

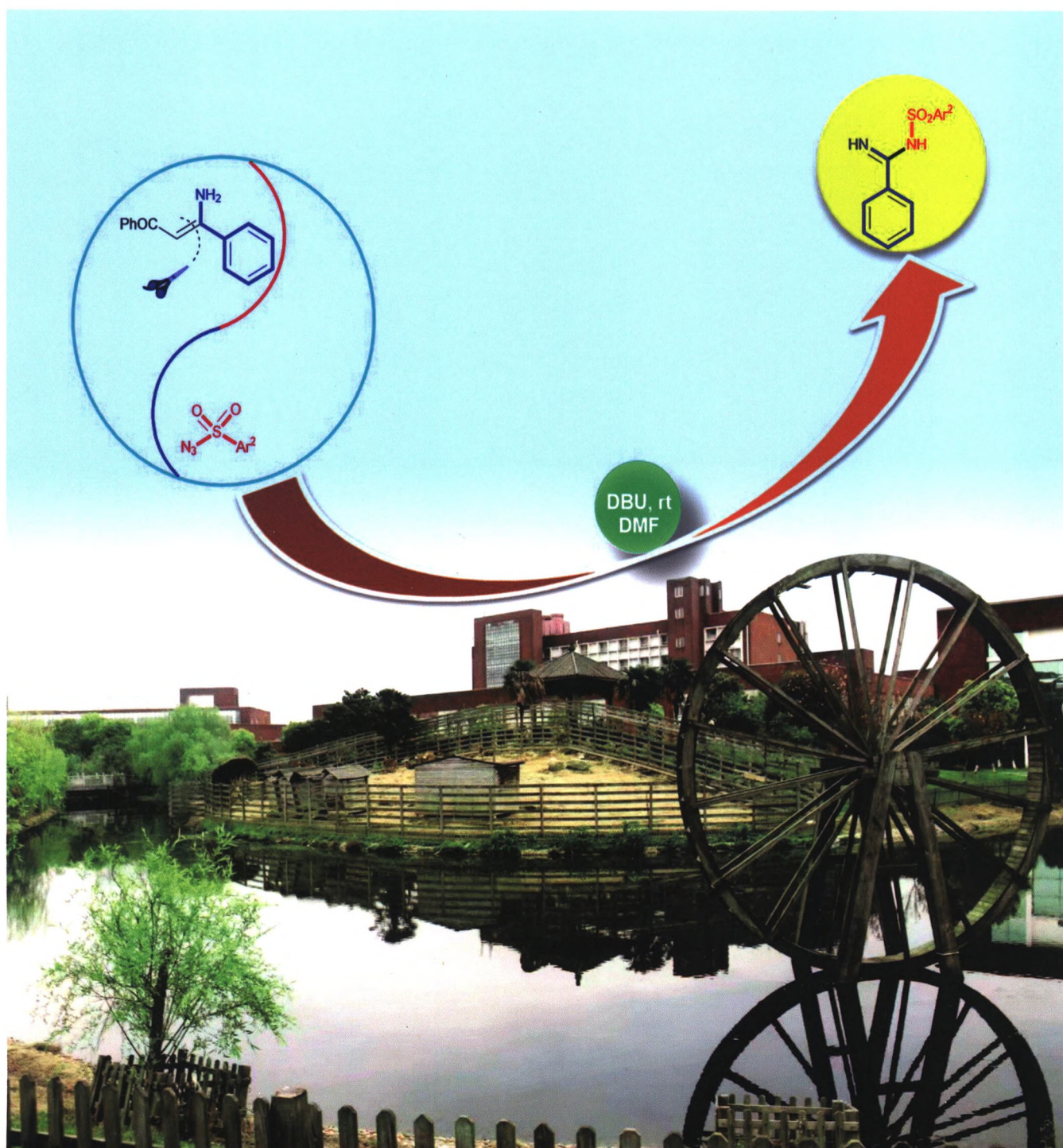
有机化学

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(YOUJI HUAXUE)

第 40 卷 第 3 期 (总 376 期) 2020 年 3 月

目 次

综述与进展

多环型 Xanthone 类天然产物的合成研究进展	谢 涛 何海兵* 高栓虎*	(551)
可见光直接促进的过渡金属催化交叉偶联反应研究进展	李祯龙 金 健* 黄莎华*	(563)
胺及其衍生物的非酰基化动力学拆分	唐 亮 李雪薇 谢 芳* 张万斌	(575)
无过渡金属体系下 C(sp ²)-H 键的自由基反应构建 C-N 键研究进展	吴 燕 陈锦杨* 李 强 魏文廷*	(589)
可见光诱导烷基羧酸及其衍生物的脱羧偶联反应研究进展	周明东* 覃丕涛 经理珂 孙 京* 杜海武	(598)
含有氢键供体大环化合物的构筑及其功能研究进展	黄国保 陈志林 韦贤生 陈 钰 李秀英 仲 辉* 谭明雄*	(614)
配体促进的过渡金属催化芳烃远程 <i>meta</i> -C-H 键官能化反应研究进展	田万发 李 娜 彭子赫 冯丽华 麦 曦 贺永勤*	(625)

研究论文

无金属和氧化剂温和条件下碱促进的烯胺酮碳-碳双键断裂合成 NH ₂ -结构脒类化合物	王国栋 郭艳辉 万结平*	(645)
镍催化环丁酮酯与芳基锌试剂的 Negishi 偶联反应	帅 斌 李兆明 裴 晖 方 萍* 梅天胜*	(651)
开环葫芦脲在水中对芳烃和芳醛的增溶和对脞大环形成的促进作用	刘旭波 林佳乐 王 辉 张丹维* 黎占亭*	(663)
胡椒基咪唑盐类化合物的合成及生物活性研究	刘正芬 字玉金 张琳琳 方永晟 沈艳珍 李 艳 羊晓东* 张洪彬*	(669)
云南蕊木茎中的抗炎吡啶生物碱	解天珍 赵云丽 马伟光 王易芬 于浩飞 王 蓓 魏 鑫 黄之锴 朱培凤 刘亚平* 罗晓东*	(679)
钌(II)-催化偶氮苯与乙醛酸乙酯的环化反应构建 3-羧酸酯吡唑	陈 训* 王 颖 王烁今 孔杜林 文丽君 翟锐锐 赵 珂 白丽丽* 李友宾	(688)
无催化剂条件下 4-羟基烷基-2-炔酸乙酯与 <i>N</i> -杂环芳基甲基- <i>N</i> -2,2-二氟乙基-1-胺的串联反应	赵 宇 王卫伟 刘鑫磊 耿 瑞 王明安*	(694)
Zn/Y 双金属接力催化: 一锅法分子内环异构化/分子间阿尔德-烯反应构建 α -羟基酰胺噻唑衍生物	张 硕 楼建芳* 王佳睿 宋子贺 彭 丹 王 峰 闫志旺 崔仕麒 刘一帆 牟秋红 李金辉	(704)

* 通讯联系人.

