

Organic & Biomolecular Chemistry

An international journal of synthetic, physical and biomolecular organic chemistry

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IN THIS ISSUE

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Cover

See Tadashi Ema *et al.*, pp. 6299–6308.

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Inside cover

See Wen-Xiong Zhang, Zhenfeng Xi *et al.*, pp. 6266–6270.

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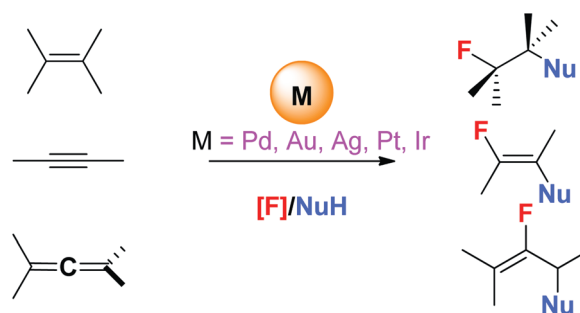
EMERGING AREA

6243

Transition metal-catalyzed fluorination of multi carbon–carbon bonds: new strategies for fluorinated heterocycles

Guosheng Liu*

Recent progress in transition metal-catalyzed fluorination is reviewed with an emphasis on the selective fluorination of multi carbon–carbon bonds.



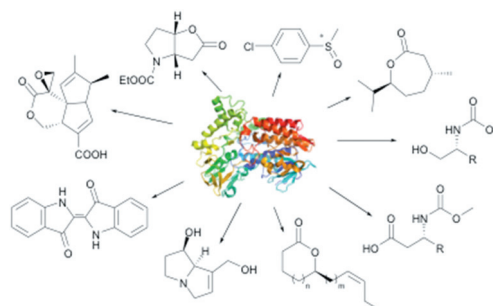
PERSPECTIVE

6249

Discovery, application and protein engineering of Baeyer–Villiger monoxygenases for organic synthesis

Kathleen Balke, Maria Kadow, Hendrik Mallin, Stefan Saß and Uwe T. Bornscheuer*

This perspective article summarizes how enzyme discovery, protein engineering and optimization of reaction systems contributed to synthetic applications of Baeyer–Villiger monoxygenases.



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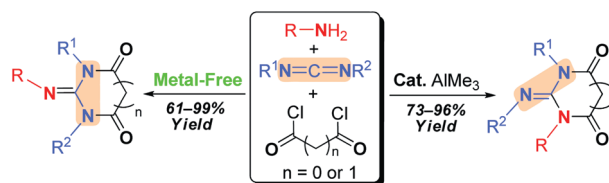
COMMUNICATIONS

6266

Metal-free synthesis of cyclic di-oxoguanidines via one-pot sequential transformation of amines, carbodiimides and acyl dichlorides

Fei Zhao, Yang Wang, Wen-Xiong Zhang* and Zhenfeng Xi

Two sequential transformations from the same starting materials are achieved under metal-free or AlMe₃-catalyzed conditions providing structurally different cyclic di-oxoguanidines.

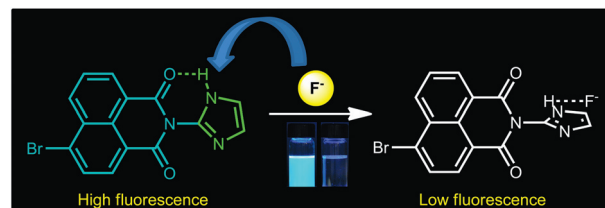


6271

A new *N*-imidazolyl-1,8-naphthalimide based fluorescence sensor for fluoride detection

Junqi Wang, Lingyun Yang, Chen Hou and Haishi Cao*

An *N*-imidazolyl-1,8-naphthalimide behaves as a fluoride sensor with high selectivity in CH₂Cl₂ owing to disruption of the intramolecular H-bond by fluoride.

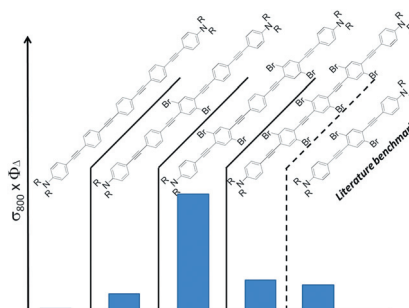


6275

Influence of bromine substitution pattern on the singlet oxygen generation efficiency of two-photon absorbing chromophores

Pierre-Henri Lanoë, Thibault Gallavardin, Aurore Dupin, Olivier Maury, Patrice L. Baldeck, Mikael Lindgren, Cyrille Monneré* and Chantal Andraud*

Position of bromine substituents along the backbone of quadrupolar chromophores determines their two-photon induced singlet oxygen generation efficiency.



