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(YOUJI HUAXUE)

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* 通讯联系人.

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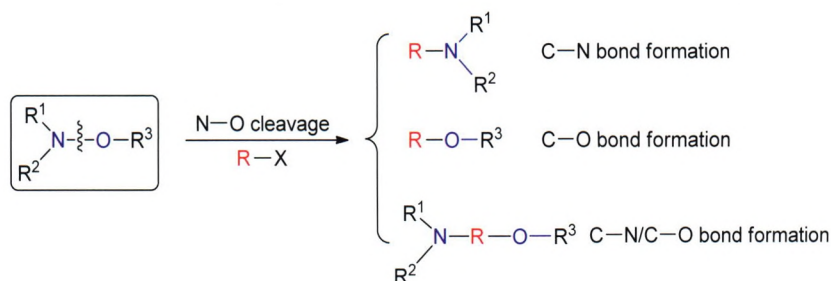
Vol. 39 No. 11 November 2019

On the Cover

The recent advances in copper-catalyzed N—O bond cleavage strategy to introduce N- or O-functional groups into the target molecules are reviewed by Lei, Li, and Mo on page 2989. Copper catalysts have been not only widely used to construct C—N and C—O bonds but also successfully applied in the total synthesis of natural products and pharmaceuticals through N—O bond cleavage.

REVIEWS

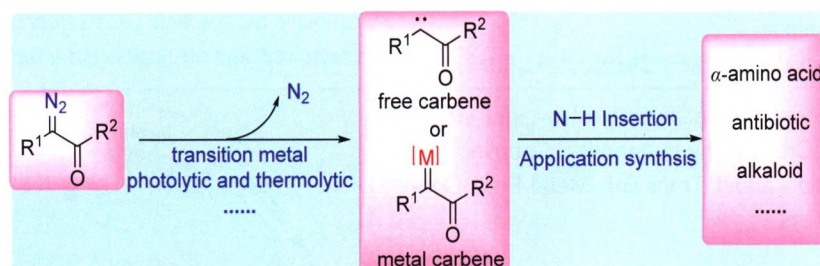
Recent Advances in Copper-Catalyzed N—O Cleavage Strategy



Lei, Lu; Li, Chengjing; Mo, Dongliang*
Chin. J. Org. Chem. **2019**, 39(11), 2989

The new development of copper-catalyzed N—O bond cleavage to construct C—N, C—O and C—N/C—O bonds and its application in the total synthesis of natural products and pharmaceuticals in recent years are reviewed.

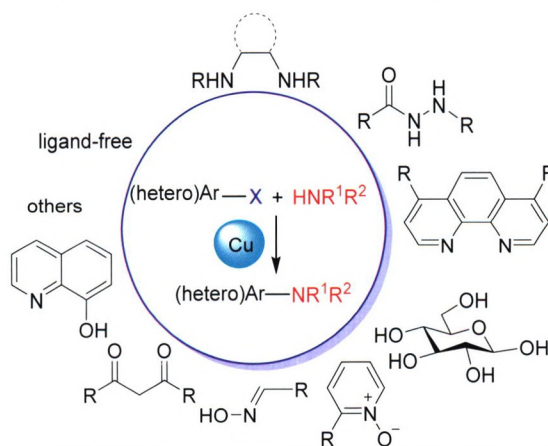
Progress in N—H Insertion Reaction of α -Diazocarbonyl Compounds



Feng, Jiajun; Yi, Xiangyan; Fu, Yaofeng; Yu, Yang; Huang, Fei*
Chin. J. Org. Chem. **2019**, 39(11), 3013

The research progress in the insertion reaction of α -diazocarbonyl compounds into N—H bonds under transition metal, organic small molecules, biomacromolecule or photolytic and thermolytic conditions, including the reaction mechanism and synthesis applications, is summarized. Finally, the prospects of this reaction are also discussed.

Research Progress in Ligand-Assisted Copper-Catalyzed C—N Cross-Coupling Reaction in Aqueous Media or Pure Water



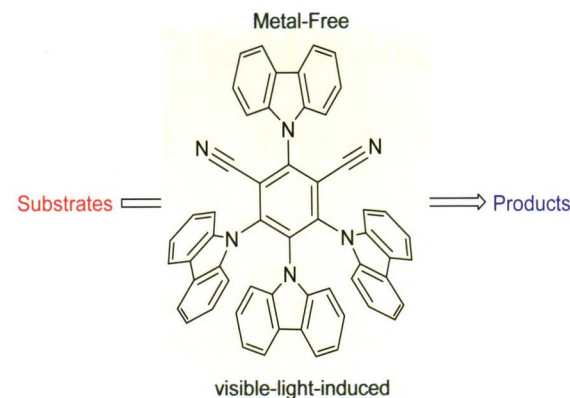
Xie, Jianwei*; Wang, Xiaochuang; Wu, Fengtian; Zhang, Jie
Chin. J. Org. Chem. **2019**, 39(11), 3026

According to the structure of ligands, the progress of ligand-assisted copper-catalyzed C—N cross-coupling reaction in aqueous media or pure water is summarized. In addition, ligand-free copper-catalyzed C—N coupling reactions in aqueous media or pure water are also reviewed.

CONTENT

Application of Photosensitizer 2,4,5,6-Tetrakis(carbazol-9-yl)-1,3-dicyanobenzene in Photo-induced Transition-Metal-Free Organic Synthesis

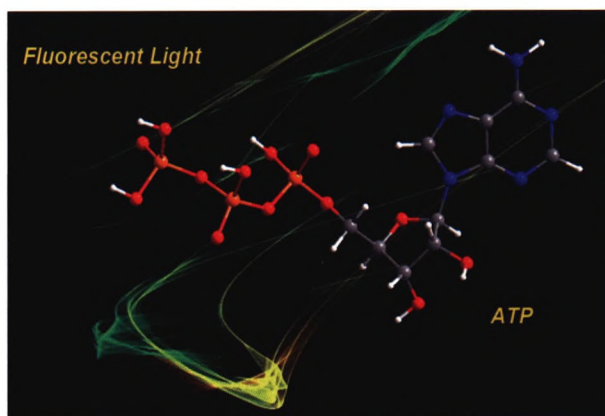
Chen, Jinyang*; Li, Yuhang; Mei, Lan; Wu, Hongyu
Chin. J. Org. Chem. **2019**, 39(11), 3040



The recent progress on the transition-metal-free photosynthesis under visible-light catalyzed by 2,4,5,6-tetrakis(carbazol-9-yl)-1,3-dicyanobenzene (4CzIPN) is reviewed, and the application of 4CzIPN for inducing radicals from different precursors (including silicon reagent, carboxylic acid and its derivatives, sulfur-containing reagent and fluorine reagent) is included.

