



Cite this: *Chem. Commun.*, 2017, 53, 7553

Received 25th May 2017,
Accepted 13th June 2017

DOI: 10.1039/c7cc04022a

rsc.li/chemcomm

Pd(0)-Catalyzed intramolecular arylative dearomatization of β -naphthols†

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An efficient Pd(0)-catalyzed intramolecular arylative dearomatization of β -naphthols is described. Using Q-Phos as a ligand, the arylative dearomatization reaction proceeded smoothly affording excellent yields and chemoselectivity even when the catalyst loading was reduced to 0.1 mol%. This method offers an efficient access to a series of structurally diverse spirocarbocycles. Preliminary investigation indicates that an enantioselective reaction is feasible in the presence of a chiral phosphoramidite ligand.

Spirocarbocycles often exist in diverse natural products, biologically active molecules¹ and useful chemical structures² (Fig. 1). Therefore, it has attracted the intense attention of organic chemists to synthesize the unique structure of spirocarbocycles. Due to the congested quaternary carbon centers existed in spirocarbocycles, it has been a long-standing challenge to develop convenient synthetic methodologies. The recent progress in organometallic catalysis provides new solutions for the synthesis.³ However, highly efficient synthetic routes to obtain structurally diverse spirocarbocycles from readily available starting materials are still in great demand.

Phenol and its derivatives are important chemical starting materials and are widely used in organic synthesis.⁴ The recent development on the dearomatization of phenol and derivatives provides a novel route for the construction of spirocarbocycles.^{5,6} Of particular note, Pd-catalyzed cross-coupling type dearomatization reactions of anilines,⁷ phenols,⁸ indoles,⁹ pyrroles,¹⁰ pyridines¹¹ and furans¹² have been reported to construct interesting and useful structures. We envisaged that through an intramolecular design, spirocarbocycles can be easily obtained *via* the Pd-catalyzed arylative dearomatization of phenol derivatives. However, the chemoselectivity between the *O*-arylation and *C*-arylation reaction pathways is of great challenge in Pd-catalyzed dearomatization reactions especially when a quaternary carbon

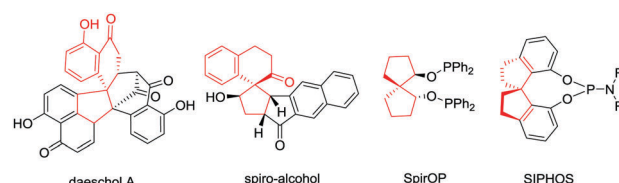
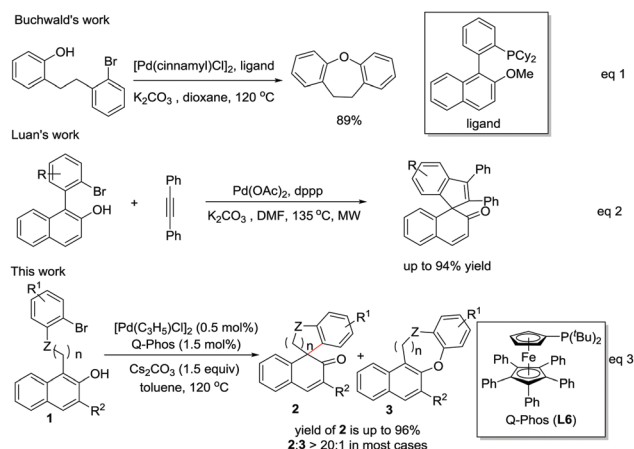


Fig. 1 Biologically active spirocarbocycle-based compounds and ligands.



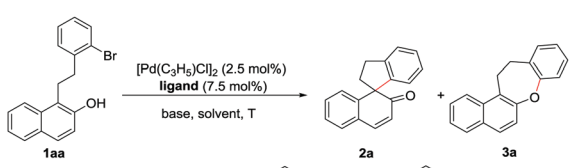
Scheme 1 Pd-catalyzed arylation of phenols and naphthols.

