Generation and reaction of benzylammonium *N*-methylides with *N*-cyanomethyl or *N*-(ethoxycarbonylalkyl) groups

Chen Zhang, Yasuhiro Maeda, Naohiro Shirai and Yoshiro Sato*

Faculty of Pharmaceutical Sciences, Nagoya City University, Tanabe-dori, Mizuho-ku, Nagoya 467, Japan

***N*-Cyanomethyl-, *N*-ethoxycarbonylmethyl- and *N*-(3-ethoxycarbonylpropyl)-*N*-methylbenzylammonium *N*-methylides 6a,b and 22 have been generated by fluoride ion-induced desilylation of the corresponding *N*-(trimethylsilylmethyl)benzylammonium salts 5a,b and 21. Sommelet–Hauser 7, 23 and Stevens 8, 24 rearrangement products were obtained from 6a and 22, but not from 6b.**

Introduction

Ammonium ylides are generally prepared by removal of an α-proton from quaternary ammonium cations with strong bases.[1] However, the selective formation of *N*-methylides from methylammonium salts with electron-withdrawing groups is difficult because an adjacent acidic hydrogen is removed more quickly than an *N*-methyl hydrogen. For example, reaction of *N*-cyanomethyl-*N*,*N*-dimethylbenzylammonium chloride **1a** with sodium methoxide gave a mixture of *N*,*N*-dimethyl-α-cyano-2-methylbenzylamine **3a** (Sommelet–Hauser rearrangement product) and 2-dimethylamino-3-phenylpropiononitrile **4a** (Stevens rearrangement product) (Scheme 1, Table 1, entry 1).[2] Reaction of *N*-ethoxycarbonylmethyl-*N*,*N*-dimethylbenzylammonium chloride **1b** with potassium *tert*-butoxide gave 2-dimethylamino-3-phenylpropionic acid **4b** (R = CO_2H) (entry 5).[3] These are all isomerization products of *N*-cyanomethylide **2a** and *N*-ethoxycarbonylmethylide **2b**.

Fluoride ion-induced desilylation of 1-trimethylsilylalkylammonium cations is suitable for regio- and stereo-selective *N*-methylide formation.[4,5] Under these conditions, it is possible to generate *N*-methylides with functional groups. We report here the generation and reaction of benzylammonium *N*-methylides with a *N*-cyanomethyl, *N*-ethoxycarbonylmethyl or *N*-(3-ethoxycarbonylpropyl) group.

Results and discussion

N-Cyanomethyl- and *N*-ethoxycarbonylmethyl-*N*-methyl-*N*-(trimethylsilylmethyl)benzylammonium bromides **5a** and **5b** were prepared by quaternization of *N*-methyl-*N*-(trimethylsilylmethyl)benzylamine with bromoacetonitrile or ethyl bromoacetate. Treatment of **5a** with caesium fluoride in DMF at room temperature gave a mixture of **3a**, *N*-cyanomethyl-*N*-methyl-2-methylbenzylamine **7a** (Sommelet–Hauser rearrangement product) and *N*-(2-cyanoethyl)-*N*-methylbenzylamine **8a** (Stevens rearrangement product) (entry 3). When the reaction was carried out at 0 °C, *N*-methyl-*N*-(trimethylsilylmethyl)-α-cyano-2-methylbenzylamine **11a** was formed (entry 2). However, the formation of **3a** and **11a** decreased at 60 °C (entry 4). Thus, at lower temperature, the rearrangement of the *N*-methylide **6a** to **7a** and **8a** competed with intramolecular proton transfer to **2a** and intermolecular proton transfer to **10a**, which are precursors of **3a** and **11a**, respectively. Thus, rearrangement of **6a** proceeded preferentially at 60 °C, while the ratio of **8a** to **7a** increased with increasing temperature.[1a]

The reaction of *N*,*N*-dimethyl-*N*-cyanomethyl-α-(trimethylsilyl)benzylammonium bromide **13** with caesium fluoride gave selectively 3-dimethylamino-3-phenylpropiononitrile **15** (Stevens rearrangement product) (Scheme 2). The lack of **15** in the reaction products of **5a** with caesium fluoride shows that there is no ylide conversion from **6a** to **14**.

