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有机化学

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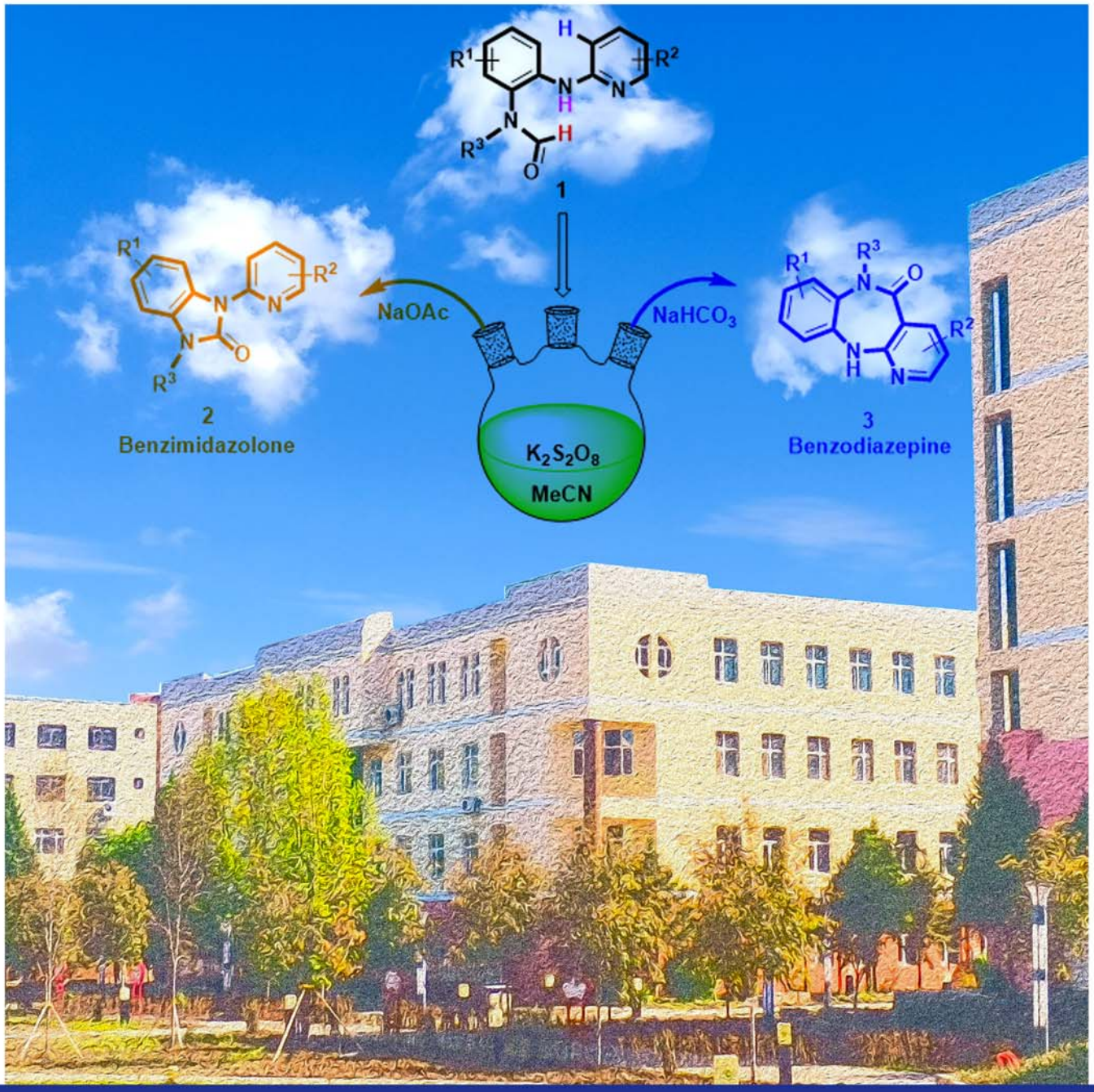

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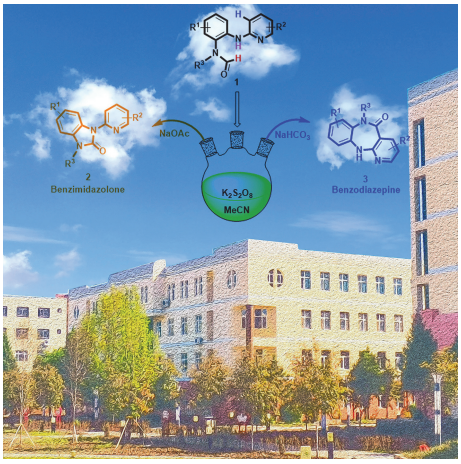
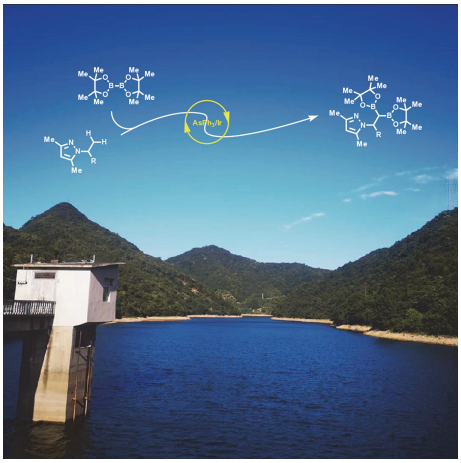
Cover Picture: Basicity-Tuned Selectivity: Synthesis of Benzimidazolone and Benzodiazepine from *N*-Alkyl-*N*-(2-(pyridin-2-ylamino)phenyl)formamides



A base-controlled strategy for the selective preparation of benzimidazolone and pyrido-benzodiazepine derivatives was developed by Wang, Guo, Tao, Liu, Zhao, Liu and Dai on page 1146. The combined use of $K_2S_2O_8$ and NaOAc as system promote the formation of benzimidazolones, however, benzodiazepines are obtained when system is switched to $K_2S_2O_8$ and $NaHCO_3$.

