai-Liang Zhu and Yong-Hao Ye*

A series of 1,2,3-triazole phenylhydrazone derivatives were designed and synthesized as antifungal agents.



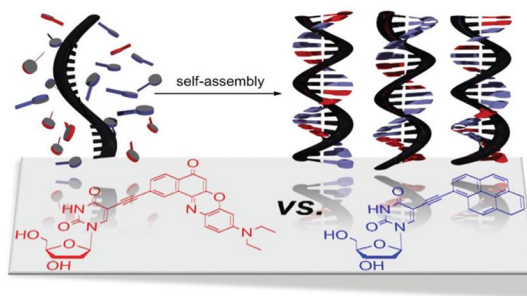
5p	EC ₅₀
<i>Rhizoctonia solani</i>	0.18 μg/mL
<i>Sclerotinia sclerotiorum</i>	2.28 μg/mL
<i>Fusarium graminearum</i>	1.01 μg/mL
<i>Phytophthora capsici</i>	1.85 μg/mL

487

Mixed non-covalent assemblies of ethynyl nile red and ethynyl pyrene along oligonucleotide templates

Philipp Ensslen, Yannic Fritz and Hans-Achim Wagenknecht*

The mixture makes the difference: the assemblies of ethynyl pyrene and ethynyl nile red along oligonucleotide templates are highly ordered and yield dual fluorescence.

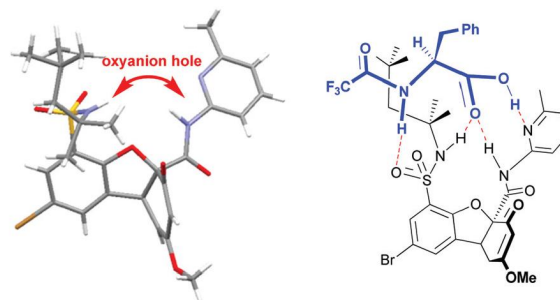


493

Chiral recognition with a benzofuran receptor that mimics an oxyanion hole

Ángel L. Fuentes de Arriba, Ángel Gómez Herrero, Omayra H. Rubio, Laura M. Monleón, Luis Simón Rubio, Victoria Alcázar, Francisca Sanz and Joaquín R. Morán*

Chiral resolution of a novel tripodal oxyanion-hole receptor with application in amino acid derivatives enantioselective extraction.

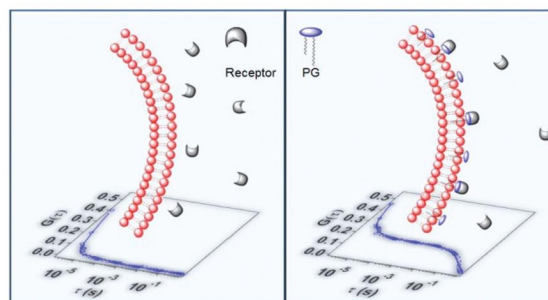


502

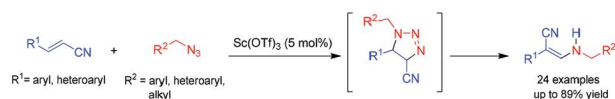
Developing a targeting system for bacterial membranes: measuring receptor-phosphatidylglycerol interactions with ¹H NMR, ITC and fluorescence correlation spectroscopy

Amanda Alliband, Zifan Wang, Christopher Thacker, Douglas S. English* and Dennis H. Burns*

We report the development of a potential targeting system for bacterial membranes containing phosphatidylglycerol.



513

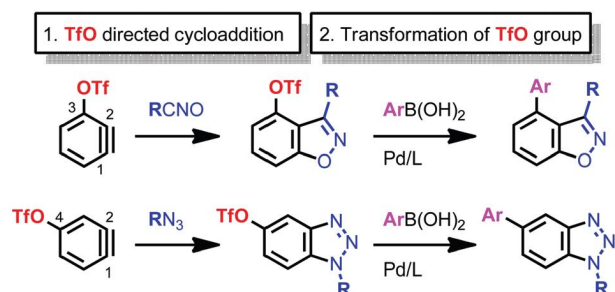


Sc(OTf)₃-mediated 1,3-dipolar cycloaddition–ring cleavage–rearrangement: a highly stereoselective access to Z-β-enaminonitriles

Ying-chun Wang, Yu-yang Xie, Xian-chun Tan, Heng-shan Wang* and Ying-ming Pan*

In this work, a novel and highly stereoselective synthesis of Z-β-enaminonitriles from azides and α,β-unsaturated nitriles is reported.