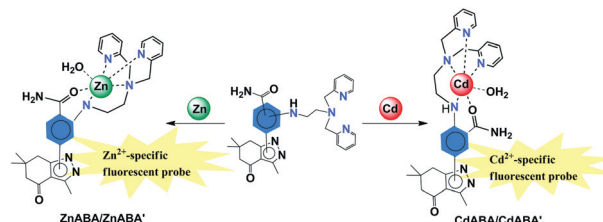
PAPERS

6279

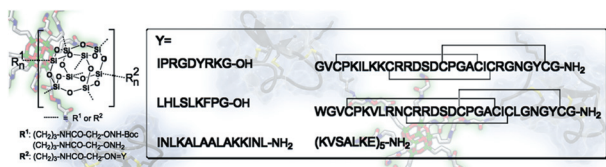
Tetrahydroindazolone substituted 2-aminobenzamides as fluorescent probes: switching metal ion selectivity from zinc to cadmium by interchanging the amino and carbonyl groups on the fluorophore

Jia Jia, Qin-Chao Xu, Ri-chen Li, Xi Tang, Ying-Fang He, Meng-Yu Zhang, Yuan Zhang and Guo-Wen Xing*

Three Cd²⁺ or Zn²⁺-specific fluorescent probes have been developed with *N,N*-bis(2-pyridylmethyl)ethylenediamine as chelator and 2-aminobenzamide as fluorophore.



6287

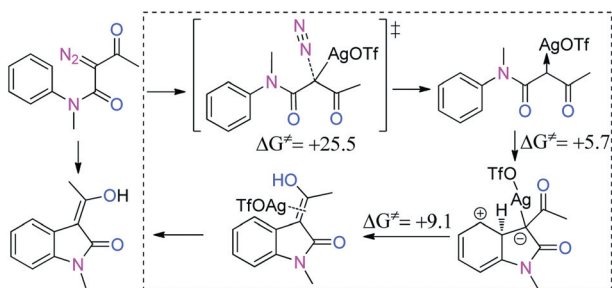


From pico to nano: biofunctionalization of cube-octameric silsesquioxanes by peptides and miniproteins

Sebastian Fabritz, Sebastian Hörner, Doreen Könning, Martin Empting, Michael Reinwarth, Christian Dietz, Bernhard Glotzbach, Holm Frauendorf, Harald Kolmar* and Olga Avrutina*

Conjugation of bioorthogonal aminoxy cube-octameric silsesquioxanes with bioactive peptides and miniproteins as well as AFM analysis of resulting constructs.

6294

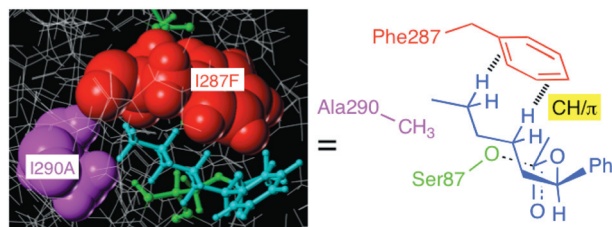


Theoretical study on the mechanism of Ag-catalyzed synthesis of 3-alkylideneoxindoles from *N*-aryl- α -diazoamides: a Lewis acid or Ag-carbene pathway?

Zhe Li* and Hai-Xiang Gao

An Ag-carbene pathway is more feasible than a Lewis acid pathway for the title reaction with N₂ dissociation as the turnover determining step.

6299



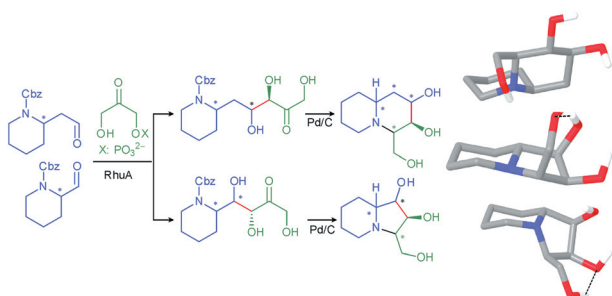
$E > 200$ (50% conv. in 2.5 h) for I287F/I290A double mutant
 [$E = 5$ (23% conv. in 41 h) for wild-type enzyme]

Redesign of enzyme for improving catalytic activity and enantioselectivity toward poor substrates: manipulation of the transition state

Tadashi Ema,* Yasuko Nakano, Daiki Yoshida, Shusuke Kamata and Takashi Sakai

Catalytic activity and enantioselectivity of lipase toward poor substrates bearing bulky substituents on both sides have been dramatically improved.

