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电子转移活化和 1,2-硼迁移相结合实现酰胺脱氧硅基化

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摘要 报道了在 SmI_2/Mg 存在下, 使用膦配体促进的芳基酰胺与硅硼试剂的直接脱氧硅基化反应, 以中等至优异的收率和良好的官能团兼容性, 得到一系列具有高价值的 α-氨基硅烷化合物. 这一策略成功的关键在于电子转移诱导活化酰胺和 1,2-硼迁移的融合, 通过添加双膦配体 Xantphos 能够提高反应的效率. 该反应具有操作简单和条件温和的优点, 且所需原料易得, 产物价值高.

关键词 酰胺; 电子转移; 硅硼试剂; 1,2-硼迁移; α-氨基硅烷

Merging Electron Transfer Activation with 1,2-Metalate Migration: Deoxygenative Silylation of Amides

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Abstract A direct deoxygenative silylation of amides with silylboronate reagents is developed in the presence of SmI_2/Mg, affording a variety of high value-added α-aminosilane compounds in moderate to excellent yields with good functional group compatibility. The key to the success of this strategy lies in the merging of activation of amides induced by electron transfer with 1,2-metalate migration. The addition of Xantphos ligand can improve the reaction efficiency. The reactions are operationally simple and proceed under mild conditions, the raw materials are easily available and the products are highly valuable.

Keywords amide; electron transfer; silylboronate reagent; 1,2-metalate migration; α-aminosilane

1 Introduction

Amides are vital functional structures which are used in the construction of proteins and widely found in nature products and drugs. Meanwhile, amides as important organic intermediates are required to be converted into target compounds via subsequent reactions.[1] However, the conversion of amides to synthetically useful amines remains a longstanding challenge due to the low electrophilicity of carbonyl carbon (Scheme 1a). In recent years, several methodologies relating to the activation of amides have been reported,[2-7] wherein stoichiometric activators are used[3] or selective partial reduction catalyzed by transition metals is involved,[5-6] converting the amides into electrophilic active intermediates to achieve deoxygenative functionalization of such compounds. Alternatively, transforming the amides to the α-aminocarbene intermediates would be extremely appealing for further reaction owing to versatile reactivities of the intermediates. SmI_2 is a highly versatile, commercially available and readily prepared electron transfer (ET) reductant.[8] Early in 1992, Ogawa, Sonoda and co-workers[9] demonstrated Sm/SmI_2-mediated deoxygenative self-dimerization of amides via α-aminocarbene intermediates (Scheme 1b). In this context, Procter and the co-workers[10] have pioneered the transformations of amides by electron-transfer reduction of amide-type carbonyls in the presence of Sm(II). More recently, our group[11] reported the Sm(Mg)/SmI_2 promoted deoxygena-

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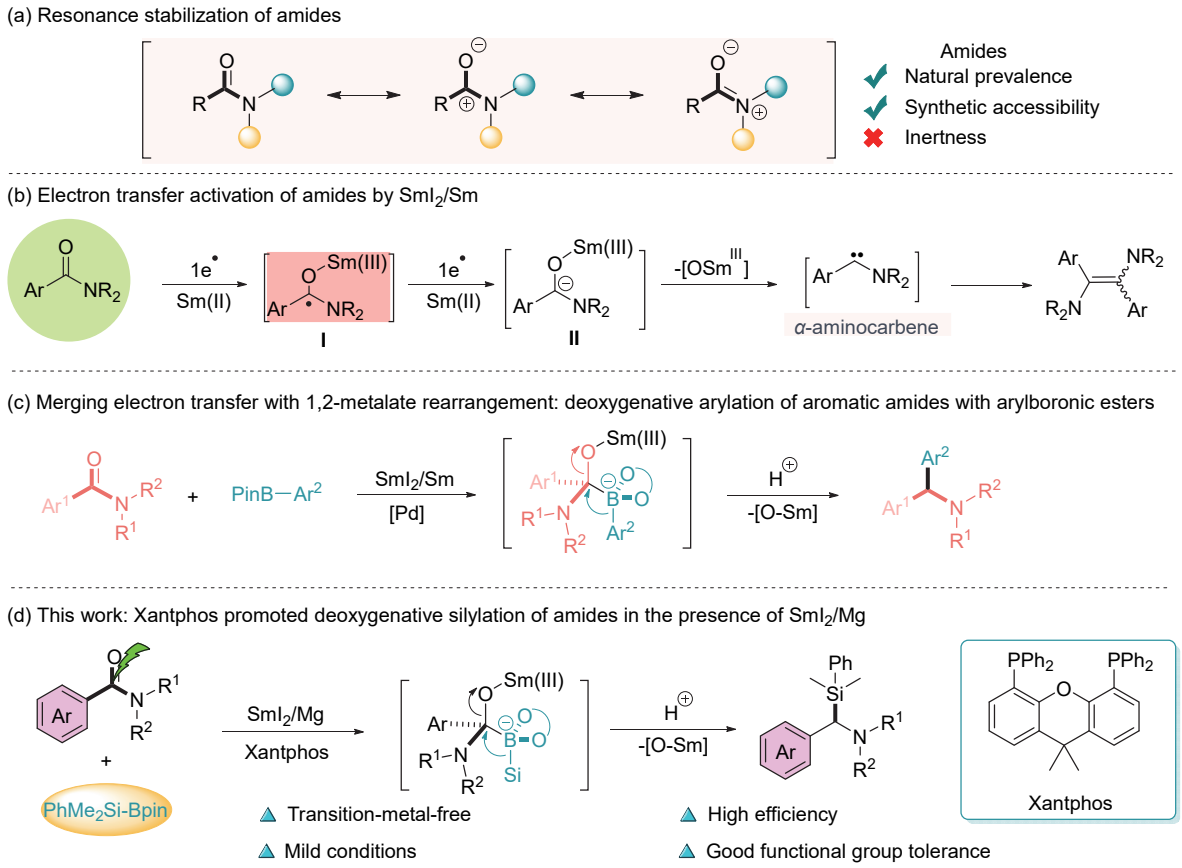

Scheme 1 Deoxygenative functionalization of amides

tive functionalization of relatively inert amides using arylboronic esters, polyfluorobenzene and hydrogermanes, respectively. Notably, an attractive deoxygenative arylation of amides with arylboronic esters were successfully achieved by the merging of electron transfer activation with 1,2-metalate migration (Scheme 1c).[11a,12]

Organosilicon compounds provide unique opportunities for biological studies, material chemistry and medicinal applications due to their unique properties.[13] Specifically, *α*-aminosilanes as a subclass of organosilicon compounds have garnered considerable interest because such compounds can play roles of mimics of natural *α*-amino acids in several protease inhibitors and have been merged into peptide isosteres.[14-16] Therefore, the development of mild and high-efficient methods to synthesis *α*-aminosilanes is highly desirable. The early methods of the synthesis of *α*-aminosilanes were mainly achieved by nucleophilic addition of imines with Si-Li reagents, which restricted the range of substrates scope owing to the utilization of active organometallic reagents.[17] Using Si-Bpin as silylation reagent, Oestreich and co-workers[18] successfully developed catalytic synthesis of *α*-aminosilanes via copper catalysts, showing the potential of silylboranes as silylation reagents. Recently, Studer group[19] developed an approach to *α*-aminosilanes via multicomponent radical cross-coupling mediated by visible light. Regarding to the deoxygenative silylation of amides, our group[20] reported a highly efficient synthesis of *α*-aminosilanes from inert amides with silanes via a proposed *α*-aminocarbene insertion into Si—H bond in the presence of Sm/SmI_2 and Pd additive. In view of our research group's long-term exploration and research in reductive transformation of amides,[11,21] herein, we report a phosphine ligand promoted Mg/SmI_2-mediated deoxygenative silylation of amides with Si-Bpin reagent for the synthesis of *α*-aminosilanes via the combination of electron transfer activation of amides with 1,2-metalate migration (Scheme 1d). Electron transfer induced activation of amide is proposed to be merged with tetracoordinated boron-mediated 1,2-Si group migration, which is crucial for the success of the transformation.[12] Notably, Song and co-workers[22] reported a similar transformation enabled by Sm/SmI_2, wherein a nickel salt was essential as an additive. To the best of our knowledge, the current work is the first example of Sm(Mg)/SmI_2 system using a phosphine ligand as an efficient additive to promote the efficiency of the reaction.