520

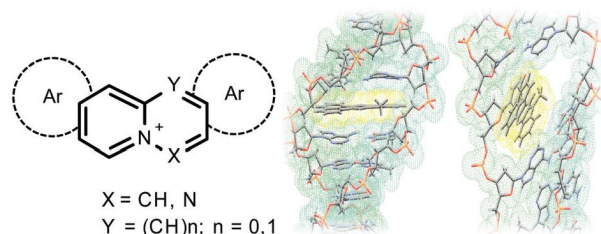


Trifluoromethanesulfonyloxy-group-directed regioselective (3 + 2) cycloadditions of benzynes for the synthesis of functionalized benzo-fused heterocycles

Takashi Ikawa,* Hideki Kaneko, Shigeaki Masuda, Erika Ishitsubo, Hiroaki Tokiwa and Shuji Akai*

Highly regioselective (3 + 2) cycloadditions of 3- and 4-TfO-benzynes with 1,3-dipoles followed by cross-coupling reactions provided multisubstituted benzo-fused heterocycles.

527

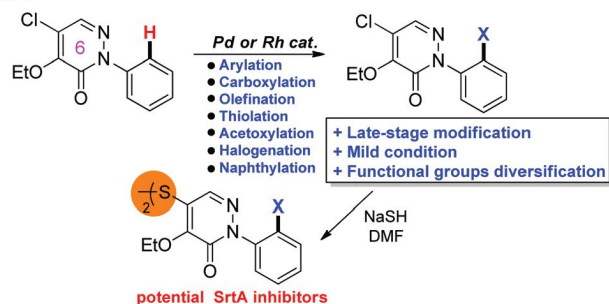


Targeting DNA with small molecules: a comparative study of a library of azonia aromatic chromophores

Rosa M. Suárez, Pedro Bosch, David Sucunza,* Ana M. Cuadro, Alberto Domingo, Francisco Mendicuti and Juan J. Vaquero*

The fluorescence, DNA binding and DNA sequence selectivity properties of a library of azonia aromatic cations have been studied.

539



Late-stage diversification of biologically active pyridazinones via a direct C–H functionalization strategy

Wei Li, Zhoulong Fan, Kaijun Geng, Youjun Xu* and Ao Zhang*

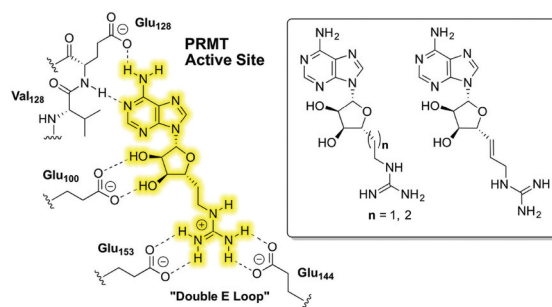
Divergent *ortho*-selective C–H functionalization was successfully established using a pyridazinone moiety as an internal directing group.

549

Synthesis and evaluation of protein arginine *N*-methyltransferase inhibitors designed to simultaneously occupy both substrate binding sites

Matthijs van Haren, Linda Quarles van Ufford, Ed E. Moret and Nathaniel I. Martin*

PRMT inhibitors designed to simultaneously occupy both substrate binding sites display potent activity and surprising selectivity.

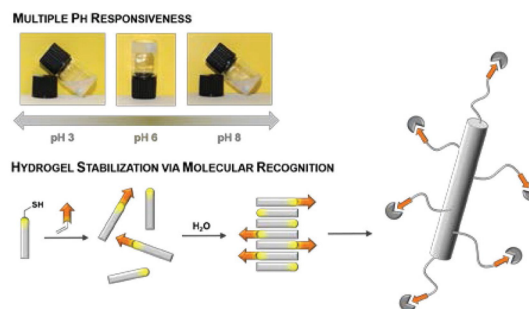


561

pH response and molecular recognition in a low molecular weight peptide hydrogel

Stefanie C. Lange, Jan Unsleber, Patrick Drücker, Hans-Joachim Galla, Mark P. Waller and Bart Jan Ravoo*

The preparation and characterization of a tripeptide based hydrogel, which possesses characteristic rheological properties, is pH responsive and can be functionalized at its thiol function is reported.

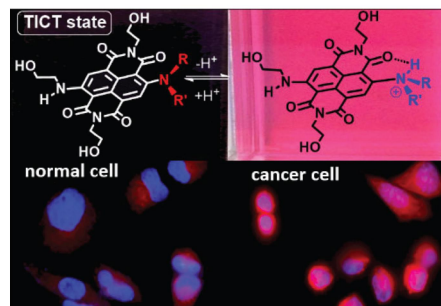


570

Naphthalene diimides as red fluorescent pH sensors for functional cell imaging

Filippo Doria,* Marco Folini, Vincenzo Grande, Graziella Cimino-Reale, Nadia Zaffaroni and Mauro Freccero*

Water-soluble naphthalene diimides have been designed and synthesized as cell permeable pH "turned-on" fluorescent sensors for cellular applications.

