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Organic & Biomolecular Chemistry

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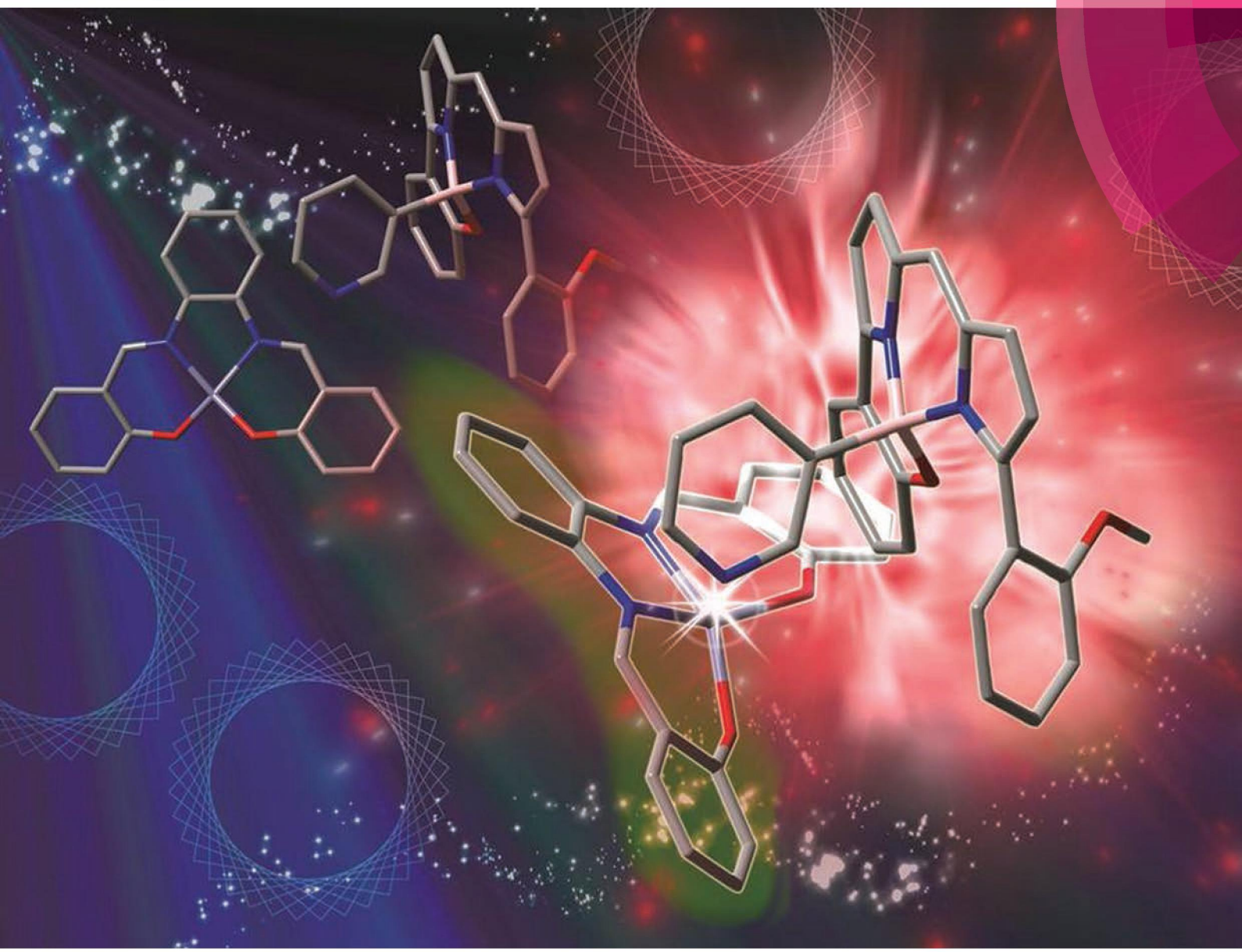

ISSN 1477-0520

ROYAL SOCIETY OF CHEMISTRY

PAPER

Tatsuya Nabeshima *et al.*

A facile and high-yield formation of dipyrrin-boronic acid dyads and triads: a light-harvesting system in the visible region based on the efficient energy transfer

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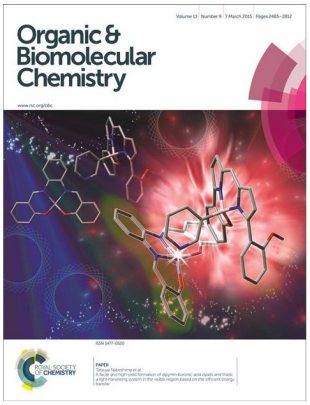
IN THIS ISSUE

ISSN 1477-0520 CODEN OBCRAK 13(9) 2485–2812 (2015)



Cover
See Tatsuya Nabeshima *et al.*, pp. 2574–2581.

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EDITORIAL

2499

Editorial: Supramolecular chemistry in water

Anthony P. Davis, Stefan Kubik and Antonella Dalla Cort

This themed collection highlights some of the exciting research in supramolecular chemistry in water.