钴催化双齿导向基辅助的 1-萘胺衍生物与醇的区域选择性碳氢键烷氧基化反应	张梦帆 李瑞鹏 杨震* 冯若昆*	(714)
铜促进 8-氨基喹啉导向的芳基酰胺的二甲硫基化反应	郭圆圆 刘振伟 朱明祥 李琳琳 李敬亚 邹大鹏* 吴豫生* 吴养洁*	(724)
1,3,4-噁二唑和 1,3,4-噻二唑衍生物的设计、合成和生物活性研究	郝龙家 黎永良 邓明高 陈安超 杜志云 董长治 陈惠雄*	(731)
银催化 2-芳基吡啶与马来酰亚胺 C—H 键烷基化反应	皮超 曲亚平 崔秀灵* 吴养洁*	(740)
基于不同功能的苯并噻二唑为受体单元和低聚噻吩为给体单元的 D-A-D-A-D 型有机小分子光伏材料的理论计算研究	王丽辉* 白锁柱 李东勇 周宏	(748)
氰基亚胺与亚甲基二氢苯并呋喃二酮类化合物的[3+2]环加成反应: 螺吡唑类化合物的合成	史望宇 徐嘉擎 毛比明 贾皓 黄家兴* 郭红超*	(756)
基于 1-(4-己氧)苯和 2,4,6-三苯基-1,3,5-三嗪功能化苄单元的聚合物蓝光材料的合成与表征	姜鸿基* 何煦 李雄	(763)

研究简报

含取代噻唑结构的新型吡唑脒衍生物的合成与生物活性	朱玥 郑丹丹 倪亚丹 李金峰 周环宇 胡兰萍* 李玲 鞠剑峰* 陈家正 李宏 石玉军 戴红*	(774)
双酰胺类化合物的合成及生物活性研究	杨森 邹文倩 陶敏 马天扬 刘幸海 翁建全 谭成侠*	(782)
N-(2-(5-(3-溴-1-(3-氯吡啶-2-基)-1 <i>H</i> -吡唑-5-基)-1,3,4-噁二唑-2-基)-4-氯-6-甲基苯基)酰胺类新化合物的合成及生物活性	张燕 尚俊峰 李欢 刘航 宋海斌 王宝雷* 李正名*	(787)
新型 1,3-二取代酞嗪酮类衍生物的合成及抗肿瘤活性研究	张路野 张洋 包崇男 杨鹏 李二冬 孟姪琪 崔飞 周蕊 黄诗雨 郑甲信* 单丽红* 刘宏民* 张秋荣*	(794)
含氮原子桥联吡咯基稀土金属双核配合物的合成及催化ε-己内酯的开环聚合反应	吴杰 张秀丽 张丽军* 韦芸 周双六*	(801)

亮点述评

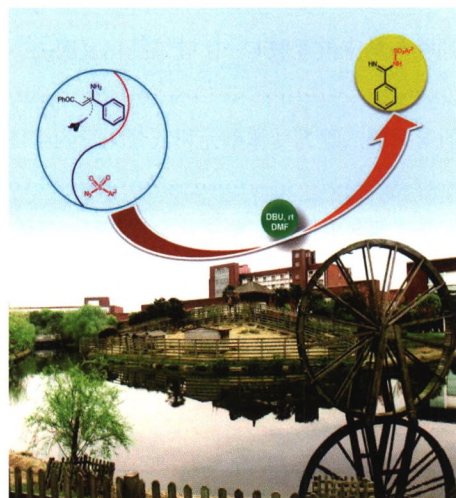
手性环丙烯构建策略用于对映选择性合成偕二氟亚甲基三元环化合物	欧阳瑶 卿凤翎*	(806)
利用 1,2-叠氮迁移策略合成 β-氟烷基叠氮化合物	吴镇 朱晨*	(808)
钯催化芳基三氟甲磺酸酯与炔烃的不对称 Heck 偶联反应	蔡援 施世良*	(810)
钯催化三亚甲基甲烷的不对称[3+4]环加成反应: 高区域选择性、高非对映选择性和高对映选择性构筑苯并呋喃[3,2- <i>b</i>]吡啶	王美馨 邓玉华* 邵志会*	(812)
Cu(I)催化的不对称直接插炔 Aldol 反应构建手性 2,3-联烯醇	冉光尧 陈应春*	(814)

Chinese Journal of Organic Chemistry

Vol. 40 No. 3 March 2020

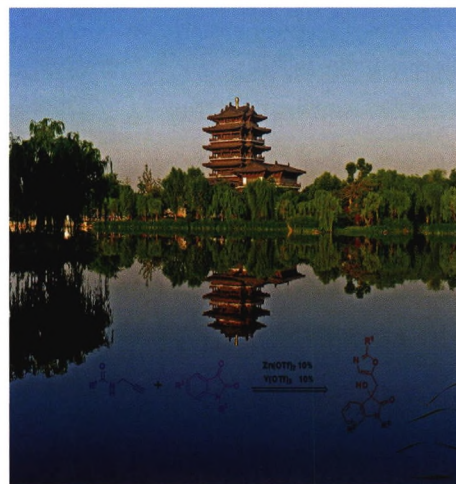
Cover Picture: Base-Promoted, Metal- and Oxidant-Free C=C Bond Cleavage in Enaminones for Ambient Synthesis of NH₂-Amidines

The synthesis of NH₂-functionalized sulfonyl amidines by the reactions of enaminones and sulfonyl azides is reported by Wang, Guo and Wan on page 645. By means of cleaving the enaminones C=C double, the synthesis of NH₂-functionalized N-sulfonyl amidines has been realized via the promotion of only DBU, disclosing the first metal-free method toward NH₂-sulfonyl amidine synthesis.



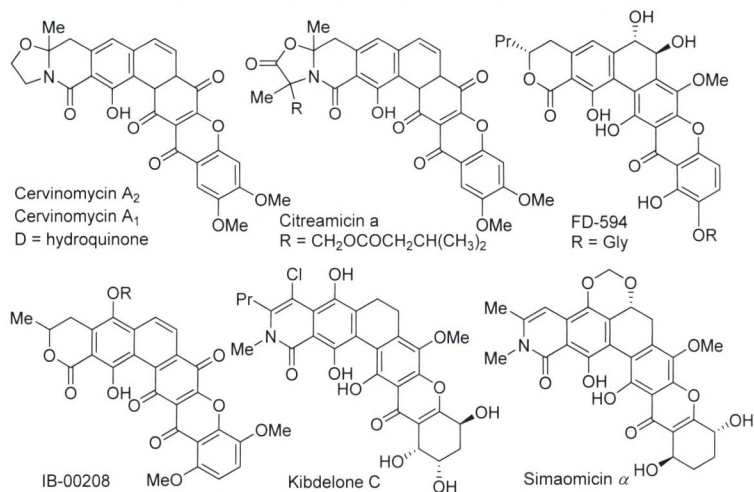
Inside Cover: Zn/Y Bimetallic Relay Catalysis: One Pot Intramolecular Cycloisomerization/Intermolecular Alder-Ene Reaction toward Oxazole α -Hydroxy Amide Derivatives

A novel bimetallic relay catalytic system of Zn(OTf)₂/Y(OTf)₃ has been successfully developed. A variety of oxazole α -hydroxy amide derivatives were obtained from easily available *N*-(propargyl)arylamides and various 1-benzylindoline-2,3-dione derivatives through intramolecular cycloisomerization/intermolecular Alder-ene reaction under mild conditions by Zhang, Lou, Wang, Song, Peng, Wang, Yan, Cui, Liu, Mu and Li on page 704.