center is needed to be formed at the *ortho*-position of the phenolic hydroxyl group. For instance, the group of Buchwald reported that only the *O*-arylation product is obtained when an *ortho*-substituted phenol is subjected to Pd(0) catalysis (Scheme 1, eqn (1)).^{8b} Recently, Luan and co-workers^{8e} reported an elegant microwave-assisted Pd(0)-catalyzed alkyne migratory insertion and β -naphthol dearomatization cascade process, in which excellent chemoselectivity was achieved (Scheme 1, eqn (2)). However, to our knowledge, Pd(0)-catalyzed intramolecular direct arylative dearomatization of β -naphthols has not been reported. Herein, we describe an efficient Pd(0)-catalyzed intramolecular arylative dearomatization of β -naphthols affording all-carbon

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† Electronic supplementary information (ESI) available. CCDC 1534422. For ESI and crystallographic data in CIF or other electronic format see DOI: 10.1039/c7cc04022a



Table 1 Optimization of the reaction conditions^a


Entry	Ligand	Base	Solvent	T (°C)	Time (h)	2a yield ^b (%)	2a : 3a ^c
1	L1	K ₂ CO ₃	Toluene	120	8	0	ND
2	PPh ₃	K ₂ CO ₃	Toluene	120	8	24	> 20 : 1
3	L2	K ₂ CO ₃	Toluene	120	8	11 ^d	55 : 45
4	L3	K ₂ CO ₃	Toluene	120	8	0	< 20 : 1
5	L4	K ₂ CO ₃	Toluene	120	8	56 ^d	> 20 : 1
6	L5	K ₂ CO ₃	Toluene	120	8	56 ^d	> 20 : 1
7	L6	K ₂ CO ₃	Toluene	120	8	68 ^d	4 : 1
8	L6	Na ₂ CO ₃	Toluene	120	8	Trace	ND
9	L6	K ₃ PO ₄	Toluene	120	8	64	19 : 1
10	L6	DBU	Toluene	120	10	13	ND
11	L6	Cs ₂ CO ₃	Toluene	120	1	91 ^d	19 : 1
12 ^e	L6	Cs ₂ CO ₃	Toluene	120	5	95 ^d	> 20 : 1
13 ^f	L6	Cs ₂ CO ₃	Toluene	120	31	95 ^d	> 20 : 1
14	L6	Cs ₂ CO ₃	Toluene	40	7	92 ^d	> 20 : 1
15	L6	Cs ₂ CO ₃	DCM	40	7	93 ^d	> 20 : 1
16	L6	Cs ₂ CO ₃	THF	40	7	92	> 20 : 1
17	L6	Cs ₂ CO ₃	CH ₃ CN	40	7	20	ND
18	L6	Cs ₂ CO ₃	Dioxane	40	7	95	> 20 : 1

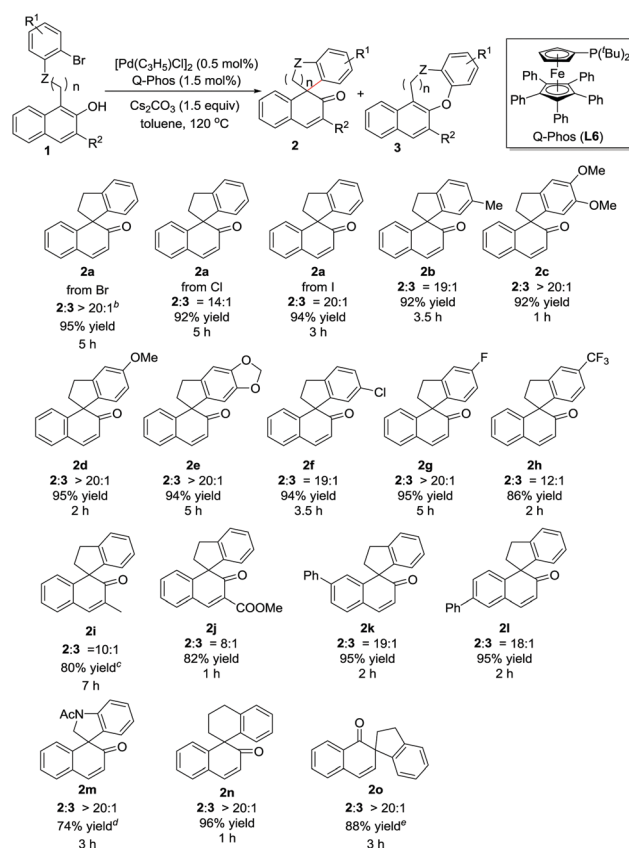
^a Reaction conditions: **1aa** (0.2 mmol), [Pd(C₃H₅)Cl]₂ (0.005 mmol), ligand (0.015 mmol), base (0.3 mmol) in solvent (1.0 mL), T °C. ^b Determined by ¹H NMR using CH₂Br₂ (0.2 mmol) as an internal standard. ^c Determined by ¹H NMR of the crude products. ^d Isolated yield. ^e [Pd(C₃H₅)Cl]₂ (0.5 mol%) and **L6** (1.5 mol%) were used. ^f [Pd(C₃H₅)Cl]₂ (0.1 mol%) and **L6** (0.3 mol%) were used.