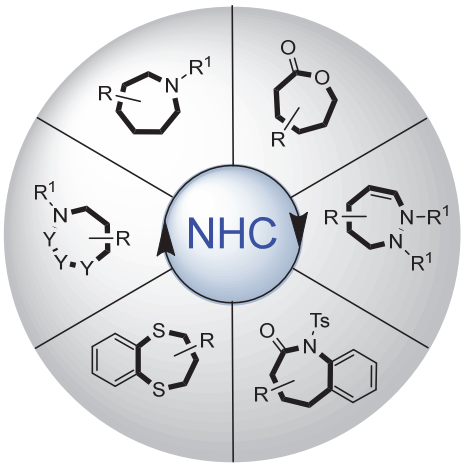
Inside Cover: Synthesis of 1,1-Diboron Alkanes via Diborylation of Unactivated Primary C(sp^3)—H Bonds Enabled by $AsPh_3$/Iridium Catalysis



A novel Iridium-catalyzed dual borylation of unactivated primary C(sp^3)—H bonds have been developed by Senmiao Xu and coworkers on page 1101. The current method could tolerate a variety of functional groups, affording corresponding 1,1-diboron alkanes in moderate to good yields. Finally, a gram-scale C—H borylation and diversifications of obtained borylated products were also demonstrated.

REVIEWS

Recent Advances for the Construction of Seven-Membered Ring Catalyzed by N-Heterocyclic Carbenes

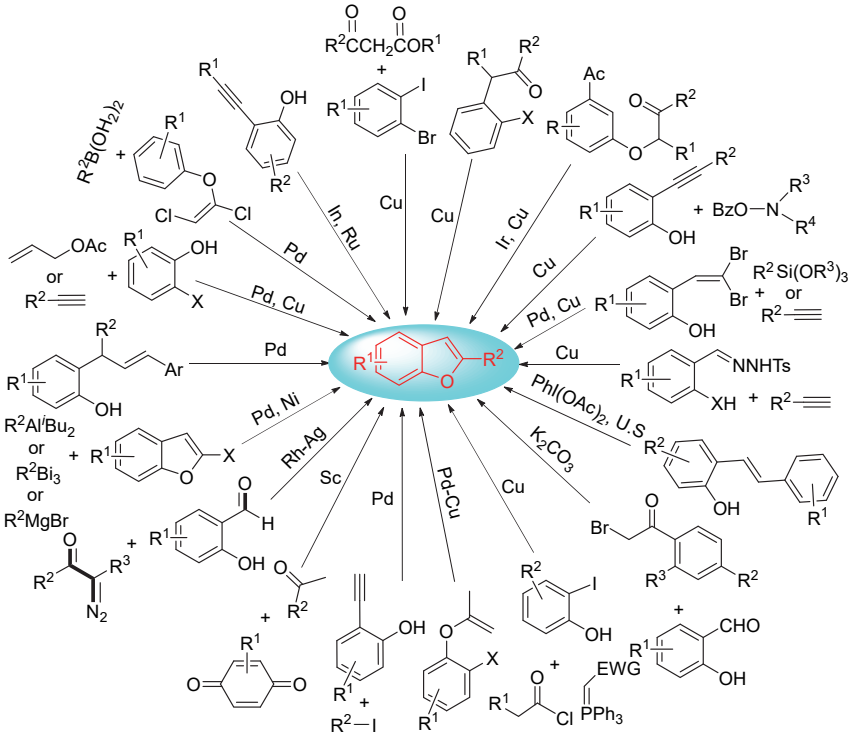


The seven-membered rings represent an important structural motif. The development of efficient strategies for the synthesis of seven-membered rings is very desirable. This review highlights the developments and new advances for the construction of seven-membered rings catalyzed by N-Heterocyclic carbene (NHC).

Yao, Ting; Li, Jiayan; Wang, Jiaming; Zhao, Changgui*
Chin. J. Org. Chem. **2022**, *42*(4), 925

CONTENT

Recent Progress in the Synthesis of Substituted Benzo[*b*]furan Derivatives

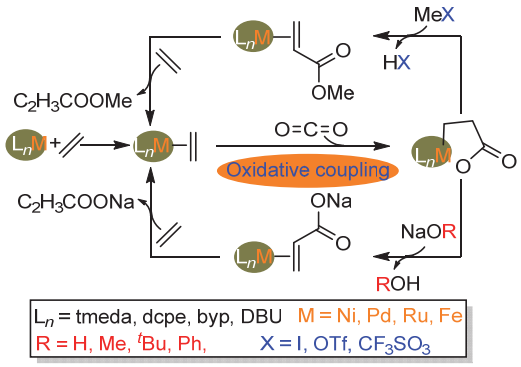


R^1 = H, Cl, Br, Me, MeO, CH_3CO, CN, NO_2, Ph; R^2, R^3, R^4 = alkyl, alkenyl, aryl; X = Cl, Br, I, O, NH, SMe; EWG = CO_2R, COAr, CN

The recent progress in the synthesis and applications of substituted benzo[*b*]furan is reviewed, involving various reaction systems. Finally, the future development and application of them are also prospected.

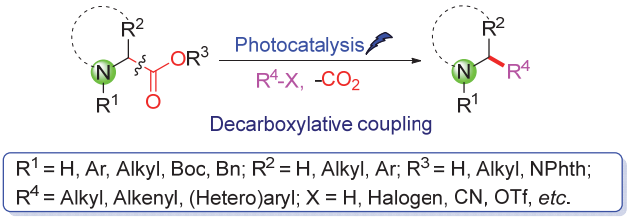
Zhang, Zhihao; Jiang, Xin; Li, Qinghan*
Chin. J. Org. Chem. **2022**, *42*(4), 945

Advances in the Production of Acrylic Acid and Its Derivatives by CO_2/C_2H_4 Coupling



The recent advances on transition metal complex catalyzed CO_2/C_2H_4 coupling reaction are summarized on the basis of catalyst activity and reaction mechanisms, including both the experiment and theoretical calculation. Furthermore, the challenges and perspectives in the catalytic transformation of CO_2/C_2H_4 are also discussed.

