benzophenone ketyl. t-BuOH and pyridine were distilled from calcium hydride. 1-(4-Formylbutanoyl)pyrrolidine (3) was prepared by the Swern oxidation[7] of 1-(4-hydroxybutanoyl)pyrrolidine which was readily obtained from γ-butyrolactone and pyrrolidine.

**1-(4-Acetoxy-5-(phenylsulfonyl)eicosanoyl)pyrrolidine (4).** To a THF solution (10 mL) of the sulfone **2** (689 mg, 1.88 mmol) was added *n*-BuLi (1.5 N hexane solution, 1.50 mL, 2.25 mmol) at 0 °C. After being stirred for 1 h at this temperature, the solution was cooled to −78 °C. To the resulting pale yellow suspension was added the aldehyde **3** (378 mg, 2.44 mmol) in THF (5 mL). The color of the suspension turned white after the reaction mixture had been stirred for 30 min at −78 °C. The reaction mixture was extracted with 0.3 N HCl–ethyl acetate. The organic layer was dried ($MgSO_4$) and evaporated to give a crude oil of 1-(4-hydroxy-5-(phenylsulfonyl)eicosanoyl)pyrrolidine (939 mg), which was subjected to acetylation with acetic anhydride (3 mL)–pyridine (3 mL) at room temperature. Usual workup and column chromatography on silica gel (5:1 hexane–ethyl acetate) afforded **4** (677 mg, 64%): $^1H$ NMR (60 MHz) ($CCl_4$) δ 0.70–2.32 (m, 39 H), 1.78, 1.83 (s, 3 H), 2.99–3.52 (m, 5 H), 4.88–5.22 (m, 1 H), 7.38–7.97 (m, 5 H).

**Synthesis of Trichonine (1).** To a t-BuOH solution (15 mL) of **4** (360 mg, 0.65 mmol) was added t-BuOK (220 mg, 1.95 mmol) in t-BuOH (4 mL) at room temperature. After being stirred for 12 h, the reaction mixture was combined with water and ethyl acetate. The separated organic layer was dried ($MgSO_4$) and evaporated. Column chromatography of the residue on activated alumina gave the following three isomers. (2*Z*,4*E*)-**1** (eluent 20:1 hexane–ethyl acetate): 10 mg (4%); mp 45–49 °C; $^1H$ NMR (500 MHz) ($CDCl_3$) δ 0.79 (m, 3 H), 0.98–1.50 (m, 26 H), 1.77–1.89 (m, 4 H), 2.04–2.26 (m, 2 H), 3.37–3.40 (m, 4 H), 5.68 (d, 1 H, *J* = 11.4 Hz), 5.88 (dt, 1 H, *J* = 15.4 and 7.0 Hz), 6.30 (dd, 1 H, *J* = 11.4 and 11.0 Hz), 7.20 (dd, 1 H, *J* = 15.4 and 11.0 Hz); HRMS calcd for $C_{24}H_{43}NO$ 361.3345, obsd 361.3348. (2*E*,4*Z*)-**1** (eluent 10:1 hexane–ethyl acetate): 20 mg (8%); mp 49–51 °C; $^1H$ NMR (500 MHz) ($CDCl_3$) δ 0.80 (m, 3 H), 0.95–1.64 (m, 26 H), 1.67–1.87 (m, 4 H), 2.11–2.29 (m, 2 H), 3.28–3.48 (m, 4 H), 5.69 (dt, 1 H, *J* = 9.9 and 7.7 Hz), 6.04 (dd, 1 H, *J* = 12.1 and 9.9 Hz), 6.08 (d, 1 H, *J* = 14.7 Hz), 7.52 (dd, 1 H, *J* = 14.7 and 12.1 Hz); HRMS calcd for $C_{24}H_{43}NO$ 361.3345, obsd 361.3345. (2*E*,4*E*)-**1** (eluent 10:1 hexane–ethyl acetate): 165 mg (70%); mp 74–75 °C; $^1H$ NMR (500 MHz) ($CDCl_3$) δ 0.80 (t, 3 H, *J* = 11.6 Hz), 1.15–1.35 (m, 26 H), 1.84 (br s, 4 H), 2.04–2.10 (m, 2 H), 3.45 (br s, 4 H), 6.00 (dt, 1 H, 15.1 and 7.1 Hz), 6.01 (d, 1 H, 14.4 Hz), 6.11 (dd, 1 H, *J* = 15.1 and 11.5 Hz), 7.20 (dd, 1 H, *J* = 14.4 and 11.5 Hz; HRMS calcd for $C_{24}H_{43}NO$ 361.3345, obsd 361.3346.