spirocarbocycle structures in excellent yields and chemoselectivity (Scheme 1, eqn (3)).

Initially, 1-(2-bromophenethyl)-2-naphthol (**1aa**) was chosen as a model substrate to examine different ligands under palladium catalysis. The results are summarized in Table 1. No desired dearomatized product was obtained when the diphosphine ligand dppf (**L1**) was used (Table 1, entry 1), while the utilization of PPh₃ led to the formation of the desired dearomatized product (**2a**) in 24% NMR yield (Table 1, entry 2). With the Buchwald-type ligand RuPhos (**L2**), **2a** was isolated in 11% yield together with the isolation of the *O*-arylation product in 9% yield (Table 1, entry 3). When the sterically bulky (di-*t*-Bu)XPhos (**L3**) was used as a ligand, only the *O*-arylation product was obtained (Table 1, entry 4). The utilization of the *rac*-Feringa ligand (**L4**) or SIPr-HBF₄ (**L5**) led to the exclusive formation of **2a** (56% yield, Table 1, entries 5 and 6). With Q-Phos (**L6**) as a ligand, **2a** could be obtained in 68% yield with **3a** isolated in 17% yield. Subsequently, several bases were examined (Table 1, entries 8–11). To our delight, the stronger inorganic base Cs₂CO₃ almost exclusively gave the desired

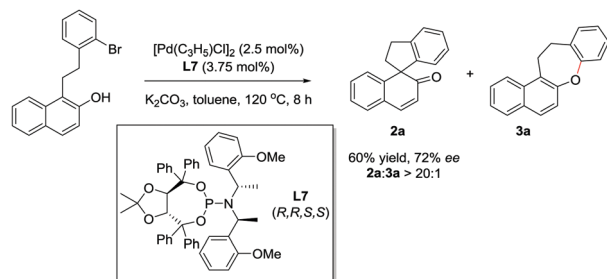
dearomatized product (**2a**) in 91% yield within 1 hour. Lowering the catalyst loading to 0.5 mol% and 0.1 mol% led to slightly higher yields in both cases but a longer reaction time was needed (Table 1, entries 12 and 13). Notably, the reaction underwent smoothly even at 40 °C (Table 1, entry 14). The reaction in varied solvents, such as DCM, dioxane and THF, all gave satisfactory yields at 40 °C (92–95% yields, Table 1, entries 15–18). Considering the efficiency and convenience of the experiments, the optimized conditions were obtained as the following: 0.5 mol% [Pd(C₃H₅)Cl]₂, 1.5 mol% Q-Phos (**L6**), and 1.5 equiv. of Cs₂CO₃ in toluene at 120 °C (Table 1, entry 12).

Next, we turned our attention to examine the substrate scope of this reaction. As shown in Scheme 2, substrates by cleaving different C–X bonds all gave their corresponding products in excellent yields and chemoselectivity (X = Cl, 92% yield, **2a** : **3a** = 14 : 1; X = Br, 95% yield, **2a** : **3a** > 20 : 1; X = I, 94% yield, **2a** : **3a** = 20 : 1). Then, a wide range of substituted aryl bromides bearing either electron-donating or electron-withdrawing groups was tested. In all cases, the intramolecular dearomatization reaction proceeded smoothly to afford their corresponding products (**2b–2g**) in excellent yields (92–95%) and chemoselectivity. The structure of **2c** was confirmed by X-ray crystallographic analysis.