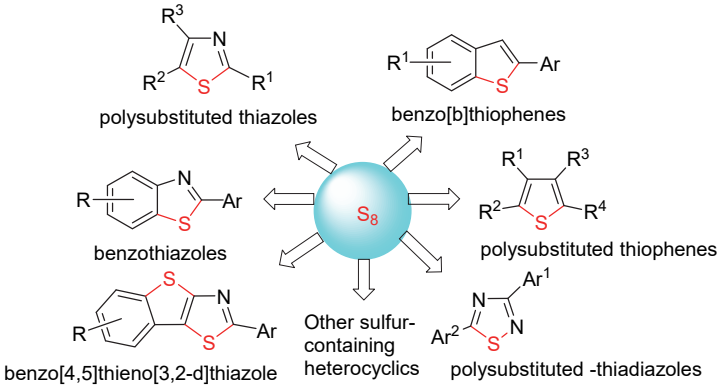
Zhu, Youcai; Ding, Xinxin; Sun, Li; Liu, Zhen*
Chin. J. Org. Chem. **2022**, *42*(4), 965



The use of visible-light to promote decarboxylative coupling of *α*-amino acids to construct various nitrogen-containing compounds has been considered as an attractive synthetic strategy. This review highlights the recent progress in photocatalytic decarboxylative coupling reactions of *α*-amino acid derivatives with various partners.

Hu, Jiayu; Zhu, Zhiqiang*; Xie, Zongbo; Le, Zhanggao*
Chin. J. Org. Chem. **2022**, *42*(4), 978

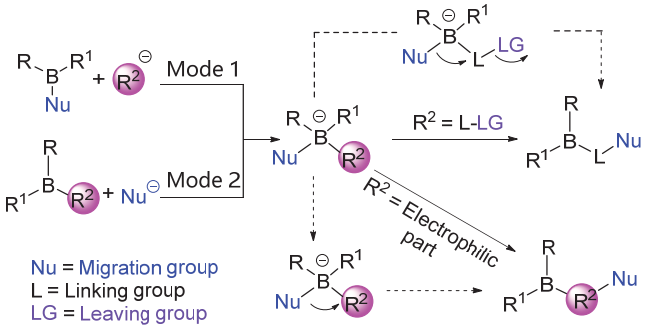
Elemental Sulfur: An Excellent Sulfur-Source for Synthesis of Sulfur-Containing Heterocyclics



Xiao, Liwei*; Liu, Guangxian; Ren, Ping; Wu, Tongtong; Lu, Yuwei; Kong, Jie
Chin. J. Org. Chem. **2022**, *42*(4), 1002

The synthesis of sulfur-containing heterocycles from elemental sulfur has become a research hotspot. The recent research progress of synthesis of sulfur-containing heterocyclic employing sulfur as sulfur-source is reviewed.

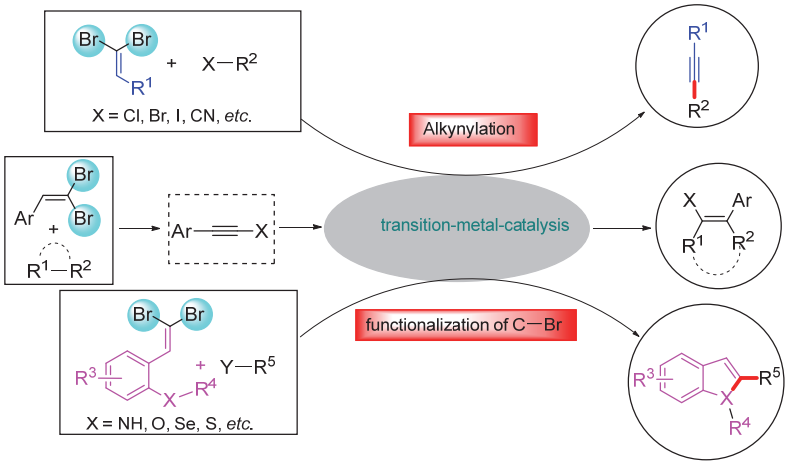
Recent Progress on 1,2-Metallate Shift Reactions Based on Tetracoordinate Boron Intermediates



Zhang, Feng; Zhou, Lu; Yang, Kai; Song, Qiuling*
Chin. J. Org. Chem. **2022**, *42*(4), 1013

In view of the versatile transformations, migration reactions of organoboron compounds have attracted great attention from chemists in recent years due to their high efficiency and mild reaction conditions, which are widely utilized for rapid constructions of carbon-carbon and carbon-heteroatom bonds. Recent progress on 1,2-migration reactions based on tetracoordinate boron intermediates is summarized according to various reaction conditions and bond formations.

Transition-Metal Catalyzed Coupling Reactions of *gem*-Dibromoviny Derivatives

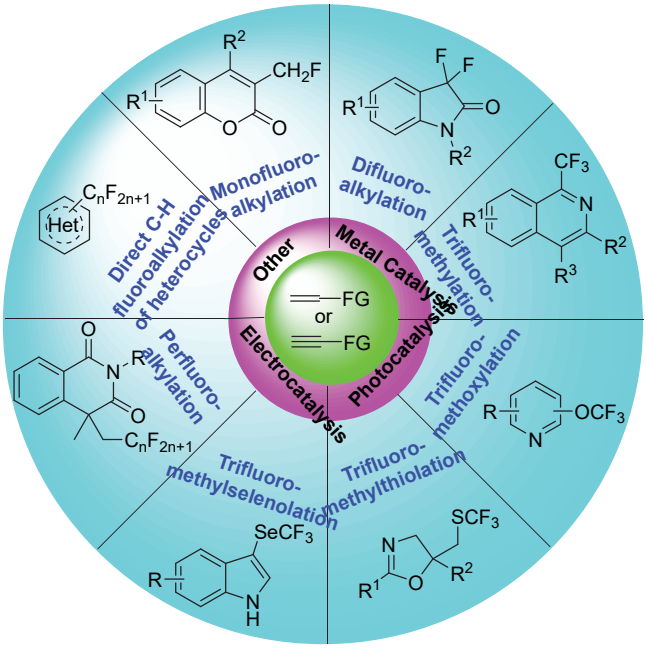


Liang, Luqi; Zhang, Lizhi*; Peng, Yongli*; Liu, Hui*
Chin. J. Org. Chem. **2022**, *42*(4), 1033

Through transition metal catalysis, *gem*-dibromovinyl derivatives can be transformed into benzo-fused heterocycles with important physiological activities. The coupling reactions involving transition-metal catalyzed *gem*-dibromovinyl derivatives are summarized and the reaction mechanisms for most transformations are discussed in detail.