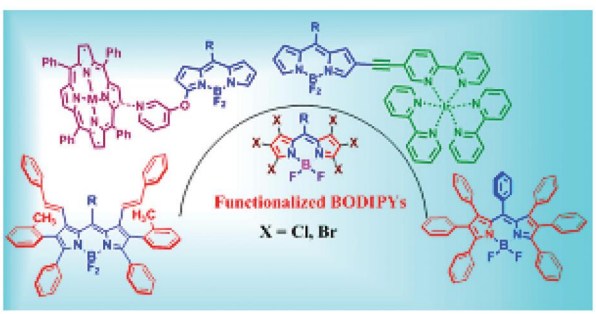
REVIEWS

2501

Halogenated boron-dipyrromethenes: synthesis, properties and applications

Vellanki Lakshmi, Malakalapalli Rajeswara Rao and Mangalampalli Ravikanth*

Synthesis and properties of halogenated boron-dipyrromethenes and their applications in developing various BODIPY systems are described in this review.



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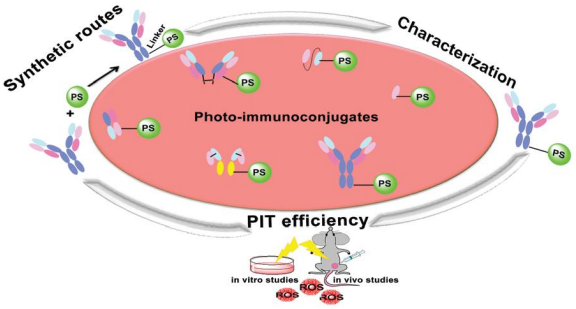
REVIEWS

2518

Antibodies armed with photosensitizers: from chemical synthesis to photobiological applications

Patricia M. R. Pereira, Barbara Korsak, Bruno Sarmento, Rudolf J. Schneider, Rosa Fernandes and João P. C. Tomé*

Targeting photosensitizers to cancer cells by conjugating them with specific antibodies, able to recognize and bind to tumor-associated antigens, is today one of the most attractive strategies in photodynamic therapy (PDT).

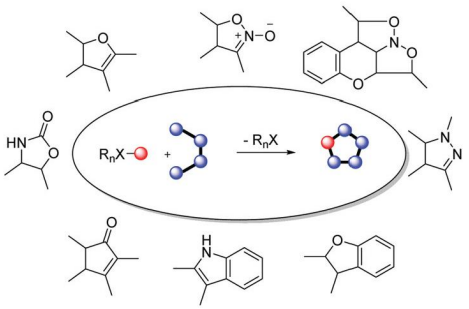


2530

Ylide formal [4 + 1] annulation

Chunyin Zhu,* Ya Ding and Long-Wu Ye*

Ylide [4 + 1] annulation: beyond small-ring formation.



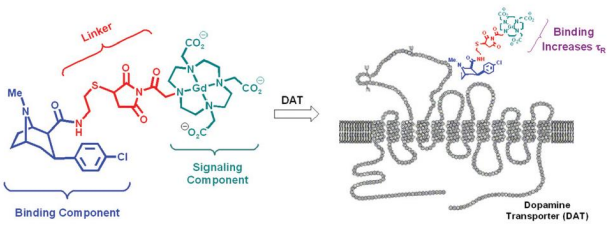
COMMUNICATIONS

2537

Synthesis of a β-CCT-lanthanide conjugate for binding the dopamine transporter

Gregory R. Naumiec, Grace Lincourt, Jeremy P. Clever, Michael A. McGregor, Abraham Kovoor and Brenton DeBoef*

The development of a β-CCT-lanthanide conjugate that binds the dopamine transporter (DAT) with high affinity (K_d = 303 nM) is described. This molecular probe could be used for *in vivo* or *in vitro* studies of the DAT *via* MRI, PET or SPECT.

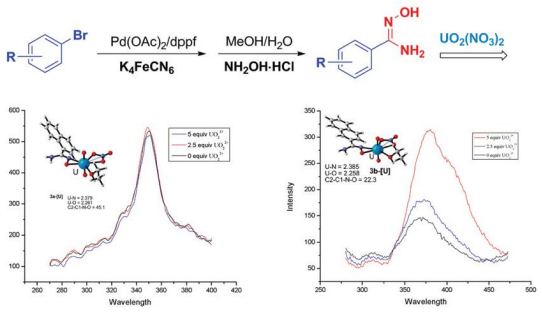


2541

"One-pot" synthesis of amidoxime *via* Pd-catalyzed cyanation and amidoximation

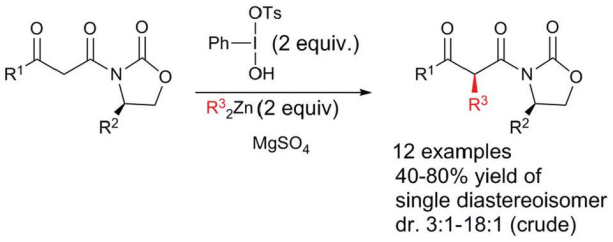
Chu-Ting Yang, Jun Han, Jun Liu, Mei Gu, Yi Li, Jun Wen, Hai-Zhu Yu, Sheng Hu* and Xiaolin Wang*

"One-pot" synthesis of amidoxime was developed for studies on the interactions between amidoxime and uranyl.