Scheme 2 The reaction substrate scope. ^a Reaction conditions: **1** (0.8 mmol), [Pd(C₃H₅)Cl]₂ (0.004 mmol), **L6** (0.012 mmol), Cs₂CO₃ (1.2 mmol) in toluene (4.0 mL), 120 °C. Isolated yield. ^b Determined by ¹H NMR of the crude products. ^c **1** (0.4 mmol), Cs₂CO₃ (0.6 mmol) in toluene (2.0 mL). ^d **1** (0.2 mmol), Cs₂CO₃ (0.3 mmol) in toluene (1.0 mL), 60 °C. ^e **1** (0.2 mmol), [Pd(C₃H₅)Cl]₂ (0.005 mmol), **L6** (0.015 mmol), Cs₂CO₃ (0.3 mmol) in toluene (1.0 mL).



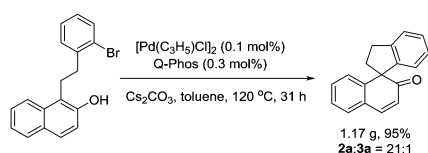


Scheme 3 Pd(0)-Catalyzed asymmetric intramolecular arylation dearomatization of β -naphthols.

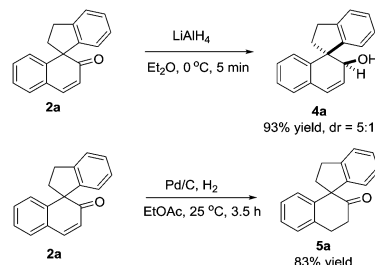
The reaction of the CF_3 -substituted substrate gave the desired product (**2h**) in good yield with slightly decreased chemoselectivity (86% yield, **2h:3h** = 12:1). Then the influence of the substituent group on the β -naphthol moiety was investigated. 3-Substituted (Me, CO_2Me) naphthols were transformed into dearomatized products (**2i** and **2j**) in good yields and chemoselectivity (80% yield, **2i:3i** = 10:1; 82% yield, **2j:3j** = 8:1). Fortunately, the substituents on the other positions (6-Ph and 7-Ph) of the β -naphthol moiety did not influence the chemoselectivity to give **2k** and **2l** in excellent yields and chemoselectivity (95% yield, **2k:3k** = 19:1; 95% yield, **2l:3l** = 18:1). Apart from substrates bearing an all-carbon tether, the substrate with an *N*-linked tether (**1m**) was also compatible, affording the desired product (**2m**) in 74% yield and excellent chemoselectivity (**2m:3m** > 20:1) under relatively milder conditions (at 60 °C). For substrate **1n** with an extended carbon chain, the desired dearomatized product with a 6-membered ring formation (**2n**) was obtained smoothly in 96% yield and excellent chemoselectivity (**2n:3n** > 20:1). Interestingly, the reaction of the α -naphthol substrate gave the dearomatized product (**2o**) in 88% yield with a **2o:3o** ratio of 20:1.

The asymmetric reaction was also explored. Several commercially available chiral phosphine ligands were screened. TADDOL-derived phosphoramidite (**L7**) was found to be the optimal ligand. In the presence of 2.5 mol% $[\text{Pd}(\text{C}_3\text{H}_5)\text{Cl}]_2$ and 3.75 mol% **L7**, the reaction of **1aa** could give **2a** in 60% yield and 72% ee with excellent chemoselectivity (**2a:3a** > 20:1, Scheme 3, see the ESI† for details).

To further demonstrate the utility of this method, a gram-scale reaction and several transformations of the 2-naphthalenone product have been carried out. The intramolecular dearomatization of **1aa** on a 5.0 mmol scale gave the desired product **2a** in 95% yield and excellent chemoselectivity (**2a:3a** = 21:1) while the catalyst loading could be further reduced to 0.1 mol% (Scheme 4). In addition, the ketone group of the product (**2a**) could be



Scheme 4 Gram-scale reaction.



Scheme 5 Transformations of the 2-naphthalenone product.

selectively reduced by LiAlH_4 to afford the alcohol **4a** in 93% yield (*dr* = 5:1), while the carbon–carbon double bond could be reduced by Pd/C catalyzed hydrogenation to afford **5a** in 83% yield (Scheme 5).

In summary, we have developed a Pd-catalyzed intramolecular dearomatization of β -naphthols to construct an all-carbon quaternary stereocenter at the *ortho*-position of the hydroxyl group with excellent yields and chemoselectivity. A series of structurally diverse spirocarbocycles were obtained efficiently and conveniently. Further studies on the application of the current method and development of more efficient catalytic asymmetric reactions are currently underway in our laboratory.

