

Organic & Biomolecular Chemistry

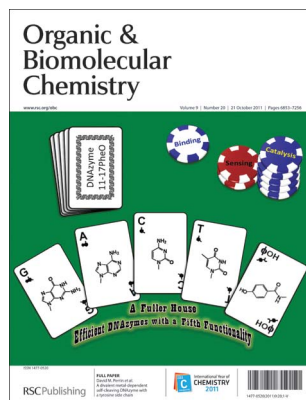
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ISSN 1477-0520 CODEN OBCRAK 9(20) 6853–7256 (2011)



Cover

See David M. Perrin *et al.*, pp. 6949–6954.

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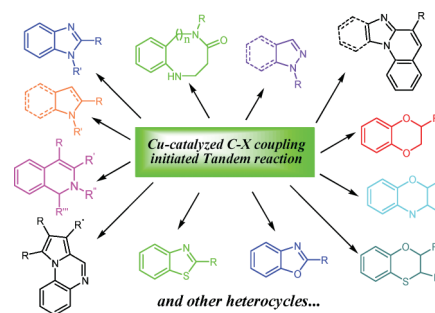
PERSPECTIVE

6873

Tandem reactions initiated by copper-catalyzed cross-coupling: A new strategy towards heterocycle synthesis

Yunyun Liu* and Jie-Ping Wan

In this review, recent research progress in heterocycle syntheses using tandem reactions initiated by copper-catalyzed coupling transformations, including C–N, C–O, C–S as well as C–C coupling processes are summarized.



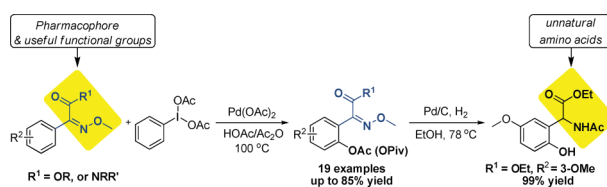
COMMUNICATIONS

6895

Palladium-catalyzed C–H acetoxylation of 2-methoxyimino-2-aryl-acetates and acetamides

Liang Wang, Xu-Dong Xia, Wei Guo, Jia-Rong Chen and Wen-Jing Xiao*

The transformation features excellent regioselectivity, wide substrate scope, and moderate to good yields. The product can be easily converted into naturally unprecedented α -amino acids in excellent yields.



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Organic & Biomolecular Chemistry (print: ISSN
1477-0520; electronic: ISSN 1477-0539) is published 24 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
Distribution Services, c/o Portland Customer Services,
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2011 Annual (print+electronic) subscription price: £3726;
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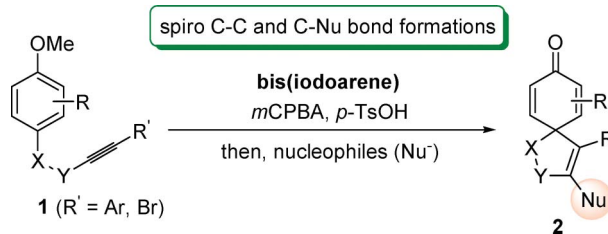
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6899

New synthesis of spirocycles by utilizing *in situ* forming hypervalent iodine species

Toshifumi Dohi, Tomofumi Nakae, Yohei Ishikado, Daishi Kato and Yasuyuki Kita*

An effective spirocyclization procedure for installing nucleophiles *via* iodonium(III) salts has been developed using the combination of bis(iodoarene) and *m*CPBA.

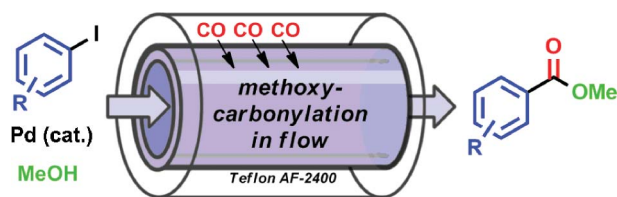


6903

Teflon AF-2400 mediated gas-liquid contact in continuous flow methoxycarbonylations and in-line FTIR measurement of CO concentration

Peter Koos, Ulrike Gross, Anastasios Polyzos, Matthew O'Brien, Ian Baxendale and Steven V. Ley*

The gas permeable membrane Teflon AF-2400 affords homogeneous CO solutions for continuous-flow methoxycarbonylations of aryl halides; in-line FTIR allows monitoring of CO solution levels.

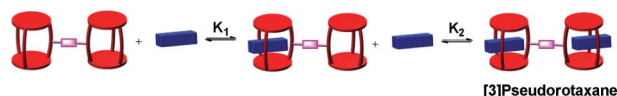


6909

Contrasting biscryptand/dimethyl paraquat [3]pseudorotaxanes: statistical *vs.* anticooperative complexation behavior

Zhenbin Niu and Harry W. Gibson*

Via the self-assembly of two bis(*meta*-phenylene)-32-crown-10-based cryptands with dimethyl paraquat, novel [3]pseudorotaxanes were formed statistically and anticooperatively, respectively.

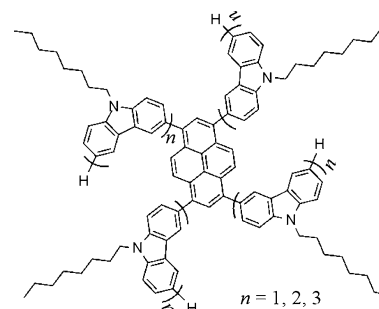


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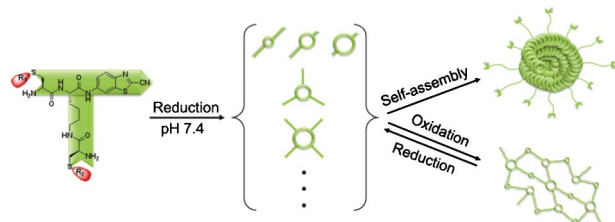
Fluorescent pyrene-centered starburst oligocarbazoles with excellent thermal and electrochemical stabilities

Ming-Guang Ren, Hui-Jun Guo, Fei Qi and Qin-Hua Song*

A series of pyrene-centered starburst oligocarbazoles have been synthesized and well characterized. As OLED materials, their superior properties will enhance device stability and lifetime.



