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Synthesis and Reactivity of 1-Substituted 2-Fluoro- and 2,2-Difluoroaziridines

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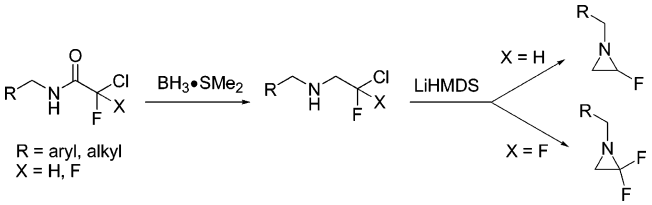

A straightforward synthesis toward 2-fluorinated aziridines was developed via ring closure of β-fluorinated β-chloroamines, which were obtained via reduction of the corresponding α-fluorinated amides by borane. When 1-benzyl-2-fluoroaziridine was treated with methanol, reaction occurred at the 2-position, giving rise to *N*-benzyl-2,2-dimethoxyethylamine, while in the case of 1-benzyl-2,2-difluoroaziridine the 3-position was attacked, giving rise to *N*-benzyl-2-methoxyacetamide. These reactions point to the divergent reactive behavior of monofluoro- and difluoroaziridines.

Fluorinated amines have been the subject of considerable research because these compounds are highly interesting building blocks for pharmaceutical and agrochemical purposes.[1,2] This increasing interest is a consequence of the special properties of fluorine to drug design.[3] Next to β-fluorinated and perfluorinated amines, α-fluorinated amines have also been synthesized previously.[4,5] However, α-fluoroamines generally require an electron-withdrawing group at nitrogen for reasons of stability. In the case of saturated azaheterocyclic α-fluoroamines, C-fluorinated aziridines occupy a special place because of the intriguing combination of a strained ring system with an α-fluoroamine moiety. Only recently, the profound effect of fluorine on the reactivity of aziridines was studied by means of ab initio calculations, where it was found that a nucleophilic induced ring opening of the aziridine ring was predicted to occur much faster as compared to nonfluorinated analogues.[6] In contrast to 2-chlorinated aziridines, only very few literature data are available concerning 2-fluoroaziridines, most of which have been synthesized via carbene or nitrene additions to suitable imines or olefins.[7−11] Moreover, almost no 2,2-difluoroaziridines are known.[12−17] Surprisingly, 2,2-difluoroaziridines could not be synthesized via difluorocarbene additions to imines, while analogous reactions using dichloro- or chlorofluorocarbenes gave rise to the corresponding 2,2-dihalogenated aziridines.[18] Very recently, we developed an efficient pathway for the synthesis of 3-substituted 2-fluoro- and 2,2-difluoroaziridines via ring closure of suitable β-halogenated amines.[19] However, this methodology can only be applied for the synthesis of fluorinated aziridines with substitution at the 3-position. We herein describe an alternative synthetic method toward β-chloro-β-fluoroamines and the application of the latter compounds in the synthesis of new 3-unsubstituted fluorinated aziridines, which can be used as building blocks for further organic transformations and which provide experimental proof related to recently reported theoretical predictions.[6] It should be noted that the latter fluorinated aziridines cannot be synthesized via currently known methods such as fluorocarbene additions to C=N-bonds or starting from α-fluorinated imines, as this would imply the use of imines derived from formaldehyde or fluoroacetaldehyde, respectively, which cannot be handled readily.

To enable the synthesis of 3-unsubstituted 2-fluoroaziridines **5**, suitable β-chloro-β-fluoroamines **3** were required as starting compounds for ring-closure reactions (Scheme 1). Therefore, commercially available ethyl chlorofluoroacetate **1** was treated with primary amines in dichloromethane at room temperature for 16 h resulting in the corresponding α-chloro-α-fluorocarboxylic amides **2**. 2-Chloro-2-fluorocarboxylic amides **2**[20,21] were treated with a variety of reducing agents to obtain new chlorofluoroamines **3**. While the use of $LiAlH_4$ or chloroalane ($AlClH_2$) in THF or diethyl ether at room temperature gave no reaction or resulted in tarry reaction mixtures when reflux