Recent Progress in Fluorescent Probes for Adenosine Triphosphate Based on Small Organic Molecules

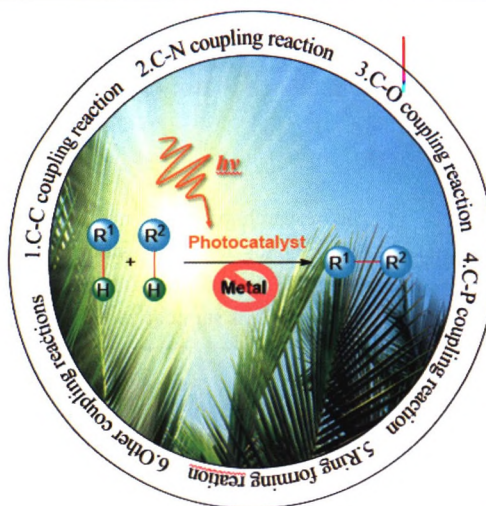
Zhang, Jidong*; Zhang, Jun; Yan, Zhan; Xie, Juanping
Chin. J. Org. Chem. **2019**, 39(11), 3051



Based on organic small molecule fluorescence sensors, the recent progress in research of adenosine triphosphate (ATP) fluorescence sensors in molecular design and application is reviewed, and the prospects for their development are discussed.

Recent Advances in Visible-Light-Induced Cross Dehydrogenation Coupling Reaction under Transition Metal-Free Conditions

Kong, Yaolei; Xu, Wenxiu; Ye, Feixia; Weng, Jianquan*
Chin. J. Org. Chem. **2019**, 39(11), 3065

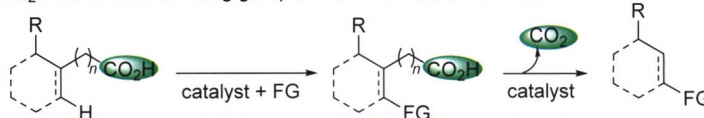


Visible-light-induced cross dehydrogenation coupling reaction under transition metal-free conditions was widely concerned due to the advantages of cleanliness, safety as well as high step and atom economy. Classified by the type of bonding, the applications of these reactions in organic synthesis are reviewed, and their future outlook is also discussed.

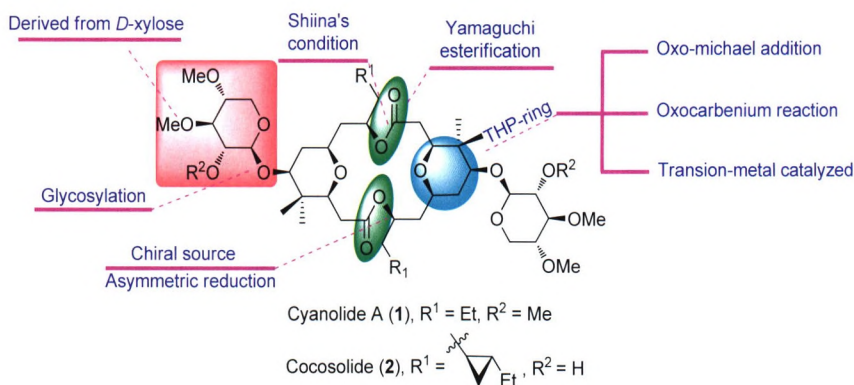
Progress in Transition Metal Catalyzed C—H Functionalization Directed by Carboxyl Group

Luo, Feihua*
Chin. J. Org. Chem. **2019**, 39(11), 3084

CO₂H as decious directing group in C-H bond functionalization

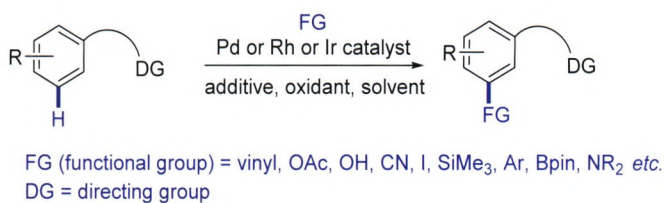


The recent progress in transition metal catalyzed C—H functionalization directed by carboxyl group according to different coupling modes is summarized, and the representative reaction mechanism is briefly described. Existing problems with a brief outlook in this field are also presented.

Synthetic Studies toward Natural Occur-
red Cyanolide A and Cocosolide

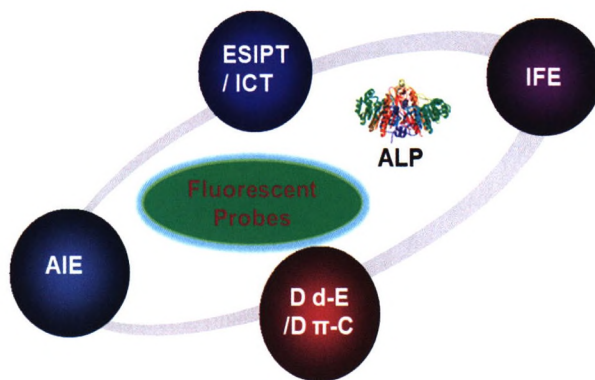
Zhang, Liu; Zhang, Mengfan; Qi, Chenze;
Yang, Zhen*
Chin. J. Org. Chem. **2019**, 39(11), 3105

Cyanolide A (1) and cocosolide (2) were isolated from Papua New Guinea and Guam, respectively. The synthesis of cynaolide A (1) and cocosolide (2) is reviewed based on the construction methods of tetrahydropyran ring, which involves oxo-Michael addition, oxo-carbenium cyclization and transition-metal catalyzed cyclization reactions.