COMMUNICATIONS

2546

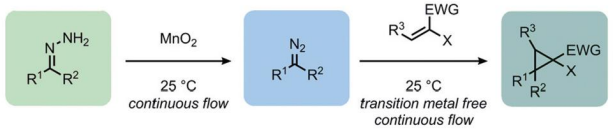


Oxidative asymmetric umpolung alkylation of Evans' β-ketoimides using dialkylzinc nucleophiles

Tom A. Targel, Jayprakash N. Kumar, O. Svetlana Shneider, Sukanta Bar, Natalia Fridman, Shimon Maximenko and Alex M. Szpilman*

Umpolung alkylation of Evans' auxiliary substituted β-ketoimides affords the diastereomerically pure products in yields ranging from 40 to 80%.

2550

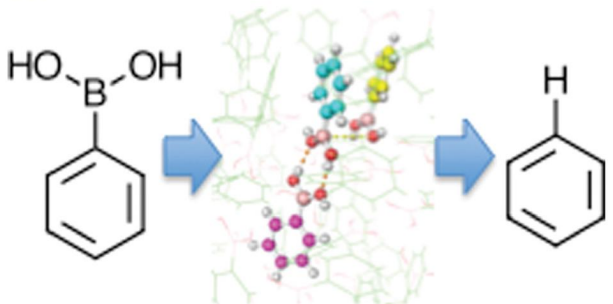


Cyclopropanation using flow-generated diazo compounds

Nuria M. Roda, Duc N. Tran, Claudio Battilocchio, Ricardo Labes, Richard J. Ingham, Joel M. Hawkins and Steven V. Ley*

A practical and mild protocol for the cyclopropanation of unstabilised diazo compounds is reported.

2555

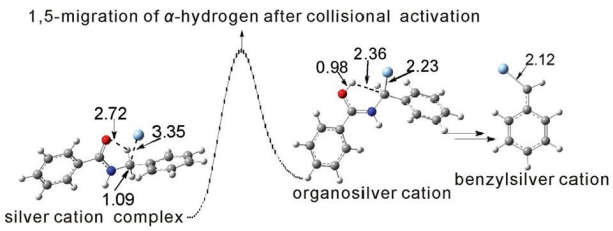


A mechanistic proposal for the protodeboronation of neat boronic acids: boronic acid mediated reaction in the solid state

Gary Noonan* and Andrew G. Leach*

Boronic acids that undergo protodeboronation as solids are stable in solution: the solid state organizes them for reaction.

2561



Gas phase chemistry of *N*-benzylbenzamides with silver(I) cations: characterization of benzylsilver cation

Hezhi Sun, Zhe Jin, Hong Quan, Cuirong Sun* and Yuanjiang Pan*

Benzylsilver cations are synthesized in the gas phase from the collisional dissociation of argentinated *N*-benzylbenzamides, when the carbonyl oxygen nucleophilically attacks an α-hydrogen.

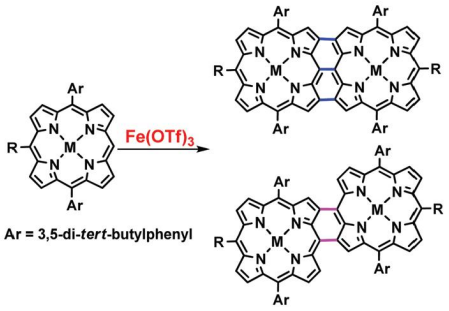
COMMUNICATIONS

2566

Synthesis of directly fused porphyrin dimers through $Fe(OTf)_3$-mediated oxidative coupling

Chuan-Mi Feng, Yi-Zhou Zhu,* Shao-Chun Zhang, Yun Zang and Jian-Yu Zheng*

A practical and general $Fe(OTf)_3$-mediated oxidative coupling method was developed for the synthesis of doubly or triply linked porphyrin dimers.

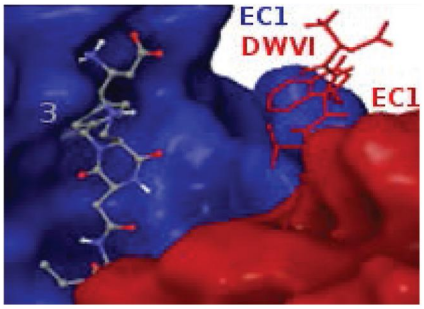


2570

Computational design of novel peptidomimetic inhibitors of cadherin homophilic interactions

Fabio Doro, Cinzia Colombo, Chiara Alberti, Daniela Arosio, Laura Belvisi, Cesare Casagrande, Roberto Fanelli, Leonardo Manzoni, Emilio Parisini, Umberto Piarulli, Elena Luison, Mariangela Figini, Antonella Tomassetti* and Monica Civera*

Peptidomimetic **3** mimicking the DWVI adhesive sequence of the cadherin EC1 domain inhibits cadherin-mediated adhesion in epithelial ovarian cancer cells.