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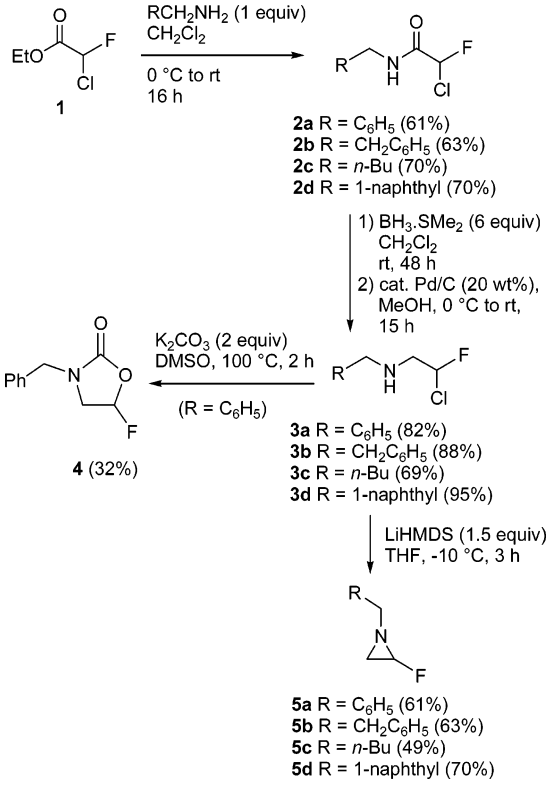
SCHEME 1. Synthesis of 2-Fluoroaziridines 5

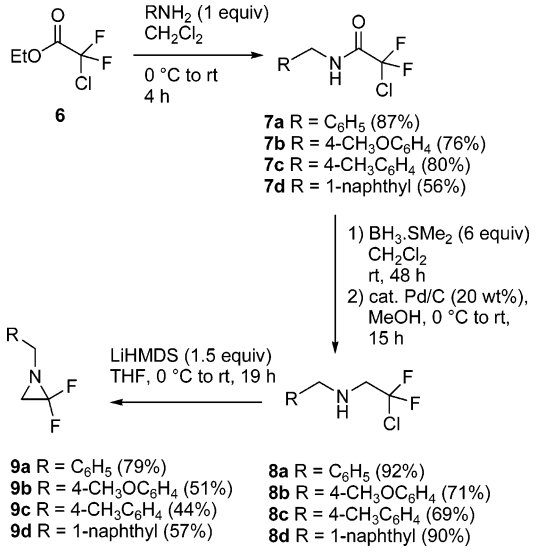


temperatures were used, the reduction of the amide using an excess of borane dimethyl sulfide complex in dichloromethane yielded the desired amines **3a**−**d** after treatment with Pd/C in methanol. The latter procedure is required to convert the formed stable amine−borane complex to the free amine,[22] which is not possible by standard treatments with aqueous 2 M HCl or 2 M NaOH. Analogous reactions using borane in THF or catechol borane in refluxing toluene did not improve the yield. The present method provides useful chlorinated β-fluoroamines.

The resulting new amines **3** were treated with various bases to induce an intramolecular substitution of chlorine. Although previous results indicated that heating of α-substituted β-halogenated amines with K_2CO_3 in DMSO could be used to synthesize 3-substituted 2-fluoroaziridines,[19] the reaction of *N*-benzyl-*N*-(2-chloro-2-fluoroethyl)amine **3a** with K_2CO_3 at 100 °C yielded the corresponding fluorinated oxazolidinone **4** instead. Probably, the formation of compound **4** is initiated by a nucleophilic attack of amine **3a** across the carbonyl moiety of CO_2 (from K_2CO_3), followed by ring closure. Experiments in which 2-fluoroaziridine **5a** was heated in DMSO to 100 °C, in the presence of an excess of K_2CO_3, mainly yielded starting material **5a** along with decomposition products. This experiment suggests that the formation of **4** probably does not proceed via a CO_2 insertion into aziridine **5a**.