Advances on Directing-Group Assisted
meta-C—H Functionalization Catalyzed
by Transition Metal

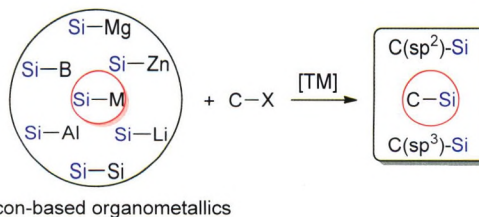
Wu, Mei*; Huang, Xinping; Zhang, Haibing;
Li, Pengfei
Chin. J. Org. Chem. **2019**, 39(11), 3114

The *meta*-C—H functionalization assisted by directing group such as template directed, secondary interaction and transient mediator is summarized in detail. The existing problems and limitations of this field are also included. Finally, the development trend of this area is prospected.

Research Progress in the Fluorescent
Probes for Alkaline Phosphatase

Zhang, Jidong*; Liu, Hongze; Meng, Li
Chin. J. Org. Chem. **2019**, 39(11), 3132

The fluorescent probes for alkaline phosphatase with different luminous mechanism are summarized, and the development tendency of the sensing ensembles is prospected.

Cross-Coupling of C—Si Bond by Using
Silyl Reagents

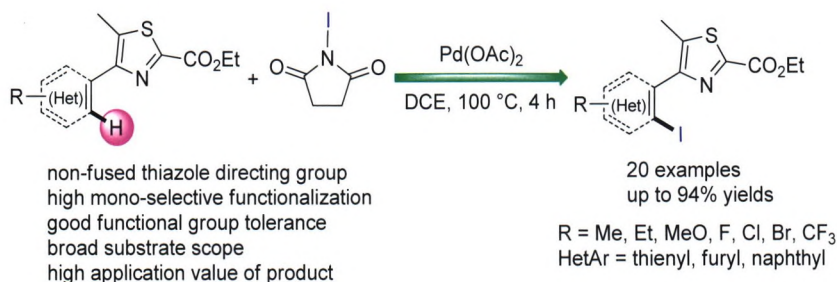
Wang, Mingfeng; Yu, Maodong; Wang, Wen-
shu; Lin, Weili; Luo, Feixian*
Chin. J. Org. Chem. **2019**, 39(11), 3145

The recent progress on the cross-coupling of C—Si bond by using silyl reagents is summarized. The application of silyl reagents in cross-coupling for C—Si bond formation including silyl boranes, organosilyl magnesium, organosilyl zinc, unassymetric disilanes, organosilyl aluminum and organosilyl lithium reagents is mainly discussed.

CONTENT

ARTICLES

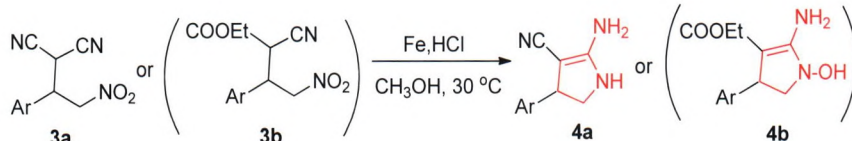
Palladium-Catalyzed Thiazole-Directed *mono*-Selective C(sp²)—H Bond Iodination Reaction



Xing, Lihao; Shao, Lingyan*; Fu, Xiaopan;
Deng, Kezuan; Yang, Jinyue; Ji, Yafei*
Chin. J. Org. Chem. **2019**, 39(11), 3154

A palladium-catalyzed *ortho*-C(sp²)—H bond iodination of 4-arylthiazoles has been developed. Through screening of directing groups and optimization of reaction parameters, the most efficient reaction conditions for mono-*ortho*-position iodination were obtained, which were applied to synthesize a series of 4-(2-iodoaryl)thiazoles with broad scope of 4-arylthiazole substrates. At last, plausible Pd^{II}/Pd^{IV} mechanism was proposed.

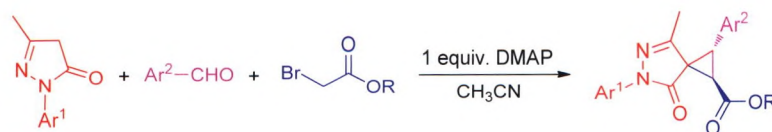
Synthesis of 4,5-Dihydropyrrole from γ -Nitro-nitrile



Wen, Ting; Kang, Meng; Chen, Zhanguo*
Chin. J. Org. Chem. **2019**, 39(11), 3162

A new method for the synthesis of 4,5-dihydropyrrole derivatives from γ -nitrocarbo-nitrile is established. Using the mixed system of reduced iron powder and concentrated hydrochloric acid as reducing agent and methanol as solvent, the nitro group of γ -nitronitrile was reduced to amino group at 30 °C. And then the amino-groups reacted with cyano groups to form target compounds via intramolecular nucleophilic addition reaction and rearrangement reaction.

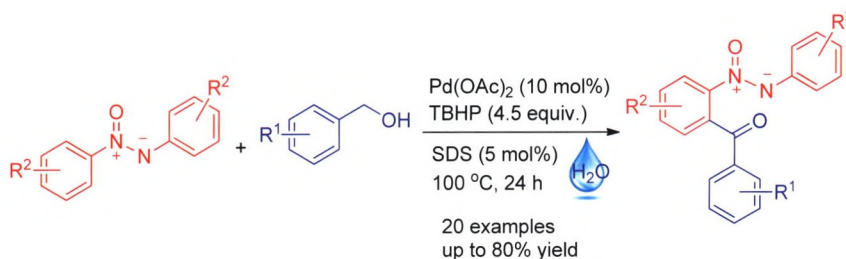
High-Selective One-Pot Synthesis of Spirocyclopropane Pyrazolones Promoted by 4-Dimethylaminopyridine



Liang, Jie; Ma, Huifang; Ablajan, Keyume*
Chin. J. Org. Chem. **2019**, 39(11), 3169

An efficient 4-dimethylaminopyridine-promoted highly stereoselective synthesis of multi-substituted spirocyclopropane pyrazolones was developed via one-pot cascade reaction. A variety of spirocyclopropane compounds were obtained in high yield with excellent diastereoselectivities.