6917

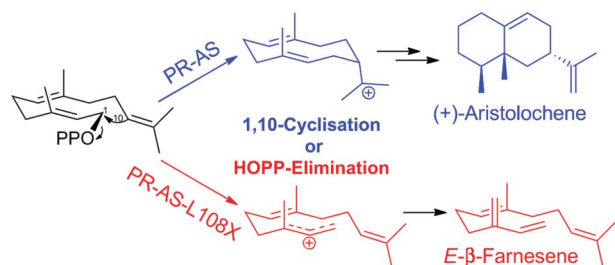


Multifunctional small molecule for controlled assembly of oligomeric nanoparticles and crosslinked polymers

Yun Deng, Shuang Liu, Kun Mei, An-ming Tang, Chun-yan Cao and Gao-lin Liang*

One multifunctional small molecule can fulfil all the transformations between monomers, oligomers, nanoparticles, and polymers when applied to a condensation reaction.

6920

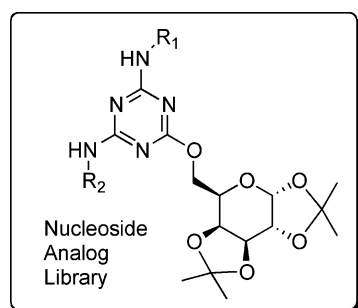


Templating effects in aristolochene synthase catalysis: elimination versus cyclisation

Juan A. Faraldos, Verónica González, Michael Senske and Rudolf K. Allemann*

Leucine108 of aristolochene synthase plays an essential structural role in maintaining the quasi-cyclic catalytic conformation of FDP.

6924

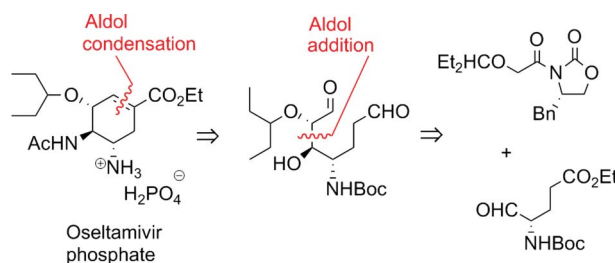


Combinatorial synthesis of galactosyl-1,3,5-triazines as novel nucleoside analogues

Shenliang Wang, Woo Sirl Lee, Hyung-Ho Ha and Young-Tae Chang*

Fast parallel synthesis of novel nucleoside analogues in high purity without extensive reaction conditions or tedious purification.

6927



An aldol approach to the enantioselective synthesis of (–)-oseltamivir phosphate

Milos Trajkovic, Zorana Ferjancic* and Radomir N. Saicic*

A formal asymmetric synthesis of the antiviral agent (–)-oseltamivir phosphate is achieved using two aldol reactions as key steps.

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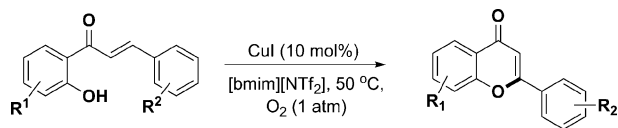
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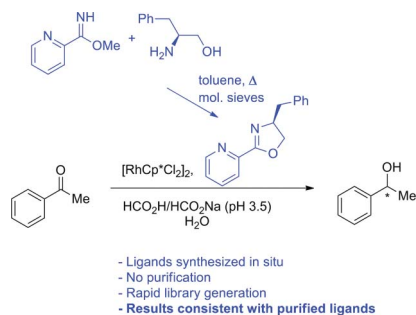


Ionic liquid mediated Cu-catalyzed cascade oxa-Michael-oxidation: efficient synthesis of flavones under mild reaction conditions

Zhiyun Du, Huifen Ng, Kun Zhang, Huaqiang Zeng and Jian Wang*

A highly efficient synthetic method of CuI-catalyzed cascade oxa-Michael-oxidation using chalcones as substrates and mediated by the ionic liquid [bmim][NTf₂] at a low temperature was disclosed.

6934

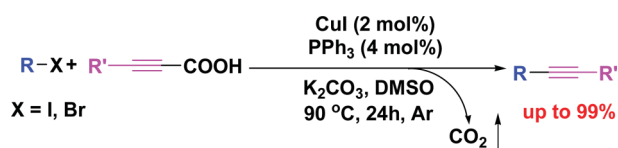


Rapid, *in situ* synthesis of bidentate ligands: chromatography-free generation of catalyst libraries

Robin Frauenlob, Martha M. McCormack, Carolyn M. Walsh and Enda Bergin*

Ligand libraries have been rapidly formed by combining aldehydes/imidates with amino compounds, and then screened directly in catalytic reactions without purification. This proved a convenient method of evaluating catalyst performance.

6938

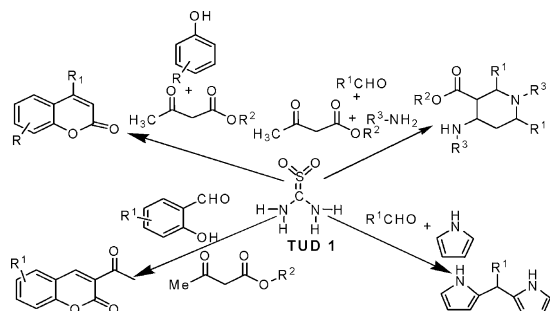


Highly effective copper-catalyzed decarboxylative coupling of aryl halides with alkynyl carboxylic acids

Xiaoming Qu, Tingyi Li, Peng Sun, Yan Zhu, Hailong Yang and Jincheng Mao*

A highly effective copper-catalyzed decarboxylative coupling of alkynylcarboxylic acids with various aryl and alkyl halides at 2 mol% loading of copper is described.