REVIEWS

Synthetic Progress of Polycyclic Xanthone

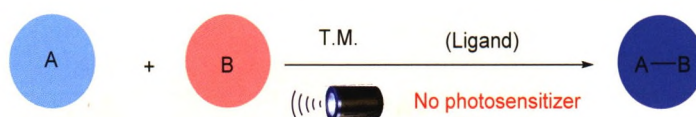


Xie, Tao; He, Haibing*; Gao, Shuanhu*
Chin. J. Org. Chem. **2020**, 40(3), 551

Recent progress in the synthesis of polycyclic xanthones: cervinomycin A₁/A₂, FD-594 aglycon, IB-00208 aglycone, kibdelones, simaomicin α and kigamicin is reviewed.

CONTENT

Recent Advances in Transition Metal-Catalyzed Cross-Coupling Reactions Directly Promoted by Visible Light

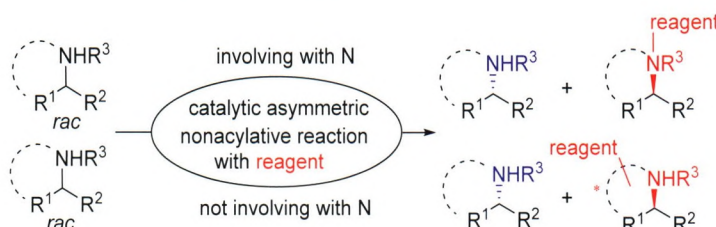


T.M. : Cu, Ni, Co, Fe, Pd, Rh, Ru, Au
cross coupling : Suzuki-Miyaura, Kumada, Negishi, Stille, Heck, Ullmann, Sonogashira, Hiyama

The recent progress in transition metal (Cu, Ni, Co, Fe, Pd, Rh, Ru, Au) catalyzed cross-coupling reactions directly promoted by visible light is reviewed. In this field, chemists have implemented a series of coupling reactions including Suzuki-Miyaura, Heck, Sonogashira, Stille, Kumada, Negishi, Ullmann and Hiyama reactions.

Li, Zhenlong; Jin, Jian*; Huang, Shahua*
Chin. J. Org. Chem. **2020**, 40(3), 563

Catalytic Kinetic Resolution of Amines and Their Derivatives by Non-acylation Reaction



Tang, Liang; Li, Xuewei; Xie, Fang*; Zhang, Wanbin
Chin. J. Org. Chem. **2020**, 40(3), 575

This review aims to introduce the development of the non-acylation KR of amines for the synthesis of enantiopure amines.

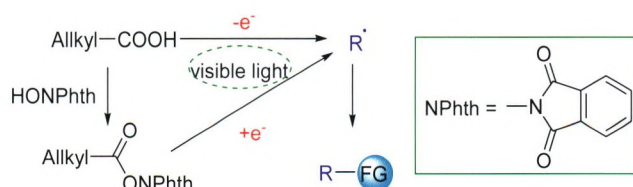
Progress in C—N Bond Formation Involving C(sp²)—H Bond through Transition-Metal-Free Radical Reactions



The C—N bond is widely found in medicinal molecules, natural products and functional materials. Recently, remarkable progress has been made in construction of C—N bond involving C(sp²)—H bond through transition-metal-free radical reactions. Due to the relatively mild reaction condition and high reactivity, it provides a novel approach to construct C—N bond. In this review, the recent developments in this area are summarized on the basis of different nitrogen sources.

Wu, Yan; Chen, Jinyang*; Li, Qiang; Wei, Wenting*
Chin. J. Org. Chem. **2020**, 40(3), 589

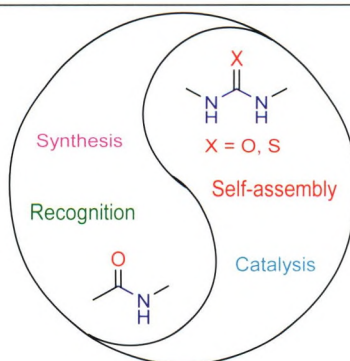
Progress in Photoinduced Decarboxylative Radical Cross-Coupling of Alkyl Carboxylic Acids and Their Derivatives



The recent progress in photoinduced decarboxylative radical cross-coupling of alkyl carboxylic acids and their derivatives is reviewed. They are often easy to generate alkyl radical by photoredox catalysis under mild conditions for building various chemical bonds in organic chemistry.

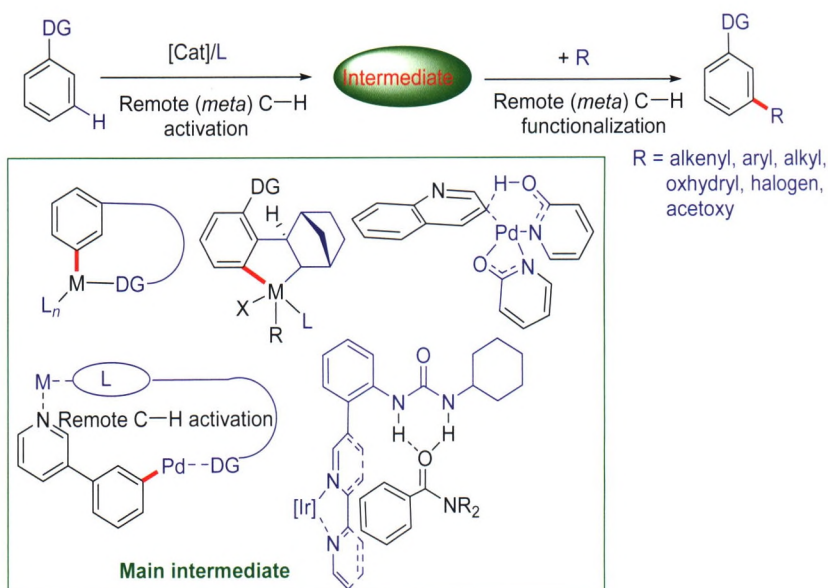
Zhou, Mingdong*; Qin, Pitao; Jing, Like; Sun, Jing*; Du, Haiwu
Chin. J. Org. Chem. **2020**, 40(3), 598

Recent Progress on the Construction and Function of Macrocyclic Compounds Containing Hydrogen Bond Donors



The recent progress on the synthetic methods of macrocyclic compounds based on (thio) urea, amide and its molecular recognition in 2010~2019 is summarized. It is hoped that this review can be referred to synthesis and applications of this kind of macrocyclic compounds.