Palladium-Catalyzed Regioselective *ortho*-Acylation of Azoxybenzenes under Aqueous Conditions



Chen, Xiaopei*; Ma, Zhiwei; Wang, Chuan-
chuan; Liu, Juntao; Wu, Jinsong
Chin. J. Org. Chem. **2019**, 39(11), 3176

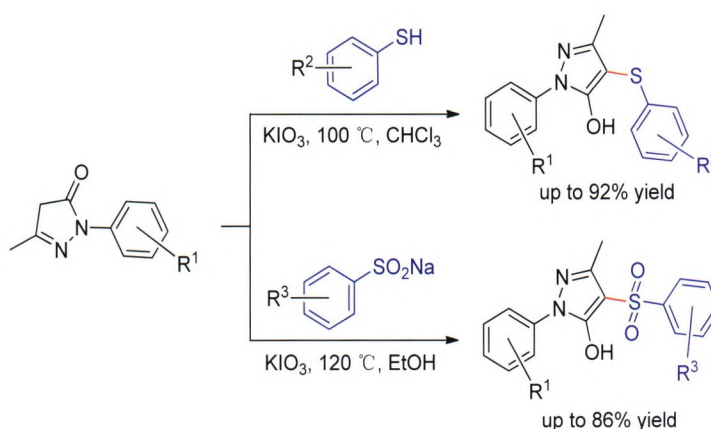
A facile and efficient protocol for palladium-catalyzed *ortho*-acylation of azoxybenzenes has been developed under aqueous conditions. In this process, the alcohols were oxidized into the corresponding aldehydes *in situ*, which coupled with azoxybenzenes with excellent regioselectivity, affording the acylated azoxybenzenes in moderate to good yields. A variety of functional groups were tolerated in this procedure.

Synthesis of 3-Ethanoyl/Aroylacetylindoles in Water



4-Dodecylbenzenesulfonic acid (DBSA) catalyzed hydrolysis reaction of β -ethylthio- β -indolyl- α,β -unsaturated ketones (**1**) in water to yield 3-ethanoyl/aroylacetylindoles (**2**) was studied. It showed that the hydrolysis of **1** in water smoothly occurred in the presence of 10 mol% DBSA in reflux, and **2** was obtained in excellent yield.

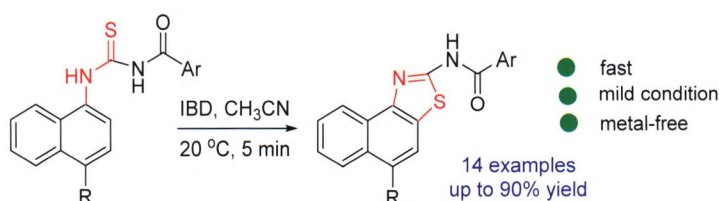
Hu, Xiaoyu; Yu, Haifeng*; Wang, Wenju*; Jiang, Siao; Liu, Qi; He, Jie
Chin. J. Org. Chem. **2019**, 39(11), 3183

Direct Synthesis of Sulfonated or Sulfonylated Pyrazolones Mediated by KIO_3 and Their Anti-microbial Activity

A facile and efficient method for the synthesis of sulfonated or sulfonylated pyrazolones catalyzed by KIO_3 was established. A variety of desired products were obtained in moderate to high yields. This methodology could be conducted under mild reaction conditions without requiring any metal or oxidant. Control experiments showed that the mechanism of this reaction was different from previous KIO_3 -catalyzed reactions. Some of these desired products showed high inhibitory activity against *V. mali* and *B. cinerea*.

Dong, Daoqing; Chen, Wenjing; Chen, Demao; Li, Lixia; Li, Guanghui; Wang, Zuli*; Deng, Qi; Long, Shu
Chin. J. Org. Chem. **2019**, 39(11), 3190

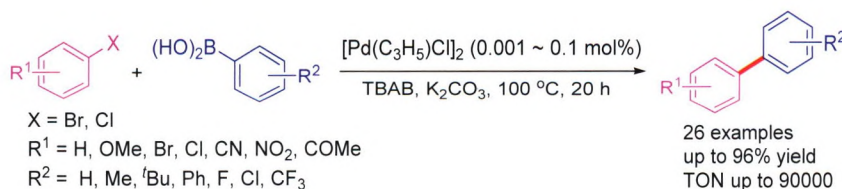
Metal-Free Rapid Synthesis of 2-Aroyl-amino Naphtho[1,2-d]thiazoles



A novel and practical method to synthesize naphtho[1,2-d]thiazole derivatives under metal-free conditions has been developed. This protocol provides a quick, efficient and mild approach to various 2-aryl-amino naphtho[1,2-d]thiazole compounds with a broad range of functional groups with up to 90% isolated yields in 5 min.

Liu, Tianbao*; Peng, Yanfen; Gui, Meifang; Zhang, Min*
Chin. J. Org. Chem. **2019**, 39(11), 3199

An Efficient Palladium Nanoparticles Catalytic System for Suzuki Coupling Reactions



A simple and highly efficient palladium nanoparticles catalytic system was successfully used in the Suzuki reaction of aryl halides and arylboronic acids. A high turnover number of 90000 was achieved at the catalyst loading as low as 0.001 mol%. This catalytic system exhibited good stability and longevity, and a broad scope of substrates was tolerated.

Li, Hengchao; Zhao, Ling; Liu, Yan; Zhang, Xia; Li, Wangbing; Jing, Linhai; Huang, Jin*; Wang, Wei*
Chin. J. Org. Chem. **2019**, 39(11), 3207

CONTENT

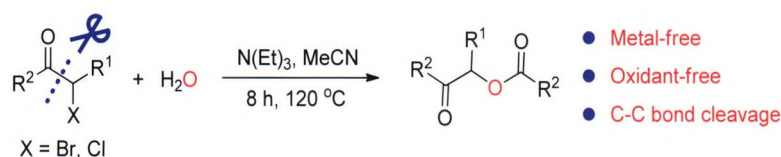
Pd(OAc)₂/CuI-Catalyzed Tandem Reaction for Synthesis of Polysubstituted 3-Chalcogenylindoles



Tandem Pd(OAc)₂/CuI catalyzed coupling/cyclization/chalcogenylation reaction of gem-dibromovinylanilines with boronic acids and dichalcogenides has been developed, which provides a new synthetic approach to 3-sulfenyl- and 3-selenyl-indoles. Various functional groups such as methoxyl, halides, trifluoromethyl and 2-thienyl groups in the substrates are tolerated.