6943



Thiourea dioxide promoted efficient organocatalytic one-pot synthesis of a library of novel heterocyclic compounds

Sanny Verma, Subodh Kumar, Suman L. Jain and Bir Sain*

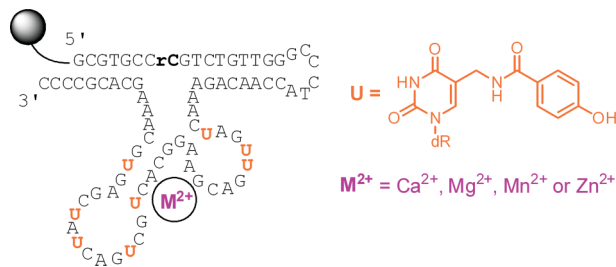
A simple, highly efficient organocatalytic approach using thiourea dioxide (TUD) as catalyst for the selective synthesis of a library of heterocyclic compounds in excellent yields is described.

6949

A divalent metal-dependent self-cleaving DNzyme with a tyrosine side chain

Curtis H. Lam, Christopher J. Hipolito, Marcel Hollenstein and David M. Perrin*

In vitro selection with a phenol-modified dUTP gave rise to the first example of a DNzyme with a tyrosine-like side chain. The catalytic sequence, Dz11-17PheO, efficiently catalyzed the self-cleavage of an embedded RNA linkage in the presence of divalent metal cations.

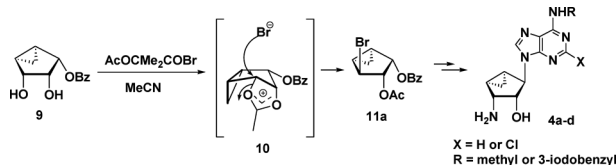


6955

Regio- and stereoselective synthesis of truncated 3'-aminocarbanucleosides and their binding affinity at the A₃ adenosine receptor

Mun Ju Choi, Girish Chandra, Hyuk Woo Lee, Xiyan Hou, Won Jun Choi, Khai Phan, Kenneth A. Jacobson and Lak Shin Jeong*

The stereoselective synthesis of truncated 3'-aminocarbanucleosides *via* a stereo- and regioselective conversion of a diol to bromoacetate and their binding affinity towards the human A₃ adenosine receptor are described.

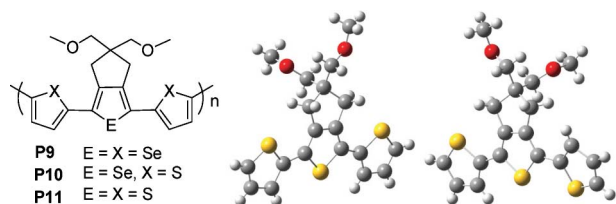


6963

Cyclopenta[*c*]selenophene based cooligomers and their polymers: comparative study with thiophene analogues

Soumyajit Das, Anjan Bedi, G. Rama Krishna, C. Malla Reddy and Sanjio S. Zade*

Selenophene and thiophene capped cyclopenta[*c*]selenophenes were synthesized and compared with thiophene analogues. Alternate polymers of cyclopenta[*c*]selenophenes (CPS)/cyclopenta[*c*]thiophene (CPT) and thiophene/selenophene possess the energy of HOMO and LUMO significantly lower than that of homopolymers of CPS and CPT, however, possess higher band gap than PCPS.

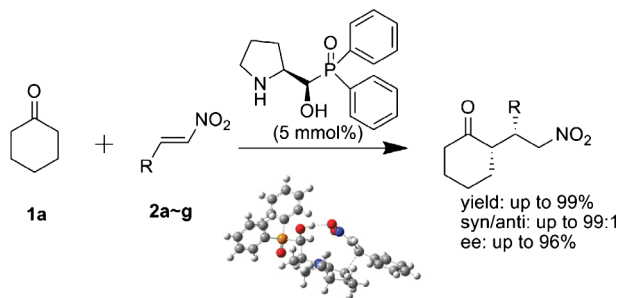


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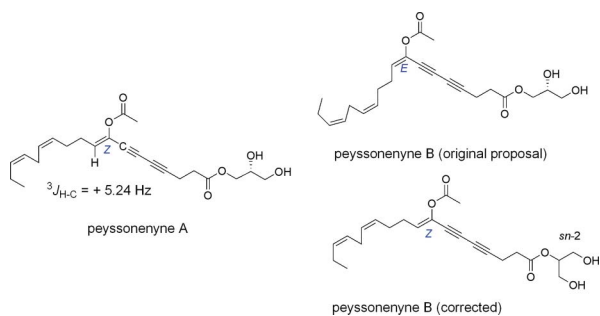
Chiral phosphoproline-catalyzed asymmetric Michael addition of ketones to nitroolefins: an experimental and theoretical study

Zhiping Zeng, Ping Luo, Yao Jiang, Yan Liu, Guo Tang, Pengxiang Xu, Yufen Zhao* and G. Michael Blackburn*

High regio- and stereo-selective Michael addition catalyzed by one novel chiral phosphoproline is studied experimentally and theoretically.



6979

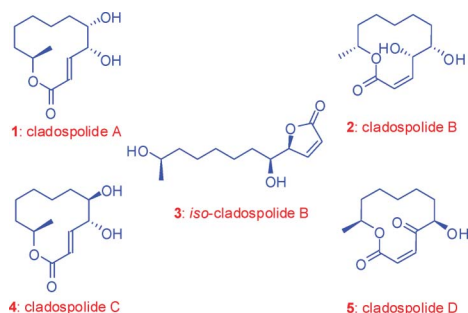


Total synthesis of the proposed structures of the DNA methyl transferase inhibitors peyssonenyynes, and structural revision of peyssonenyne B

Patricia García-Domínguez, Ilaria Lepore, Cathie Erb, Hinrich Gronemeyer, Lucia Altucci, Rosana Álvarez* and Ángel R. de Lera*

Not a geometric isomer of peyssonenyne A, but a positional isomer, was determined to be the correct structure of peyssonenyne B.