CONTENT

Construction of Fluoro-containing Heterocycles Mediated by Free Radicals



Chen, Ning; Lei, Jia; Wang, Zhichuan; Liu, Yingjie*; Sun, Kai*; Tang, Shi
Chin. J. Org. Chem. **2022**, *42*(4), 1061

With the rapid development of transition metal catalysis, photocatalysis and electrocatalytic radical reactions, radical chemistry has shown endless potential in the field of organic synthesis. In this paper, monofluoroalkylation, difluoroalkylation, trifluoromethylation, trifluoroalkoxy/sulfur/selenylation, perfluoroalkylation of unsaturated hydrocarbons and direct C—H fluoroalkylation of heterocycles are discussed from the aspects of transition metal catalysis, photocatalysis and electrocatalysis.

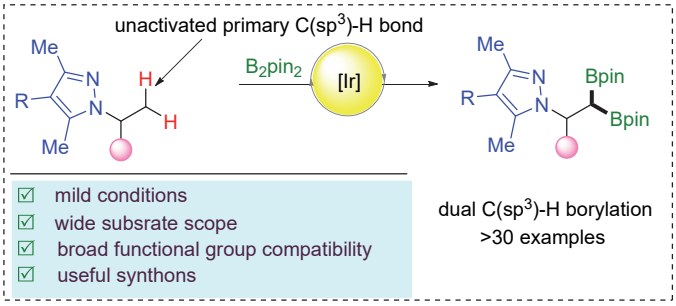
Recent Progress in the Synthesis of 2-Benzofuran-1(3*H*)-one

Gong, Tingting; Chen, Zhibin; Liu, Miaochang; Cheng, Jiang*
Chin. J. Org. Chem. **2022**, *42*(4), 1085

2-Benzofuran-1(3*H*)-ones (isobenzofuranons) are multi-function precursors toward a variety of cyclic compounds as isobenzofuranon structure is ubiquitous in many natural products. Thus, the preparation of isobenzofuranons has attracted great attention. Our and other research groups' work on the synthesis of isobenzofuranone is summarized, which is divided into three aspects, transition metal catalytic synthesis, acid-base catalytic synthesis and other synthesis methods.

ARTICLES

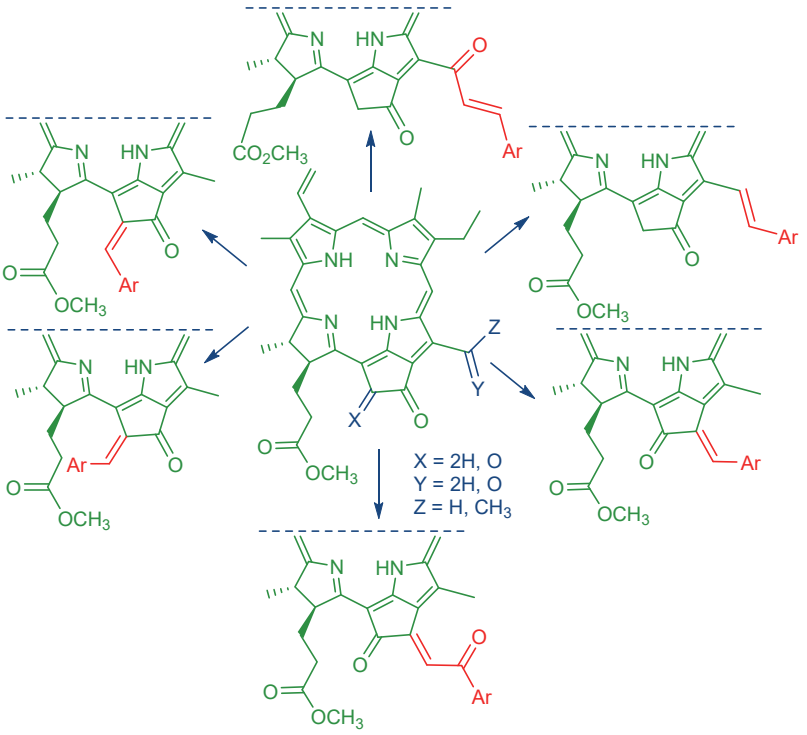
Synthesis of 1,1-Diboron Alkanes via Diborylation of Unactivated Primary C(sp^3)—H Bonds Enabled by AsPh$_3$/Iridium Catalysis



Liu, Wenqi; Shen, Zhenlu; Xu, Senmiao*
Chin. J. Org. Chem. **2022**, *42*(4), 1101

The AsPh$_3$/iridium catalyzed diborylation of unactivated primary C(sp^3)—H bonds using pyrazoles as directing groups was disclosed. This method could tolerate a variety of functional groups, affording a vast array of 1,1-diboron alkanes in moderate to good yields. The synthetic utility of the current method on a gram-scale reaction for further functionalization was also demonstrated.

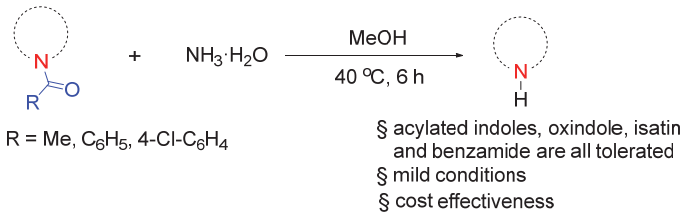
Regio- and Stereoselective Aryl(aroyl)-methylenenation of Pyropheophorbide and Syntheses of Chlorophyllous Chlorin Derivatives



Based on the cross-aldol condensation, the regio- and stereoselective aryl(aroyl)methylenations of pyropheophorbide-*a* were accomplished on the exocyclic ring and at 12-position. A series of unreported chlorin derivatives containing the aryl(aroyl)ketene unit were synthesized. The formation mechanisms, the stereoisomerism and the optical properties of the aryl(aroyl)methylenated products were discussed.