2 Results and discussion

Initially, we began our studies with the reaction of amide **1a** with $PhMe_2Si$-Bpin **2a** in the presence of Sm (2.0 equiv.) and SmI_2 (2.0 equiv.) at 80 ℃ for 24 h. Intriguingly, the desired product *α*-aminosilane **3aa** was obtained in 70% yield (Table 1, Entry 1). This result confirmed the feasibility of the conversion. Without Sm powder, the reaction only resulted in 21% yield of **3aa** (Entry 2). To our delight, switching the Sm powder to cheaper

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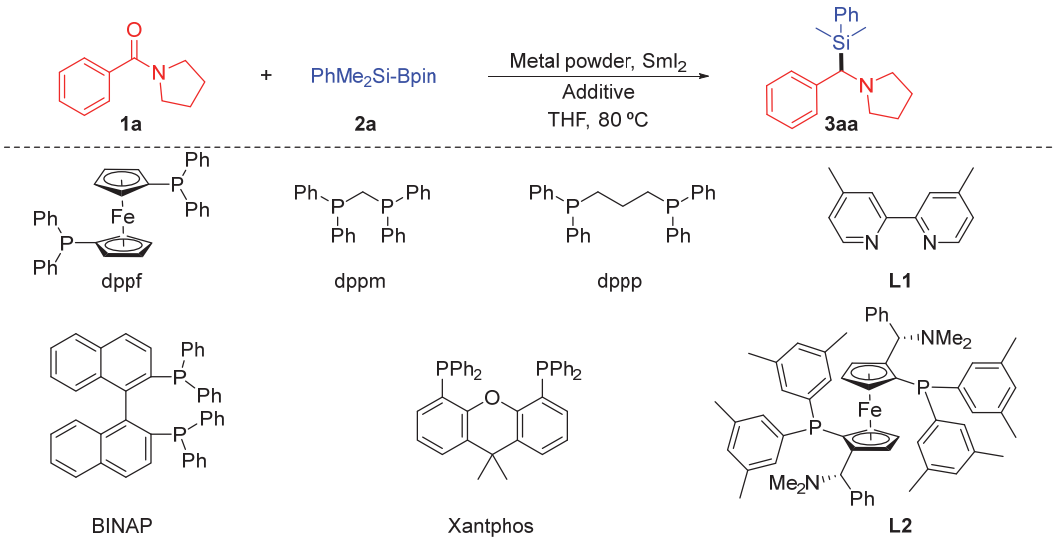
Mg, the reaction resulted in an increment of the yield of **3aa** up to 81% (Entry 3). In our previous studies,[11,23] various metal additives were investigated to improve the yields. Given the possibilities of the ligands to influence the reactivities of SmI_2[24], ligand was added to promote the efficiency of this reaction. After adding various phosphine ligands including bis(diphenylphosphino)methane (dppm), 1,3-bis(diphenylphosphino)propane (dppp), bis(diphenylphosphino)ferrocene (dppf), 9,9-dimethyl-4,5-bis(diphenylphosphino)xanthene (Xantphos), PPh_3, (±)-2,2'-bis(diphenylphosphino)-1,1'-binaphthyl (BINAP), **L1** and **L2** (Entries 4～11), Xantphos was found to be the best additive, affording the product **3aa** in 98% isolated yield (Entry 7). Subsequently, the amount of each reactant was screened (Entries 12～15). When the amount of SmI_2 was decreased to 1.0 equiv., the reaction gave the product **3aa** in 67% yield (Entry 12). Further decreasing the amount of Xantphos to 0.05 equiv. resulted in slightly lower yield of **3aa** (69% yield, Entry 13). Catalytic amount of SmI_2 (20 mol%) was also investigated together with Mg (5.0 equiv.) and Xantphos (0.05 equiv.) in the reaction, but only 34% yield of **3aa** was obtained (Entry 14).

With the optimized reaction conditions in hand, the generality of this reaction was explored (Table 2). A range of aromatic amides with different substituents at various positions all participated in this cross-coupling process to provide the corresponding *α*-aminosilanes in good to excellent yields. As shown in Table 2, we were pleased to find that the cross-coupling process was effective for amides from a diverse array of secondary alkyl amines. For example, amides form cyclic amines varying in ring size including six- and seven membered amines could be successfully used for this cross-coupling, and the corresponding products *α*-aminosilanes **3ba** and **3ca** were obtained in yields of 83% and 79%, respectively. Besides, an amide formed from thiomorpholine was also compatible and the targeted *α*-aminosilane **3da** was obtained in 85% yield. Amides generated from different piperazines performed well in this

Table 1 Optimization of the deoxygenative siliconization of amide[a]



Entry	Metal powder (equiv.)	SmI_2 (equiv.)	Ligand (0.1 equiv.)	Yield[b]/%
1	Sm (2.0)	2.0	—	70
2	—	2.0	—	21
3	Mg (2.0)	2.0	—	81
4	Mg (2.0)	2.0	dppm	74
5	Mg (2.0)	2.0	dppp	45
6	Mg (2.0)	2.0	dppf	77
7	Mg (2.0)	2.0	Xantphos	98 (98)[c]
8	Mg (2.0)	2.0	PPh_3	47
9	Mg (2.0)	2.0	**L1**	71
10	Mg (2.0)	2.0	**L2**	73
11	Mg (2.0)	2.0	BINAP	81
12	Mg (2.0)	1.0	Xantphos	67
13	Mg (2.0)	2.0	Xantphos[d]	69
14	Mg (5.0)	0.2	Xantphos	34

[a] Reaction conditions: the mixture of **1a** (0.1 mmol, 1.0 equiv.), **2a** (0.1 mmol), metal powder (0.2 mmol.), SmI_2 (0.2 mmol, 0.1 mol/L in THF) and additive was stirred at 80 ℃ for 24 h. [b] GC yield. [c] Isolated yield. [d] Xantphos (0.05 mmol.).