6988

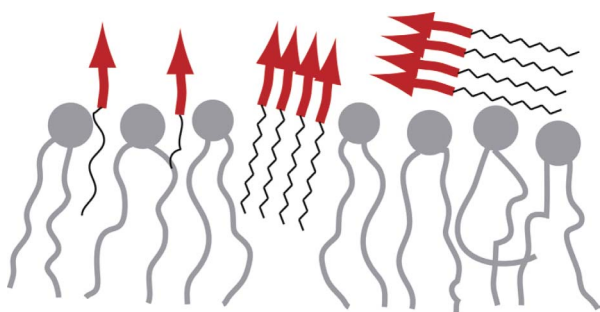


A flexible and unified strategy for syntheses of cladospolides A, B, C, and iso-cladospolide B

Debjani Si, Narayana M. Sekar and Krishna P. Kaliappan*

A general and straightforward approach towards the cladospolide family of natural products utilising Julia–Kocienski olefination and Yamaguchi lactonization is delineated.

6998

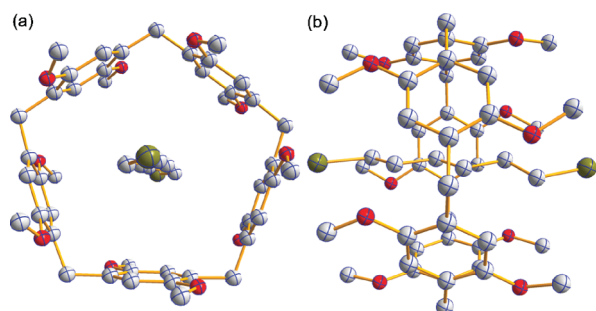


The interaction of lipid modified pseudopeptides with lipid membranes

Holger A. Scheidt, Annemarie Sickert, Thomas Meier, Nicola Castellucci, Claudia Tomasini* and Daniel Huster*

The conformational preferences of two lipopeptides $C_n^2H_{(2n+1)}CO-L-Phe-D-Oxd-OBn$ or $C_n^2H_{(2n+1)}CO-D-Phe-L-Oxd-OBn$ with $n = 5$ or 11 have been analyzed with solid-state 2H and ^{13}C NMR techniques.

7007



Efficient synthesis of copillar[5]arenes and their host–guest properties with dibromoalkanes

Luzhi Liu, Derong Cao,* Yi Jin, Hongqi Tao, Yuhui Kou and Herbert Meier*

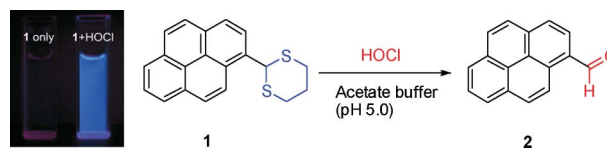
Pillar[5]arenes show strong host–guest interactions with dibromoalkanes.

7011

Signaling of hypochlorous acid by selective deprotection of dithiolane

Jiyoung Hwang, Myung Gil Choi, Jihee Bae and Suk-Kyu Chang*

The selective signaling of hypochlorous acid by dithiolane-protected pyrene-aldehyde **1** was investigated. Dithiolane probe was efficiently deprotected by hypochlorous acid to its corresponding aldehyde, which resulted in a prominent UV-vis and turn-on type fluorescence signaling.

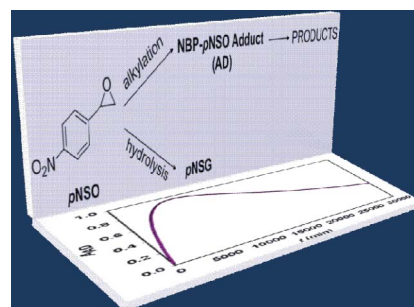


7016

Reactivity of *p*-nitrostyrene oxide as an alkylating agent. A kinetic approach to biomimetic conditions

Marina González-Pérez, Rafael Gómez-Bombarelli, M. Teresa Pérez-Prior, José A. Manso, Isaac F. Céspedes-Camacho, Emilio Calle and Julio Casado*

The alkylating capacity of *p*-nitrostyrene oxide is similar to those of very effective mutagenic agents. The instability of the adduct formed reduces its effectiveness.

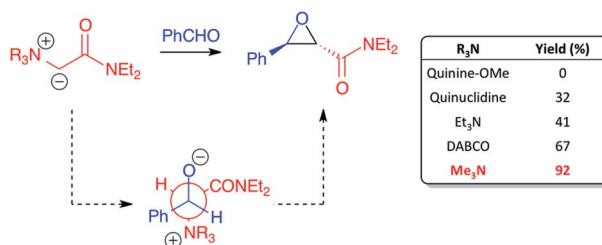


7023

Identification of the best-suited leaving group for the diastereoselective synthesis of glycidic amides from stabilised ammonium ylides and aldehydes

Richard Herchl, Martin Stiftinger and Mario Waser*

Trimethylamine was identified as the best-suited leaving group for the *trans*-selective epoxide formation using amide-stabilised ammonium ylides.

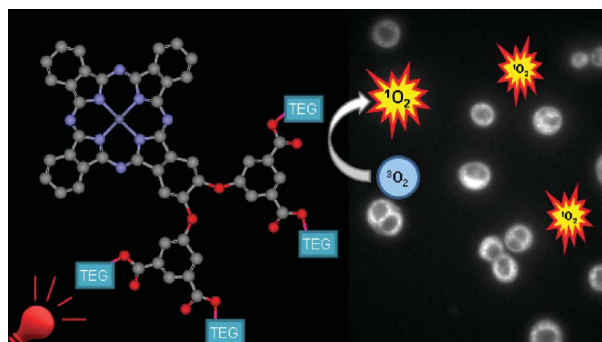


7028

Facile synthesis of pegylated zinc(II) phthalocyanines via transesterification and their *in vitro* photodynamic activities

Ming Bai, Pui-Chi Lo, Jing Ye, Chi Wu, Wing-Ping Fong and Dennis K. P. Ng*

Several pegylated "3 + 1" zinc(II) phthalocyanines have been prepared and studied for their photocytotoxicity and ability to form surfactant-free nanoparticles.