Gao, Na; Chu, Xiaohui; Liu, Yang; Li, Jiazhu*; Wang, Jinjun*
Chin. J. Org. Chem. **2022**, *42*(4), 1111

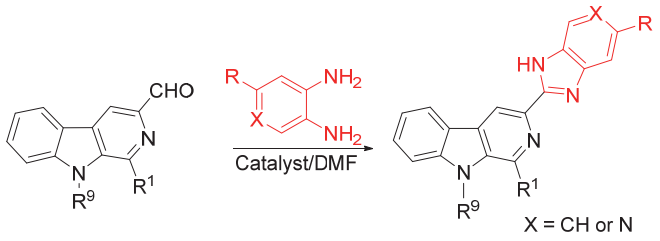
A Practical Transamidation Strategy for the N-Deacylation of Amides



A general solution to the N-deacylation of amides with $NH_3 \cdot H_2O$ under mild and scalable conditions was reported. A range of drugs and drug derivatives could be deacylated to release free amines in excellent yields. The good functional group compatibility, combined with operational simplicity, excellent yield and cost effectiveness of all reagents, makes this protocol a prime candidate for N-deacylation of amides.

Han, Qun; Xu, Kun*; Tian, Faning; Huang, Shengyang*; Zeng, Chengchu*
Chin. J. Org. Chem. **2022**, *42*(4), 1123

Design, Synthesis, and Antitumor Activity of *β*-Carboline-Benzimidazole Hybrids

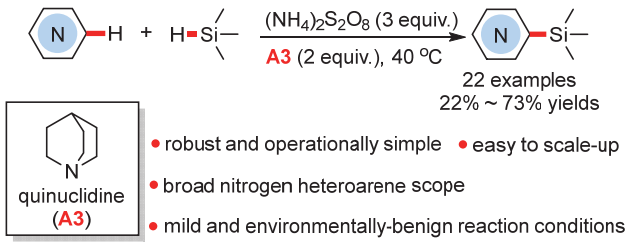


In a continuing effort to develop novel *β*-carbolines endowed with better pharmacological profiles, a series of 1,9-disubstituted *β*-carboline-benzimidazole hybrids with various substituents were designed and synthesized from the starting material *L*-tryptophan and aldehydes.

Zhu, Siyu; Huo, Xinyu; Ma, Qin; Chen, Wei; Zhang, Jie*; Guo, Liang*
Chin. J. Org. Chem. **2022**, *42*(4), 1129

CONTENT

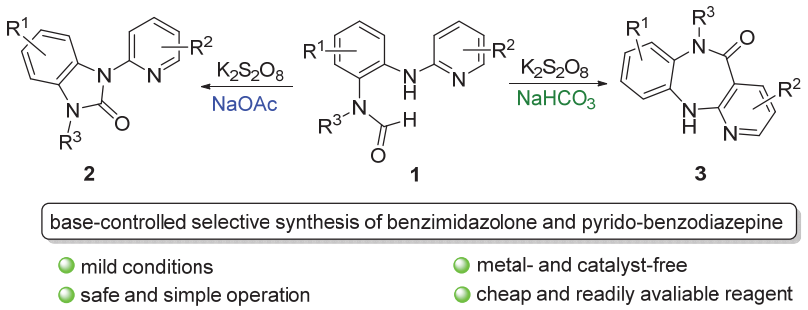
Research of Quinuclidine-Promoted C—H silylation of Electron-Deficient Nitrogen Heteroarenes



Pan, Peng; Yuan, Qiyang; Liu, Shihui; Zhao, Jianhong*; Zhang, Yongqiang*
Chin. J. Org. Chem. **2022**, *42*(4), 1136

A novel quinuclidine-promoted C—H silylation reaction using ammonium persulfate as the oxidant and hydrosilane as the silyl sourse has been reported.

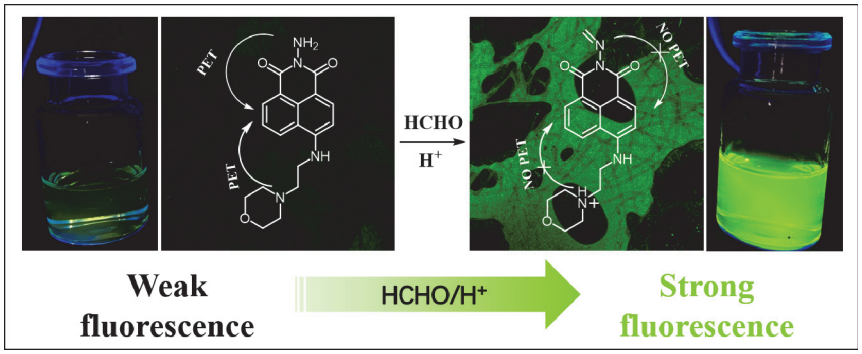
Basicity-Tuned Selectivity: Synthesis of Benzimidazolone and Benzodiazepine from *N*-Alkyl-*N*-(2-(pyridin-2-ylamino)-phenyl)formamides



Wang, Yubin; Guo, Cheng; Tao, Sheng; Liu, Jichang; Zhao, Jigang; Liu, Ning*; Dai, Bin*
Chin. J. Org. Chem. **2022**, *42*(4), 1146

A base-controlled strategy for the selective synthesis of benzimidazolone and pyrido-benzodiazepine derivatives was developed.

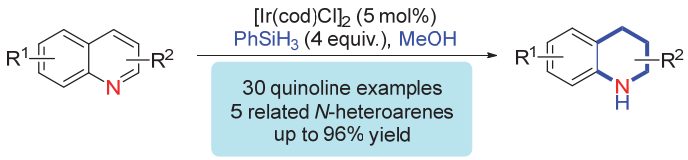
Synthesis and Study of Performance for An Enhanced Formaldehyde Fluorescent Probe



Lu, Huixu; Tang, Yonghe; Zhou, Hongmei; Lin, Weiying*
Chin. J. Org. Chem. **2022**, *42*(4), 1163

The new highly selective fluorescence probe **HSU-FA** was designed and synthesized for monitoring formaldehyde (FA) in the solution and air.