Table 1 Reaction of *N*-cyanomethyl- or *N*-ethoxycarbonylmethyl-*N*-methylbenzylammonium bromide **5a**,**b** with CsF for 3 h in DMF

		R	Reaction temp./°C	Total yield (%)	Product ratio[a]					
					3	**4**	**7**	**8**	**11**	**12**
1	**1a**[b]	CN	35	72	85	15	0	0	0	0
2	**5a**	CN	0	61	17	00	19	31	33	0
3	**5a**	CN	room temp.	68	22	00	26	52	0	0
4	**5a**	CN	60	64	0	00	33	67	0	0
5	**1b**[c]	CO_2Et	80	33[d]	0	100	0	0	0	0
6	**5b**	CO_2Et	room temp.	53	0	30	8	0	0	62
7	**5b**	CO_2Et	60	59	0	45	7	0	0	48

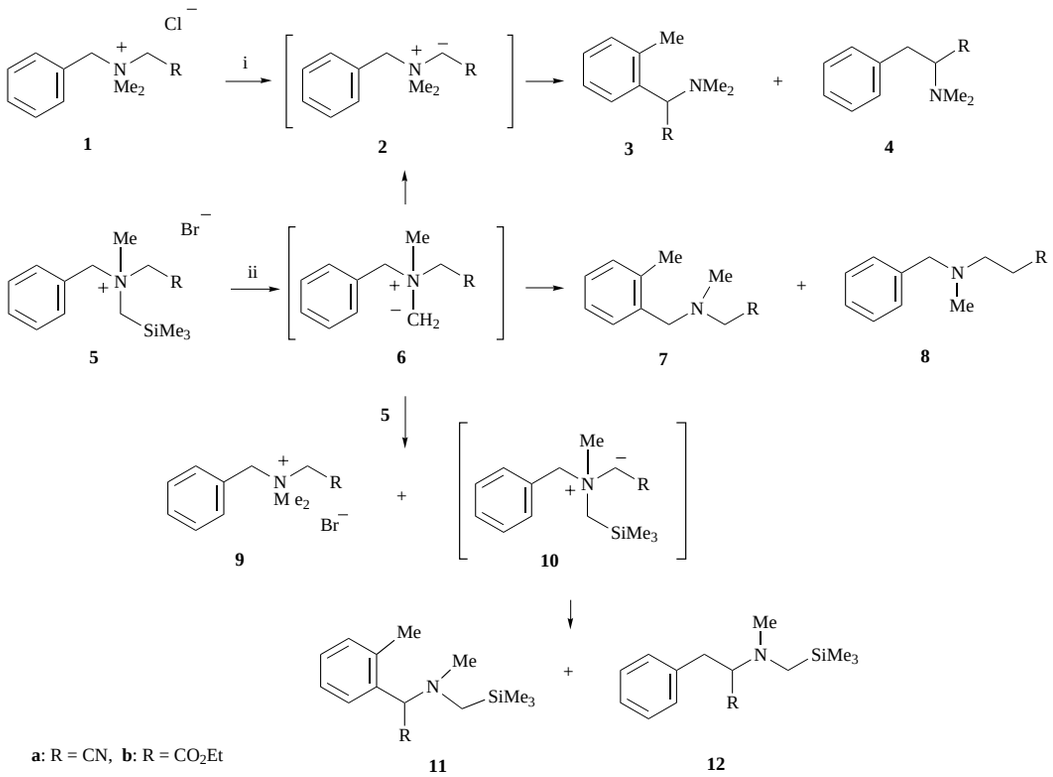
[a] Ratio of the products determined by integration of the ^1H signals of the 500 MHz NMR spectrum. [b] Ref. 2. [c] Ref. 3. [d] 2-Dimethylamino-3-phenylpropionic acid.