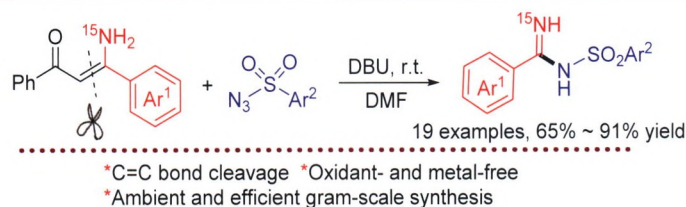
Huang, Guobao; Chen, Zhilin; Wei, Xian-sheng; Chen, Yu; Li, Xiuying; Zhong, Hui*; Tan, Mingxiong*
Chin. J. Org. Chem. **2020**, 40(3), 614

Recent Advances in Liangd-Promoted
Transition-Metal-Catalyzed Remote *meta*-C—H
Functionalization of Arenes

Tian, Wanfa; Li, Na; Peng, Zihé; Feng, Lihua; Mai, Xi; He, Yongqin*
Chin. J. Org. Chem. **2020**, 40(3), 625

The recent progress on ligand accelerated transition-metal-catalyzed remote *meta*-selective C—H bond functionalization of arenes is summarized, and the limitations of the research field and the prospects for future development are presented.

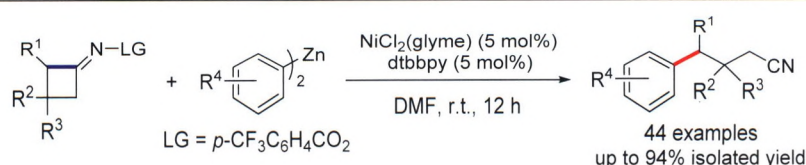
ARTICLES

Base-Promoted, Metal- and Oxidant-Free
C=C Bond Cleavage in Enaminones for
Ambient Synthesis of NH₂-Amidines

Wang, Guodong; Guo, Yanhui; Wan, Jieping*
Chin. J. Org. Chem. **2020**, 40(3), 645

The synthesis of NH₂-based amidines is realized under metal- and oxidant-free conditions via the reactions of NH₂-enaminones and sulfonyl azides, resulting in the fast access to structurally divergent amidine products at room temperature.

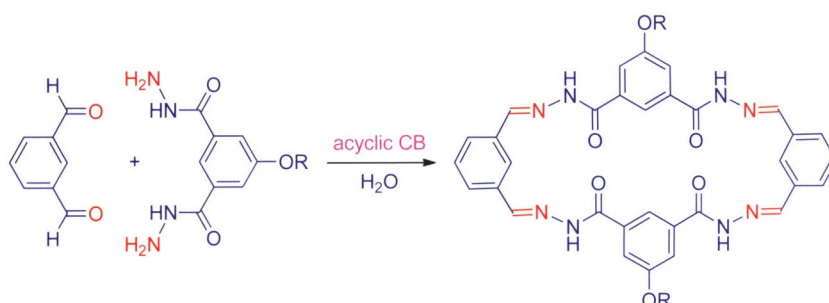
Nickel-Catalyzed Negishi Coupling of Cyclobutanone Oxime Esters with Aryl Zinc Reagents



Shuai, Bin; Li, Zhao-Ming; Qiu, Hui; Fang, Ping*; Mei, Tian-Sheng*
Chin. J. Org. Chem. **2020**, 40(3), 651

A nickel-catalyzed Negishi coupling of cyclobutanone oxime esters with aryl zinc reagents has been developed, in which Ni catalysis serves as dual roles for iminyl radical generation and cross-coupling of alkyl radical and aryl zinc reagent.

Water-Solubilization of Acyclic Cucurbiturils for Arenes and Aromatic Aldehydes and the Promotion for the Generation of Two Hydrazone-Based Macrocycles



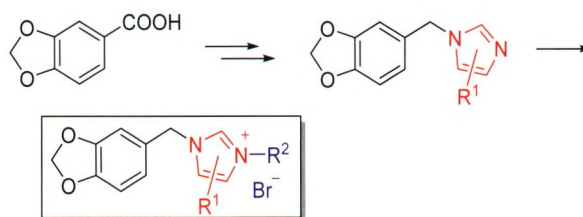
Liu, Xu-Bo; Lin, Jia-Le; Wang, Hui; Zhang, Dan-Wei*; Li, Zhan-Ting*
Chin. J. Org. Chem. **2020**, 40(3), 663

Water-soluble acyclic cucurbiturils have been used to increase the water-solubility of hydrophobic arenes and aromatic aldehydes. The solubilization has been further applied to promote the formation of two hydrazone-based macrocycles in water.

CONTENT

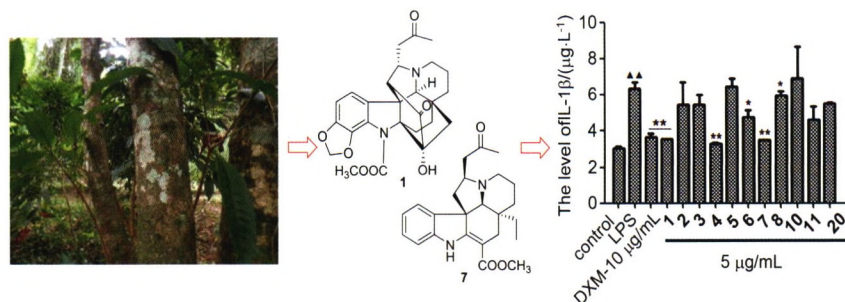
Synthesis and Cytotoxic Activity of Novel Hybrid Compounds between Piperonyl and Imidazolium Salts

Liu, Zhengfen; Zi, Yujin; Zhang, Linlin; Fang, Yongsheng; Shen, Yanzhen; Li, Yan; Yang, Xiaodong*; Zhang, Hongbin*
Chin. J. Org. Chem. **2020**, 40(3), 669



A series of novel hybrid compounds between piperonyl and imidazolium salts have been prepared and evaluated *in vitro* against a panel of human tumor cell lines.

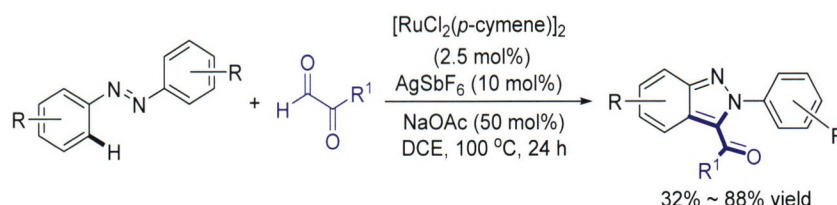
Anti-Inflammatory Indole Alkaloids from the Stems of *Kopsia officinalis*



Xie, Tian-Zhen; Zhao, Yun-Li; Ma, Wei-Guang; Wang, Yi-Fen; Yu, Hao-Fei; Wang, Bei; Wei, Xin; Huang, Zhi-Pu; Zhu, Pei-Feng; Liu, Ya-Ping*; Luo, Xiao-Dong*
Chin. J. Org. Chem. **2020**, 40(3), 679

Seven new monoterpenoid indole alkaloids, kopsioffines A~G, together with twenty known alkaloids, were isolated from the stems of *Kopsia officinalis*. The anti-inflammatory activities of all alkaloids were evaluated on lipopolysaccharide-stimulated RAW 264.7 cells by the inhibiting the production of IL-1 β , PGE2 and TNF- α .