After an evaluation of different bases (KOH in aqueous THF, LDA, or LiHMDS in THF) to synthesize aziridine **5a**, it was concluded that optimal yields were obtained when LiHMDS was used in THF at −10 °C. These reaction conditions resulted in new 1-substituted-2-fluoroaziridines **5a**−**d** which were purified by column chromatography on silica gel or via distillation. 2-Fluoroaziridines **5** were stable for several days at room

SCHEME 2. Synthesis of 2,2-Difluoroaziridines 9



temperature and for months at −20 °C. Indeed, from the few available data it could be concluded that fluorinated aziridines are more stable as compared to the chlorinated or brominated analogues.[23]

To extend this synthetic pathway toward the elusive class of 2,2-difluoroaziridines, 2-chloro-2,2-difluoroamides **7** were reduced toward β,β,β-trihalogenated amines **8** using borane dimethyl sulfide (Scheme 2). 2-Chloro-2,2-difluoroamides **7** were prepared by reaction of ethyl chlorodifluoroacetate **6** with primary amines. Indeed, when the trihaloamines **8** were treated with LiHMDS in THF, novel 2,2-difluoroaziridines were isolated after chromatography or distillation. Remarkably, no sharp signal could be detected in the ^{19}F NMR spectrum of the 2,2-difluoroaziridines **9a**−**d**, although other spectral data (^{13}C NMR and MS) clearly revealed that the two fluorine atoms were present. When the ^{19}F NMR spectrum of difluoroaziridine **9d** was recorded at room temperature, a very broad singlet (from −114.0 to −134.0 ppm, relative to $CFCl_3$) was observed. In addition, when the ^{19}F NMR spectrum was recorded at elevated temperatures (60 °C in DMSO-d_6), no clear NMR signal appeared. However, recording the spectrum at −40 °C gave rise to a splitting of the broad singlet to two doublets at −110.1 and −136.6 ppm, with a coupling constant J = 83.2 Hz. The latter chemical shift is in good agreement with spectral data of 3-substituted 2,2-difluoroaziridines, which we described recently.[19] The above-described magnetic resonance characteristics of the fluorine atoms at the 2-position of compounds **9a**−**d** are caused by the fact that the rate of inversion at the nitrogen atom (at 25 °C) is comparable to the time scale at which the NMR spectrum is recorded, while at −40 °C, this inversion rate is slowed down resulting in two different ^{19}F NMR signals for both fluorine atoms.[16,24] In addition, elemental analysis data of the crystalline compound **9d** (Anal. Calcd for $C_{13}H_{11}F_2N$: C, 71.22; H, 5.06; N, 6.39. Found: C, 71.27; H, 5.06; N, 6.44) further supported the correct structure assignment.

Having in hand this new method for the synthesis of fluorinated aziridines, a preliminary reactivity study was

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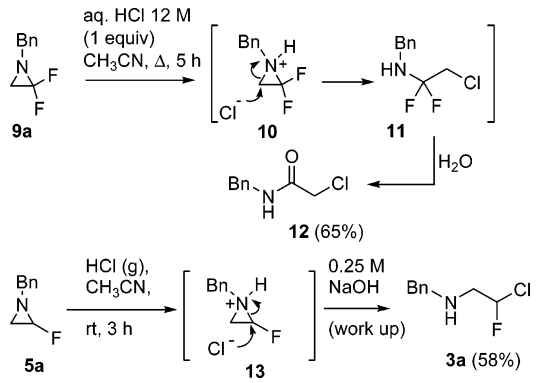
SCHEME 3. Reaction of Fluoroaziridines 5a and 9a with Hydrogen Chloride



performed to study the reaction of the obtained aziridines with nucleophiles (with and without *N*-protonation, Scheme 3 and 4). Apart from a very recent study of 3-fluoro-2,2-diphenylaziridines, which were synthesized via fluorocarbene additions to benzophenone imines, no data on the chemical properties of fluorinated aziridines have been published.[25] The latter study revealed that 3-fluoro-2,2-diphenylaziridines undergo a ring opening when reacted with aqueous HCl resulting in 2-hydroxy-2,2-diphenylacetaldehyde. In order to evaluate the reactivity of 3-unsubstituted difluoro- and monofluoroaziridines, 1-benzyl-2,2-difluoroaziridine **9a** was treated with aqueous 12 M HCl (Scheme 3). It is proposed that this reaction resulted in a protonation of the nitrogen atom, followed by an attack of chloride at the 3-position. After ring opening, the intermediate α,α-difluoroamine **11** gives rise to *N*-benzyl-2-chloroacetamide **12**[26] via an intermediate α-chloroimidoyl fluoride and hydrolysis. The same reaction with monofluoroaziridine **5a** resulted in an attack of chlorine at the 2-position after protonation of the nitrogen atom. Because the reaction conditions using aqueous 12 M HCl yielded amine **3a** in very low yield (<10%), hydrogen chloride gas was bubbled through a solution of aziridine **5a** in CH_3CN. This reaction resulted in amine **3a** in a better yield (58%) after basic workup. The difference in reactivity between di- and monofluoroaziridines **9a** and **5a** is related to sterical hindrance in combination with the specific electronic properties of fluorinated aziridines.