We thank the National Key Research and Development Program of China (2016YFA0202900), the National Basic Research Program of China (2015CB856600), the NSFC (21332009, 21361140373, and 21421091), the Program of Shanghai Subject Chief Scientist (16XD1404300), and the Chinese Academy of Sciences (XDB20000000, QYZDY-SSW-SLH012) for generous financial support.

Notes and references

- (a) Y.-L. Zhang, J. Zhang, N. Jiang, Y.-H. Lu, L. Wang, S.-H. Xu, W. Wang, G.-F. Zhang, Q. Xu, H. M. Ge, J. Ma, Y.-C. Song and R.-X. Tan, *J. Am. Chem. Soc.*, 2011, **133**, 5931; (b) Z. Xi, G. B. Jones, G. Qabaja, J. Wright, F. Johnson and I. H. Goldberg, *Org. Lett.*, 1999, **1**, 1375.
- (a) A. S. C. Chan, W.-H. Hu, C.-C. Pai, C.-P. Lau, Y.-Z. Jiang, A.-Q. Mi, M. Yan, J. Sun, R.-L. Lou and J.-G. Deng, *J. Am. Chem. Soc.*, 1997, **119**, 9570; (b) J.-H. Zhang, J. Liao, X. Cui, K.-B. Yu, J.-G. Deng, S.-F. Zhu, L.-X. Wang, Q.-L. Zhou, L.-W. Chung and T. Ye, *Tetrahedron: Asymmetry*, 2002, **13**, 1363; (c) J.-H. Xie and Q.-L. Zhou, *Acc. Chem. Res.*, 2008, **41**, 581.
- (a) A. Fürstner, *Chem. Soc. Rev.*, 2009, **38**, 3208; (b) V. A. D'yakonov, O. A. Trapeznikova, A. de Meijere and U. M. Dzhemilev, *Chem. Rev.*, 2014, **114**, 5775; (c) L. Fensterbank and M. Malacria, *Acc. Chem. Res.*, 2014, **47**, 953.
- M. Weber, M. Weber and M. Kleine-Boymann, "Phenol" in *Ullmanns Encyclopedia of Industrial Chemistry*, Wiley-VCH, Weinheim, 2004.
- For recent reviews, see: (a) A. R. Pape, K. P. Kaliappan and E. P. Kündig, *Chem. Rev.*, 2000, **100**, 2917; (b) S. Quideau, L. Pouységu and D. Deffieux, *Synlett*, 2008, 467; (c) L. Pouységu and S. Quideau, *Tetrahedron*, 2010, **66**, 2235; (d) S. P. Roche and J. A. Porco Jr., *Angew. Chem., Int. Ed.*, 2011, **50**, 4068; (e) C.-X. Zhuo, W. Zhang and S.-L. You, *Angew. Chem., Int. Ed.*, 2012, **51**, 12662; (f) Q. Ding, Y. Ye and R. Fan, *Synthesis*, 2013, 1; (g) C.-X. Zhuo, C. Zheng and S.-L. You, *Acc. Chem. Res.*, 2014, **47**, 2558; (h) W.-T. Wu, L. Zhang and S.-L. You, *Chem. Soc. Rev.*, 2016, **45**, 1570; (i) W. Sun, G. Li, L. Hong and R. Wang, *Org. Biomol. Chem.*, 2016, **14**, 2164; (j) C. Zheng and S.-L. You, *Chem*, 2016, **1**, 830; (k) R. B. Bedford, in *Asymmetric Dearomatization Reactions*, ed. S.-L. You, Wiley-VCH, Verlag GmbH & Co. KGaA, 2016, ch. 10, pp. 229–246; (l) W.-T. Wu, L. Zhang and S.-L. You, *Acta Chim. Sin.*, 2017, **75**, 419.