7033

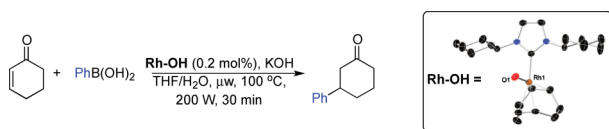


An efficient approach to pyrazolo[5,1-*a*]isoquinolin-2-amines *via* a silver(I)-catalyzed three-component reaction of 2-alkynylbenzaldehyde, sulfonohydrazide, and nitrile

Xingxin Yu, Qin Yang, Honglei Lou, Yiyuan Peng* and Jie Wu*

A three-component reaction of 2-alkynylbenzaldehyde, sulfonohydrazide, and nitrile catalyzed by silver triflate under mild conditions is reported, which generates pyrazolo[5,1-*a*]isoquinolin-2-amines in good to excellent yields.

7038

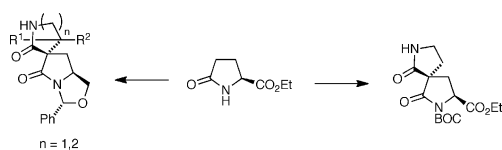


Well-defined [Rh(NHC)(OH)] complexes enabling the conjugate addition of arylboronic acids to α,β -unsaturated ketones

Byron J. Truscott, George C. Fortman, Alexandra M. Z. Slawin and Steven P. Nolan*

The synthesis and catalytic activity of three well-defined monomeric rhodium(I) hydroxide complexes bearing *N*-heterocyclic carbene (NHC) ligands are reported.

7042

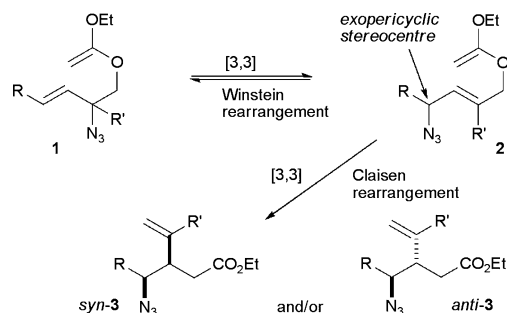


Spirocyclic systems derived from pyroglutamic acid

Andrew R. Cowley, Thomas J. Hill, Petr Kocis, Mark G. Moloney,* Robert D. Stevenson and Amber L. Thompson

The synthesis and likely conformational structure of rigid spirocyclic bislactams and lactam-lactones derived from pyroglutamic acid, and their suitability as lead structures for applications in drug development programmes using cheminformatic analysis, has been investigated.

7057



Claisen rearrangements of equilibrating allylic azides

Donald Craig,* John W. Harvey, Alexander G. O'Brien and Andrew J. P. White

Equilibrating mixtures of allylic azide-containing allylic alcohols or allylic 2-tolylsulfonylacetic esters undergo Johnson–Claisen or Ireland–Claisen rearrangement reactions to give unsaturated γ -azidoesters and -acids, respectively.

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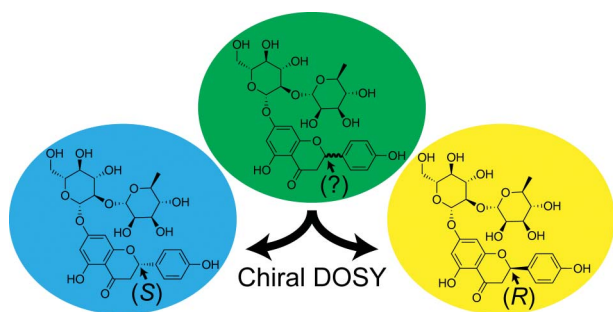
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7062

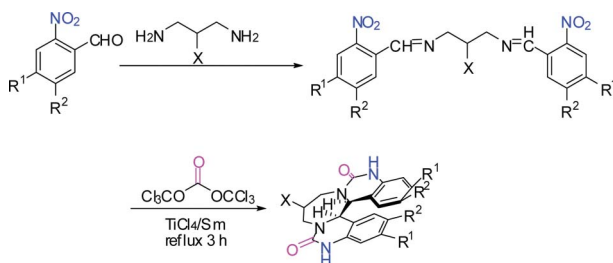


Resolving natural product epimer spectra by matrix-assisted DOSY

Ralph W. Adams, Juan A. Aguilar, Julia Cassani, Gareth A. Morris and Mathias Nilsson*

Chiral matrix-assisted DOSY allows the separation of signals from different epimers based on differential interaction with β -cyclodextrin.

7065

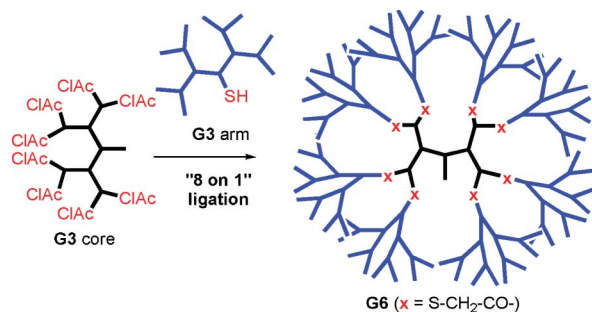


One pot synthesis of *cis*-bispyrimidodiazepinone derivatives via low-valent titanium reagent (TiCl_4/Sm)

Guolan Dou and Daqing Shi*

An efficient and convenient method for the preparation of *cis*-bispyrimidodiazepinone derivatives has been described.

7071

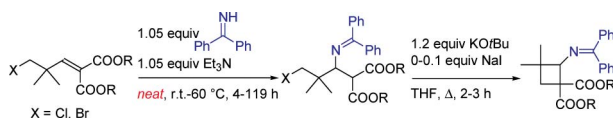


Peptide dendrimer enzyme models for ester hydrolysis and aldolization prepared by convergent thioether ligation

Nicolas A. Uhlich, Tamis Darbre and Jean-Louis Reymond*

Peptide dendrimers bearing four or eight chloroacetyl groups at their N-termini underwent multiple thioether ligation with G2 and G3 peptide dendrimers with a cysteine residue at their focal point, to give G4, G5 and G6 dendrimers containing up to 341 amino acids.