In a second reaction, both aziridines **9a** and **9** were treated with sodium methoxide in methanol (Scheme 4). In the case of difluoroaziridine **9a**, a ring opening can occur, induced by nucleophilic attack of methoxide at the 3-position (pathway a). The resulting intermediate **14** is unstable in methanol and results in the formation of imidate **15**, which was easily converted to 2-methoxyacetamide **16**[27] by hydrolysis. An alternative reaction mechanism (pathway b) proceeds via an initial expulsion of fluoride by nitrogen, followed by a nucleophilic attack at the 2-position of the intermediate azirinium ion **17**. A second expulsion of fluoride results in methoxyazirinium species **19** which can undergo either nucleophilic attack of methoxide at the 3-position in a second-order process or a ring opening toward intermediate **20** which is subsequently trapped by methanol. Attempts to react 1-benzyl-2-fluoroaziridine **5a** with sodium methoxide under the same reaction conditions as described above, yielded a complex reaction mixture in which only a small amount of 2,2-dimethoxyamine **22** could be detected in the ^{1}H NMR spectrum. However, when the aziridine **5a** was simply heated in methanol in a sealed vessel to 100 °C, a clean conversion toward *N*-benzyl-2,2-dimethoxyethylamine **22**,[28] which is a result of the nucleophilic attack of methanol at the 2-position, was obtained. Also in this case an alternative reaction pathway (pathway b) is proposed via initial expulsion of a fluoride anion and reaction of the intermediate azirinium fluoride **23** with methanol. The resulting 2-methoxyaziridine can also yield in this case dimethoxyamine **22** via a second-order process or via a first-order reaction by ring opening of aziridine **24** to **25**, followed by reaction with methanol.

In conclusion, the ring closure of β-chloro-β-fluorinated amines has proven to be an efficient and straightforward method for the synthesis of the elusive class of 2-fluorinated aziridines. The latter compounds are remarkably stable for months at −20 °C and can easily be used for various reactions without thermal degradation. Upon treatment of 2-fluoroaziridines with nucleophiles, it was observed that reaction occurred at the 2-position, which is the most electrophilic carbon atom, while in the case of 2,2-difluoroaziridines, the nucleophilic attack took place at the 3-position.

Experimental Section

***N*-Benzyl-2-chloro-2-fluoroacetamide 2a**.[20] This procedure can be used as a general procedure for the synthesis of amides **2a**−**d** and **7a**−**d**. To a solution of 1.00 g (7.12 mmol) of ethyl 2-chloro-2-fluoroacetate in 25 mL of CH_2Cl_2 was added 0.76 g (1 equiv, 7.12 mmol) of benzylamine at 0 °C. The cooling bath was removed and the mixture was stirred at room temperature for 16 h. The solvents were evaporated in vacuo, and the resulting crude amide **2a** was recrystallized from a 9:1 mixture of hexane and diethyl ether. Yield: 61%. Mp: 56.2−56.7 °C (no literature data reported).[20] ^{1}H NMR (300 MHz, $CDCl_3$): δ 4.47 (1H, dd, J = 15.2, 5.8 Hz), 4.52 (1H, dd, J = 15.2, 5.8 Hz), 6.30 (1H, d, J = 50.3 Hz), 6.70 (1H, s(b)), 7.25−7.39 (5H, m). ^{13}C NMR (75 MHz, $CDCl_3$): δ 43.6, 94.1 (d, J = 256.1 Hz), 2 × 127.9, 128.0, 2 × 128.9, 136.9, 164.3 (d, J = 21.9 Hz). ^{19}F NMR (282 MHz, $CDCl_3$): δ −143.8 (1F, d, J = 50.3 Hz). IR (KBr): ν_{max} 3305, 1671, 1549, 1059 cm^{-1}. MS ES+ (*m*/*z*): 202/204 (M + H$^+$, 100). Copies of ^{1}H NMR and ^{13}C NMR spectra are included in Chapter III of the Supporting Information. Experimental details for analogous compounds **2b**−**d** and **7a**−**d** are included in the Supporting Information.