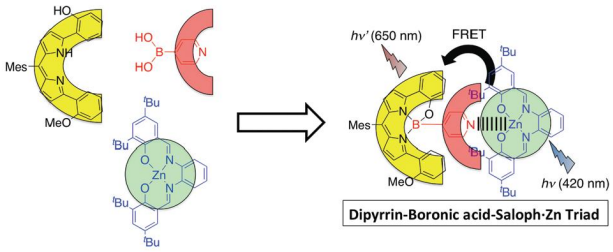
PAPERS

2574

A facile and high-yield formation of dipyrrin-boronic acid dyads and triads: a light-harvesting system in the visible region based on the efficient energy transfer

Masaki Yamamura, Shinya Yazaki, Motofumi Seki, Yasunori Matsui, Hiroshi Ikeda and Tatsuya Nabeshima*

Artificial light-harvesting systems, Ar,O-BODIPY dyads and triads conjugated with a light harvester, were synthesized in high yield by the reaction of an N_2O_2-type dipyrrin with boronic acids.

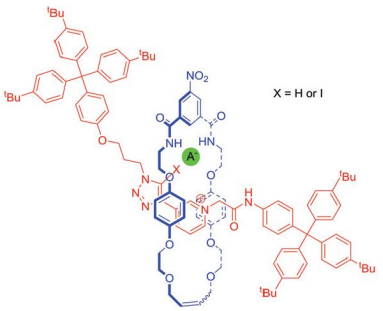


2582

Contrasting anion recognition behaviour exhibited by halogen and hydrogen bonding rotaxane hosts

Stuart P. Cornes, Charles H. Davies, David Blyghton, Mark R. Sambrook and Paul D. Beer*

A [2]rotaxane anion host that switches selectivity from dihydrogen phosphate to the halides upon substituting a hydrogen bond donor group for a halogen bond donor group within the axle component is described.



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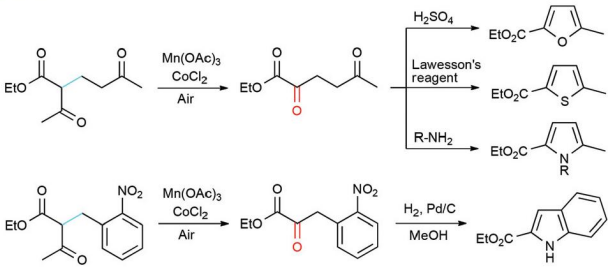
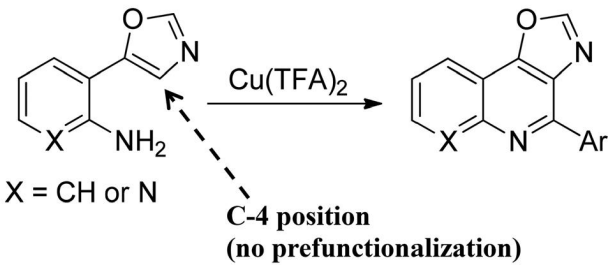


Tandem catalytic oxidative deacetylation of acetoacetic esters and heteroaromatic cyclizations

Yeming Ju, Di Miao, Ruiyang Yu and Sangho Koo*

Coupling of ethyl acetoacetate with MVK and with 2-nitrobenzyl bromides, Mn(III)/Co(II)-catalyzed deacetylation, and heterocyclizations provide pyrroles and indoles, respectively.

2600

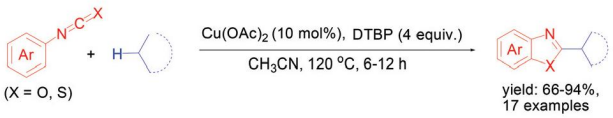


Synthesis of 4-substituted oxazolo[4,5-*c*]quinolines by direct reaction at the C-4 position of oxazoles

Mahesh Akula, Yadagiri Thigulla, Connor Davis, Mukund Jha and Anupam Bhattacharya*

$Cu(TFA)_2$ catalysed synthesis of 4-arylsubstituted oxazolo-[4,5-*c*]quinolines/[1,8] naphthyridines has been described *via* a modified Pictet–Spengler method, without prefunctionalization of the unreactive 4th position of oxazoles.

2606

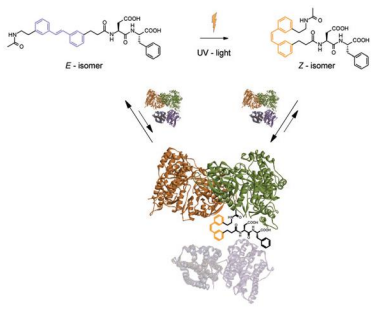


Direct construction of 2-alkylbenzo-1,3-azoles *via* C–H activation of alkanes for C–C and C–X (X = O, S) bond formation

Arvind K. Yadav and Lal Dhar S. Yadav*

2-Alkylated benzo(oxa)thiazoles were prepared directly from simple alkanes and aryl isocyanates/isothiocyanates in a one-pot procedure.