The reaction of **5b** with caesium fluoride at room temperature or 60 °C gave mainly ethyl 2-dimethylamino-3-phenylpropionate **4b** and ethyl 2-[methyl(trimethylsilylmethyl)amino]-3-phenylpropionate **12b**, while the yield of *N*-ethoxycarbonylmethyl-*N*-methyl-2-methylbenzylamine **7b** was very low (entries 6, 7). Thus, ylide conversion from **6b** into **2b** and proton transfer from **6b** to **10b** occurred more quickly than rearrangement of **6b**.

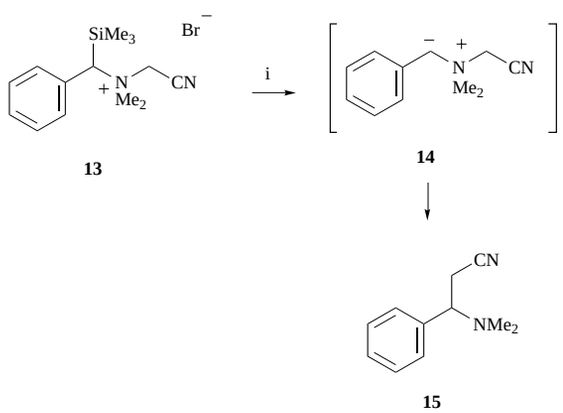
Vedejs and Martinez[6] reported that cinnamyl(ethoxycarbonylmethyl)sulfonium methylide **17** generated at 20 °C in acetonitrile isomerized mainly to a [2,3] sigmatropic migration product **18** rather than undergo proton transfer to **19** which was subsequently converted into **20** (Scheme 3). The difference in chemical behaviour of **6b** and **17** may be the result of the difference between of *N*-methylide and *S*-methylide.

The reaction of *N*-(3-ethoxycarbonylpropyl)-*N*-methyl-*N*-(trimethylsilylmethyl)benzylammonium hexafluorophosphate **21** with caesium fluoride gave ethyl 4-[methyl(2-methylbenzyl)amino]butyrate **23** (Sommelet–Hauser rearrangement product) and ethyl 4-[methyl(2-phenylethyl)amino]butyrate **24** (Stevens product) in good yield (ratio, 2:1) at 0 °C with a small amount of bibenzyl **25** (Scheme 4).

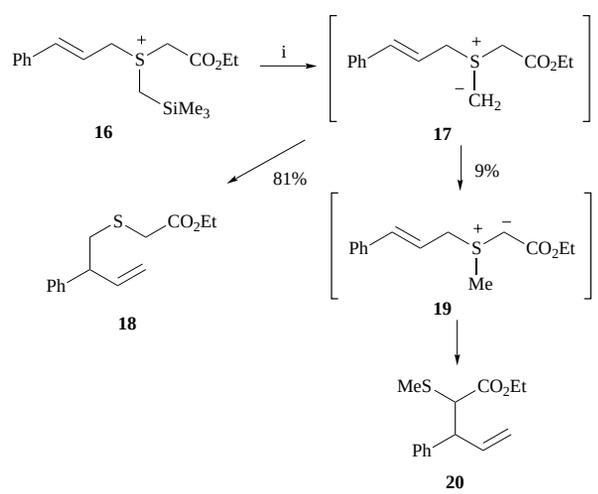
Thus, the rearrangement of benzylammonium *N*-methylides was successful for **6a** and **22**, but not for **6b**. It is interesting that the main rearrangement product of **2a** was a Sommelet–Hauser rearrangement product **3a** while that of **2b** was a Stevens product **4b**. We are currently attempting to identify the precise reason for this difference.



Scheme 1 *Reagents and conditions*: i, for **a**; R = CN, NaOMe, Et_2O, 30–35 °C; for **b**; R = CO_2Et, $KOBu^t$, C_6H_6, reflux, 6 h; ii, CsF, DMF, room temp., 3 h



Scheme 2 *Reagents and conditions*: i, CsF, DMF, room temp., 3 h



Scheme 3 *Reagents and conditions*: i, CsF, MeCN, 20 °C, 16 h

Experimental

All reactions were carried out under N_2. DMF was dried by distillation from BaO under reduced pressure. CsF was dried over P_2O_5 at 190 °C under reduced pressure. Distillation was carried out using a Kugelrohr distillation apparatus. All melting and boiling points are uncorrected.