***N*-Benzyl-2-chloro-2-fluoroethanamine 3a.** *N*-Benzyl-2-chloro-2-fluoroacetamide **2a** (1.01 g; 5.00 mmol) was dissolved in 25 mL of CH_2Cl_2, and subsequently 1.42 mL (3 equiv, 15.00 mmol) of borane dimethyl sulfide complex was added. The resulting mixture was stirred at room temperature. After 24 h, a second amount of 1.42 mL (3 equiv) of borane dimethyl sulfide was added, and stirring was continued for another 24 h. The reaction was quenched by gently adding 50 mL of aqueous methanol (1:1) while the reaction temperature was maintained at 25 °C by the use of an ice bath (exothermic reaction: evolution of hydrogen gas), the mixture was extracted with dichloromethane (5 × 50 mL). After drying ($MgSO_4$) and evaporation of the solvent, the obtained crude mixture was dissolved in 50 mL of methanol and 0.20 g of Pd/C (20 wt %) was added at 0 °C. The mixture was stirred for 15 h at room temperature. After filtration of the catalyst and evaporation of the solvent, amine **3a** was purified via column chromatography over a

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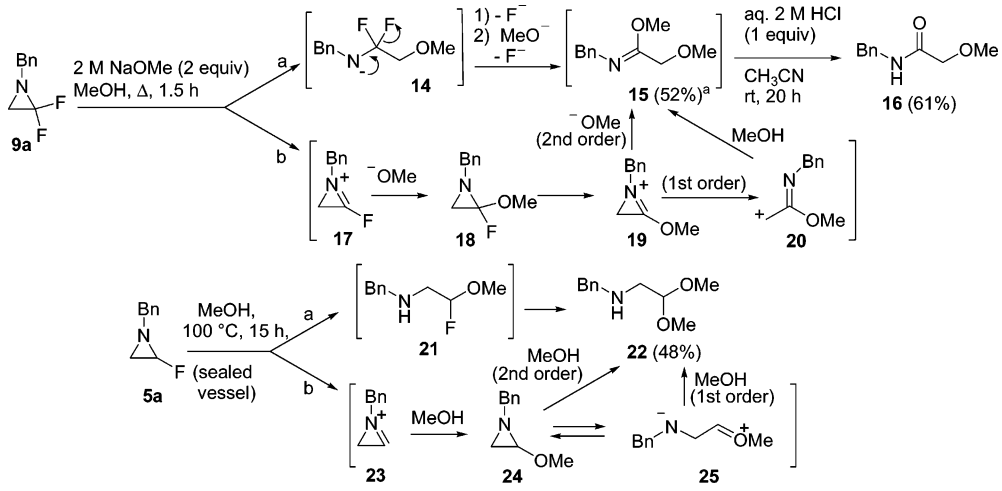
SCHEME 4. Reaction of Fluoroaziridines 5a and 9a with Sodium Methoxide or Methanol[a]



[a] Compound **15** could not be obtained in pure form due to hydrolysis (purity 89%).

short plug of silica gel (hexane/EtOAc/Et_3N 96/3.9/0.1, $R_f = 0.10$). Yield: 82%. ^{1}H NMR (300 MHz, $CDCl_3$): δ 1.70 (1H, s(b)), 3.07 (1H, ddd, J = 4.2, 13.8, 21.7 Hz), 3.21 (1H, ddd, J = 6.1, 13.8, 14.3 Hz), 3.87 (2H, s), 6.20 (1H, ddd, J = 4.2, 6.1, 50.7 Hz), 7.24−7.38 (5H, m). ^{13}C NMR (75 MHz, $CDCl_3$): δ 53.3, 55.3 (d, J = 21.9 Hz), 101.6 (d, J = 242.3 Hz), 127.5, 2 × 128.2, 2 × 128.7, 139.7. ^{19}F NMR (282 MHz, $CDCl_3$): δ −137.8 (1F, dt(b), J = 50.7, 18.0 Hz). IR (NaCl): ν_{max} 3300, 1603, 1496, 1454 cm^{-1}. MS ES+ (*m*/*z*): 188/190 (M + H$^+$, 100). Copies of ^{1}H NMR and ^{13}C NMR spectra are included in Chapter III of the Supporting Information. Spectral data for amines **3b**−**d** and **8a**−**d** are included in the Supporting Information.