Liu, Ruiting; Li, Zhen; Wang, Shengke; Zhou, Xigeng*
Chin. J. Org. Chem. **2019**, 39(11), 3215

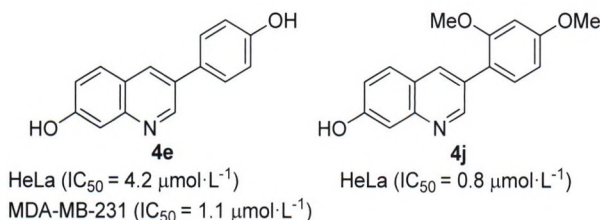
Triethylamine Promoted the C—C Bond Cleavage of α -Halo Ketones: α -Acetoxyaryl Ketone Synthesis



A novel amine-promoted C—C bond cleavage of α -halo ketones was reported. A variety of α -acetoxyaryl ketone compounds were prepared from commercially available α -halo ketones in good to excellent yields.

Wang, Maorui; Wu, Yuzheng; Yao, Jian; Deng, Li; Pan, Yingming; Huang, Kebin*; Tang, Haitao*
Chin. J. Org. Chem. **2019**, 39(11), 3223

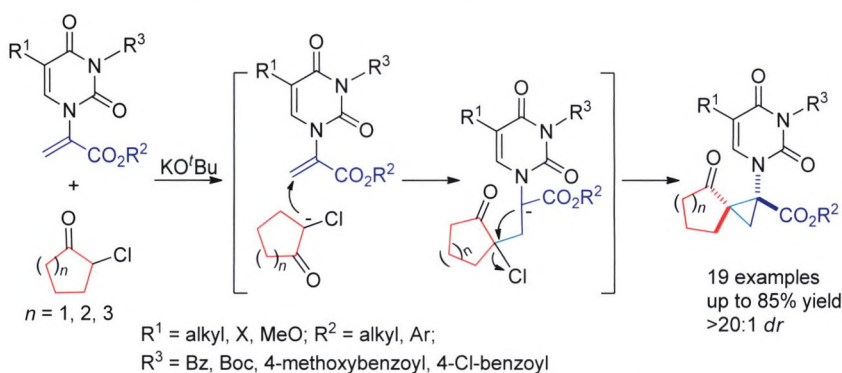
Antitumor and Topoisomerase II α Inhibitory Activities of 3-Aryl-7-hydroxyquinolines



Twenty-one 3-aryl-7-hydroxyquinolines were designed and synthesized by scaffold hopping of the lead compound CS1. These compounds were evaluated for their inhibitory activity against Topo II α activity in DNA relaxation assays, and evaluated for the antitumor activity in *in vitro* growth inhibition assays against human triple negative breast cancer MDA-MB-231 cells and human cervical cancer HeLa cells.

Hu, Yuan; Li, Zhenyu; Ding, Yanjiao; Li, Zhiying; Liu, Zhiyong; Shen, Yuemao*
Chin. J. Org. Chem. **2019**, 39(11), 3230

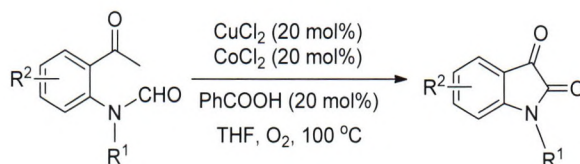
Efficient Synthesis of Spirocyclic Nucleosides via Michael Addition-Initiated Intermolecular Cyclopropanation Reaction



• Spirocyclic nucleoside analogs • Simple and practical conditions • Broad substrate scope

An efficient route to synthesize 2'-spiro[2-oxocyclopentyl]cyclopropyl nucleoside analogues via KOtBu promoted Michael addition-initiated cyclopropanation reactions of α -thymine acrylates with α -chloro-cycloalkanones was successfully developed. A wide range of C(2')-spirocyclic modified nucleoside analogs were obtained with excellent diastereoselectivities and good yields (up to 85%).

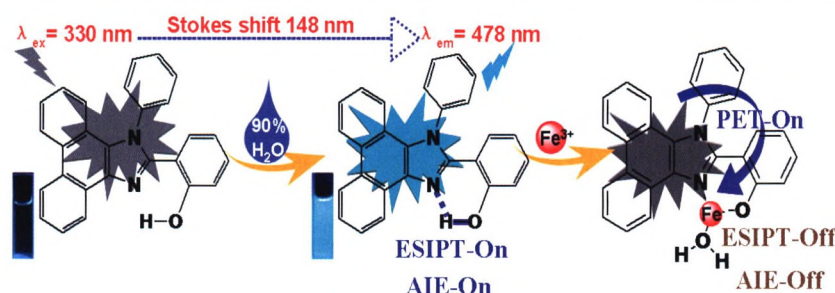
Hao, Erjun; Zhang, Qing; Zhang, Qiying*; Qu, Guirong; Yang, Xining; Guo, Haiming*
Chin. J. Org. Chem. **2019**, 39(11), 3237

Copper-Catalyzed Aerobic Oxidation
Strategy: A Concise Route to Isatin

Ahmad, Muhammad Siddique; Zhu, Yamin;
Guo, Yunlong; Zhang, Saisai; Shen, Zeng-
ming*

Chin. J. Org. Chem. **2019**, 39(11), 3244

A copper-catalyzed decarbonylation cyclization to form isatins using oxygen as terminal oxidant is developed. This complementary way offers a new protocol for the synthesis of isatins through $\text{C}(\text{sp}^3)\text{—H}$ bond functionalization in $\text{Cu}/\text{O}_2/\text{Co}$ system. This system shows good reactivity and compatibility.