2612



Phototriggerable peptidomimetics for the inhibition of *Mycobacterium tuberculosis* ribonucleotide reductase by targeting protein–protein binding

Christoffer Karlsson, Magnus Blom, Miranda Johansson (neé Varedian), Anna M. Jansson, Enzo Scifo, Anders Karlén, Thavendran Govender and Adolf Gogoll*

Peptidomimetic inhibitors with photomodulable affinity for the R1–R2 subunit association site were designed based on the R2-subunit C-terminal.

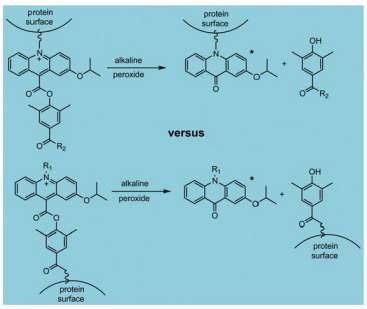
PAPERS

2622

A comparison of chemiluminescent acridinium dimethylphenyl ester labels with different conjugation sites

Anand Natrajan* and David Wen

Chemiluminescent acridinium esters containing conjugation sites at the acridinium nitrogen show increased light yield when labeled to proteins.

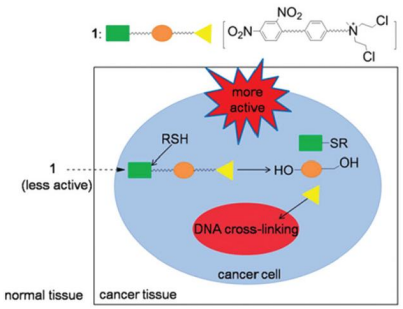


2634

Synthesis and biological studies of the thiols-triggered anticancer prodrug for a more effective cancer therapy

Yuanzhen Xu, Jianjun Chen, Ya Li, Shoujiao Peng, Xueyan Gu, Meng Sun, Kun Gao* and Jianguo Fang*

Thiols-triggered anticancer prodrug **1** produces an active DNA alkylating agent mechlorethamine leading to DNA damage and finally cell death.

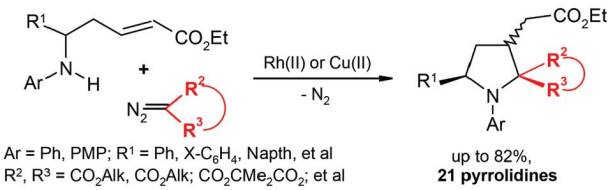


2640

Domino [4 + 1]-annulation of α,β-unsaturated δ-amino esters with Rh(II)–carbenoids – a new approach towards multi-functionalized *N*-aryl pyrrolidines

J. J. Medvedev, O. S. Galkina, A. A. Klinkova, D. S. Giera, L. Hennig, C. Schneider and V. A. Nikolaev*

Catalytic decomposition of diazoesters using Rh(II)- and Cu(II)-complexes in the presence of δ-amino-α,β-unsaturated esters produces multi-functionalized pyrrolidines.

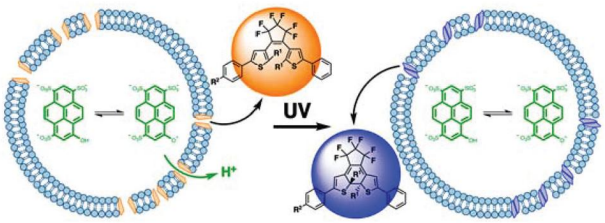


2652

Photocontrol of ion permeation in lipid vesicles with amphiphilic dithienylethenes

Yamuna S. Kandasamy, Jianxin Cai, Alisha Beler, M.-S. Jemeli Sang, Patrick D. Andrews and R. Scott Murphy*

Asymmetrical amphiphilic dithienylethenes have been prepared and photocontrol of ion permeation was observed in lipid vesicles.



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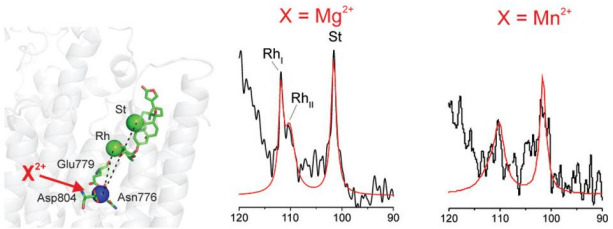
2664

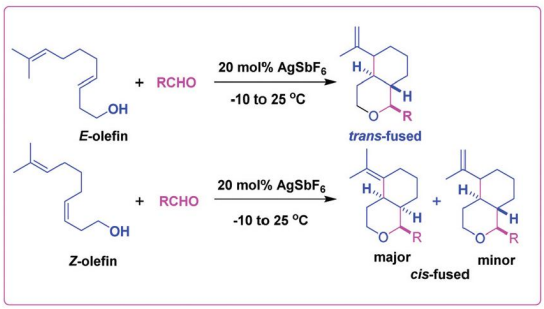


Ligand orientation in a membrane-embedded receptor site revealed by solid-state NMR with paramagnetic relaxation enhancement

Christopher A. P. Whittaker, Simon G. Patching, Mikael Esmann and David A. Middleton*

Paramagnetic relaxation-enhanced solid-state NMR reveals a ouabain analogue with an inverted orientation in the Na, K-ATPase inhibitory site.