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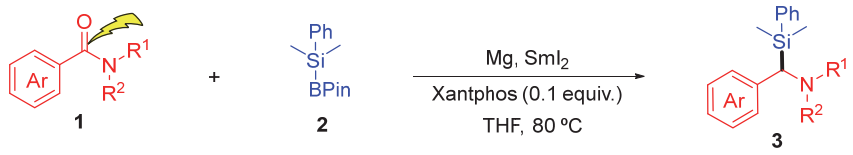
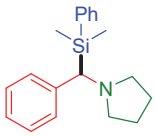
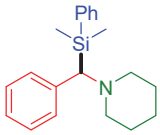
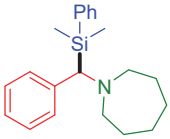
Table 2 Substrate scope of the deoxygenative silylation of amides

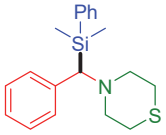


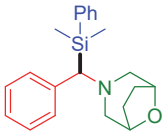
(a) For substituent groups of N atom

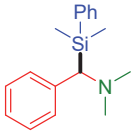

3aa, 86%

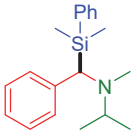

3ba, 83%

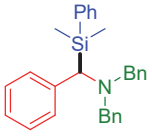

3ca, 79%

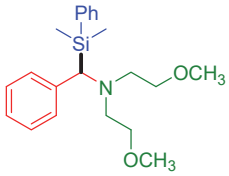

3da, 85%

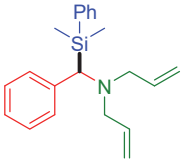

3ea, 88%

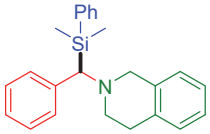

3fa, 89%

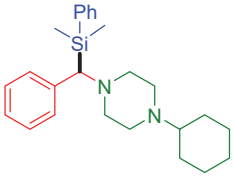

3ga, 63%

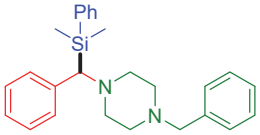

3ha, 45%

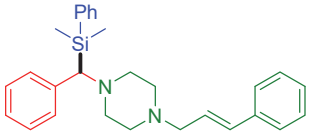

3ia, 80%


3ja, 51%

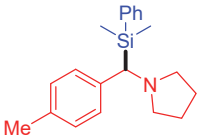

3ka, 82%

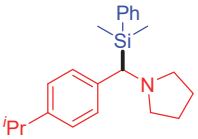

3la, 64%


3ma, 97%


3na, 62%

(b) For aromatic rings


3oa, 85%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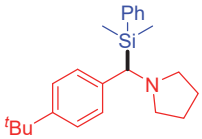
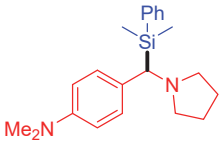

3qa, 89%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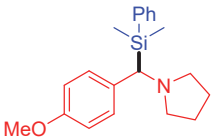
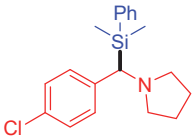

3sa, 71%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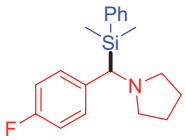
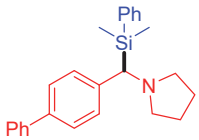
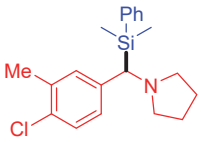

3ua, 71%


3va, 84%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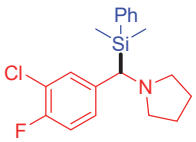
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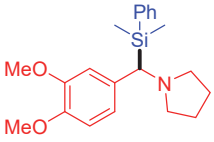
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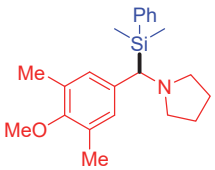
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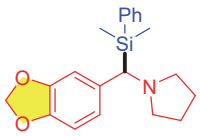
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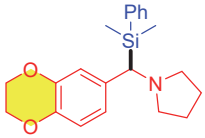
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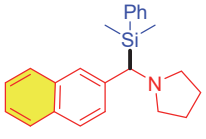
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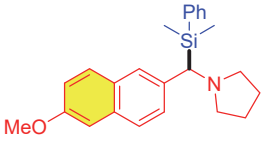
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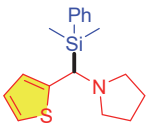
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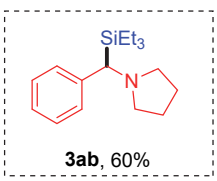
3ab, 60%

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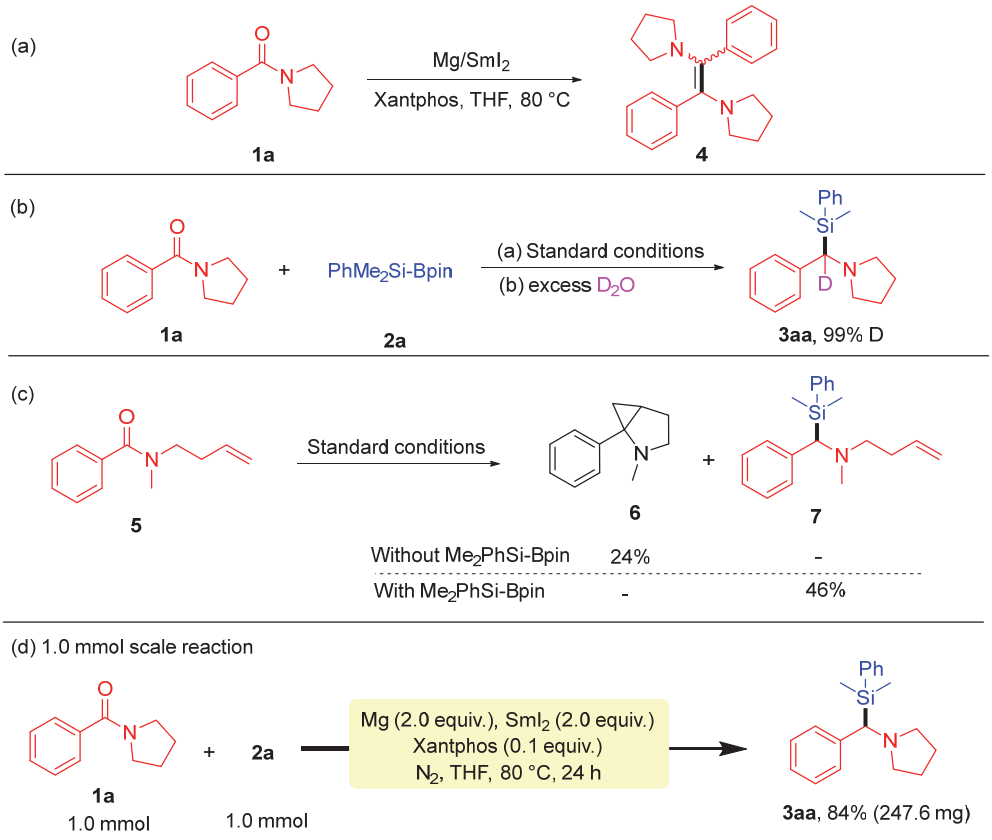
reaction, affording the desired products **3la**～**3na** in 62%～97% yields. Remarkably, the reaction of **2a** with amides bearing strained ring worked well under the standard conditions, affording **3ea** in 88% yield. Amides from noncyclic secondary amines displaying linear alkyl substituents such as methyl, isopropyl, benzyl *etc.* participated in this reaction to afford the desired *α*-aminosilanes **3fa**～**3ia** with moderate to good yields (45%～89%). To our delight, this reaction is compatible with alkenyl to give **3ja** in 51% yield. Amides from tetrahydroisoquinoline provided **3ka** in 82% yield. These results demonstrated that the procedure worked well with aryl amides derived from secondary amines, irrespective of cyclic or acyclic amines. Subsequently, the substrate scope of the aromatic ring in amides was also examined in the reactions with silylborane **2a**. As shown in Scheme 2b, various electron donating groups substituents on phenyl group of amides such as methyl, isopropyl, tertbutyl, *N*,*N*-dimethyl, OMe all performed well, affording the corresponding products **3oa**～**3sa** in 62%～89% yields. However, using electron-deficient amides with CF_3 or CN groups at the *para* position, the reactions only afforded trace amount of the desired products. Moreover, this method showed a high tolerance to several important functional groups, such as Cl and F on the arene under the standard conditions, affording the corresponding products **3ta**～**3ua** in yields of 71%～96%. Furthermore, 3,4-disubstituted and 3,4,5-trisubstituted phenylcarboxyamide **1w**～**1ab** were converted successfully to the corresponding products **3wa**～**3aba** in 58%～81% yields. Notably, the substrate scope with respect to the naphthalene and aromatic amide component could be further expanded to heterocyclic amides, affording the corresponding products **3aca**～**3aea** in moderate to good yields (67%～81%). Secondary amides and alkyl amides were found to be not applicable in the standard reaction conditions of note, Et_3Si-BPin **2b** could be engaged in this reaction with amide **1a** to afford the corresponding *α*-aminosilane **3ab** in 60% yield.