7085



Synthesis of novel β -aminocyclobutanecarboxylic acid derivatives by a solvent-free aza-Michael addition and subsequent ring closure

Tamara Meiresonne, Sven Mangelinckx and Norbert De Kimpe*

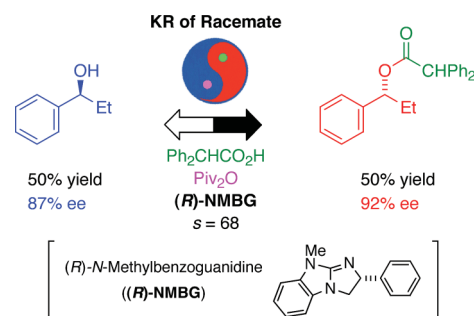
A solvent-free aza-Michael addition leading to new substituted β -aminocyclobutanecarboxylic acid derivatives and the reactivity of these donor-acceptor substituted cyclobutanes is described.

7092

(R)-(+)-N-Methylbenzguanidine ((R)-NMBG) catalyzed kinetic resolution of racemic secondary benzylic alcohols with free carboxylic acids by asymmetric esterification

Kenya Nakata and Isamu Shiina*

New guanidine-type catalyst ((R)-NMBG) promotes the asymmetric esterification of racemic secondary benzylic alcohols with carboxylic acids to provide chiral esters and alcohols with high enantioselectivities.

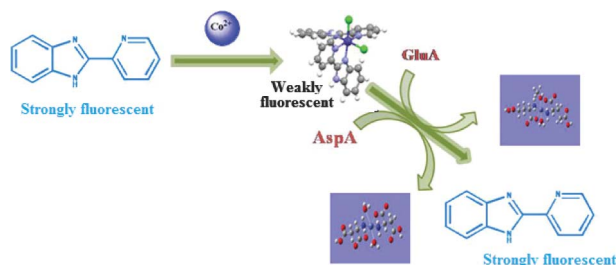


7097

2-(2-Pyridyl) benzimidazole based Co(II) complex as an efficient fluorescent probe for trace level determination of aspartic and glutamic acid in aqueous solution: A displacement approach

Sudipta Das, Subarna Guha, Arnab Banerjee, Sisir Lohar, Animesh Sahana and Debasis Das*

X-Ray structurally characterized [Co(II)-2-(2-Pyridyl) benzimidazole] complex used as a turn-on fluorescent probe for trace level determination of aspartic and glutamic acid *via* displacement approach in aqueous solution.

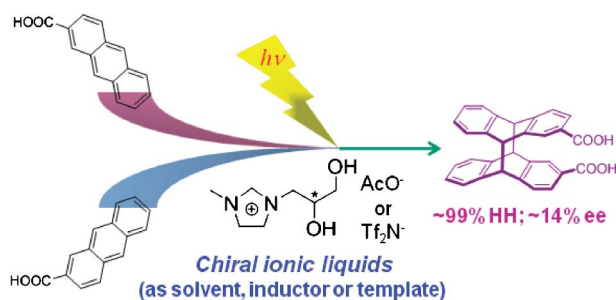


7105

Chiral ionic liquid-mediated photochirogenesis. Enantiodifferentiating photocyclodimerization of 2-anthracenecarboxylic acid

Gaku Fukuhara,* Takahiro Okazaki, Marco Lessi, Masaki Nishijima, Cheng Yang, Tadashi Mori, Andrea Mele, Fabio Bellina, Cinzia Chiappe* and Yoshihisa Inoue*

Chiral ionic liquid-mediated photocyclodimerization of 2-anthracenecarboxylic acid gave *head-to-head* (HH) cyclodimers in 99% yield and *anti*-HH in 14% enantiomeric excess.

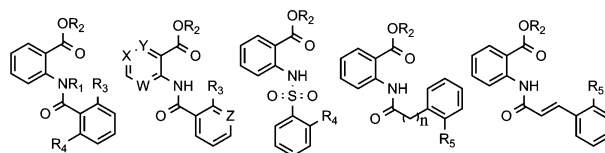


7113

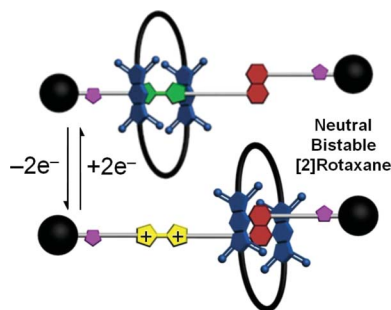
Anthranilic acid-based inhibitors of phosphodiesterase: Design, synthesis, and bioactive evaluation

Yih-Dih Cheng, Tsong-Long Hwang, Han-Hsiang Wang, Tai-Long Pan, Chin-Chung Wu, Wen-Yi Chang, Yi-Ting Liu, Tzu-Chi Chu and Pei-Wen Hsieh*

Forty-six anthranilic acid derivatives were synthesized and their anti-inflammatory effects and underlying mechanisms were investigated in human neutrophils.



7126

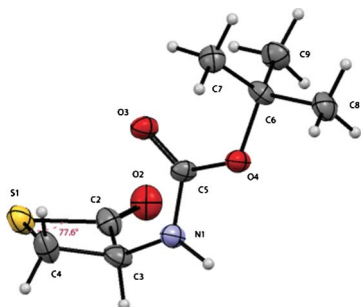


A neutral redox-switchable [2]rotaxane

J.-C. Olsen, A. C. Fahrenbach, A. Trabolsi, D. C. Friedman, S. K. Dey, C. M. Gothard, A. K. Shveyd, T. B. Gasa, J. M. Spruell, M. A. Olson, C. Wang, H.-P. Jacquot de Rouville, Y. Y. Botros and J. F. Stoddart*

A neutral [2]rotaxane with redox addressability: A bistable neutral [2]rotaxane composed of a dumbbell component containing electron-rich tetrathiafulvalene and 1,5-dioxynaphthalene units along with a ring incorporating electron-deficient pyromellitic diimide units is shown to switch upon oxidation of tetrathiafulvalene.