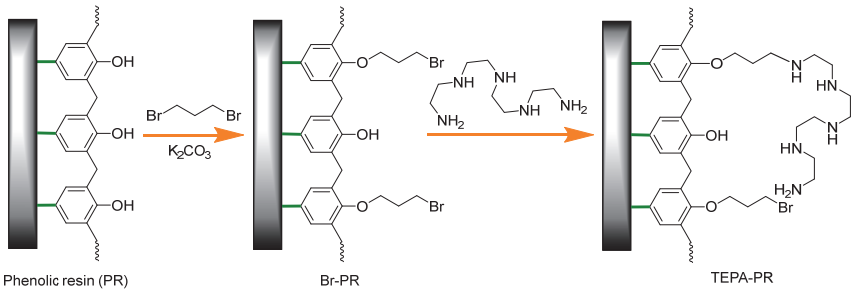
Hydrosilanes as Hydrogen Source: Iridium-Catalyzed Hydrogenation of *N*-Heteroarenes



Zhang, Miaomiao; Han, Bo*; Ma, Haojie; Zhao, Liang; Wang, Jijiang; Zhang, Yuqi*
Chin. J. Org. Chem. **2022**, *42*(4), 1170

A catalytic reduction of *N*-heteroarenes employing low-cost and air-stable hydrosilane was demonstrated under mild conditions. This reaction is scalable and tolerable for sensitive functional groups, such as bromide, chloride, fluoride, ester, carboxylic acid, cyanogroup and nitro groups. This catalytic system provides a convenient, environmentally friendly and pragmatic method to obtain a variety of 1,2,3,4-tetrahydroquinoline derivatives under mild reaction conditions.

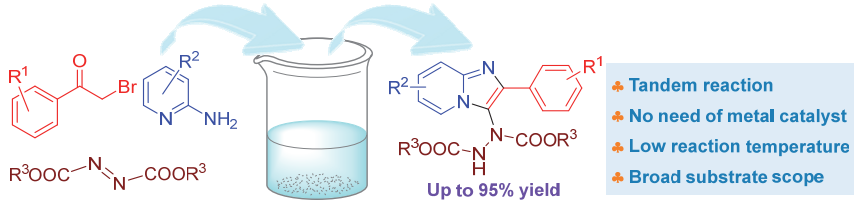
Tetraethylenepentamine Functionalized Phenolic Resin as Highly Active Acid-Base Bifunctional Catalyst for Knoevenagel Condensation Reaction



The tetraethylenepentamine functionalized phenolic resin (TEPA-PR) for Knoevenagel condensation reaction has been prepared by aminating a commercially available phenolic resin (PR) and tetraethylenepentamine. The TEPA-PR can efficiently catalyze Knoevenagel condensation reaction of aromatic aldehydes with high yields of a wide range of products.

Xiao, Jian; Wu, Zhiying; Chen, Ziyi; Zhao, Pengfei; Liu, Chunyan*
Chin. J. Org. Chem. **2022**, *42*(4), 1179

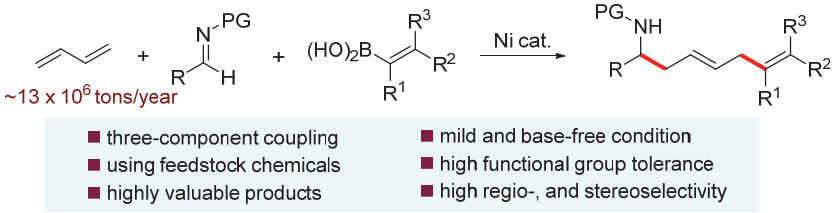
An Efficient Three-Component Tandem Approach for the Synthesis of Imidazoheterocycle-Hydrazine Derivatives under Mild Conditions



An three-component tandem reaction for the synthesis of imidazo[1,2-*a*]pyridine-hydrazines was accomplished with the easily available formyl methyl bromides, pyridin-2-amines and azodiformates. The approach features simple operation, mild conditions (transition-metal-free and low reaction temperature) as well as good tolerance of substrates.

Qiao, Huijie*; Yang, Liting; Chen, Ya; Wang, Jialin; Sun, Wuxuan; Dong, Haobo; Wang, Yunwei
Chin. J. Org. Chem. **2022**, *42*(4), 1188

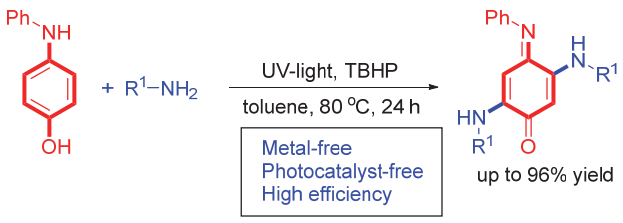
Ni-Catalyzed Three-Component Coupling Reaction of Butadiene, Aldimines and Alkenylboronic Acids



A highly regio- and *trans*-selective Ni-catalyzed three-component coupling reaction of alkenylboronic acids, aldimines, and 1,3-butadiene is reported. This reaction represents the first general synthetic method to homoallylic amines bearing a skipped diene fragment and a rare example for modular 1,4-dicarbofunctionalization of 1,3-butadiene, an abundant feedstock chemical.

Zhang, Yurong; Wang, Han*; Mao, Yongjun; Shi, Shiliang*
Chin. J. Org. Chem. **2022**, *42*(4), 1198

Construction of Diaminobenzoquinone Imines through Radical Coupling of Aminophenols with Amine Under UV-Light

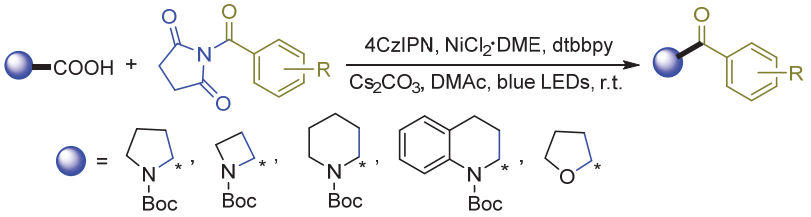


A metal-free radical coupling reaction of aminophenols with amines under UV-light has been successfully developed to synthesize diaminobenzoquinone imines in high yields, in which the commercially available and cheap *t*-butyl hydroperoxide (TBHP) was used as an oxidant. This protocol provides an easy and green approach for the construction of benzoquinone imines with potential pharmaceutical interest avoiding metal catalyst or photocatalyst.