Preliminary mechanistic studies were also conducted in an effort to probe into the mechanism for the reaction (Scheme 2). Amide **1a** under the standard conditions in the absence of **2a** resulted in the self-dimerization product **4** detected by GC-MS (gas chromatography-mass spectrometer) (Scheme 2a). Addition of D_2O to quench the reaction of **1a** and **2a** led to 99% deuteration at the benzhydryl position in the isolated product **3aa-D**, suggesting protonation should be involved in the formation of the final deoxygenative product (Scheme 2b). Under the standard conditions without addition of $PhMe_2Si$-Bpin, cyclopropanylation product **6** was generated from amide **5** with a terminal olefin, indicating that *α*-aminocarbene species should be involved in the reaction (Scheme 2c). Nevertheless, the reaction of amide **5** with **2a** generated **7** in 46% yield under the standard conditions, showing that the Si-BPin reagent can capture *α*-aminocarbene before producing cyclopropane product. To demonstrate the practicability of this



Scheme 2 Preliminary mechanistic studies of deoxygenative silylation of amides

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protocol, a 1.0 mmol scale experiment was carried out, and the desired product **3aa** was obtained in 84% isolated yield (247.6 mg) under the standard conditions (Scheme 2d). Regarding to the role of the Xantphos ligand, it was proposed that the coordination of Sm species with Xanthos may influence the reductant strength and steric profile of the active Sm intermediate.[24]

3 Conclusions

In summary, we have developed a reductive silylation process of amides using silylboranes as silyl-transfer reagents mediated by Mg/SmI_2/Xantphos system, affording a series of biologically important α-aminosilanes under mild conditions. The key to this success is the merging of electron transfer induced activation of amide with tetracoordinated boron-mediated 1,2-Si group migration, leading to the cleavage of C—O bond and the construction of the new C—Si bond. Phosphine ligand Xantphos was found beneficial to improve the reaction efficiency, probably due to the effect of the reactivities of samarium species by the coordination with Xantphos.

4 Experimental section

4.1 General experimental information

Unless otherwise noted below, commercially available reagents were used throughout without further purification, and all reactions were performed using standard Schlenk techniques under an atmosphere of argon or in glovebox. Dry solvents were bought and stored over molecular sieves in a nitrogen atmosphere. Tetrahydrofuran (THF) was distilled from sodium/benzophenone prior to use. SmI_2 (0.1 mol/L in THF) was commercially available [Meryer (Shanghai) Chemical Technology Co., Ltd.] and was used as received. ^{1}H NMR, ^{13}C NMR and ^{19}F NMR spectra were recorded on Varian 400 MHz, Agilent 400 MHz spectrometers. Chemical shifts (δ values) were reported relative to internal TMS (^{1}H NMR) or $CDCl_3$ (^{1}H NMR) or $CDCl_3$ (^{13}C NMR), respectively. IR spectra were obtained on a Bruker Tensor 27 instruments with Bruker Platinum ATR accessory. HRMS (ESI) were determined on a JMS-T100LP AccuTOF LC-plus 4G. HRMS (EI) were determined on an Agilent Technologies 7250 Accurate-Mass Q-TOF GC/MS. GC analysis was performed on a Shimadzu GC-2014 gas chromatograph.

4.2 Procedure for deoxygenative siliconization of aromatic amides

To a sealed tube equipped with a magnetic stir bar was added **1** (0.2 mmol), **2** (0.2 mmol, 1.0 equiv.), Mg (9.6 mg, 0.4 mmol), SmI_2 (0.1 mol/L in THF, 4.0 mL, 0.4 mmol), and Xantphos (11.6 mg, 0.02 mmol, 10.0 mol%) in a nitrogen filled glovebox. The mixture was stirred at 80 ℃ for 24 h. After cooling to ambient temperature, it was filtered through silica, subsequently the combined solvent was removed under reduced pressure. Purification by column chromatography on silica gel using a mixture of petroleumether/ethyl acetate as eluent to give the product **3**.

1-((Dimethyl(phenyl)silyl)(phenyl)methyl)pyrrolidine (**3aa**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)=30/1～4/1], yellow brown oil, 50.5 mg, 86% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.52～7.50 (m, 2H), 7.36～7.30 (m, 3H), 7.23～7.22 (m, 4H), 7.17～7.11 (m, 1H), 3.09 (s, 1H), 2.42～2.39 (m, 4H), 1.69～1.66 (m, 4H), 0.41 (s, 3H), 0.05 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 143.0, 139.0, 133.9 , 128.7, 128.0, 127.8, 127.5, 125.4, 65.0, 55.5, 23.4, −2.0, −5.0.

1-((Dimethyl(phenyl)silyl)(phenyl)methyl)piperidine (**3ba**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)=30/1～4/1], yellow oil, 51.5 mg, 83% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.49～7.36 (m, 2H), 7.30～7.18 (m, 3H), 7.17～7.08 (m, 4H), 7.07～7.01 (m, 1H), 3.09 (s, 1H), 2.35～2.17 (m, 4H), 1.43～1.33 (m, 4H), 1.29～1.22 (m, 2H), 0.25 (s, 3H), −0.08 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 142.4, 139.5, 133.8, 128.6, 127.8, 127.5, 125.4, 66.3, 55.7, 26.5, −1.3, −4.6.

1-((Dimethyl(phenyl)silyl)(phenyl)methyl)azepane (**3ca**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)=30/1～4/1], yellow oil, 51.1 mg, 79% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.56～7.43 (m, 2H), 7.38～7.28 (m, 3H), 7.25～7.18 (m, 4H), 7.16～7.11 (m, 1H), 3.57 (s, 1H), 2.74～2.62 (m, 4H), 1.60～1.41 (m, 8H), 0.37 (s, 3H), 0.07 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 142.8, 139.3, 133.9, 128.7, 128.6, 127.7, 127.5, 125.5, 65.4, 55.8, 29.0, 27.0, −1.6, −4.3.

4-((Dimethyl(phenyl)silyl)(phenyl)methyl)thiomorpholine (**3da**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)=80/1], brown solid, 55.6 mg, 85% yield. m.p. 91～93 ℃; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.54～7.38 (m, 2H), 7.35～7.24 (m, 3H), 7.23～7.16 (m, 2H), 7.16～7.08 (m, 3H), 3.31 (s, 1H), 2.71～2.63 (m, 4H), 2.56～2.45 (m, 4H), 0.30 (s, 3H), −0.02 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 141.2, 138.8, 133.7, 128.9, 128.7, 128.0, 127.7, 125.8, 65.9, 55.9, 28.2, −1.4, −4.7. IR (neat) ν: 2906, 2872, 1425, 1246, 1070, 921, 814, 699 cm^{-1}. HRMS (ESI) calcd for $C_{19}H_{26}NSSi$ $[M+H]^+$ 328.1550, found 328.1552.