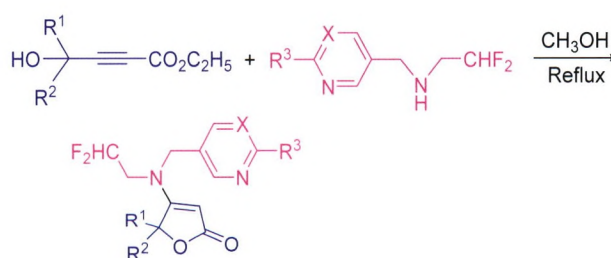
Synthesis of 3-Carboxylate Indazoles via Ru(II)-Catalyzed Annulation of Azobenzenes with Ethyl Glyoxalate



Chen, Xun*; Wang, Ying; Wang, Shuojin; Kong, Dulin; Wen, Lijun; Zhai, Ruirui; Zhao, Ke; Bai, Lili*; Li, Youbin
Chin. J. Org. Chem. **2020**, 40(3), 688

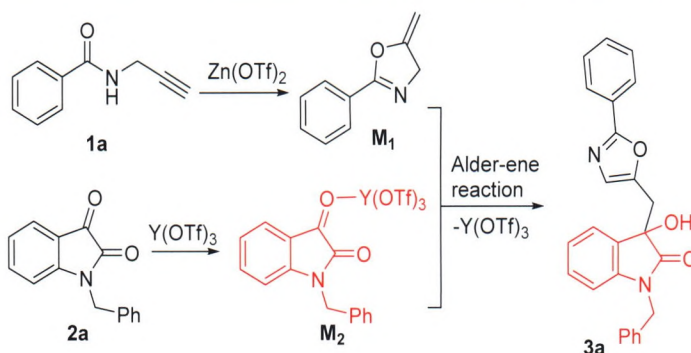
The Ru(II)-catalyzed regioselective [4+1] cycloaddition of azobenzenes with ethyl glyoxalate through C—H bond activation has been developed. This method provides a facile approach to various 3-carboxylate indazoles in moderate to good yields. Meanwhile, the kinetic isotope effect was further investigated and the results indicated that the C—H bond-breaking was possibly not involved in the rate-limiting step of this transformation.

Catalyst-Free Domino Reaction of Ethyl 4-Hydroxyalkyl-2-ynoate with *N*-Heteroaryl-methyl-*N*-2,2-difluoroethan-1-amine



Zhao, Yu; Wang, Weiwei; Liu, Xinlei; Geng, Rui; Wang, Ming'an*
Chin. J. Org. Chem. **2020**, 40(3), 694

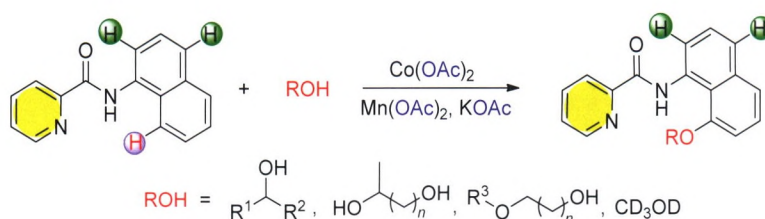
The catalyst-free domino reaction of ethyl 4-hydroxyalkyl-2-ynoate and *N*-heteroaryl-methyl-*N*-2,2-difluoroethan-1-amine was performed, and used to synthesize 4-(*N*-(2,2-difluoroethyl)(*N*-heteroaryl-methyl)amino)-5,5-disubstitutedfuran-2(5*H*)-one in methanol under the reflux condition in 39%~83% yields. Their insecticidal activities were evaluated.

Zn/Y Bimetallic Relay Catalysis: One Pot
Intramolecular Cycloisomerization/Inter-
molecular Alder-Ene Reaction toward
Oxazole α -Hydroxy Amide Derivatives

Zhang, Shuo; Lou, Jianfang*; Wang, Jiarui;
Song, Zihé; Peng, Dan; Wang, Feng; Yan,
Zhiwang; Cui, Shiqi; Liu, Yifan; Mu, Qiu-
hong; Li, Jinhui

Chin. J. Org. Chem. **2020**, 40(3), 704

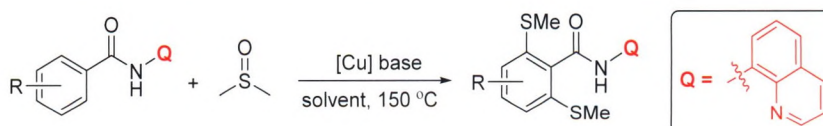
A novel tandem metal relay catalytic system of Zn/Y has been successfully developed. By using this unprecedented Zn(OTf)₂/Y(OTf)₃ bimetallic relay catalytic system, a variety of oxazole α -hydroxy amide derivatives were obtained from easily available *N*-propargyl-arylamides and various 1-benzylindoline-2,3-dione derivatives through intramolecular cycloisomerization/intermolecular Alder-ene reaction under mild conditions.

Cobalt-Catalyzed Bidentate-Assisted Re-
gioselective C—H Alkoxylation of 1-Na-
phthylamide with Alcohols

Zhang, Mengfan; Li, Ruipeng; Yang, Zhen;
Feng, Ruokun*

Chin. J. Org. Chem. **2020**, 40(3), 714

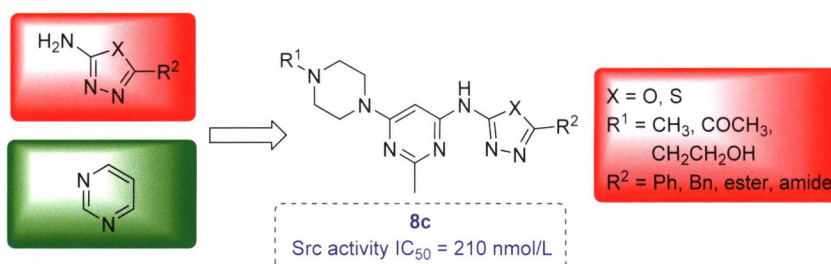
The cobalt-catalyzed regioselective C—H alkoxylation of 1-naphthylamide with alcohols through bidentate-chelation assistance has been developed. In this transformation, not only primary and secondary alcohols, but also aliphatic diols and oligoethylene glycols, which always be employed as O,O-donor ligands and reducing agents in transition metal catalyzed coupling reaction, were all tolerated under current reaction conditions.

Copper-Promoted Dimethylthiolation of
Benzamides under Assistance of 8-Amino-
quinoline Group

Guo, Yuanyuan; Liu, Zhenwei; Zhu, Ming-
xiang; Li, Linlin; Li, Jingya; Zou, Dapeng*;
Wu, Yusheng*; Wu, Yangjie*

Chin. J. Org. Chem. **2020**, 40(3), 724

Cu(II)-promoted dimethylthiolation of C(sp²)—H bonds using dimethyl sulfoxide (DMSO) as the methylthiolation source with the assistance of 8-aminoquinoline directing group have been developed. A number of dimethylthiolated benzamides were efficiently synthesized using CuSO₄·5H₂O as a promoter in moderate to good yields. In addition, this reaction system features facile conditions and no other oxidant additive was required.