7134

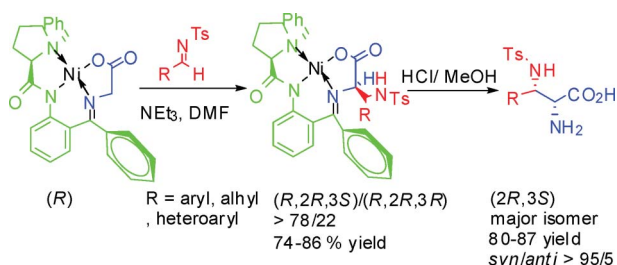


Exploring the potential of the β -thiolactones in bioorganic chemistry

Sylvain Aubry, Kaname Sasaki, Laure Eloy, Geneviève Aubert, Pascal Retailleau, Thierry Cresteil and David Crich*

Peptidyl C-terminal β -thiolactones are readily prepared, are more stable than β -lactones, and have potential applications in bioorganic and medicinal chemistry

7144

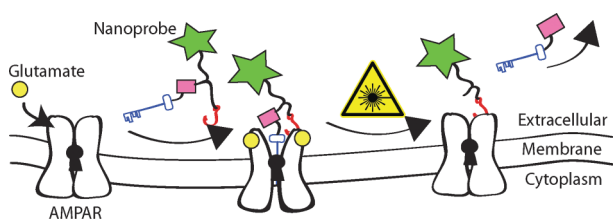


Asymmetric synthesis of α,β -diamino acid derivatives via Mannich-type reactions of a chiral Ni(II) complex of glycine with *N*-tosyl imines

Guowei Song, Meihong Jin, Zhenjiang Li* and Pingkai Ouyang

Asymmetric Mannich-type reactions of (*R*)-Ni(II) complex of glycine with *N*-tosyl imines were investigated to offer chiral α,β -diamino acids.

7151



Silent, fluorescent labeling of native neuronal receptors

Devaiah Vytla, Rosamund E. Combs-Bachmann, Amanda M. Hussey, Ismail Hafez and James J. Chambers*

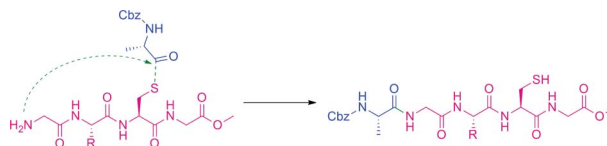
A photo-labile, minimally-perturbing strategy to label and visualize endogenous receptors on live, wild-type neurons has been developed.

7162

Microwave-assisted chemical ligation of *S*-acyl peptides containing non-terminal cysteine residues

Finn K. Hansen, Khanh Ha, Ekaterina Todadze, Aaron Lillicotch, Alexander Frey and Alan R. Katritzky*

An efficient approach for the synthesis of a series of *S*-acyl peptides containing internal cysteine residues has been developed and the chemical long-range ligation of these *S*-acyl peptides *via* 5-, 8-, 11- and 14-membered cyclic transition states has been investigated.

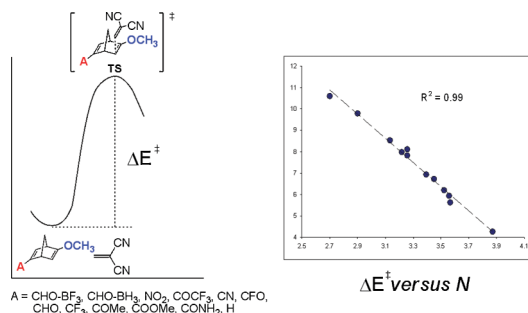


7168

The nucleophilicity *N* index in organic chemistry

Luis R. Domingo* and Patricia Pérez*

The nucleophilicity *N* index is a measure of the nucleophilicity of single and complex organic molecules displaying concurrently electrophilic and nucleophilic behaviors.

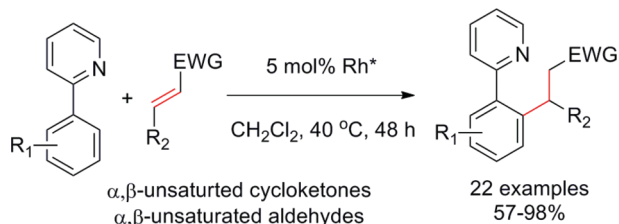


7176

Rhodium-catalyzed C–H activation and conjugate addition under mild conditions

Luo Yang, Camille A. Correia and Chao-Jun Li*

An efficient rhodium^{III}-catalyzed C–H activation and subsequent conjugate addition was achieved under mild conditions. The reaction utilized inert arenes to replace stoichiometric organometallic reagents and can tolerate various functional groups as well as air and water.

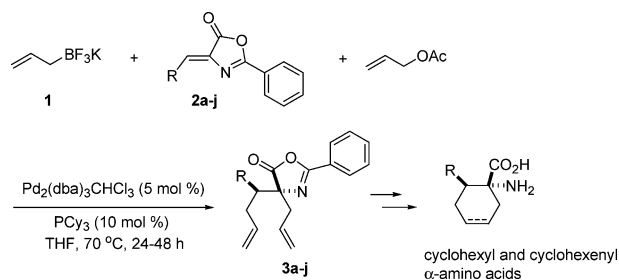


7180

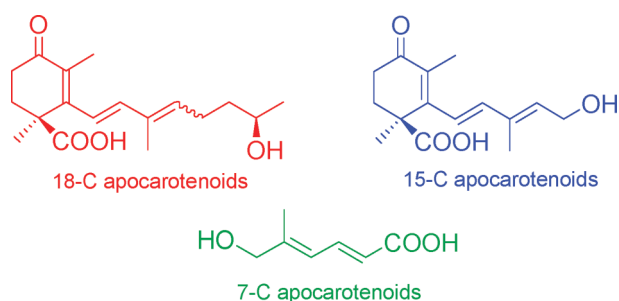
Amphiphilic allylation of arylidene-1,3-oxazol-5(4*H*)-one using bis- π -allylpalladium complexes: an approach to synthesis of cyclohexyl and cyclohexenyl α -amino acids

Afaf R. Genady and Hiroyuki Nakamura*

An efficient method for synthesis of cyclohexyl and cyclohexenyl α -amino acids has been developed *via* palladium-catalyzed three-component assemblies followed by ring-closing metathesis (RCM).