6309



Chemoenzymatic synthesis, structural study and biological activity of novel indolizidine and quinolizidine iminocyclitols

Livia Gómez, Xavier Garrabou, Jesús Joglar, Jordi Bujons, Teodor Parella, Cristina Vilaplana, Pere Joan Cardona and Pere Clapés*

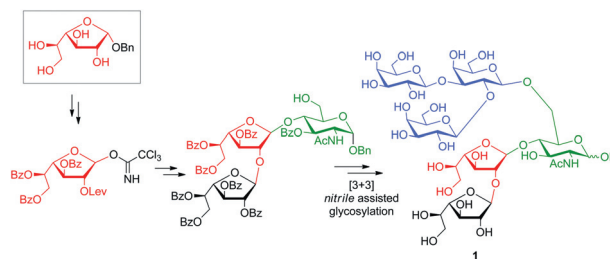
Structural analysis and inhibitory properties of stereochemical diverse indolizidine and quinolizidine iminocyclitols, prepared by a two-step chemo-enzymatic methodology, were studied.

6322

Synthesis of the *O*-linked hexasaccharide containing β -D-Galf-(1 \rightarrow 2)- β -D-Galf in *Trypanosoma cruzi* mucins

Gustavo A. Kashiwagi, Verónica M. Mendoza, Rosa M. de Lederkremer and Carola Gallo-Rodriguez*

The novel hexasaccharide **1** was synthesized by a [3 + 3]-convergent strategy employing benzyl α -D-galactofuranoside, obtained in one step from galactose, as a new internal Galf-precursor.

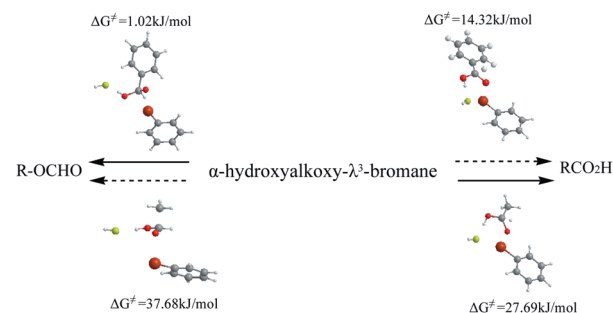


6333

Theoretical study on the hypervalent λ^3 -bromane strategy for Baeyer–Villiger oxidation of benzaldehyde and acetaldehyde: rearrangement mechanism

Hui Fu,* Shouwen Xie, Aiping Fu, Xufeng Lin, Hui Zhao and Tianxu Ye

DFT calculations described and explained well the key step of the novel BVO reaction of benzaldehyde and acetaldehyde.

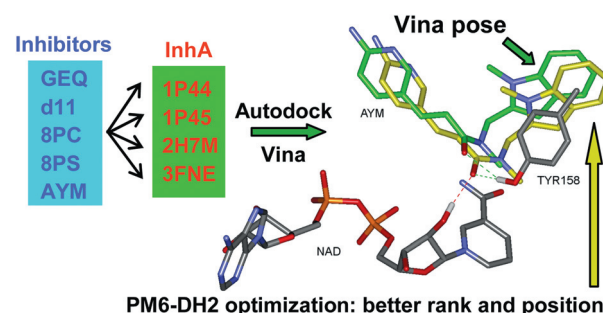


6341

Cross-docking study on InhA inhibitors: a combination of Autodock Vina and PM6-DH2 simulations to retrieve bio-active conformations

Jean-Luc Stigliani,* Vania Bernardes-Génisson,* Jean Bernadou and Geneviève Pratiel

An Autodock Vina ensemble docking of InhA inhibitors followed by a PM6-DH2 post-docking allowed to select the correct ligand poses.