Design, Synthesis and Biological Activi-
ties of Compounds Containing 1,3,4-
Oxadiazole or 1,3,4-Thiadiazole

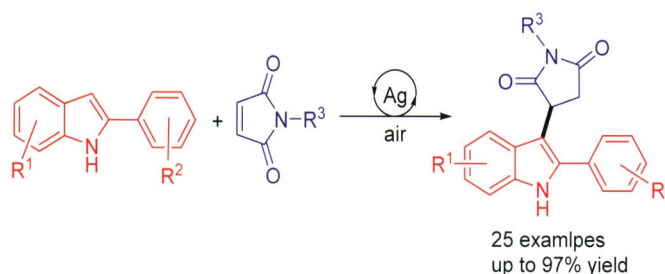
Yan, Longjia; Li, Yongliang; Deng, Minggao;
Chen, Anchao; Du, Zhiyun; Dong, Changzhi;
Chen, Huixiong*

Chin. J. Org. Chem. **2020**, 40(3), 731

A series of novel 1,3,4-oxadiazole and 1,3,4-thiadiazole derivatives were designed and synthesized. The target compounds were evaluated for antitumor activity *in vitro* on four human cancer cell lines including B-16 (skin melanoma cells), PC-3 (human prostate cancer cells), U87 (human primary glioblastoma cells) and A549 (human non-small cell lung cancer cells).

CONTENT

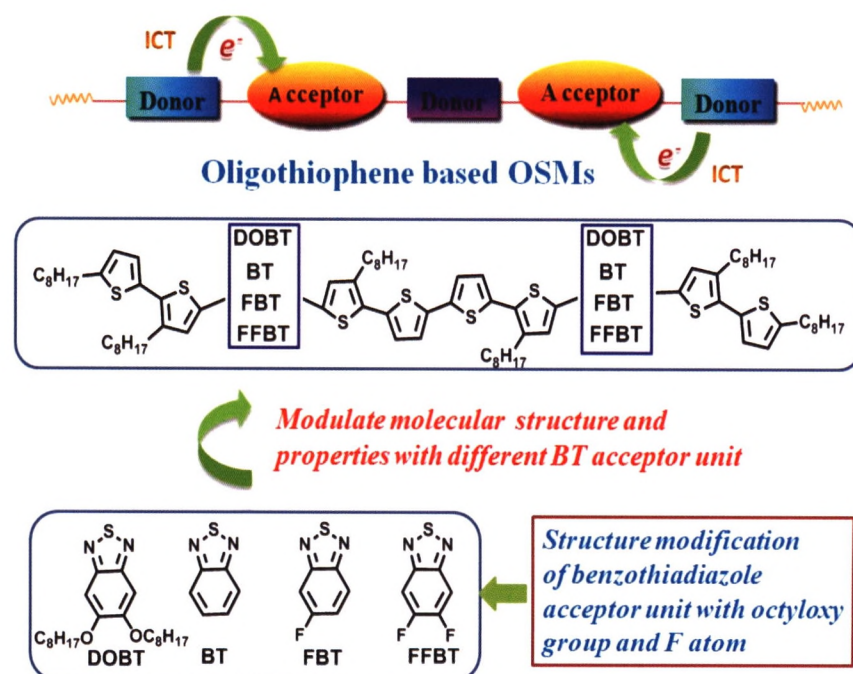
Silver-Catalyzed C—H Alkylation of 2-Arylindoles with Maleimides



Pi, Chao; Qu, Yaping; Cui, Xiuling*; Wu, Yangjie*
Chin. J. Org. Chem. **2020**, 40(3), 740

A highly efficient and regioselective C-3-alkylation of 2-arylindoles with maleimides has been developed using Ag(I) as the catalyst. 3-(2-Aryl-1*H*-indol-3-yl)pyrrolidine-2,5-diones were afforded with high yields (up to 97%) under relatively mild reaction conditions. The method features operational simplicity and using oxygen from the air as oxidant.

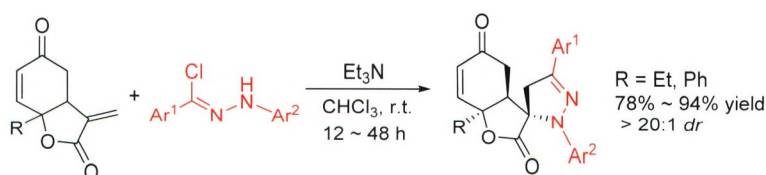
Effect of Structure Modification of Benzothiadiazole Acceptor Unit on the D-A-D-A-D Typed Oligothiophene Based Donor Materials for Organic Small Molecules Solar Cells: A Theoretical Study



Wang, Lihui*; Bai, Suozhu; Li, Dongyong; Zhou, Hong
Chin. J. Org. Chem. **2020**, 40(3), 748

Four D-A-D-A-D structured organic small molecules (OSMs) **DOBT-8T**, **BT-8T**, **FBT-8T** and **FFBT-8T** have been designed. The designed four OSMs were analyzed using density functional theory (DFT) and time dependent-DFT (TDDFT) calculations at B3LYP/6-31G(d) level.

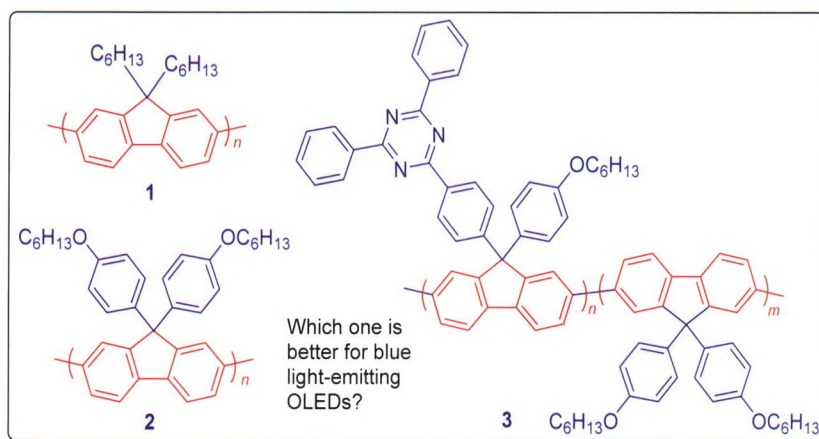
[3 + 2] Cycloaddition of Methylene-dihydrobenzofurandiones and Nitrilimines: Synthesis of Spiropyrazole Compounds



Shi, Wangyu; Xu, Jiaqing; Mao, Biming; Jia, Hao; Huang, Jiaying*; Guo, Hongchao*
Chin. J. Org. Chem. **2020**, 40(3), 756

The [3 + 2] cycloaddition of methylenedihydrobenzofurandiones and nitrilimines worked efficiently in CHCl₃ at room temperature in the presence of Et₃N, producing biologically interesting spirocyclic pyrazole derivatives in high yields (78%~94%) with excellent diastereoselectivity.