3-((Dimethyl(phenyl)silyl)(phenyl)methyl)-8-oxa-3-azabicyclo[3.2.1]octane (**3ea**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)=30/1～4/1], yellow oil, 59.4 mg, 88% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.49～7.42 (m, 2H), 7.36～7.28 (m, 3H), 7.26～7.11 (m, 5H), 4.17 (d, J=4.0 Hz, 1H), 4.10 (d, J=5.6 Hz, 1H), 3.20 (s, 1H), 2.71 (d, J=10.8 Hz, 1H), 2.50 (d, J=11.2 Hz, 1H), 2.32 (d, J=10.8 Hz, 1H), 2.08 (d, J=11.2 Hz, 1H), 2.01～1.94 (m, 1H), 1.88～1.73 (m, 3H), 0.35 (s, 3H), 0.02 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 141.6, 138.6, 133.7, 128.8, 128.4, 127.9, 127.6, 125.7, 65.0, 60.2, 59.0, 28.44, 28.36, −1.7, −4.7.

1-(Dimethyl(phenyl)silyl)-*N*,*N*-dimethyl-1-phenylmeth-

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anamine (**3fa**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)＝30/1～4/1], yellow brown oil, 47.9 mg, 89% yield. ^{1}H NMR (400 MHz, $CDCl_3$) 7.50～7.47 (m, 2H), 7.33～7.31 (m, 3H), 7.25～7.21 (m, 2H), 7.14～7.12 (m, 3H), 2.99 (s, 1H), 2.20 (s, 6H), 0.39 (s, 3H), －0.01 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) *δ*: 142.3, 139.0, 133.8, 128.8, 128.4, 128.0, 127.6, 125.7, 67.3, 47.2, －1.6, －4.8.

N-(cyclohexa-1,3-dien-1-yl(dimethyl(phenyl)silyl)methyl)-*N*-methylpropan-2-amine (**3ga**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)＝100/1～80/1], yellow oil, 37.5 mg, 63% yield. ^{1}H NMR (400 MHz, $CDCl_3$) ^{1}H NMR (400 MHz, $CDCl_3$) *δ*: 7.53～7.46 (m, 2H), 7.36～7.28 (m, 3H), 7.27～7.17 (m, 4H), 7.16～7.09 (m, 1H), 3.50 (s, 1H), 3.06～2.92 (m, 1H), 2.15～2.06 (s, 3H), 0.88 (d, *J*＝6.0 Hz, 3H), 0.80 (d, *J*＝6.4 Hz, 3H), 0.36 (s, 3H), 0.00 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) *δ*: 143.1, 139.5, 133.8, 128.6,128.5, 127.8, 127.5, 125.4, 62.6, 50.9, 35.1, 18.2, 15.9, －1.5, －4.8. IR (neat) *v*: 3067, 2958, 1427, 1253, 1118, 1045, 828, 786, 697 cm^{-1}. HRMS (ESI) calcd for $C_{19}H_{28}NSi$ $[M+H]^+$ 298.1986, found 298.1990.

N,*N*-dibenzyl-1-(dimethyl(phenyl)silyl)-1-phenylmethanamine (**3ha**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)＝50/1～20/1], white solid, 30.5 mg, 45% yield. m.p. 97～98 ℃; ^{1}H NMR (400 MHz, $CDCl_3$) *δ*: 7.44～7.40 (m, 2H), 7.38～7.26 (m, 10H), 7.25～7.20 (m, 6H), 7.16～7.08 (m, 2H), 3.92～7.80 (m, 2H), 3.66 (s, 1H), 3.42～7.32 (m, 2H), 0.37 (s, 3H), 0.28 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) *δ*: 140.0, 138.1, 138.0, 134.4, 130.8, 129.0, 128.8, 128.1, 127.9, 127.6, 126.7, 126.2, 58.2, 56.5, －2.4, －3.2.

N-((dimethyl(phenyl)silyl)(phenyl)methyl)-2-methoxy-*N*-(2-methoxyethyl)ethan-1-amine (**3ia**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)＝50/1～10/1], yellow oil, 57.2 mg, 80% yield. ^{1}H NMR (400 MHz, $CDCl_3$) *δ*: 7.50～7.48 (m, 2H), 7.35～7.30 (m, 3H), 7.26～7.22 (m, 2H), 7.19～7.12 (m, 3H), 3.73 (s, 1H), 3.37～3.27 (m, 4H), 3.26 (s, 6H), 2.87～2.80 (m, 2H), 2.78～2.71 (m, 2H), 0.36 (s, 3H), 0.20 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) *δ*: 140.6, 138.3, 134.2, 129.4, 128.9, 127.9, 127.5, 125.9, 71.6, 62.0, 58.7, 53.2, －2.3, －3.7.

N-allyl-*N*-((dimethyl(phenyl)silyl)(phenyl)methyl)prop-2-en-1-amine (**3ja**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)＝100/1～80/1], yellow oil, 30.5 mg, 48% yield. ^{1}H NMR (400 MHz, $CDCl_3$) *δ*: 7.48～7.35 (m, 2H), 7.30～7.20 (m, 3H), 7.19～7.00 (m, 5H), 5.85～5.50 (m, 2H), 5.05～4.90 (m, 4H), 3.65 (s, 1H), 3.24～3.12 (m, 2H), 2.92～2.79 (m, 2H), 0.24 (s, 3H), 0.12 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) *δ*: 134.0, 138.6, 136.4, 134.1, 129.8, 128.9, 127.8, 127.5, 125.8, 116.8, 59.0, 55.0, －2.3, －4.0. IR (neat) *v*: 3069, 2957, 2811, 1641, 1426, 1250, 1110, 917, 807, 699 cm^{-1}. HRMS (ESI) calcd for $C_{21}H_{28}NSi$ $[M+H]^+$ 322.1986, found 322.1988.

2-((Dimethyl(phenyl)silyl)(phenyl)methyl)-1,2,3,4-tetrahydroisoquinoline (**3ka**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)＝30/1～4/1], white solid, 38.8 mg, 66% yield. ^{1}H NMR (400 MHz, $CDCl_3$) *δ*: 7.56～7.53 (m, 2H), 7.338～7.32 (m, 3H), 7.26～7.25 (m, 4H), 7.19～7.16 (m, 1H), 7.11～7.02 (m, 3H), 6.79～6.77 (m, 1H), 3.59 (dd, *J*＝27.6, 15.2 Hz, 2H), 3.39 (s, 1H), 2.77～2.68 (m, 4H), 0.40 (s, 3H), 0.05 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) *δ*: 141.8, 139.1, 135.5, 134.4, 133.8, 128.9, 128.53,128.47, 128.0, 127.7, 126.5, 125.8, 125.7, 125.3, 65.1, 57.4, 51.3, 29.4, －1.4, －4.9.