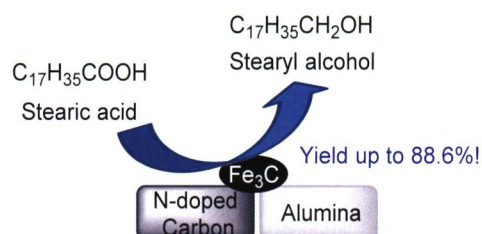
An Excited-State Intramolecular Proton
Transfer (ESIPT) Plus Aggregation Induced
Emission (AIE) Phenanthro[9,10-*d*]-
imidazole-Based Fluorescence Probe for
Detection of Fe^{3+} in Living Cells

He, Yuqian; Zhao, Bing*; Kan, Wei*; Wang,
Liyang; Song, Bo; Yin, Guangming; Bi, Ye;
Chen, Shuwen*

Chin. J. Org. Chem. **2019**, 39(11), 3250

Based on the mechanism of excited-state intramolecular proton transfer (ESIPT) plus aggregation induced emission (AIE), a fluorescence probe of phenanthro[9,10-*d*]imidazole modified by the phenolic hydroxyl (PIP-*o*-OH) had been designed, synthesized and applied in the detection of Fe^{3+} .

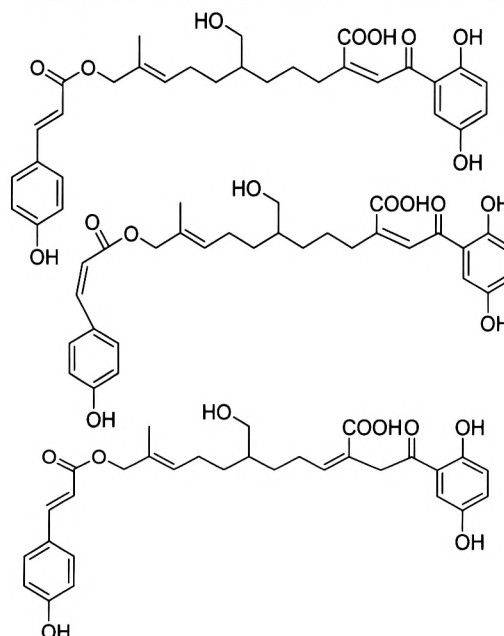
NOTES

Iron-Catalyzed Selective Hydrogenation
of Stearic Acid to Stearyl Alcohol

Li, Jiang*; Wan, Tong; Zhang, Junjie; Fu,
Yao

Chin. J. Org. Chem. **2019**, 39(11), 3258

Selective hydrogenation of stearic acid to stearyl alcohol is first achieved over heterogeneous iron catalysts.

Meroterpenoids from the Fruiting Bodies
of *Ganoderma ahmadii* Steyaret and
Their Protein Tyrosine Phosphatase 1B
Inhibitory Activities

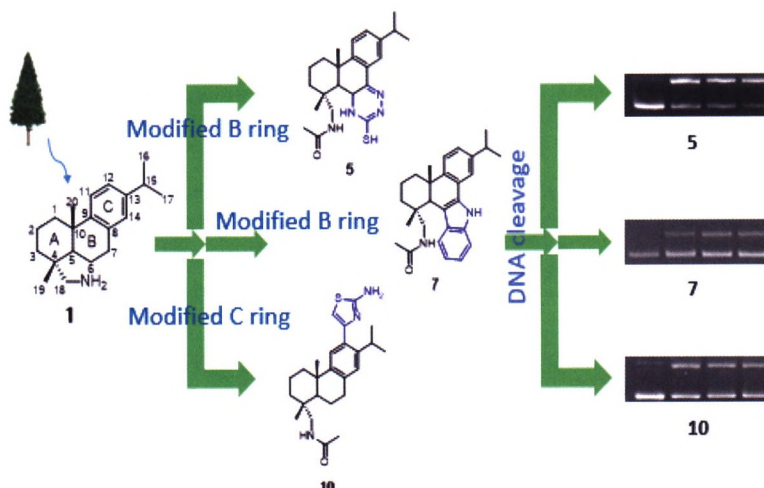
Guo, Jiaocen; Ma, Qingyun; Kong, Fang-
dong; Xie, Jingyi; Zhou, Liman; Ding,
Qiong; Wu, Yougen*; Zhao, Youxing*

Chin. J. Org. Chem. **2019**, 39(11), 3264

The extraction, isolation, structure identification and protein tyrosine phosphatase 1B (PTP1B) inhibitory activity of three new meroterpenoid compounds from the fruiting body of *Ganoderma ahmadii* Steyaret are reported. Finally, their future development and application are also prospected.

CONTENT

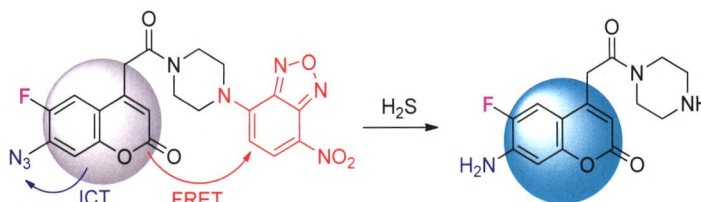
Interaction of Optically Pure Dehydroabietylamine Heterocyclic Derivatives with DNA and Preliminary Cytotoxic Activity



Three optically pure dehydroabietylamine derivatives, acetyldehydroabietylamine-6,7-(3-mercapto)-1,2,4-triazine (**5**), acetyldehydroabietylamine-6,7-indole (**7**) and 12-(2-aminothiazole)-acetyldehydroabietylamine (**10**), were obtained by introducing different aromatic heterocycles into the B and C rings of optically pure dehydroabietylamine. Optically pure dehydroabietylamine heterocyclic derivatives could interact with DNA, and scissor pBR 322 plasmid DNA into single strands, and showed synergistic antitumor effect with copper(II) salt.