Synthesis of 5-Fluoro-3-benzyloxazolidin-2-one 4. To a solution of 0.50 g (2.67 mmol) of amine **3a** in 10 mL of DMSO was added 0.74 g (5.34 mmol, 2 equiv) of K_2CO_3. The heterogeneous mixture was placed in a preheated oil bath at 100 °C. Stirring was continued for 2 h. After the reaction mixture was cooled to room temperature, 20 mL of water was added and extraction was performed using CH_2Cl_2 (3 × 50 mL). The organic layer was washed three times with brine (3 × 25 mL) and dried over $MgSO_4$. After filtration, the solvent was evaporated, yielding oxazolidinone **4**. The crude compound was purified via column chromatography (hexane/EtOAc/Et_3N 98/1.9/0.1, $R_f = 0.42$). Yield: 32%. ^{1}H NMR (300 MHz, $CDCl_3$): δ 3.46 (1H, ddd, J = 24.6, 11.3, 0.6 Hz), 3.62 (1H, ddd, J = 35.1, 11.3, 4.9 Hz), 4.49 (2H, s), 6.08 (1H, ddd, J = 64.1, 4.9, 0.6 Hz), 7.25−7.41 (5H, m). ^{13}C NMR (75 MHz, $CDCl_3$): δ 47.8, 50.6 (d, J = 26.5 Hz), 103.3 (d, J = 233.1 Hz), 2 × 128.1, 128.4, 2 × 129.1, 134.8, 155.4. ^{19}F NMR (282 MHz, $CDCl_3$): δ-117 (1F, m). IR (NaCl): ν_{max} 1785, 1763 cm^{-1}. MS ES+ *m*/*z*: 213 (M + NH_4^+, 100), 196 (M + H$^+$, 15). Copies of ^{1}H NMR and ^{13}C NMR spectra are included in Chapter III of the Supporting Information.

1-Benzyl-2-fluoroaziridine 5a. This procedure can be used as a general procedure for the synthesis of fluorinated aziridines **5a**−**d** and **9a**−**d**. To a solution of 0.50 g (2.67 mmol) of amine **3a** in 25 mL of dry THF was added 4.00 mL (4.00 mmol, 1.5 equiv) of a 1 M solution of LiHMDS in hexane at −10 °C. The reaction mixture was stirred at the same temperature for 3 h. After addition of ice−water to the mixture, the solution was poured in 25 mL of water and extracted with diethyl ether (3 × 50 mL). After drying ($MgSO_4$) and filtration of the drying agent, the solvent was evaporated in vacuo yielding crude 1-benzyl-2-fluoroaziridine **5a**. The obtained aziridine was purified via column chromatography (hexane/EtOAc/Et_3N 98/1.9/0.1, R_f = 0.13). Yield: 61%. ^{1}H NMR (300 MHz, $CDCl_3$): δ 1.47 (1H, d(b), J = 4.1 Hz), 2.20 (1H, m), 3.44 (1H, dd, J = 13.6, 3.0 Hz), 3.72 (1H, dd, J = 13.6, 1.9 Hz), 4.68 (1H, ddd, J = 71.7, 4.1, 1.1 Hz), 7.24−7.36 (5H, m). ^{13}C NMR (75 MHz, $CDCl_3$): δ 32.7 (d, J = 12.7 Hz), 59.8, 81.7 (d, J = 237.7 Hz), 127.5, 2 × 128.1, 2 × 128.6, 137.4. ^{19}F NMR (282 MHz, $CDCl_3$): δ-174.8 (1F, d(b), J = 71.7 Hz). IR (NaCl): ν_{max} 1454, 1359, 1124 cm^{-1}. MS ES+ *m*/*z*: 152 (M + H$^+$, 100). Anal. Calcd for $C_9H_{10}FN$: C, 71.50; H, 6.67; N, 9.26. Found: C, 71.84; H, 6.90; N, 9.11. Copies of ^{1}H NMR and ^{13}C NMR spectra are included in Chapter III of the Supporting Information. Spectral data for amines **5b**−**d** and **9a**−**d** are included in the Supporting Information.

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Supporting Information Available: General experimental conditions and spectroscopic data for all new compounds. This material is available free of charge via the Internet at http://pubs.acs.org.

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