1-Cyclohexyl-4-((dimethyl(phenyl)silyl)(phenyl)methyl)piperazine (**3la**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)＝30/1～4/1], yellow oil, 35.0 mg, 64% yield. ^{1}H NMR (400 MHz, $CDCl_3$) *δ*: 7.51～7.44 (m, 2H), 7.34～7.25 (m, 3H), 7.25～7.15 (m, 4H), 7.14～7.05 (m, 1H), 3.17 (s, 1H), 2.70～2.30 (m, 8H), 2.20～2.12 (m, 1H), 1.79 (dd, *J*＝36.0, 12.0 Hz, 4H), 1.62～1.56 (m, 1H), 1.22～1.02 (m, 5H), 0.32 (s, 3H), 0.00 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) *δ*: 141.9, 139.0, 133.8, 128.7, 128.6, 127.9, 127.6, 125.6, 65.5, 63.4, 54.6, 49.4, 28.86 (d, *J*＝9.6 Hz), 26.2, 25.8, －1.4, －4.6. IR (neat) *v*: 2927, 2854, 2807, 1652, 1449, 1252, 1116, 830, 699 cm^{-1}. HRMS (ESI) calcd for $C_{25}H_{37}N_2Si$ $[M+H]^+$ 393.2721, found 393.2724.

1-Benzyl-4-((dimethyl(phenyl)silyl)(phenyl)methyl)-piperazine (**3ma**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)＝30/1～4/1], yellow oil, 77.7 mg, 97% yield. ^{1}H NMR (400 MHz, $CDCl_3$) *δ*: 7.51～7.42 (m, 2H), 7.33～7.24 (m, 7H), 7.23～7.15 (m, 5H), 7.14～7.09 (m, 1H), 3.43 (dd, *J*＝38.4, 12.8 Hz, 2H), 3.17 (s, 1H), 2.56～2.22 (m, 8H), 0.32 (s, 3H), －0.03 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) *δ*: 141.9, 139.0, 138.1, 133.7, 129.2, 128.7, 128.5, 128.1, 127.9, 127.6, 126.9, 125.6, 65.5, 63.1, 54.3, 53.7, －1.3, －4.9.

1-Cinnamyl-4-((dimethyl(phenyl)silyl)(phenyl)methyl)-piperazine (**3na**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)＝30/1～4/1], brown yellow oil, 56.0 mg, 62% yield. ^{1}H NMR (400 MHz, $CDCl_3$) *δ*: 7.51～7.49 (m, 2H), 7.37～7.28 (m, 7H), 7.25～7.20 (m, 5H), 7.16～7.11 (m, 1H), 6.50～6.46 (m, 1H), 6.27～6.20 (m, 1H), 3.21 (s, 1H), 3.17～3.04 (m, 2H), 2.78～2.09 (m, 8H), 0.35 (s, 3H), 0.02 (s, 3H); ^{13}C NMR (151 MHz, $CDCl_3$) *δ*: 141.8, 138.9, 136.8, 133.7, 132.9, 128.8, 128.51, 128.48, 127.9, 127.6, 127.4, 126.6, 126.2, 125.6, 65.4, 61.0, 54.2, 53.7, －1.4, －4.8. IR (neat) *v*: 3024, 2955, 2805, 1655, 1449, 1427, 1253, 1115, 789, 696 cm^{-1}. HRMS (ESI) calcd for $C_{28}H_{35}N_2Si$ $[M+H]^+$ 427.2564, found 427.2568.

1-((Dimethyl(phenyl)silyl)(*p*-tolyl)methyl)pyrrolidine (**3oa**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)＝30/1～4/1], brown yellow oil, 52.8 mg, 85% yield. ^{1}H NMR (400 MHz, $CDCl_3$) *δ*: 7.55～7.41 (m, 2H), 7.39～7.21 (m, 3H), 7.04 (dd, *J*＝26.0, 7.6 Hz, 4H), 3.02 (s, 1H), 2.40～2.31 (m,

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4H), 2.28 (s, 3H), 1.66～1.56 (m, 4H), 0.35 (s, 3H), −0.01 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 139.9, 139.2, 134.7, 133.9, 128.7, 128.6, 127.9, 127.5, 64.5, 55.4, 23.4, 21.0, −1.8, −5.1. IR (neat) ν: 2958, 2781, 1610, 1456, 1295, 1117, 1044, 829, 782, 728, 698 cm^{-1}. HRMS (FI) calcd for $C_{20}H_{27}NSi$ $[M+H]^+$ 309.1907, found 309.1903.

1-((Dimethyl(phenyl)silyl)(4-isopropylphenyl)methyl)-pyrrolidine (**3pa**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)=30/1～4/1], yellow oil, 54.4 mg, 81% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.48～7.36 (m, 2H), 7.33～7.18 (m, 3H), 7.04 (dd, J=18.4, 8.0 Hz, 4H), 3.00 (s, 1H), 2.91～2.75 (m, 1H), 2.42～2.26 (m, 4H), 1.67～1.56 (m, 4H), 1.20 (d, J=6.8 Hz, 6H), 0.36 (s, 3H), −0.01 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 145.8, 140.1, 139.2, 133.9, 128.6, 127.8, 127.5, 125.8, 64.6, 55.5, 33.6, 24.10 (d, J=3.0 Hz), 23.4, −2.0, −5.0. IR (neat) ν: 2958, 2781, 1426, 1251, 1117, 1052, 829, 782, 699 cm^{-1}. HRMS (ESI) calcd for $C_{22}H_{32}NSi$ $[M+H]^+$ 338.2299, found 338.23301.

1-((4-(*tert*-Butyl)phenyl)(dimethyl(phenyl)silyl)methyl)-pyrrolidine (**3qa**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)=30/1～4/1], yellow oil, 62.6 mg, 89% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.50～7.44 (m, 2H), 7.35～7.27 (m, 3H), 7.17 (dd, J=40.4, 7.6 Hz, 4H), 3.07 (s, 1H), 2.51～2.33 (m, 4H), 1.73～1.63 (m, 4H), 1.32～1.27 (m, 9H), 0.42 (s, 3H), 0.04 (s, 3H); ^{13}C NMR (151 MHz, $CDCl_3$) δ: 148.1, 139.7, 139.2, 133.9, 128.6, 127.5, 127.4, 124.6, 64.5, 55.5, 34.3, 31.5, 23.4, −2.1, −4.9. IR (neat) ν: 2959, 2781, 1251, 1116, 829, 783, 698 cm^{-1}. HRMS (ESI) calcd for $C_{23}H_{34}NSi$ $[M+H]^+$ 352.2455, found 352.2458.

4-((Dimethyl(phenyl)silyl)(pyrrolidin-1-yl)methyl)-*N*,*N*-dimethylaniline (**3ra**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)=100/1～30/1], light yellow solid, 54.1 mg, 80% yield. m.p. 58～60 ℃; ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.53～7.50 (m, 2H), 7.34～7.30 (m, 3H), 7.06 (d, J=8.4 Hz, 2H), 6.65～6.63 (m, 2H), 2.97 (s, 1H), 2.91 (s, 6H), 2.42～2.29 (m, 4H), 1.68～1.59 (m, 4H), 0.36 (s, 3H), 0.02 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 148.7, 139.6, 134.0, 131.1, 128.8,128.6, 127.5, 112.5, 63.9, 55.4, 40.9, 23.4, −1.6, −5.0. IR (neat) ν: 2956, 2784, 1594, 1517, 1164, 731, 699 cm^{-1}. HRMS (ESI) $[M+H]^+$ calcd for $C_{21}H_{31}N_2Si$ 339.2251, found 339.2254.