Tu, Shuangyan; Xu, Wushang; Qi, Fen; He, Weijiang*; Fei, Baoli*
Chin. J. Org. Chem. **2019**, 39(11), 3269

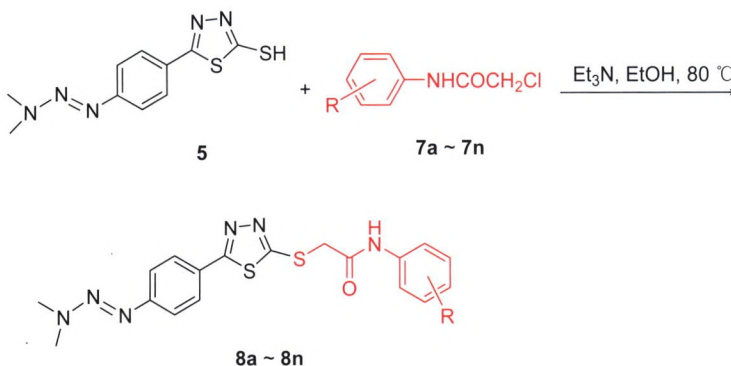
A Response Rate Matching Dual-Reactable Probe for Fluorescent Recognition of Hydrogen Sulfide



A response rate matching dual-reactable H₂S fluorescent probe was designed and synthesized by employing *ortho*-fluoro-substituted coumarin azide and 7-nitrobenzofuran-piperazine as the H₂S reactive groups and the fluorescence quenching groups. The probe showed high selectivity and sensitivity (*ca.* 3600-fold fluorescence *off-on* enhancement and nanomolar detection limit).

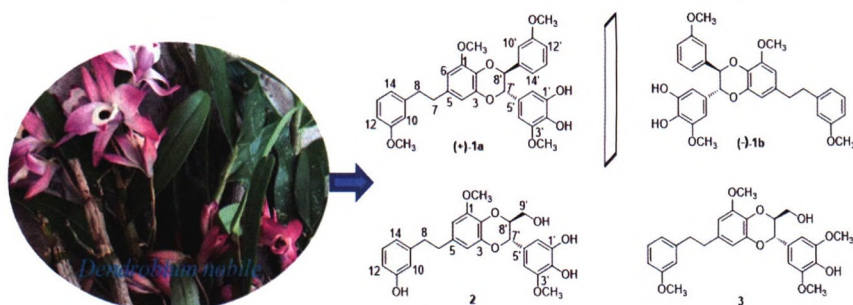
Xie, Chang; Ma, Chen; Jia, Xu; Zhang, Xueqi; Wei, Chao*; Zhang, Pingzhu; Li, Xiaoliu*
Chin. J. Org. Chem. **2019**, 39(11), 3277

Synthesis and Antitumor Activities of 1,3,4-Thiadiazole Triazene Amide Derivatives



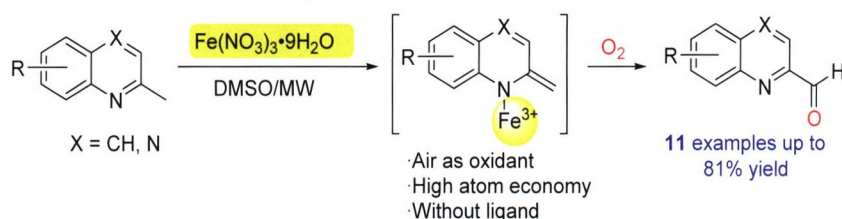
By splicing the triazene structure with 1,3,4-thiadiazole and amide, fourteen unreported 1,3,4-thiadiazole triazene amide derivatives were synthesized. By using the typical triazene drug dacarbazine as a reference, the activity detections of human esophageal cancer cells (EC109), human gastric cancer cells (MGC803) and human prostate cancer cells (PC-3) were carried out.

Chen, Yanjun; Zhang, Mingqian; Li, Ziqiu; Luo, Defu; Li, Longhui; Yu, Tingting; Long, Yue*
Chin. J. Org. Chem. **2019**, 39(11), 3283

Bibenzyl Derivatives from *Dendrobium nobile*

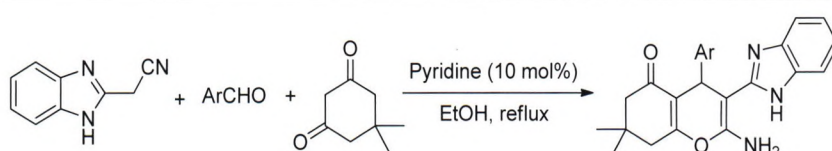
By silica gel, MCI column chromatographic and preparative high performance liquid chromatography (HPLC) technologies, three new bibenzyl derivatives were isolated from the tubes of *Dendrobium nobile*. One racemic compound was further purified by chiral HPLC to obtain a pair of enantiomers **1a** and **1b**, and the absolute configurations of the enantiomers were confirmed using electronic circular dichroism calculations. The structures of compounds **1**~**3** were identified as didendroniline A (**1**), dendroniline B (**2**) and dendroniline C (**3**).

Zhang, Maosheng; Linghu, Lang; Zhang, Jianyong; Nie, Xuqiang; Li, Xiaofei; Guo, Dale; Xiao, Shiji*
Chin. J. Org. Chem. **2019**, 39(11), 3289

Iron/O₂-Promoted C—H Bond Functionalization for the Exclusive Synthesis of 2-Quinoline Carboxaldehydes under Microwave Irradiation

Xie, Tinghui; Jiang, Xiaoying; Mi, Zhisheng; Li, Xue; Xu, Xiaohe; Bai, Renren; Shuai, Qi; Xie, Yuanyuan*
Chin. J. Org. Chem. **2019**, 39(11), 3294

An one-pot iron-catalyzed oxidative formylation of 2-methylquinolines to produce 2-quinoline carboxaldehydes under microwave irradiation has been achieved by employing O_2 as the oxygen donor.

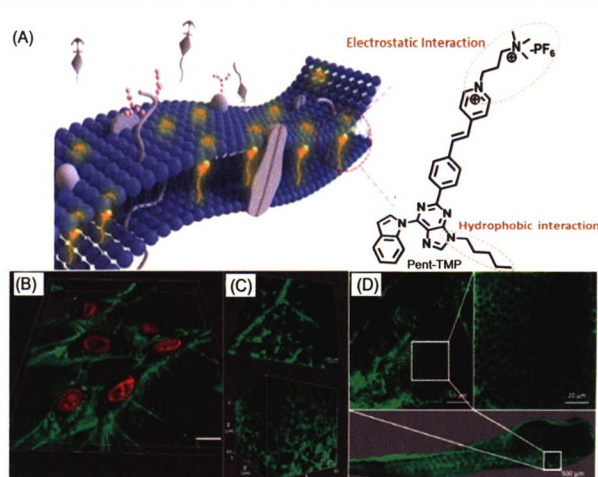
One-Pot Three-Component Synthesis of 3-(1*H*-Benzo[*d*]imidazol-2-yl)chromen Derivatives

Wang, Xiang*; Chen, Ping; Zhi, Sanjun; Hu, Huayou; Kan, Yuhe; Zhang, Zaichao*
Chin. J. Org. Chem. **2019**, 39(11), 3299

The efficient synthesis of new substituted 3-(1*H*-benzo[*d*]imidazol-2-yl)-4*H*-chromens in 48%~89% yields via one-pot, three-component reaction of 2-(1*H*-benzo[*d*]imidazol-2-yl)acetonitrile with aromatic aldehydes and 5,5-dimethylcyclohexane-1,3-dione was reported. This reaction was carried out in EtOH in the presence of pyridine under reflux conditions. All reactions were completed within 1~3 h.