N-Cyanomethyl-*N*-methyl-*N*-(trimethylsilylmethyl)benzylammonium bromide 5a

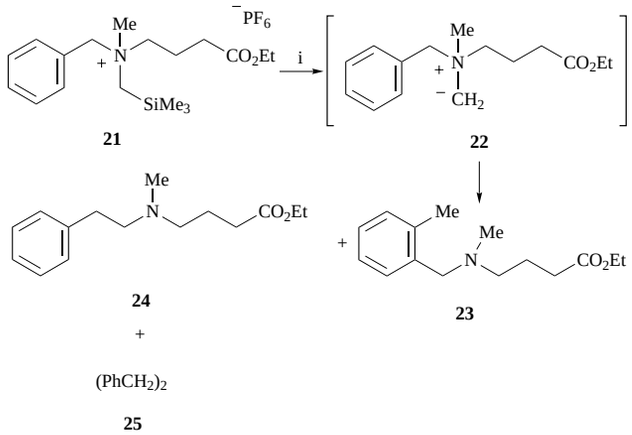
A solution of *N*-methyl-*N*-(trimethylsilylmethyl)benzylamine[7] (3.52 g, 17.0 mmol) and bromoacetonitrile (2.55 g, 20 mmol) in acetone (10 cm^3) was heated at reflux for 20 h. The precipitated crystals were separated and recrystallized from ethanol–hexane to give the *title salt* **5a** (4.8 g, 75%), mp 144 °C (Found: C, 51.2; H, 7.0; N, 8.6. $C_{14}H_{23}BrN_2Si$ requires C, 51.4; H, 7.1; N, 8.6%); δ_H(270 MHz; $CDCl_3$) 0.42 (9 H, s), 3.45 and 3.53 (2 H, ABq, *J* 15.1), 3.50 (3 H, s), 4.93 and 5.34 (2 H, ABq, *J* 12.4), 5.31 and 5.52 (2 H, ABq, *J* 16.5) and 7.49–7.74 (5 H, m); ν_{max}(KBr)/cm^{-1} 2250, 1468, 1254, 856 and 737.

N-Ethoxycarbonylmethyl-*N*-methyl-*N*-(trimethylsilylmethyl)-benzylammonium bromide 5b

In a manner similar to that described above, *N*-methyl-*N*-(trimethylsilylmethyl)benzylamine (3.52 g, 17.0 mmol) and ethyl bromoacetate (2.90 g, 17.1 mmol) were allowed to react to give the *title salt* **5b** (5.50 g, 86%), mp 155–156 °C (Found: C, 51.3; H, 7.5; N, 4.1. $C_{16}H_{28}BrNO_2Si$ requires C, 51.3; H, 7.5; N, 3.7%); δ_H(270 MHz; $CDCl_3$) 0.31 (9 H, s), 1.32 (3 H, t, *J* 7.2), 3.37 and 3.77 (2 H, ABq, *J* 14.9), 3.44 (3 H, s), 4.28 (2 H, q, *J* 7.2), 4.49 and 4.80 (2 H, ABq, *J* 17.2), 5.01 and 5.44 (2 H, ABq, *J* 12.2) and 7.56–7.45 (5 H, m); ν_{max}(KBr)/cm^{-1} 1751, 1209 and 854.

Reaction of 5a with CsF

Compound **5a** (690 mg, 2 mmol) was placed in a 20 cm^{-3} flask equipped with a magnetic stirrer, a septum and a test tube which was connected to the flask by a short piece of rubber