Synthesis and Characterization of Blue Light Emitting Polymer Based on 2,4,6-Triphenyl-1,3,5-triazine and 1-(4-Hexyloxy)benzene Functionalized Fluorene Units



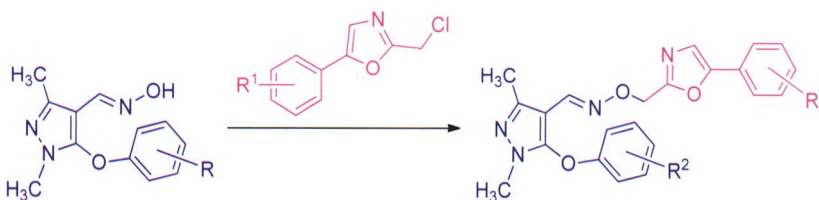
Polymer **3** was synthesized by using 4-(4-hexyloxy)-benzene and 2,4,6-triphenyl-1,3,5-triazine functionalized fluorene monomers through Suzuki cross-coupling polymerization. It exhibited much higher thermal, color purity and photo-stability, which made it to have great potential as emitter for blue light organic light emitting diodes (OLEDs).

Jiang, Hongji*; He, Xu; Li, Xiong
Chin. J. Org. Chem. **2020**, 40(3), 763

NOTES

Synthesis and Biological Activities of Novel Pyrazole Oxime Derivatives Bearing an Oxazole Moiety

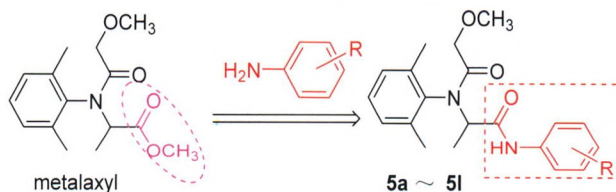
Zhu, Yue; Zheng, Dandan; Ni, Yadan; Li, Jinfeng; Zhou, Huanyu; Hu, Lanping*; Li, Ling; Ju, Jianfeng*; Chen, Jiazheng; Li, Hong; Shi, Yujun; Dai, Hong*
Chin. J. Org. Chem. **2020**, 40(3), 774



A series of novel pyrazole oxime derivatives bearing oxazole unit were synthesized, and their bioactivities were tested.

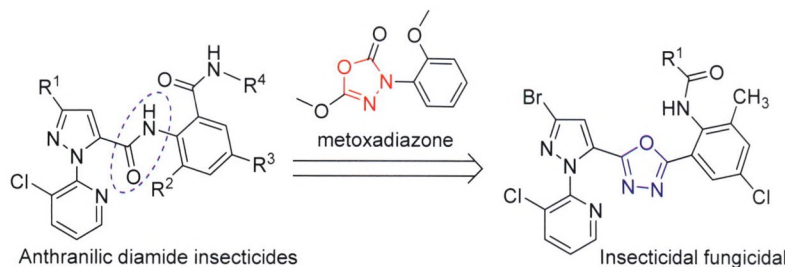
Synthesis and Biological Activity of Diamide Compounds

Yang, Sen; Zou, Wenqian.; Tao, Min; Ma, Tianyang; Liu, Xinghai; Weng, Jianquan; Tan, Chengxia*
Chin. J. Org. Chem. **2020**, 40(3), 782



A series of novel diamide compounds were designed and synthesized by using metalaxyl as leading compound, as well as the principle of active substructure combination. The preliminary bioassay showed that compound **5j** showed 85% inhibition rate against *Pseudoperonospora cubensis* at 50 mg/L, most of the target compounds had good insecticidal activity against *Oriental armyworm* at 500 mg/L.

Synthesis and Biological Activities of Novel *N*-(2-(5-(3-Bromo-1-(3-chloropyridin-2-yl)-1*H*-pyrazol-5-yl)-1,3,4-oxadiazol-2-yl)-4-chloro-6-methylphenyl) Amides



Zhang, Yan; Shang, Junfeng; Li, Huan; Liu, Hang; Song, Haibin; Wang, Baolei*; Li, Zhengming*
Chin. J. Org. Chem. **2020**, 40(3), 787

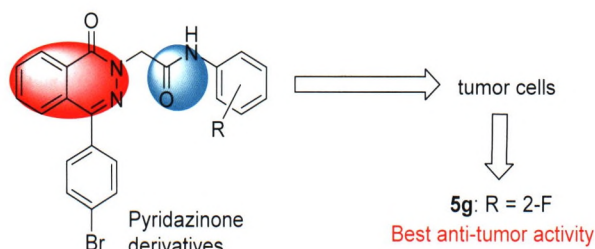
The synthesis, structure and insecticidal and fungicidal activities of novel *N*-(2-(5-(3-bromo-1-(3-chloropyridin-2-yl)-1*H*-pyrazol-5-yl)-1,3,4-oxadiazol-2-yl)-4-chloro-6-methylphenyl) amides were reported. The results provided significant reference information for new agrochemical innovations.

CONTENT

Synthesis and Antitumor Activity of Novel 1,3-Disubstituted Pyridazinone Derivatives

Zhang, Luye; Zhang, Yang; Bao, Chongnan; Yang, Peng; Li, Erdong; Meng, Yaqi; Cui, Fei; Zhou, Rui; Huang, Shiyu; Zheng, Jiaxin*; Shan, Lihong*; Liu, Hongmin*; Zhang, Qiurong*

Chin. J. Org. Chem. **2020**, 40(3), 794

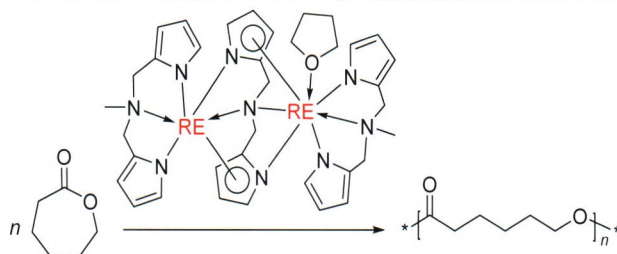


In order to find more efficient and low toxicity antitumor drugs, a series of novel 1,3-disubstituted pyridazinone derivatives were synthesized and evaluated for their antiproliferative activities against four human cancer cell lines (MCF-7, PC-3, SW-620 and HGC-27) *in vitro*.

Synthesis and Catalytic Activity of Dinuclear Rare-earth Metal Complexes with Nitrogen-containing Bridged Dipyrrolyl Ligand towards Ring-opening Polymerization of ϵ -Caprolactone

Wu, Jie; Zhang, Xiuli; Zhang, Lijun*; Wei, Yun; Zhou, Shuangliu*

Chin. J. Org. Chem. **2020**, 40(3), 801



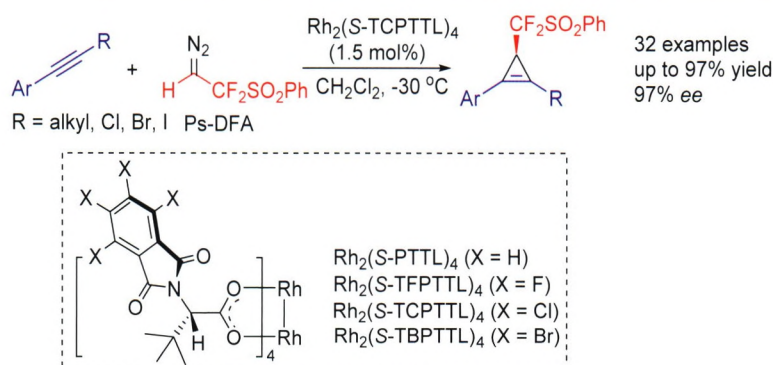
Nitrogen-containing bridged dipyrrolyl rare-earth metal complexes were synthesized and characterized. The complexes showed high catalytic activities on the ring-opening polymerization of ϵ -caprolactone.