HIGHLIGHTS

Visual Imaging of Plasma Membrane: New Application for Aggregation Induced Emission (AIE) Probe

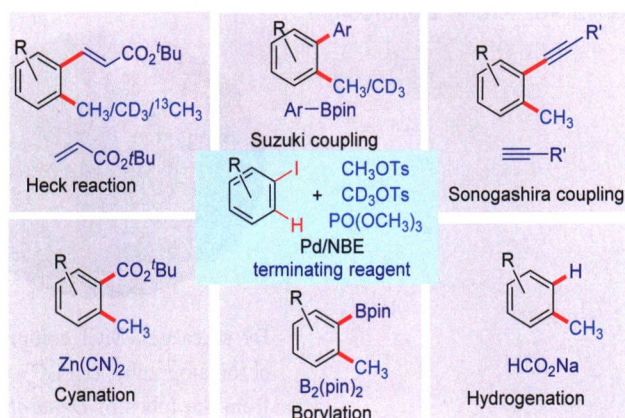


Yang, Jiea; Li, Zhen*
Chin. J. Org. Chem. **2019**, 39(11), 3304

CONTENT

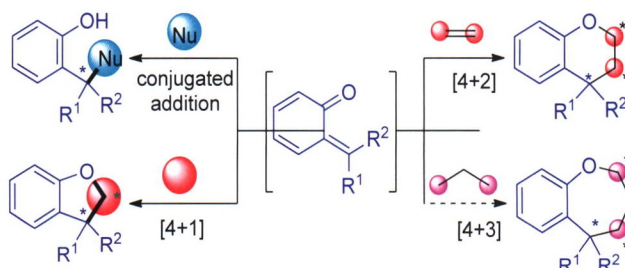
A Modular C—H Methylation Reaction via Catellani Strategy

Tong, Huarong; He, Gang; Chen, Gong*
Chin. J. Org. Chem. **2019**, 39(11), 3306



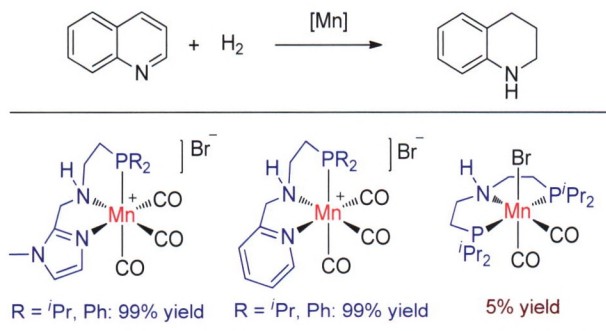
Catalytic Asymmetric [4+3] Cyclizations of 2-Indolylmethanols with *ortho*-Quinone Methides

Liu, Lu*; Zhang, Junliang*
Chin. J. Org. Chem. **2019**, 39(11), 3308



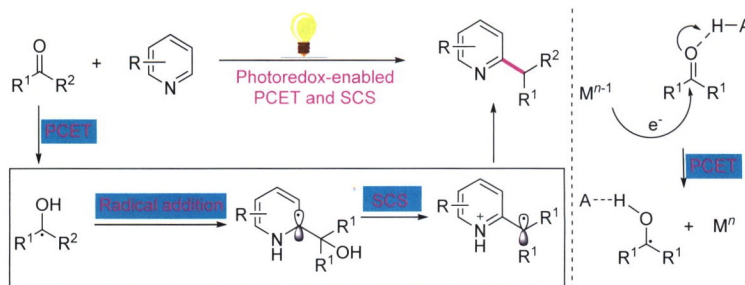
Ligand Effect in Manganese-Catalyzed Hydrogenation: Mechanism and Application Studies

He, Yanmei; Fan, Qinghua*
Chin. J. Org. Chem. **2019**, 39(11), 3310



Ketones and Aldehydes as Alkyl Radical Equivalents for Direct C—H Alkylation of Heteroarenes

Cheng, Xiaokai; Lu, Zhan*
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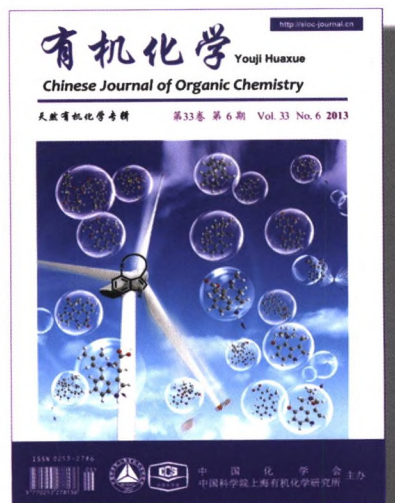
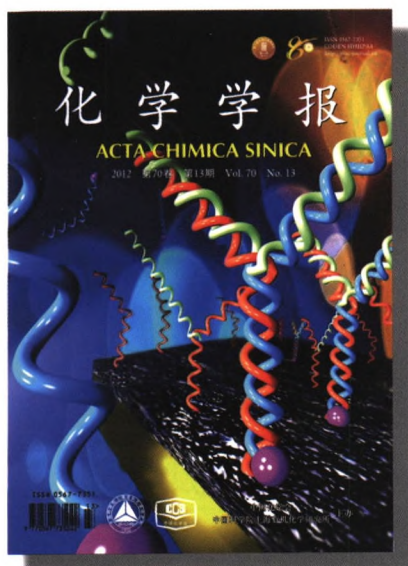
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