Scheme 4 *Reagents and conditions*: i, CsF, DMF, room temp., 3 h

tubing. CsF (912 mg, 6.0 mmol) was placed in the test tube. The apparatus was dried under reduced pressure and flushed with N_2. DMF (10 cm^3) was added to the flask with a syringe and then CsF was added from the test tube. The mixture was stirred for 3 h at 0 °C, room temp. or 60 °C (see Table 1), after which it was quenched with 1% aqueous $NaHCO_3$ (100 cm^3) and extracted with Et_2O. The extract was washed with water, dried ($MgSO_4$), filtered and concentrated under reduced pressure. The residue was distilled at 95–125 °C (0.6 mmHg) and the distillate (213 mg, 238 mg or 222 mg) was chromatographed on a silica gel column with hexane–Et_2O (1.5:1) as the eluent to give *N*,*N*-dimethyl-α-cyano-2-methylbenzylamine[2] **3a**, *N*-cyanomethyl-*N*-methyl-2-methylbenzylamine **7a**, *N*-(2-cyanoethyl)-*N*-methylbenzylamine[8] **8a** and *N*-methyl-*N*-trimethylsilylmethyl-α-cyano-2-methylbenzylamine **11a**, which were purified by redistillation. The product ratio was determined from the proton ratios of an ^{1}H NMR spectrum of the mixture.

Compound **3a**: bp 90–92 °C (0.4 mmHg); δ_H(270 MHz; $CDCl_3$) 2.31 (6 H, s), 2.39 (3 H, s), 4.84 (1 H, s) and 7.20–7.54 (4 H, m).

Compound **7a**: bp 75–80 °C (0.5 mmHg) (Found: C, 75.6; H, 8.1; N, 15.9. $C_{11}H_{14}N_2$ requires C, 75.8; H, 8.1; N, 16.1%); δ_H(270 MHz; $CDCl_3$) 2.36 (3 H, s), 2.43 (3 H, s), 3.42 (2 H, s), 3.59 (2 H, s) and 7.14–7.53 (4 H, m); ν_{max}(film)/cm^{-1} 2951, 2234, 1460, 1034 and 754.

Compound **8a**: bp 115 °C (0.7 mmHg); the structure was confirmed by comparison with an authentic sample prepared from *N*-methylbenzylamine with 3-bromopropiononitrile.[8]

Compound **11a**: bp 90–95 °C (0.3 mmHg) (Found: C, 68.1; H, 9.1; N, 11.1. $C_{14}H_{22}N_2Si$ requires C, 68.2; H, 9.0; N, 11.4%); δ_H(270 MHz; $CDCl_3$) −0.01 (9 H, s), 1.93 and 2.04 (2 H, ABq, *J* 14.0), 2.24 (3 H, s), 2.39 (3 H, s), 4.85 (1 H, s) and 7.18–7.53 (4 H, m); ν_{max}(film)/cm^{-1} 2957, 2230, 1250, 856 and 750.

Reaction of 5b and CsF

In a manner similar to that described above, **5b** (788 mg, 2 mmol) was treated with CsF (912 mg, 6 mmol) in DMF (10 cm^3) at room temp. or 60 °C (see Table 1). The Et_2O extract was distilled at 90–115 °C (0.2 mmHg) and the distillate (281 mg or 302 mg) was chromatographed on a silica gel column with hexane–Et_2O (4:1) to give ethyl 2-dimethylamino-3-phenylpropionate[9] **4b**, ethyl [methyl(2-methylbenzyl)amino]acetate **7b** and ethyl 2-[methyl(trimethylsilylmethyl)amino]-3-phenylpropionate **12b**, which were purified by redistillation. The product ratio was determined from the proton ratios of an ^{1}H NMR spectrum of the mixture.

Compound **7b**: bp 90–95 °C (0.35 mmHg) (Found: C, 70.2; H, 8.5; N, 6.5. $C_{13}H_{19}NO_2$ requires C, 70.5; H, 8.7; N, 6.3%); δ_H(270 MHz; $CDCl_3$) 1.27 (3 H, t, *J* 7.3), 2.37 (3 H, s), 2.38 (3 H, s), 3.26 (2 H, s), 3.66 (2 H, s), 4.16 (2 H, q, *J* 7.3) and 7.10–7.30 (4 H, m); ν_{max}(film)/cm^{-1} 1738, 1182, 1046 and 745.

