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SCI 收录引用检索证明报告

姓　名	王沐荣（WANG MR）	电　话	13598609350
单　位	新乡医学院		
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Palladium-catalyzed allylic allenylation of homoallyl alcohols with propargylic carbonates†

Ping-Xin Zhou, *[a] Murong Wang,‡[a] Xiang Li,‡[a] Xueyan Du,[a] Xiaozhe Yang,[a] Han Wang,[a] Tangqiang Sun,*[c] Feng Ren*[a,b] and Yong-Min Liang [d]

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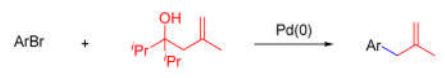
A palladium-catalyzed reaction of homoallyl alcohols with propargylic carbonates was realized *via* oxidative addition/decarboxylation/retro-allylation. The reaction proceeds with a wide substrate scope by using readily available starting materials, which provides a new and efficient method to synthesize a series of tri- and tetra-substituted allyl allenes.

Introduction

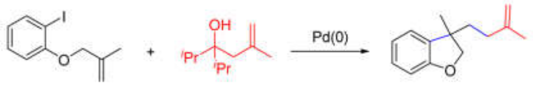
Transition metal-catalyzed organic reactions involving the cleavage of C–C single bonds have attracted much attention as these transformations offer unique opportunities to quickly synthesize complex molecules through an intriguing and straightforward strategy.[1] Over the past decades, efficient approaches for the cleavage of C–C single bonds have been elegantly developed.[2] Remarkably, in 2006, Yorimitsu and Oshima reported a unique and interesting strategy for allylation of aryl halides with homoallyl alcohols as the allylation reagents (Scheme 1a).[3] Because this C–C bond cleavage proceeds *via* a favorable six-membered chairlike transition state, the regio- and stereochemical information on the homoallyl alcohols can be transferred onto the products, which is difficult to achieve by the palladium-catalyzed reaction of aryl halides with allylic metal reagents. This fact greatly enhances the synthetic utility of homoallyl alcohol derivatives as appropriate equivalents of well-defined allylic metal reagents. Recently, our group developed a palladium-catalyzed Heck cyclization/allylation with homoallyl alcohols *via* retro-allylation (Scheme 1b).[4] We further developed a palladium/norbornene-catalyzed *ortho*-amination/allylation of aryl iodides for the construction of *ortho*-aminated allylbenzene by using homoallyl alcohols as the allylation reagent.[5] Although considerable advances of the palladium-catalyzed retro-allylation with homoallyl alcohols, this well-established protocol is restricted to allylic arylation and allylic alkylation.

On the other hand, allene moieties are found in many natural products and bioactive molecules.[6] Furthermore, allenes can undergo diverse transformations, enabling a rapid molecular complexity increase in a single reaction step.[7] Thus, the development of efficient methods for the synthesis of allenes has attracted considerable attention over the past decades.[8] The palladium-catalyzed coupling reaction of propargylic alcohol derivatives with organometallic reagents or nucleophiles are one of the most powerful and straightforward protocols for the construction of functionalized allenes, which was extensively studied by Ma and others groups. Up to now, the syntheses of aryl allenes,[9] allenenes,[10] alkyl allenes,[11] allenylboronate,[12] cyanoallenes,[13] allenylphosphonates[14] and allenynes[15] have been achieved by this strategy. Although considerable progress has been achieved in palladium-catalyzed coupling reaction of propargylic alcohol derivatives with nucleophiles, it is also highly desirable to introduce newly

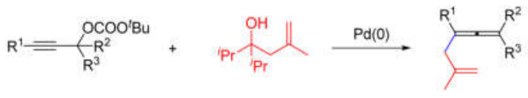
(a) Palladium-catalyzed allylic arylation of homoallyl alcohols



(b) Our previous work:Palladium-catalyzed allylic alkylation of homoallyl alcohols



(c) This work: Palladium-catalyzed allylic allenylation of homoallyl alcohols



Scheme 1 Palladium-catalyzed reaction of homoallyl alcohols.

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[a] *School of Basic Medical Sciences, Xinxiang Medical University, Xinxiang, China. E-mail: zhoupingxin518@163.com, 1215377283@qq.com*

[b] *Henan International Joint Laboratory of Immunity and Targeted Therapy for liver-intestinal Tumors, Xinxiang Medical University, Xinxiang, China*

[c] *College of Pharmacy, Xinxiang Medical University, Xinxiang, China*

[d] *State Key Laboratory of Applied Organic Chemistry, Lanzhou University, Lanzhou, China*

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‡ These authors contributed equally to this work.