7190

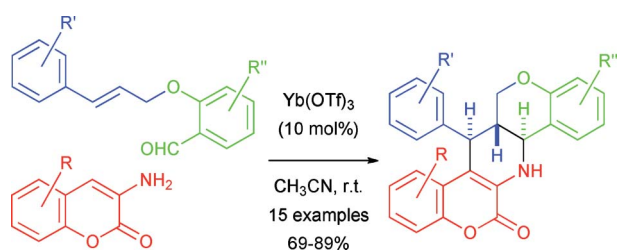


New apocarotenoids and β -carotene cleavage in *Blakeslea trispora*

Alejandro F. Barrero,* M. Mar Herrador, Pilar Arteaga, Jesús Gil, Jose-Antonio González, Eugenio Alcalde and Enrique Cerdá-Olmedo*

Three families of apocarotenoids in *Blakeslea trispora* are originated from the double oxidative cleavage of β -carotene during sexual interaction.

7196

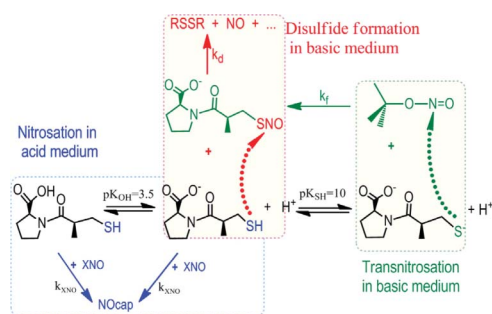


Intramolecular Povarov reactions involving 3-aminocoumarins

Amit A. Kudale, David O. Miller, Louise N. Dawe and Graham J. Bodwell*

3-Aminocoumarins react with benzaldehydes bearing a pendant electron rich dienophile in an intramolecular Povarov reaction to afford pentacyclic heterocycles in good yield and high diastereoselectivity.

7207

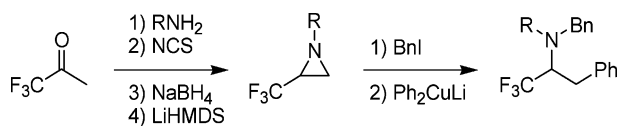


S-Nitrosocaptopril formation in aqueous acid and basic medium. A vasodilator and angiotensin converting enzyme inhibitor

Alexia Sexto and Emilia Iglesias*

S-Nitrosocaptopril forms in acid solutions of nitrite, and in basic solutions of *t*-butylnitrite; its decomposition mechanism explains NOcap vasodilator and ACE inhibitor actions.

7217



Straightforward synthesis of 1-alkyl-2-(trifluoromethyl)aziridines starting from 1,1,1-trifluoroacetone

Sara Kenis, Matthias D'hooghe,* Guido Verniest, Vinh Duc Nguyen, Tuyet Anh Dang Thi, Tuyen Van Nguyen and Norbert De Kimpe*

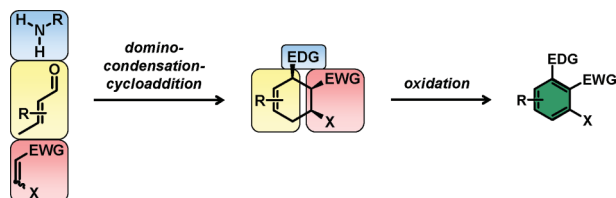
1-Alkyl-2-(trifluoromethyl)aziridines were prepared starting from 1,1,1-trifluoroacetone *via* imination, α -chlorination, hydride reduction and ring closure, followed by regioselective aziridine ring opening to provide primary β -iodo amines.

7224

Practical three-component synthesis of crowded arenes with donor–acceptor substitution

Robert Fichtler, Jörg-M. Neudörfl and Axel Jacobi von Wangelin*

A sequential combination of three-component condensation–cycloaddition reactions and oxidations allows for the facile synthesis of various substituted aniline and anthranilic acid derivatives under practical conditions.

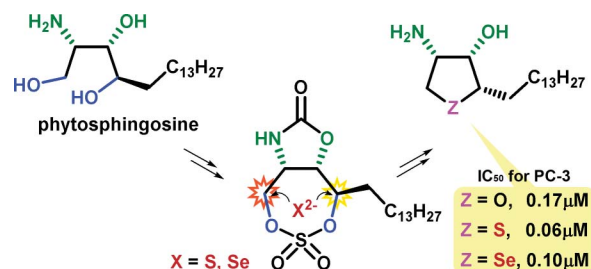


7237

Syntheses of sulfur and selenium analogues of pachastrissamine *via* double displacements of cyclic sulfate

Hongjun Jeon, Hoon Bae, Dong Jae Baek, Young-Shin Kwak, Deukjoon Kim and Sanghee Kim*

Sulfur and selenium containing bioisosteric analogues of pachastrissamine were prepared from a 7-membered cyclic sulfate intermediate by sequential intermolecular and intramolecular S_N2 displacement reactions.

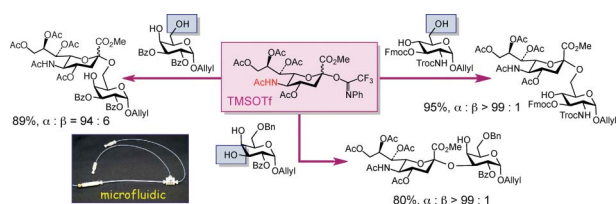


7243

Reinvestigation of the C5-acetamide sialic acid donor for α -selective sialylation: practical procedure under microfluidic conditions

Yosuke Uchinashi, Masahiro Nagasaki, Jiazhou Zhou, Katsunori Tanaka* and Koichi Fukase*

The readily available C5-acetamide donor was investigated for its use in α -sialylation under microfluidic conditions.

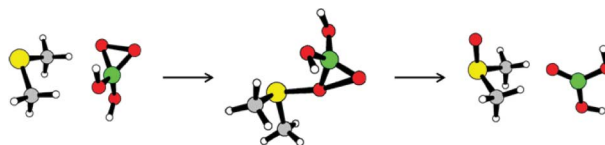


7249

Dioxaborirane: a highly reactive peroxide that is the likely intermediate in borate catalysed electrophilic reactions of hydrogen peroxide in alkaline aqueous solution

Marcus C. Durrant, D. Martin Davies and Michael E. Deary*

Reaction of dioxaborirane with dimethyl sulfide.



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