1-((Dimethyl(phenyl)silyl)(4-methoxyphenyl)methyl)-pyrrolidine (**3sa**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)=20/1～4/1], yellow oil, 46.1 mg, 71% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.58～7.46 (m, 2H), 7.40～7.27 (m, 3H), 7.26～7.17 (m, 4H), 7.16～7.09 (m, 1H), 3.18 (s, 1H), 2.42～2.24 (m, 4H), 1.53～1.43 (m, 4H), 1.41～1.23 (m, 3H), 0.35 (s, 3H), 0.02 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 142.4, 139.5, 133.8, 128.6, 127.8, 127.5, 125.4, 66.3, 55.7, 26.5, 24.4, −1.3, −4.6. IR (neat) ν: 2956, 2781, 1607, 1507, 1243, 1117, 1035, 781, 699 cm^{-1}. HRMS (ESI) calcd for $C_{20}H_{28}ONSi$ $[M+H]^+$ 326.1935, found 326.1937.

1-((4-Chlorophenyl)(dimethyl(phenyl)silyl)methyl)-pyrrolidine (**3ta**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)=30/1～4/1], brown yellow oil, 63.4 mg, 96% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.51～7.43 (m, 2H), 7.41～7.30 (m, 3H), 7.16 (dd, J=24.8, 8.0 Hz, 4H), 3.08 (s, 1H), 2.46～2.33 (m, 4H), 1.73～1.63 (m, 4H), 0.40 (s, 3H), 0.10 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 141.7, 138.4, 133.9, 130.7, 129.2, 128.9, 127.9, 127.6, 64.3, 55.3, 23.4, −2.3, −4.8. IR (neat) ν: 2959, 2783, 1486, 1427, 1249, 1192, 1115, 1088, 1013, 828, 804, 777, 731, 699 cm^{-1}. HRMS (ESI) calcd for $C_{19}H_{25}NClSi$ $[M+H]^+$ 330.1439, found 330.1438.

1-((Dimethyl(phenyl)silyl)(4-fluorophenyl)methyl)pyrrolidine (**3ua**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)=20/1～4/1], yellow oil, 44.5 mg, 71% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.51～7.40 (m, 2H), 7.38～7.27 (m, 3H), 7.19～7.03 (m, 2H), 7.01～6.75 (m, 2H), 3.05 (s, 1H), 2.44～2.33 (m, 4H), 1.72～1.62 (m, 4H), 0.38 (s, 3H), 0.08 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 160.9 (d, J=243.4 Hz), 138.7 (d, J=3.0 Hz), 138.6, 133.9, 129.1 (d, J=8.1 Hz), 128.8, 127.6, 114.5 (d, J=21.0 Hz), 64.1, 55.4, 23.4, −2.3, −4.8; ^{19}F NMR (376 MHz, $CDCl_3$) δ: −118.17. IR (neat) ν: 2957, 2784, 1601, 1504, 1427, 1253, 1221, 1118, 829, 786, 698 cm^{-1}. HRMS (ESI) calcd for $C_{19}H_{25}NFSi$ $[M+H]^+$ 314.1735, found 314.1735.

1-([1,1'-Biphenyl]-4-yl(dimethyl(phenyl)silyl)methyl)-pyrrolidine (**3va**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)=30/1～4/1], yellow oil, 62.2 mg, 84% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.66～7.57 (m, 2H), 7.57～7.36 (m, 6H), 7.36～7.28 (m, 4H), 7.27 ～7.20 (m, 2H), 3.13 (s, 1H), 2.53～2.31 (m, 4H), 1.78～1.58 (m, 4H), 0.42 (s, 3H), 0.09 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 142.3, 141.1, 138.9, 138.1, 133.9, 128.8, 128.6, 128.3 127.5, 126.8, 126.4, 109.96, 64.7, 55.5, 23.4, −2.0, −4.8. IR (neat) ν: 2957, 2785, 1485, 1252, 1117, 828, 696 cm^{-1}. HRMS (ESI) calcd for $C_{25}H_{30}NSi$ $[M+H]^+$ 372.2142, found 372.2145.

((4-Chloro-3-methylphenyl)(dimethyl(phenyl)silyl)methyl)pyrrolidine (**3wa**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl acetate)=30/1～4/1], brown yellow oli, 54.3 mg, 80% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.49～7.42 (m, 2H), 7.39～7.26 (m, 3H), 7.23～7.09 (m, 1H), 7.07～6.84 (m, 2H), 3.00 (s, 1H), 2.44～2.34 (m, 4H), 2.29 (s, 3H), 1.72～1.62 (m, 4H), 0.38 (s, 3H), 0.07 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 141.6, 138.5, 135.1, 133.9, 130.9, 130.4, 128.8, 128.3, 127.5, 126.6, 64.4, 55.4, 23.4, 20.0, −2.3, −4.8. IR (neat) ν: 2957, 2786, 1479, 1406, 1253, 1118, 1045, 828, 786, 698 cm^{-1}. HRMS (ESI) calcd for $C_{20}H_{27}NClSi$ $[M+H]^+$ 344.1596, found 344.1596.

1-((3-Chloro-4-fluorophenyl)(dimethyl(phenyl)silyl)-methyl)pyrrolidine (**3xa**): Purified by flash column chromatography [eluent: *V*(petroleum ether)∶*V*(ethyl ace-

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tate)=30/1～4/1], yellow oil, 40.4 mg, 58% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.44～7.38 (m, 2H), 7.37～7.27 (m, 3H), 7.24～7.10 (m, 1H), 7.09～6.78 (m, 2H), 3.00 (s, 1H), 2.46～2.29 (m, 4H), 1.75～1.60 (m, 4H), 0.38 (s, 3H), 0.11 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 155.9 (d, J=247.5 Hz), 139.1 (d, J=238.4 Hz), 133.9, 129.2 (d, J=42.4 Hz), 127.6, 127.2 (d, J=7.1 Hz), 120.1 (d, J=18.2 Hz), 115.7 (d, J=21.2 Hz), 63.9, 55.3, 23.4, −2.7, −4.6; ^{19}F NMR (376 MHz, $CDCl_3$) δ: −120.53 (dd, J=15.0, 7.5 Hz). IR (neat) ν: 2957, 2790, 1498, 1252, 1118, 1055, 828, 787, 698 cm^{-1}. HRMS (ESI) calcd for $C_{19}H_{24}NClFSi$ $[M+H]^+$ 348.1345, found 348.1343.

1-((3,4-Dimethoxyphenyl)(dimethyl(phenyl)silyl)methyl)pyrrolidine (**3ya**): Purified by flash column chromatography [eluent: V(petroleum ether)∶V(ethyl acetate)=20/1～4/1], yellow oli, 47.1 mg, 59% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.43～7.35 (m, 2H), 7.28～7.19 (m, 3H), 6.71～6.55 (m, 3H), 3.79 (s, 3H), 3.66 (s, 3H), 2.92 (s, 1H), 2.41～2.28 (m, 4H), 1.68～1.55 (m, 4H), 0.29 (s, 3H), 0.04 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 148.3, 146.7, 138.8, 135.5, 134.0, 128.7, 127.5, 119.8, 111.2, 110.3, 55.7, 55.6, 55.4, 23.4, −2.1, −4.4. IR (neat) ν: 2955, 2781, 1510, 1255, 1117, 1027, 821, 782, 699 cm^{-1}. HRMS (ESI) calcd for $C_{21}H_{30}O_2NSi$ $[M+H]^+$ 356.2040, found 356.2038.