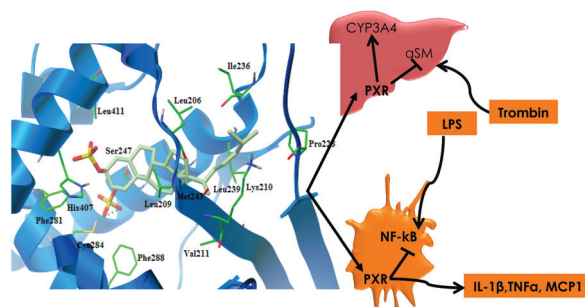


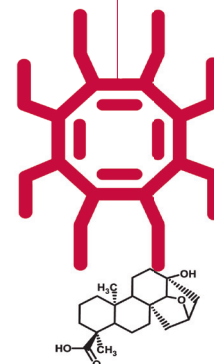
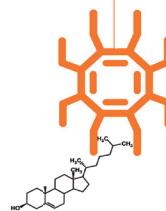
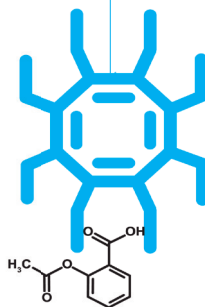
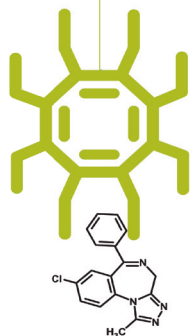
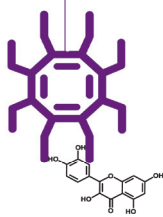
6350

Modification in the side chain of solomonsterol A: discovery of cholestan disulfate as a potent pregnane-X-receptor agonist

Valentina Sepe, Raffaella Ummarino, Maria Valeria D'Auria, Gianluigi Lauro, Giuseppe Bifulco, Claudio D'Amore, Barbara Renga, Stefano Fiorucci and Angela Zampella*

Seven synthetic analogues of the PXR (pregnane-X-receptor) marine agonist solomonsterol A were prepared by total synthesis. The study discloses cholestan disulfate as a simplified new potent PXR agonist.





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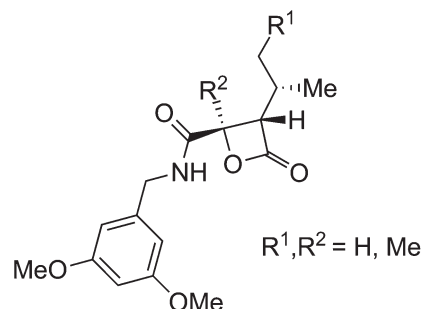
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6363

Synthesis and biological activity of simplified belactosin C analogues

Armin de Meijere,* Vadim S. Korotkov, Alexander V. Lygin, Oleg V. Larionov, Viktor V. Sokolov, Tine Graef and Mazen Es-Sayed

Successful biochemical studies of the natural products belactosin A and C and their acylated congeners have shown a β -lactonecarboxamide moiety to be a possible core structure of powerful proteasome inhibitors.

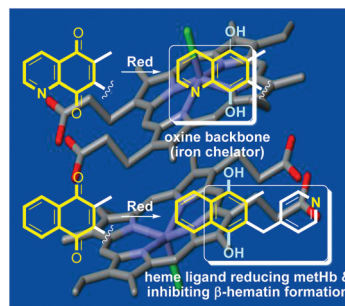


6375

Synthesis and biological evaluation of 1,4-naphthoquinones and quinoline-5,8-diones as antimalarial and schistosomicidal agents

Don Antoine Lanfranchi, Elena Cesar-Rodo, Benoît Bertrand, Hsin-Hung Huang, Latasha Day, Laure Johann, Mourad Elhabiri, Katja Becker, David L. Williams and Elisabeth Davioud-Charvet*

3-Picolinyl-menadione and quinoline-5,8-diones derivatives were evaluated for their antimalarial and antischistosomal effects.

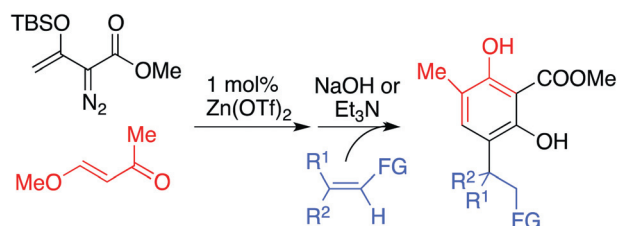


6388

Michael addition/pericyclization/rearrangement – a multicomponent strategy for the synthesis of substituted resorcinols

Yu Liu and Michael P. Doyle*

The combination of methyl 3,7-dioxo-2-diazo-4-octenoate from the zinc triflate catalyzed Mukaiyama–Michael reaction of methyl 3-*tert*-butylsilyloxy-2-diazobutenoate and 4-methoxy-3-buten-2-one with Michael acceptors in the presence of a catalytic amount of base provides convenient access to highly substituted resorcinol derivatives.