HIGHLIGHTS

New Strategy for the Construction of Chiral Cyclopropenes: Enantioselective Synthesis of *Gem*-difluoromethylenated Three-Membered Carbocycles

Ouyang, Yao; Qing, Fengling*

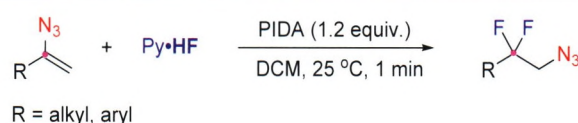
Chin. J. Org. Chem. **2020**, 40(3), 806



Synthesis of β -Difluoroalkyl Azides via 1,2-Azide Migration

Wu, Zhen; Zhu, Chen*

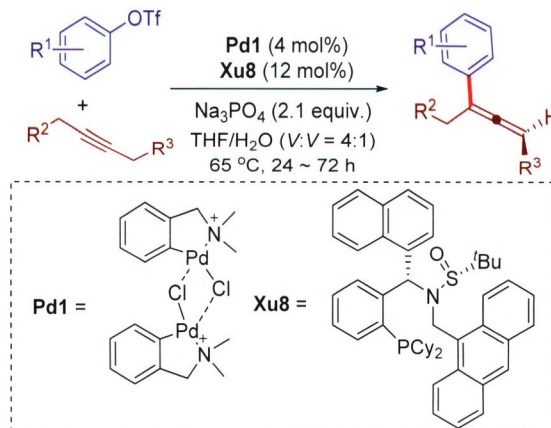
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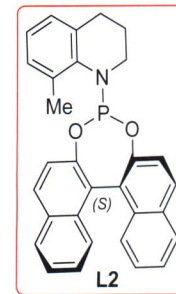
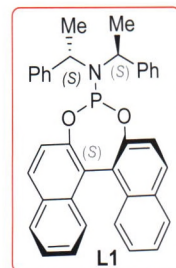
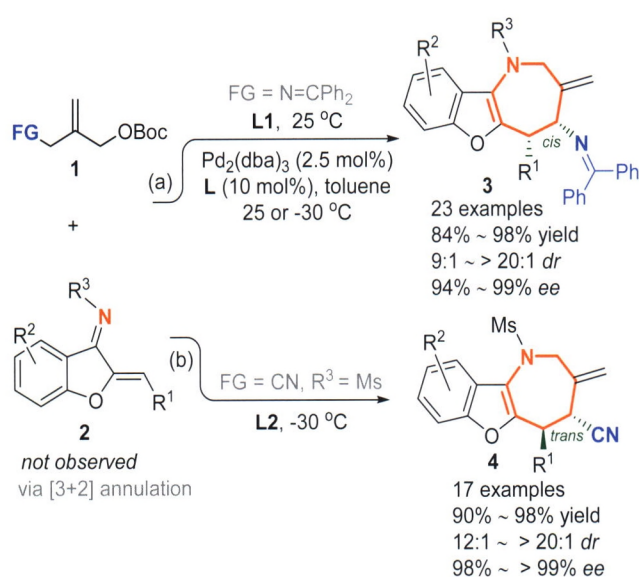
Palladium-Catalyzed Asymmetric Heck Coupling of Aryl Triflates and Alkynes

Cai, Yuan; Shi, Shi-Liang*

Chin. J. Org. Chem. **2020**, 40(3), 810

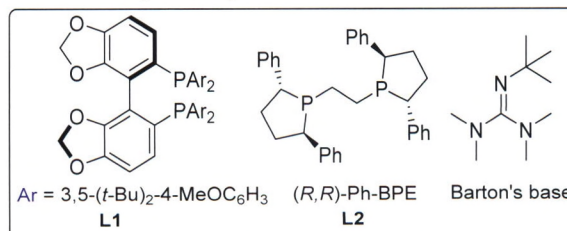
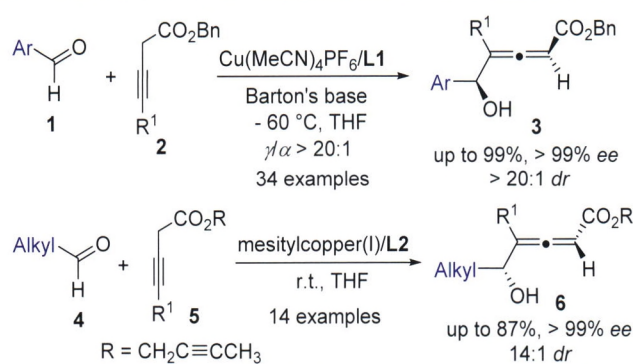


Pd-Catalyzed Asymmetric [3 + 4] Cycloaddition of Trimethylenemethane: Highly Regio-, Diastereo-, and Enantioselective Construction of Benzofuro[3,2-*b*]azepines



Wang, Meixin; Deng, Yuhua*; Shao, Zhihui*
Chin. J. Org. Chem. **2020**, 40(3), 812

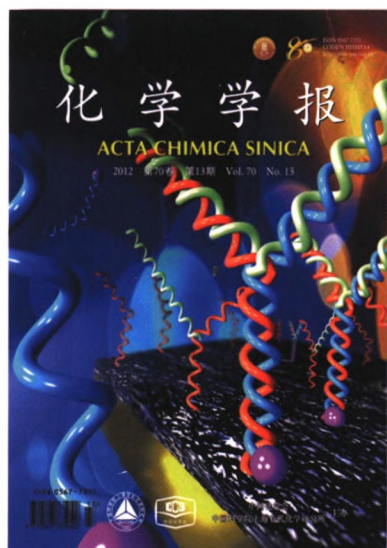
Construction of Chiral 2,3-Allenols through Copper(I)-Catalyzed Asymmetric Direct Alkynylogous Aldol Reaction



Ran, Guangyao; Chen, Yingchun*
Chin. J. Org. Chem. **2020**, 40(3), 814

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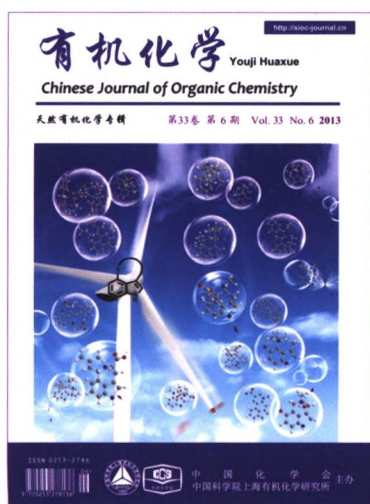
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