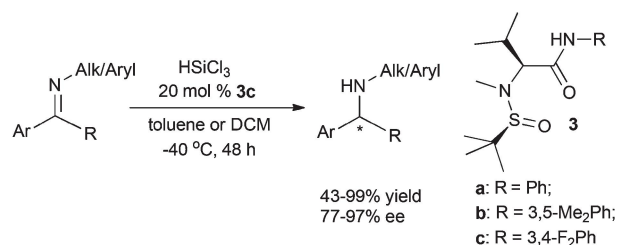


577

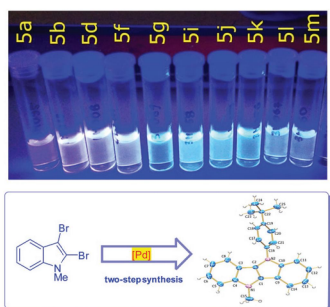
L-Valine derived chiral *N*-sulfinamides as effective organocatalysts for the asymmetric hydrosilylation of *N*-alkyl and *N*-aryl protected ketimines

Chao Wang,* Xinjun Wu, Li Zhou and Jian Sun*

L-Valine derived *N*-sulfinamides have been developed as efficient enantioselective Lewis basic organocatalysts for the asymmetric reduction of *N*-aryl and *N*-alkyl ketimines with trichlorosilane.



583

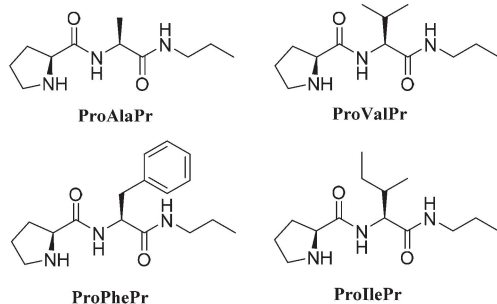


Novel synthesis of 5-methyl-5,10-dihydroindolo[3,2-*b*]indoles by Pd-catalyzed C–C and two-fold C–N coupling reactions

Tran Quang Hung, Sören Hancker, Alexander Villinger, Stefan Lochbrunner, Tuan Thanh Dang,* Aleksej Friedrich, Wolfgang Breitsprecher and Peter Langer*

A series of 5,10-dihydroindolo[3,2-*b*]indoles was successfully prepared by an efficient two-step strategy.

592

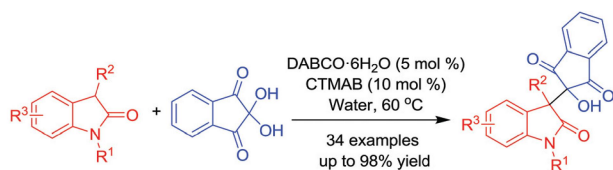


Structural insight into the aggregation of L-prolyl dipeptides and its effect on organocatalytic performance

Cristina Berdugo, Beatriu Escuder* and Juan F. Miravet*

NMR and organocatalytic studies of four dipeptides derived from L-proline are described.

601

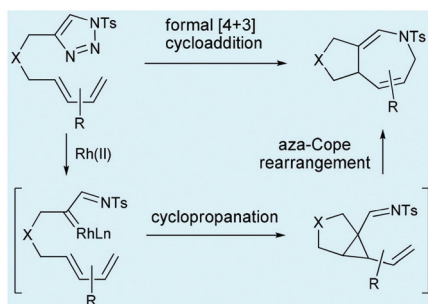


A highly efficient and eco-friendly method for the synthesis of 1,3-indandione ring-fused 3-oxindoles bearing two contiguous quaternary stereocenters via an aldol reaction in aqueous media

Xiong-Li Liu,* Bo-Wen Pan, Wen-Hui Zhang, Chao Yang, Jun Yang, Yang Shi, Ting-Ting Feng, Ying Zhou* and Wei-Cheng Yuan

The synthesis and antitumor activity evaluation of 1,3-indandione ring-fused 3-oxindoles.

612



Rhodium(II)-catalyzed intramolecular formal [4 + 3] cycloadditions of dienyltriazoles: rapid access to fused 2,5-dihydroazepines

Yu Tian, Yuanhao Wang, Hai Shang, Xudong Xu and Yefeng Tang*

Rh(II)-catalyzed intramolecular formal [4 + 3] cycloadditions of dienyltriazoles have been developed, which enable the efficient synthesis of various fused 2,5-dihydroazepines.

620

Cyclen-based cationic lipids containing a pH-sensitive moiety as gene delivery vectors

Zheng Huang, Yan-Hong Liu, Yi-Mei Zhang, Ji Zhang,*
Qiang Liu and Xiao-Qi Yu*

Imidazole-functionalized cationic lipids with a cyclen headgroup were synthesized, and the structure–activity relationship in gene delivery mediated by these lipids was discussed.