Xu, Limei; Lu, Linyan; Cai, Jinzhong; Feng, Yadong*; Cui, Xiuling*
Chin. J. Org. Chem. **2022**, *42*(4), 1210

CONTENT

Fluorescent Dye/Nickel Synergistic Catalytic Decarboxylative Carbonylation Reaction



The recent progress in the synthesis of α-amino ketone-containing derivatives is reviewed. The process and mechanism of fluorescent dye/nickel synergistic catalytic decarboxylative carbonylation reaction applied to prepare α-amino ketone-containing derivatives are mainly discussed. Factors affecting the aforementioned synergistic reaction are also explored. Finally, the future application of the synthetic method is prospected.

Yu, Weiguo*; Wang, Lingna; Yu, Xiaocong; Luo, Shuping*
Chin. J. Org. Chem. **2022**, *42*(4), 1216

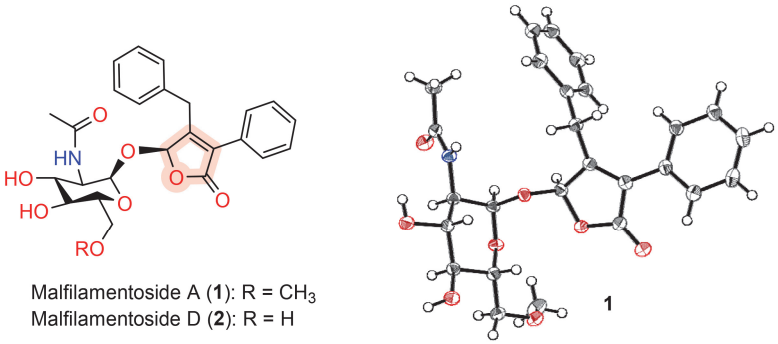
NOTES

Discovery of a New Polycyclic Tetramate Macrolactam Clifednamide K

A new polycyclic tetramate macrolactam clifednamide K (**2**) was isolated from the recombinant strain SR111-*cbmA-OX1-cftC*. The antibacterial activity of compounds **1** and **2** was tested by filter paper disc diffusion assay.

Luo, Jie; Yan, Yaqian; Wang, Haoxin; Li, Yaoyao*
Chin. J. Org. Chem. **2022**, *42*(4), 1224

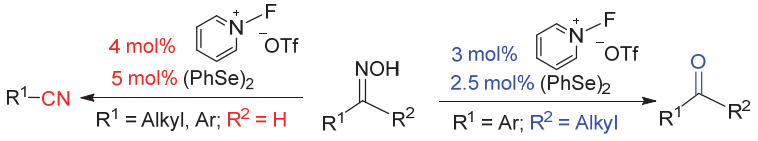
Configurational Assignment of Malfilamentoside A and a New Furanone Glycoside Malfilamentoside D



Two aromatic furanone glycoside malfilamentosides A (**1**) and D (**2**), were identified. The planar structures of malfilamentosides A (**1**) and D (**2**) were elucidated. The absolute configuration of malfilamentoside A (**1**) was unambiguously resolved for the first time by a single crystal X-ray diffraction analysis and the stereochemistry of malfilamentoside D (**2**) was determined by comparing electronic circular dichroism (ECD) spectra of **1** and **2**.

Xu, Huixin; Wang, Lu; Zhang, Liping; Liu, Wei; Zhang, Qingbo; Zhang, Haibo; Zhang, Changsheng; Zhang, Wenjun*
Chin. J. Org. Chem. **2022**, *42*(4), 1229

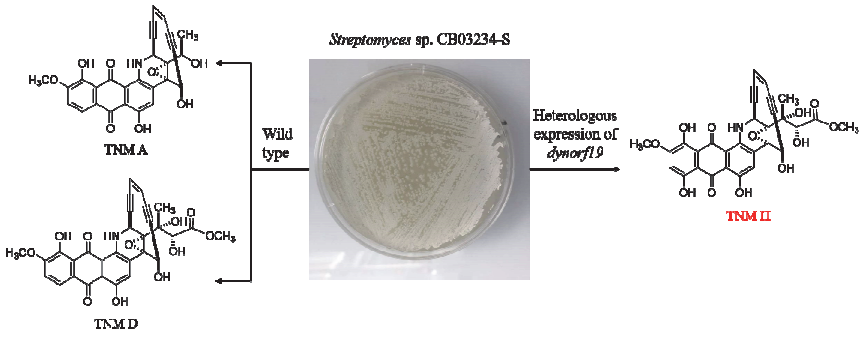
Organoselenium-Catalyzed Conversion of Oximes to Nitriles or Ketones



Phenylselenenic acid and phenylseleninic acid were formed *in situ* in the rection of diphenyl diselenide with 1-fluoropyridine trifluoromethane sulfonate (FP-OTf) in air, which catalyzed the conversions of aldoximes or ketoximes to corresponding nitriles or ketones, respectively. The proper ratios of diphenyl diselenide to FP-OTf were different in two reactions. The yields of nitriles or ketones were 57%～94%.

Wang, Liming; Li, Ke; Zhang, Wanxuan*
Chin. J. Org. Chem. **2022**, *42*(4), 1235

Discovery of Tiancimycin Congeners from *Streptomyces* sp. CB03234-S

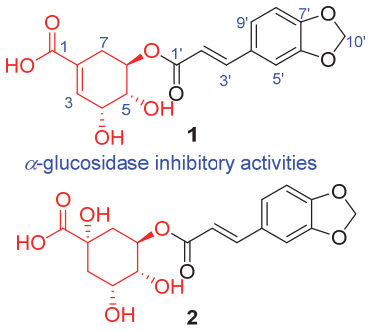


Xue, Lu; Zhang, Lihua; Zhang, Chengyu; Zhao, Xin; Dang, Weifan; Wang, Zhaoxin; Wang, Chunhua; Suo, Tongchuan; Yan, Xiaohui*

Chin. J. Org. Chem. **2022**, *42*(4), 1241

A new enediyne, tiancimycin H, was discovered by heterologous expression of two cytochrome P450 hydroxylase genes from the dynemicin pathway in the tiancimycins A and D high-producing strain, *Streptomyces* sp. CB03234-S.