- 6 For selected recent examples: (a) T. Dohi, A. Maruyama, N. Takenaga, K. Senami, Y. Minamitsuji, H. Fujioka, S. B. Caemmerer and Y. Kita, *Angew. Chem., Int. Ed.*, 2008, **47**, 3787; (b) J. K. Boppisetti and V. B. Birman, *Org. Lett.*, 2009, **11**, 1221; (c) S. Quideau, G. Lyvinec, M. Marguerit, K. Bathany, A. Ozanne-Beaudenon, T. Buffeteau, D. Cavagnat and A. Chénédé, *Angew. Chem., Int. Ed.*, 2009, **48**, 4605; (d) M. Uyanik, T. Yasui and K. Ishihara, *Angew. Chem., Int. Ed.*, 2010, **49**, 2175; (e) T. Yakura, M. Omoto, Y. Yamauchi, Y. Tian and A. Ozono, *Tetrahedron*, 2010, **66**, 5833; (f) T. Nemoto, Y. Ishige, M. Yoshida, Y. Kohno, M. Kanematsu and Y. Hamada, *Org. Lett.*, 2010, **12**, 5020; (g) A. Rudolph, P. H. Bos, A. Meetsma, A. J. Minnaard and B. L. Feringa, *Angew. Chem., Int. Ed.*, 2011, **50**, 5834; (h) T. Oguma and T. Katsuki, *J. Am. Chem. Soc.*, 2012, **134**, 20017; (i) T. Nemoto, Z. Zhao, T. Yokosaka, Y. Suzuki, R. Wu and Y. Hamada, *Angew. Chem., Int. Ed.*, 2013, **52**, 2217; (j) C.-X. Zhuo and S.-L. You, *Angew. Chem., Int. Ed.*, 2013, **52**, 10056; (k) R. J. Phipps and F. D. Toste, *J. Am. Chem. Soc.*, 2013, **135**, 1268; (l) J. Nan, Z. Zuo, L. Luo, L. Bai, H. Zheng, Y. Yuan, J. Liu, X. Luan and Y. Wang, *J. Am. Chem. Soc.*, 2013, **135**, 17306; (m) C. Bosset, R. Coffinier, P. A. Peixoto, M. El Assal, K. Miqueu, J.-M. Sotiropoulos, L. Pouységu and S. Quideau, *Angew. Chem., Int. Ed.*, 2014, **53**, 9860; (n) T. Nemoto, N. Matsuo and Y. Hamada, *Adv. Synth. Catal.*, 2014, **356**, 2417; (o) S.-G. Wang, Q. Yin, C.-X. Zhuo and S.-L. You, *Angew. Chem., Int. Ed.*, 2015, **54**, 647; (p) D. Yang, L. Wang, F. Han, D. Li, D. Zhao and R. Wang, *Angew. Chem., Int. Ed.*, 2015, **54**, 2185; (q) J. Zheng, S.-B. Wang, C. Zheng and S.-L. You, *J. Am. Chem. Soc.*, 2015, **137**, 4880; (r) X. Lian, L. Lin, G. Wang, X. Liu and X. Feng, *Chem. – Eur. J.*, 2015, **21**, 17453; (s) Q. Yin, S.-G. Wang, X.-W. Liang, D.-W. Gao, J. Zheng and S.-L. You, *Chem. Sci.*, 2015, **6**, 4179; (t) S.-G. Wang, X.-J. Liu, Q.-C. Zhao, C. Zheng, S.-B. Wang and S.-L. You, *Angew. Chem., Int. Ed.*, 2015, **54**, 14929; (u) W.-T. Wu, R.-Q. Xu, L. Zhang and S.-L. You, *Chem. Sci.*, 2016, **7**, 3427; (v) Q. Cheng, Y. Wang and S.-L. You, *Angew. Chem., Int. Ed.*, 2016, **55**, 3496; (w) H.-F. Tu, C. Zheng, R.-Q. Xu, X.-J. Liu and S.-L. You, *Angew. Chem., Int. Ed.*, 2017, **56**, 3237; (x) D. Shen, Q. Chen, P. Yan, X. Zeng and G. Zhong, *Angew. Chem., Int. Ed.*, 2017, **56**, 3242.