2669

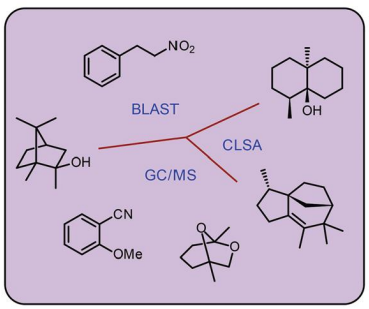


A novel Prins cascade process for the stereoselective synthesis of oxa-bicycles

B. V. Subba Reddy,* A. Venkateswarlu, B. Sridevi, Kanakaraju Marumudi, A. C. Kunwar and G. Gayatri

E- and *Z*-9-Methyldeca-3,8-dien-1-ols undergo smooth cyclization with aldehydes in the presence of 20 mol% $AgSbF_6$ under extremely mild conditions to generate the corresponding oxa-bicycles in good yields with excellent selectivity.

2673

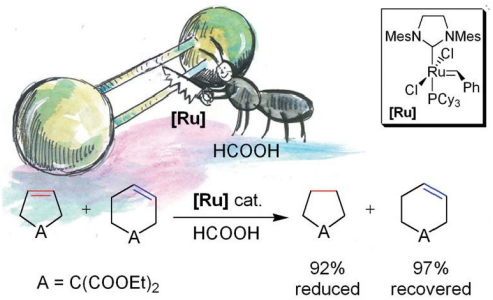


Volatiles from nineteen recently genome sequenced actinomycetes

Christian A. Citron, Lena Barra, Joachim Wink and Jeroen S. Dickschat*

The volatiles from nineteen genome sequenced actinobacteria were analysed by GC/MS and the identified terpenes were correlated to genome data.

2684



In tandem or alone: a remarkably selective transfer hydrogenation of alkenes catalyzed by ruthenium olefin metathesis catalysts

Grzegorz Krzysztof Zieliński, Cezary Samojłowicz, Tomasz Wdowik and Karol Grela*

A remarkably selective system for transfer hydrogenation of alkenes, composed of Grubbs' ruthenium metathesis catalyst and HCOONa/HCOOH, is presented. This system can also be formed directly after a metathesis reaction to effect hydrogenation in a single-pot.

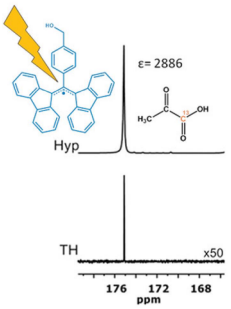
PAPERS

2689

A benzyl alcohol derivative of the BDPA radical for fast dissolution dynamic nuclear polarization NMR spectroscopy

J. L. Muñoz-Gómez, E. Monteagudo, V. Lloveras, T. Parella, J. Veciana* and J. Vidal-Gancedo*

A pyruvic acid soluble BDPA derivative, BA-BDPA, is a promising candidate as a polarizing agent for *in vivo* DNP.

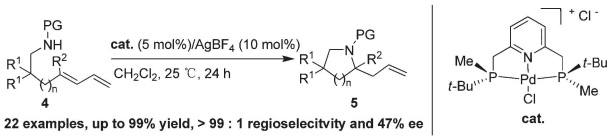


2694

P-stereogenic PNP pincer-Pd catalyzed intramolecular hydroamination of amino-1,3-dienes

Zehua Yang, Chao Xia, Delong Liu, Yangang Liu, Masashi Sugiya, Tsuneo Imamoto* and Wanbin Zhang*

A new P-stereogenic PNP pincer-Pd complex was readily prepared and was used in the asymmetric intramolecular hydroamination of amino-1,3-dienes, with the desired products being obtained in good yields and with excellent regioselectivities and up to moderate enantioselectivities.

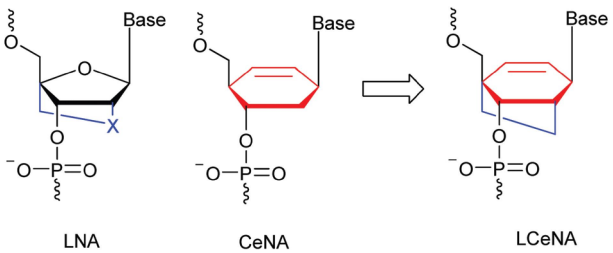


2703

Synthesis of locked cyclohexene and cyclohexane nucleic acids (LCeNA and LCNA) with modified adenosine units

Michal Šála,* Milan Dejmek, Eliška Procházková, Hubert Hřebabecký, Jiří Rybáček, Martin Dračínský, Pavel Novák, Šárka Rosenbergová, Jiří Fukal, Vladimír Sychrovský, Ivan Rosenberg and Radim Nencka*

We designed novel conformationally locked cyclohexene nucleic acid and studied their properties.