Compound **12b**: bp 90–100 °C (0.35 mmHg) (Found: C, 65.9; H, 9.3; N, 4.7. $C_{16}H_{27}NO_2Si$ requires C, 65.5; H, 9.3; N, 4.8%); δ_H(270 MHz; $CDCl_3$) 0.01 (9 H, s), 1.18 (3 H, t, *J* 7.3), 1.95 and 2.18 (2 H, ABq, *J* 15.2), 2.37 (3 H, s), 2.85 (1 H, dd, *J* 13.5, 5.9), 3.05 (1 H, dd, *J* 13.5, 8.9), 3.40 (1 H, dd, *J* 8.9, 5.9), 4.08 (2 H, m) and 7.14–7.30 (5 H, m); ν_{max}(film)/cm^{-1} 2955, 1730 and 854.

N,*N*-Dimethyl-*N*-cyanomethyl-α-(trimethylsilylbenzyl)-ammonium bromide 13

A solution of *N*,*N*-dimethyl-*N*-[α-(trimethylsilyl)benzyl]-amine[10] (950 mg, 4.6 mmol) and bromoacetonitrile (782 mg, 6.5 mmol) in DMF (10 cm^3) was stirred at room temp. for 13 h. The precipitated crystals were separated to give the *title salt* **13** (1.29 g, 85%), mp 188–189 °C (Found: C, 51.4; H, 7.0; N, 8.3. $C_{14}H_{23}BrN_2Si$ requires C, 51.4; H, 7.1; N, 8.6%); δ_H(500 MHz; $CDCl_3$) 0.32 (9 H, s), 3.42 (3 H, s), 3.69 (3 H, s), 4.90 and 5.14 (2 H, ABq, *J* 16.5), 4.93 (1 H, s), 7.42 (1 H, m), 7.54 (3 H, m) and 7.60 (1 H, m); ν_{max}(KBr)/cm^{-1} 1250 and 845.

Reaction of 13 with CsF

In a manner similar to that described for **5a**, a mixture of **13** (333 mg, 1 mmol) and CsF (800 mg, 5.3 mmol) in DMF (10 cm^3) was prepared and stirred at room temp. for 2 days, after which it was quenched with water (200 cm^3) and extracted with Et_2O (4 × 50 cm^3) and EtOAc (3 × 50 cm^3). The combined extracts were extracted with 0.5 M HCl (2 × 10 cm^3). The acid extract was washed with Et_2O, made alkaline with Na_2CO_3 and extracted with Et_2O. The extract was washed with saturated aqueous NaCl, dried ($MgSO_4$) and concentrated under reduced pressure. The residue was distilled to give 3-dimethylamino-3-phenylpropiononitrile **15** (88 mg, 50%), bp 145–155 °C (7.5 mmHg) (Found: C, 75.4; H, 8.2; N, 16.0. $C_{11}H_{14}N_2$ requires C, 75.8; H, 8.1; N, 16.1%); δ_H(500 MHz; $CDCl_3$) 2.21 (6 H, s), 2.77 (1 H, dd, *J* 7.3, 17.1), 2.81 (1 H, dd, *J* 5.8, 17.1), 3.53 (1 H, *J* 5.8, 7.3), 7.31 (3 H, m) and 7.36 (2 H, m); ν_{max}(film)/cm^{-1} 2240 and 700.

N-[3-(Ethoxycarbonyl)propyl]-*N*-methyl-*N*-(trimethylsilylmethyl)benzylammonium hexafluorophosphate 21

A mixture of *N*-methyl-*N*-(trimethylsilyl)methylamine (5.90 g, 50 mmol), ethyl 4-bromobutyrate (9.81 g, 50 mmol) and K_2CO_3 (13.83 g, 100 mmol) in benzene (50 cm^3) was heated at reflux for 20 h. The reaction mixture was then filtered and extracted with 1 M HCl (100 cm^3). The acid extract was washed with Et_2O, made alkaline with NaOH and extracted with Et_2O. The extract was washed with water, dried ($MgSO_4$) and concentrated under reduced pressure. The residue was distilled to give ethyl 4-[methyl(trimethylsilylmethyl)amino]butyrate (9.59 g, 41.5%), bp 75–77 °C (0.6 mmHg) (Found: C, 56.7; H, 10.9; N, 6.1. $C_{11}H_{25}NO_2Si$ requires C, 57.1; H, 10.9; N, 6.1%); δ_H(270 MHz; $CDCl_3$) 0.05 (9 H, s), 1.26 (3 H, t, *J* 7.3), 1.76 (2 H, dt, *J* 6.9, 7.6), 1.86 (2 H, s), 2.19 (3 H, s), 2.30 (2 H, t, *J* 6.9), 2.33 (2 H, t, *J* 7.6) and 4.12 (2 H, t, *J* 7.3); ν_{max}(film)/cm^{-1} 1738, 1250 and 856.