- 7 (a) J. Garcia-Fortanet, F. Kessler and S. L. Buchwald, *J. Am. Chem. Soc.*, 2009, **131**, 6676; (b) R. B. Bedford, C. P. Butts, M. F. Haddow, R. Osborne and R. F. Sankey, *Chem. Commun.*, 2009, 4832.
- 8 (a) S. Wiegand and H. J. Schäfer, *Tetrahedron*, 1995, **51**, 5341; (b) S. Rousseaux, J. Garcia-Fortanet, M. A. D. A. Sanchez and S. L. Buchwald, *J. Am. Chem. Soc.*, 2011, **133**, 9282; (c) R.-Q. Xu, Q. Gu, W.-T. Wu, Z.-A. Zhao and S.-L. You, *J. Am. Chem. Soc.*, 2014, **136**, 15469; (d) K. Du, P. Guo, Y. Chen, Z. Cao, Z. Wang and W. Tang, *Angew. Chem., Int. Ed.*, 2015, **54**, 3033; (e) H. Zheng, L. Bai, J. Liu, J. Nan, Z. Zuo, L. Yang, Y. Wang and X. Luan, *Chem. Commun.*, 2015, **51**, 3061; (f) L. Yang, H. Zheng, L. Luo, J. Nan, J. Liu, Y. Wang and X. Luan, *J. Am. Chem. Soc.*, 2015, **137**, 4876; (g) L. Bai, Y. Yuan, J. Liu, J. Wu, L. Han, H. Wang, Y. Wang and X. Luan, *Angew. Chem., Int. Ed.*, 2016, **55**, 6946; (h) R.-Q. Xu, P. Yang, H.-F. Tu, S.-G. Wang and S.-L. You, *Angew. Chem., Int. Ed.*, 2016, **55**, 15137; (i) Z. Zuo, H. Wang, L. Fan, J. Liu, Y. Wang and X. Luan, *Angew. Chem., Int. Ed.*, 2017, **56**, 2767; (j) G. Zhao, G. Xu, C. Qian and W. Tang, *J. Am. Chem. Soc.*, 2017, **139**, 3360; (k) R.-Q. Xu, Q. Gu and S.-L. You, *Angew. Chem., Int. Ed.*, 2017, **56**, 7252.
- 9 (a) R. B. Bedford, N. Fey, M. F. Haddow and R. F. Sankey, *Chem. Commun.*, 2011, **47**, 3649; (b) L. Zhao, Z. Li, L. Chang, J. Xu, H. Yao and X. Wu, *Org. Lett.*, 2012, **14**, 2066; (c) K.-J. Wu, L.-X. Dai and S.-L. You, *Org. Lett.*, 2012, **14**, 3772; (d) C. Shen, R.-R. Liu, R.-J. Fan, Y.-L. Li, T.-F. Xu, J.-R. Gao and Y.-X. Jia, *J. Am. Chem. Soc.*, 2015, **137**, 4936; (e) D. A. Petrone, A. Yen, N. Zeidan and M. Lautens, *Org. Lett.*, 2015, **17**, 4838; (f) D. A. Petrone, M. Kondo, N. Zeidan and M. Lautens, *Chem. – Eur. J.*, 2016, **22**, 5684; (g) K. Douki, H. Ono, T. Taniguchi, J. Shimokawa, M. Kitamura and T. Fukuyama, *J. Am. Chem. Soc.*, 2016, **138**, 14578; (h) R.-R. Liu, Y.-G. Wang, Y.-L. Li, B.-B. Huang, R.-X. Liang and Y.-X. Jia, *Angew. Chem., Int. Ed.*, 2017, **56**, 7475.
- 10 K.-J. Wu, L.-X. Dai and S.-L. You, *Chem. Commun.*, 2013, **49**, 8620.
- 11 T. Xu and H. Alper, *Org. Lett.*, 2015, **17**, 1569.
- 12 (a) B. Yin, C. Cai, G. Zeng, R. Zhang, X. Li and H. Jiang, *Org. Lett.*, 2012, **14**, 1098; (b) J. Li, H. Peng, F. Wang, X. Wang, H. Jiang and B. Yin, *Org. Lett.*, 2016, **18**, 3226; (c) J. Liu, H. Peng, L. Lu, X. Xu, H. Jiang and B. Yin, *Org. Lett.*, 2016, **18**, 6440; (d) J. Liu, X. Xu, J. Li, B. Liu, H. Jiang and B. Yin, *Chem. Commun.*, 2016, **52**, 9550; (e) J. Liu, H. Peng, Y. Yang, H. Jiang and B. Yin, *J. Org. Chem.*, 2016, **81**, 9695.