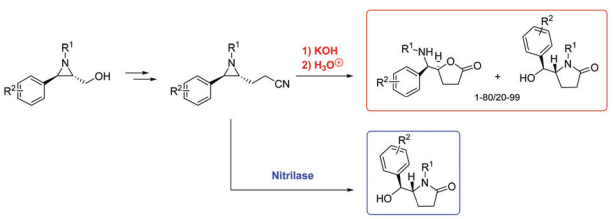


2716

Synthesis of 2-aryl-3-(2-cyanoethyl)aziridines and their chemical and enzymatic hydrolysis towards γ-lactams and γ-lactones

Karen Mollet, Lena Decuyper, Saskia Vander Meeren, Nicola Piens, Karel De Winter, Tom Desmet and Matthias D'hooghe*

Trans- and *cis*-2-aryl-3-(2-cyanoethyl)aziridines were transformed into 4-[aryl(alkylamino)methyl]butyrolactones and/or 5-[aryl(hydroxy)methyl]pyrrolidin-2-ones *via* chemical and enzymatic hydrolysis of the cyano group, followed by ring expansion.



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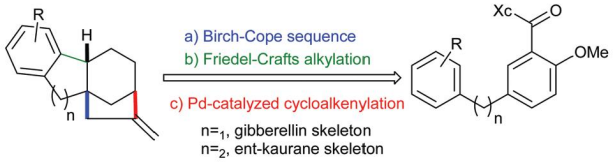
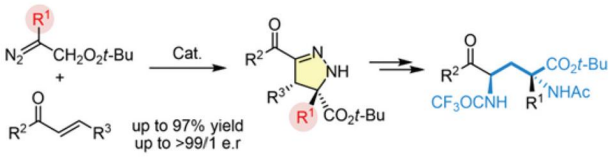


The enantioselective construction of tetracyclic diterpene skeletons with Friedel–Crafts alkylation and palladium-catalyzed cycloalkenylation reactions

Sarah J. Burke, William P. Malachowski,* Sharan K. Mehta and Roselyn Appenteng

Enantioselective synthesis of natural product-like structures from a two-step extension of the Birch–Cope sequence: intramolecular Friedel–Crafts alkylation and palladium-catalyzed cycloalkenylation.

2745

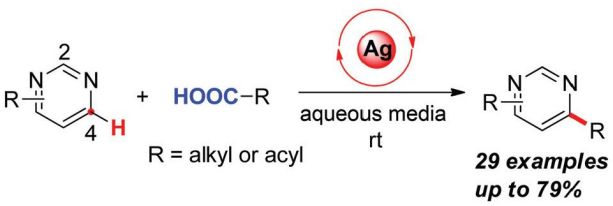


Highly enantioselective catalytic 1,3-dipolar cycloadditions of α-alkyl diazoacetates: efficient synthesis of functionalized 2-pyrazolines

Sung Il Lee, Ka Eun Kim, Geum-Sook Hwang* and Do Hyun Ryu*

Chiral oxazaborolidinium ion catalyzed 1,3-dipolar cycloaddition reaction of α-substituted diazoacetates gives functionalized 2-pyrazolines in high to excellent enantiomeric ratios.

2750

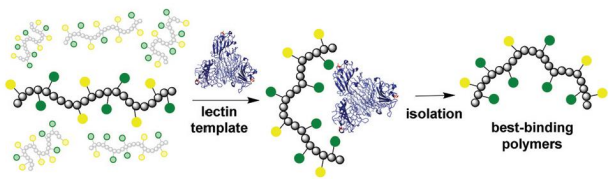


Silver catalysed decarboxylative alkylation and acylation of pyrimidines in aqueous media

Wen-Peng Mai,* Bin Sun, Li-Qin You, Liang-Ru Yang, Pu Mao,* Jin-Wei Yuan, Yong-Mei Xiao and Ling-Bo Qu*

Decarboxylative alkylation or acylation reactions of simple pyrimidines have been developed in aqueous media.

2756



Templating carbohydrate-functionalised polymer-scaffolded dynamic combinatorial libraries with lectins

Clare S. Mahon, Martin A. Fascione, Chadamas Sakonsinsiri, Tom E. McAllister, W. Bruce Turnbull and David A. Fulton*

The templation of carbohydrate-functionalised Polymer-Scaffolded Dynamic Combinatorial Libraries affords polymers possessing significantly enhanced affinities for the template, with enhancements in free energy of binding in the range of 5.2–8.8 kJ mol^{-1} observed.

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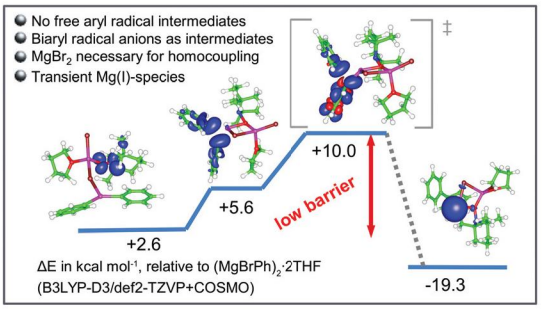
TEMPO-mediated homocoupling of aryl Grignard reagents: mechanistic studies

Sandip Murarka, Juri Möbus, Gerhard Erker, Christian Mück-Lichtenfeld* and Armido Studer*

The mechanism of TEMPO mediated oxidative homo-coupling of aryl Grignard reagents to biphenyls is investigated in detail by experimental and computational studies.