Two New Shikimic Acid Derivatives from the Flower of *Trachycarpus fortunei*

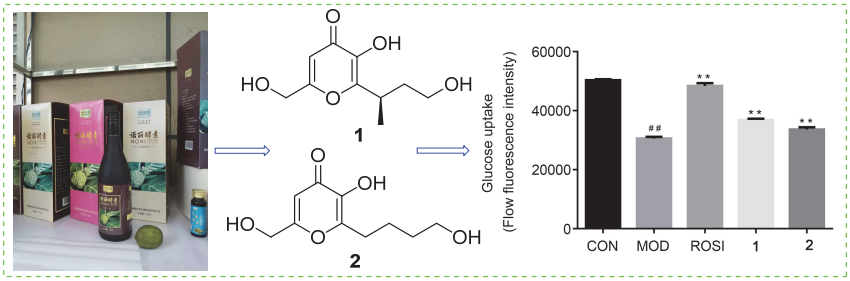


Lin, Fangxia; Wang, Zhihong; Dai, Decai; Zhou, Xueming*; Wu, Luyong*

Chin. J. Org. Chem. **2022**, *42*(4), 1248

Two new shikimic acid-derived, trachshikimics A and B, together with four known biogenetically related shikimic acid derivatives, were isolated from the flower of *Trachycarpus fortunei* (Hook.) H. Wendl. Their structures were elucidated using comprehensive spectroscopic methods and chemical reaction.

Two New Kojic Acids from *Noni* Juice with Glucose Uptake Activity in Insulin-Resistant HepG2 Cells



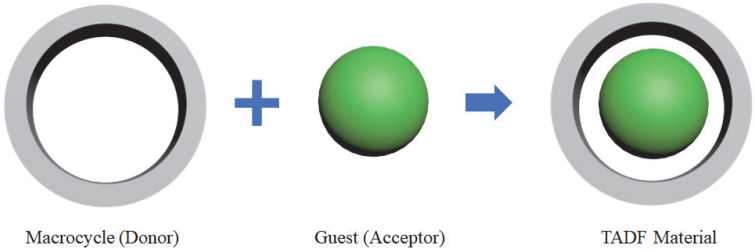
Zhang, Xuguang; Zhang, Bin; Zhou, Xueming; Yu, Zhangxin; Li, Xiaobao; Chen, Guangying*

Chin. J. Org. Chem. **2022**, *42*(4), 1252

Two new kojic acids, nonikojics A and B (**1** and **2**), were obtained from the water soluble extract of the *Noni* juice, and their chemical structures were elucidated by comprehensive modern spectroscopic techniques. Furthermore, two new compounds significantly increased glucose uptake with doses of 50 μmol/L and thus improve insulin resistance.

HIGHLIGHTS

Thermally Activated Delayed Fluorescence Materials Based on Cocrystals of Supramolecular Host and Guest

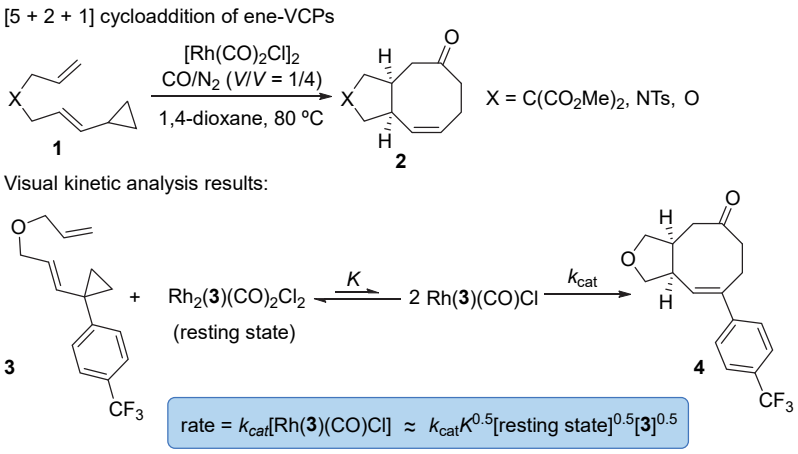


Shi, Qiang; Wang, Leyong*

Chin. J. Org. Chem. **2022**, *42*(4), 1256

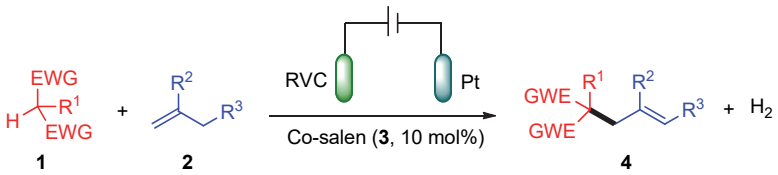
CONTENT

Visual Kinetic Analysis and Quantum Chemical Calculations Uncover the Mechanistic Insights into Rh-Catalyzed [5+2+1] Cycloaddition



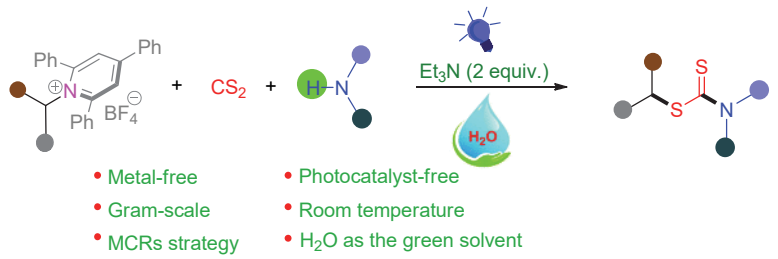
Lan, Yu; Qi, Xiaotian*
Chin. J. Org. Chem. **2022**, *42*(4), 1258

Cobalta-Electrocatalyzed Allylic C—H Alkylation



Wang, Huiqiao; Xu, Kun*
Chin. J. Org. Chem. **2022**, *42*(4), 1260

Three-Component Reaction Access to *S*-Alkyl Dithiocarbamates under Visible-Light Irradiation Conditions in Water



Yuan, Jinwei*; Yang, Liangru
Chin. J. Org. Chem. **2022**, *42*(4), 1262