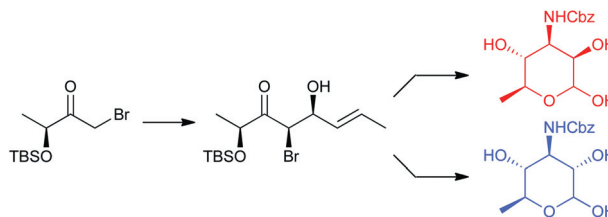


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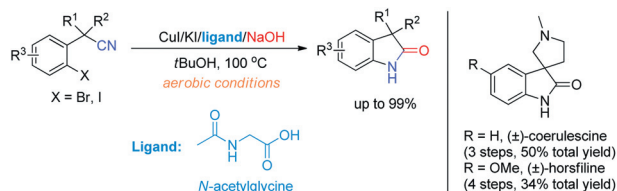
Stereoselective synthesis of protected 3-amino-3,6-dideoxyaminosugars

Joaquim Nebot, Pedro Romea* and Fèlix Urpí*

A simple titanium-mediated aldol reaction from a lactate-derived ketone enables highly stereoselective synthesis of protected dideoxyaminosugars.



6404

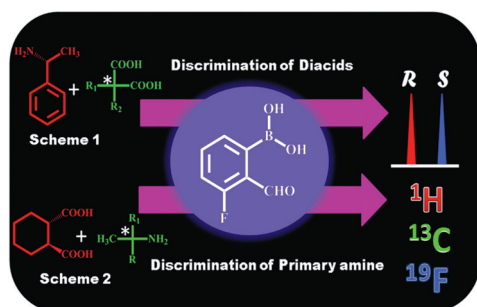


Copper-catalyzed domino coupling reaction: an efficient method to synthesize oxindoles

Jen-Chieh Hsieh,* An-Yi Cheng, Jun-Hao Fu and Ting-Wei Kang

An efficient and novel procedure for a copper catalyzed domino coupling reaction has been developed, which afforded various oxindoles in good to excellent yields with tolerance of various substituents. In addition, this method could be applied to synthesize horsfiline and coerulescine in few steps with high total yields.

6410

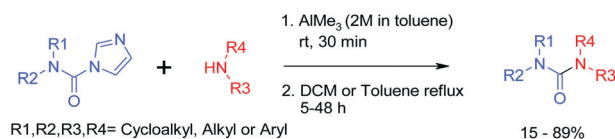


Simple and efficient methods for discrimination of chiral diacids and chiral alpha-methyl amines

Sachin R. Chaudhari and N. Suryaprakash*

The three-component chiral derivatization protocols have been developed for testing the enantiopurity of chiral diacids and primary amines by ^1H , ^{13}C and ^{19}F NMR spectroscopy.

6420

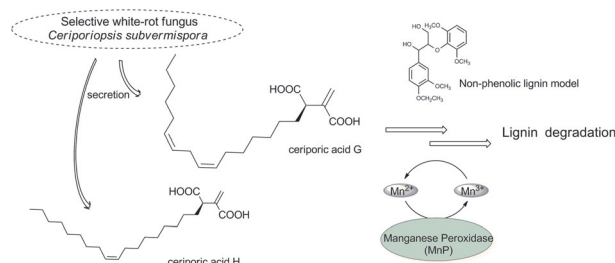


Unsymmetrical tetrasubstituted ureas from tertiary carbamoylimidazole: activation by AlMe_3

A. Velavan, S. Sumathi* and K. K. Balasubramanian*

An efficient method for the synthesis of unsymmetrical tetrasubstituted ureas from carbamoylimidazole is described. The conversion is achieved by the concurrent quarternization of the imidazole nitrogen and activation of amines with AlMe_3 .

6432



Alkadienyl and alkenyl itaconic acids (ceriporic acids G and H) from the selective white-rot fungus

Ceriporiopsis subvermispora: a new class of metabolites initiating ligninolytic lipid peroxidation

Hiroshi Nishimura, Midori Sasaki, Hirofumi Seike, Masaharu Nakamura and Takashi Watanabe*

New alkadienyl and alkenyl itaconic acids (ceriporic G and H) were isolated from wood cultures of the selective white-rot fungus *Ceriporiopsis subvermispora*. The metabolites were chemically synthesized and characterized. Ceriporic acid G degraded a recalcitrant non-phenolic lignin substructure model compound in the presence of MnP.