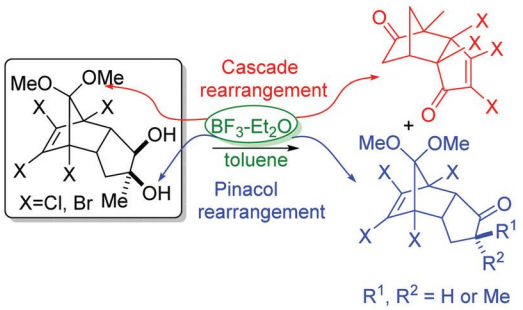


2768

BF_3-Et_2O mediated skeletal rearrangements of norbornyl appended cyclopentanediols

Chintada Nageswara Rao and Faiz Ahmed Khan*

Synthetically useful norbornyl appended cyclopentenones and pinacolone products are reported during the BF_3-Et_2O mediated skeletal rearrangement of norbornyl appended cyclopentanediols.

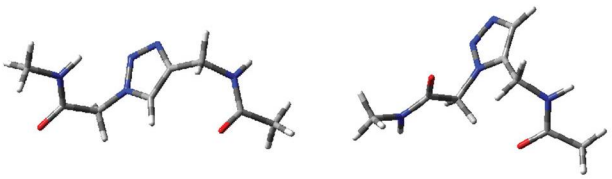


2776

Conformational properties of 1,4- and 1,5-substituted 1,2,3-triazole amino acids – building units for peptidic foldamers

Nina Kann,* Johan R. Johansson* and Tamás Beke-Somfai*

Conformational diversity of 1,4- and 1,5-substituted 1,2,3-triazole amino acids makes them promising building units for novel peptidic foldamers.

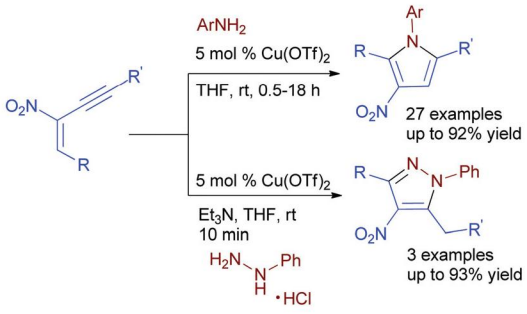


2786

Expedient synthesis of tetrasubstituted pyrroles *via* a copper-catalyzed cascade inter-/intramolecular cyclization of 1,3-enynes carry a nitro group with amines

Ganesan Bharathiraja, Mani Sengoden, Masanam Kannan and Tharmalingam Punniyamurthy*

Various tetrasubstituted pyrroles/pyrazoles have been prepared from nitro-substituted 1,3-enynes with aromatic amines/hydrazines *via* a copper-catalyzed cascade aza-Michael addition, cyclization and aromatization.



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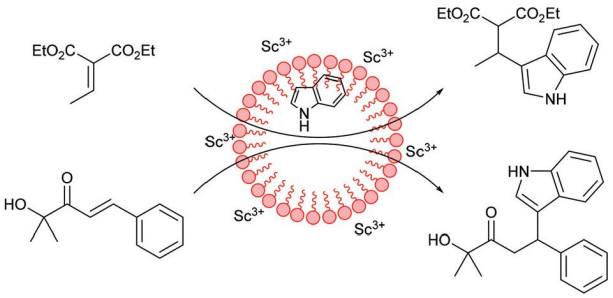
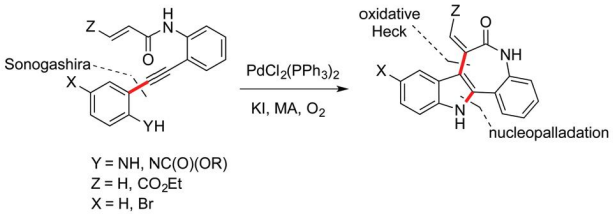


Alkylidene malonates and α,β-unsaturated α′-hydroxyketones as practical substrates for vinylogous Friedel–Crafts alkylations in water catalysed by scandium(III) triflate/SDS

Jens Oelerich and Gerard Roelfes*

Alkylidene malonates and α,β-unsaturated α′-hydroxyketones are excellent substrates for the $Sc(OTf)_3$/SDS catalysed Friedel–Crafts alkylation in water.

2800



Synthesis of 7-alkylidene-7,12-dihydroindolo-[3,2-*d*]benzazepine-6-(5*H*)-ones (7-alkylidene-paullones) by N-cyclization–oxidative Heck cascade and characterization as sirtuin modulators

J. G. Denis, G. Franci, L. Altucci, J. M. Aurrecoechea, Á. R. de Lera* and R. Álvarez*

A palladium-induced cascade of N-cyclization and oxidative Heck reaction of *o*-alkynylanilines produced 7-alkylidene-indolobenzazepinones (paullones) that have sirtuin modulation activities.