1-((Dimethyl(phenyl)silyl)(4-methoxy-3,5-dimethylphenyl)methyl)pyrrolidine (**3za**): Purified by flash column chromatography [eluent: V(petroleum ether)∶V(ethyl acetate)=30/1～4/1], yellow oil, 57.3 mg, 81% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.42～7.35 (m, 2H), 7.32～7.16 (m, 3H), 6.78～6.68 (m, 2H), 3.64 (s, 3H), 2.87 (s, 1H), 2.39～2.28 (m, 4H), 2.16 (s, 6H), 1.68～1.58 (s, 4H), 0.35 (s, 3H), 0.00 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 154.6, 139.1, 137.8, 133.9, 129.7, 128.6, 128.1, 127.4, 64.5, 59.7, 55.6, 23.3, 16.0, −2.3, −4.8. IR (neat) ν: 3068, 2954, 2780, 1481, 1426, 1250, 1219, 1140, 1014, 820, 784, 730, 698 cm^{-1}. HRMS (ESI) calcd for $C_{22}H_{32}ONSi$ $[M+H]^+$ 354.2248, found 354.2251.

1-(Benzo[d][1,3]dioxol-5-yl(dimethyl(phenyl)silyl) -methyl)pyrrolidine (**3aaa**): Purified by flash column chromatography [eluent: V(petroleum ether)∶V(ethyl acetate)=30/1～10/1], yellow oil, 54.3 mg, 80% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.58～7.46 (m, 2H), 7.43～7.27 (m, 3H), 6.79 (s, 1H), 6.64 (dd, J=20.8, 8.0 Hz, 2H), 5.92 (s, 2H), 3.00 (s, 1H), 2.44～2.29 (m, 4H), 1.73～1.59 (m, 4H), 0.39 (s, 3H), 0.07 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 147.3, 145.2, 134.0, 137.2, 133.9, 128.7, 127.5, 120.7, 108.4, 107.6, 100.6, 64.5, 55.3, 23.4, −2.0, −5.0. IR (neat) ν: 2959, 2780, 1483, 1240, 1115, 1038, 822, 699 cm^{-1}. HRMS (ESI) calcd for $C_{20}H_{26}O_2NSi$ $[M+H]^+$ 340.1727, found 340.1730.

1-((2,3-Dihydrobenzo[b][1,4]dioxin-6-yl)(dimethyl-(phenyl)silyl)methyl)pyrrolidine (**3aba**): Purified by flash column chromatography [eluent: V(petroleum ether)∶V(ethyl acetate)=30/1～4/1], yellow oil, 49.0 mg, 70% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.46～7.38 (m, 2H), 7.29～7.19 (m, 3H), 6.69～6.51 (m, 3H), 4.17～4.17～4.10 (m, 4H), 2.87 (s, 1H), 2.33～2.19 (m, 4H), 1.60～1.50 (m, 4H), 0.30 (s, 3H), −0.05 (s, 3H); ^{13}C NMR (151 MHz, $CDCl_3$) δ: 142.9, 141.2, 139.2, 136.5, 133.9, 128.7, 127.5, 121.0, 116.5, 116.4, 64.3, 64.2, 64.0, 55.3, 23.4, 1.0, −5.1. IR (neat) ν: 2960, 2781, 1586, 1501, 1427, 1282, 1253, 1116, 1067, 886, 818, 699 cm^{-1}. HRMS (ESI) calcd for $C_{21}H_{28}O_2NSi$ $[M+H]^+$ 354.1884, found 354.1887.

1-((Dimethyl(phenyl)silyl)(naphthalen-2-yl)methyl)-pyrrolidine (**3aca**): Purified by flash column chromatography [eluent: V(petroleum ether)∶V(ethyl acetate)=30/1～4/1], brown yellow oil, 55.7 mg, 81% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.85～7.73 (m, 3H), 7.66 (s, 1H), 7.58～7.52 (m, 2H), 7.50～7.41 (m, 3H), 7.40～7.29 (m, 3H), 3.30 (s, 1H), 2.51～2.41 (m, 4H), 1.77～1.67 (m, 4H), 0.46 (s, 3H), 0.08 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 140.9, 138.9, 133.9, 133.4, 132.0, 128.8, 127.6, 127.6, 127.5, 127.3, 127.1, 125.8, 125.6, 124.8, 65.3, 55.6, 23.4, −1.8, −5.0. IR (neat) ν: 3051, 2957, 1598, 1370, 1118, 824, 789, 746, 698 cm^{-1}. HRMS (ESI) calcd for $C_{23}H_{28}NSi$ $[M+H]^+$ 346.1986, found 346.1988.

1-((Dimethyl(phenyl)silyl)(6-methoxynaphthalen-2-yl)-methyl)pyrrolidine (**3ada**): Purified by flash column chromatography [eluent: V(petroleum ether)∶V(ethyl acetate)=30/1～4/1], brown yellow oil, 63.1 mg, 84% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.59～7.48 (m, 2H), 7.47～7.36 (m, 3H), 7.32～7.17 (m, 4H), 7.08～6.95 (m, 2H), 3.82 (s, 3H), 3.11 (s, 1H), 2.39～2.24 (m, 4H), 1.66～1.51 (m, 4H), 0.31 (s, 3H), −0.06 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 156.9, 139.0, 138.5, 134.0, 132.9, 129.0, 128.9, 128.8, 127.6, 127.5, 126.2, 125.7, 118.4, 105.5, 65.0, 55.6, 55.3, 23.4, 1.0, −1.8. IR (neat) ν: 2956, 1604, 1502, 1457, 1387, 1263, 1026, 812, 699 cm^{-1}. HRMS (ESI) calcd for $C_{24}H_{30}ONSi$ $[M+H]^+$ 376.2091, found 376.2091.

1-((Dimethyl(phenyl)silyl)(thiophen-2-yl)methyl)pyrrolidine (**3aea**): Purified by flash column chromatography [eluent: V(petroleum ether)∶V(ethyl acetate)=20/1～4/1], brown yellow oil, 40.3mg, 67% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.55～7.52 (m, 2H), 7.39～7.31 (m, 3H), 7.11 (d, J=5.2 Hz , 1H), 6.88～6.86 (m, 1H), 6.701～6.690 (m, 1H), 3.54 (s, 1H), 2.51～2.43 (m, 4H), 1.72～1.65 (m, 4H), 0.48 (s, 3H), 0.17 (s, 3H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 146.0, 138.4, 133.9, 128.9, 127.6, 126.0, 123.8, 123.0, 58.7, 54.7, 29.7, 23.4, −2.4, −4.7. IR (neat) ν: 2957, 2853, 1591, 1254, 1118, 830, 724, 698 cm^{-1}. HRMS (FI) calcd for $C_{19}H_{31}NSSi$ $[M+H]^+$ 309.1309, found 301.1312.

1-(Phenyl(triethylsilyl)methyl)pyrrolidine (**3ab**): Purified by flash column chromatography [eluent: V(petroleum ether)∶V(ethyl acetate)=30/1～10/1], yellow oil, 41.8 mg, 60% yield. ^{1}H NMR (400 MHz, $CDCl_3$) δ: 7.28～7.11 (m, 4H), 7.11～7.02 (m, 1H), 2.86 (s, 1H), 2.51～2.36 (m, 4H), 1.73～1.61 (m, 4H), 0.93～0.72 (m, 9H), 0.57～0.41 (m, 6H); ^{13}C NMR (101 MHz, $CDCl_3$) δ: 143.7, 128.0, 127.8, 125.2, 62.3, 55.1, 23.3, 7.6, 3.4.

Supporting Information Experimental procedures, the

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synthesis method of the starting materials, and compound characterization dat. The Supporting Information is available free of charge via the Internet at http://sioc-journal.cn/.

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