A solution of ethyl 4-[methyl(trimethylsilylmethyl)amino]-butyrate (3.0 g, 13.0 mmol) and benzyl bromide (2.29 g, 13.0 mmol) in acetone (10 cm^3) was heated at reflux for 20 h and then evaporated *in vacuo*. The residue was added to a solution of NH_4PF_6 (2.54 g, 15 mmol) in 50% aqueous MeOH (10 cm^3), stirred for 3 h and extracted with $CHCl_3$ (4 × 50 cm^3). The extract was dried ($MgSO_4$) and concentrated under reduced pressure. The residue was recrystallized from EtOH to give the *title compound* **21** (5.47 g, 96%), mp 116–117 °C (Found: C, 46.2; H, 7.1; N, 3.0. $C_{18}H_{32}F_6NO_2PSi$ requires C, 46.2; H, 6.9; N, 3.0%); δ_H(500 MHz; $CDCl_3$) 0.29 (9 H, s), 1.25 (3 H, t, *J* 7.3), 2.17 (2 H, m), 2.46 (2 H, m), 2.98 (2 H, dd, *J* 18.3, 15.3), 2.99 (3 H, s), 3.32 (2 H, m), 4.13 (2 H, q, *J* 7.3), 4.41 (1 H, d, *J* 13.4), 4.46 (1 H, d, *J* 13.4) and 7.46–7.53 (5 H, m); ν_{max}(Nujol)/cm^{-1} 1736, 1260 and 841.

Reaction of 21 and CsF

In a manner similar to that described for **5a**, a solution of **21** (880 mg, 2 mmol) and CsF (912 mg, 6.0 mmol) in DMF (10 cm^3) was stirred at 0 °C for 3 h and then worked up. The Et_2O extract was distilled at 90–125 °C (0.5 mmHg) and the distillate (329 mg) was chromatographed on a silica gel column with hexane–Et_2O (1.5:1) to give ethyl 4-[methyl(2-methylbenzyl)-amino]butyrate **23**, ethyl 4-[methyl(2-phenylethyl)amino]-butyrate **24** and bibenzyl **25**, which were purified by redistillation. The product ratio was determined from the proton ratios of an ^1H NMR spectrum of the mixture.

Compound **23**: bp 118–125 °C (0.5 mmHg) (Found: C, 72.0; H, 9.6; N, 5.5. $C_{15}H_{23}NO_2$ requires C, 72.3; H, 9.3; N, 5.6%); δ_H(500 MHz; $CDCl_3$) 1.24 (3 H, t, *J* 7.3), 1.82 (2 H, dt, *J* 7.1, 7.3), 2.15 (3 H, s), 2.32 (2 H, t, *J* 7.1), 2.35 (3 H, s), 2.40 (2 H, t, *J* 7.3), 3.43 (2 H, s), 4.10 (2 H, q, *J* 7.3) and 7.11–7.25 (4 H, m); ν_{max}(film)/cm^{-1} 1736, 1253, 1176 and 745.

Compound **24**: bp 90–110 °C (0.3 mmHg) (Found: C, 72.0; H, 9.6; N, 5.5. $C_{15}H_{23}NO_2$ requires C, 72.3; H, 9.3; N, 5.6%); δ_H(270 MHz; $CDCl_3$) 1.39 (3 H, t, *J* 7.3), 1.80 (2 H, m), 2.25 (3 H, s), 2.26 (2 H, t, *J* 7.3), 2.42 (2 H, t, *J* 7.3), 2.61 (2 H, m), 2.75 (2 H, m), 4.12 (2 H, q, *J* 7.3) and 7.18–7.28 (5 H, m); ν_{max}(film)/cm^{-1} 1739, 1258, 1023 and 706.

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