**Registry No.** (2*E*,4*E*)-**1**, 33169-28-7; (2*Z*,4*E*)-**1**, 104035-11-2; (2*E*,4*Z*)-**1**, 104035-14-5; **2**, 104035-13-4; **3**, 41193-97-9; **4**, 104035-12-3; 1-(4-hydroxy-5-(phenylsulfonyl)eicosanoyl)pyrrolidine, 104035-15-6.

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## Fluoride- or Alkoxide-Induced Reaction of 1-[(Trimethylsilyl)methyl]azoles with Carbonyl Compounds

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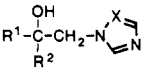
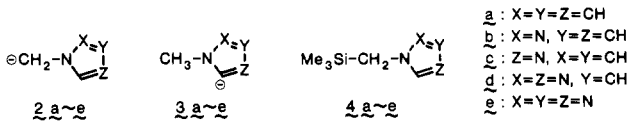
Synthetic routes to (imidazol-1-yl)- or (1,2,4-triazol-1-yl)ethanols **1** have received considerable attention recently because of the importance of their general structure in orally active antifungal azole moieties.[1] In the course of our synthetic project on orally active antifungal azoles, this structure prompted us to study the reaction of carbonyl compounds with azolylmethyl carbanions **2**.

Lithiation of 1-alkylazoles occurs at ring positions,[2] exceptionally at N-$C_\alpha$ of the alkyl radical.[3-10] 1-Methylpyrazole was lithiated at the N-methyl group in competition with the 5-position; however, 1,5-dimethylpyrazole was lithiated at the N-methyl group.[4] Katritzky et al.[7] reported that lithiation of 1-benzylpyrazole originally occurred at the $CH_2$ group and then the lithio derivative was isomerized to the thermodynamically more stable 5-isomer. Thus, if lithiation of a 1-methylazole leads to the azolylmethyl anion **2**, generation of **2** from 1-methylazoles cannot occur because of isomerization toward the thermodynamically more stable ring carbanion **3**.



**1**: X=N or CH



**a**: X=Y=Z=CH
**b**: X=N, Y=Z=CH
**c**: Z=N, X=Y=CH
**d**: X=Z=N, Y=CH
**e**: X=Y=Z=N

**2a–e** **3a–e** **4a–e**

With these points in mind, we attempted to generate carbanion **2** by fluoride- or alkoxide-induced desilylation of the corresponding 1-[(trimethylsilyl)methyl]azoles **4**. Although Tsuge et al.[11] demonstrated fluoride-induced desilylation of dimethyl 1-[(trimethylsilyl)methyl]-1,2,3-triazole-4,5-dicarboxylate to generate 1,2,3-triazolyl-1-ylmethyl anion, there had been no example of unsubstituted azolylmethyl anion **2** being generated from the corresponding **4**. We now report the fluoride- or alkoxide-induced reaction of 1-[(trimethylsilyl)methyl]azoles **4** with carbonyl compounds, with focus on the reactivities of **2** generated in this reaction.

### Results and Discussion

**Preparation of 1-[(Trimethylsilyl)methyl]azoles 4a–e.** 1-[(Trimethylsilyl)methyl]pyrrole (**4a**)[12] and 1-[(trimethylsilyl)methyl]imidazole (**4c**)[13] were prepared as described in the literature. Treatment of pyrazole and 1,2,4-triazole with (chloromethyl)trimethylsilane in the presence of potassium carbonate in dry $Me_2SO$ afforded 1-[(trimethylsilyl)methyl]pyrazole (**4b**, 51%) and 1-[(trimethylsilyl)methyl]-1,2,4-triazole (**4d**, 77%), respectively. In a similar manner, the reaction of tetrazole with (chloromethyl)trimethylsilane afforded a mixture of 1-[(trimethylsilyl)methyl]tetrazole (**4e**, 25%) and 2-[(trimethylsilyl)methyl]tetrazole (15%). The structures of these compounds were confirmed by $^1H$ NMR spectroscopy.[14]

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0022-3263/86/1951-3897$01.50/0 

**Scheme I**

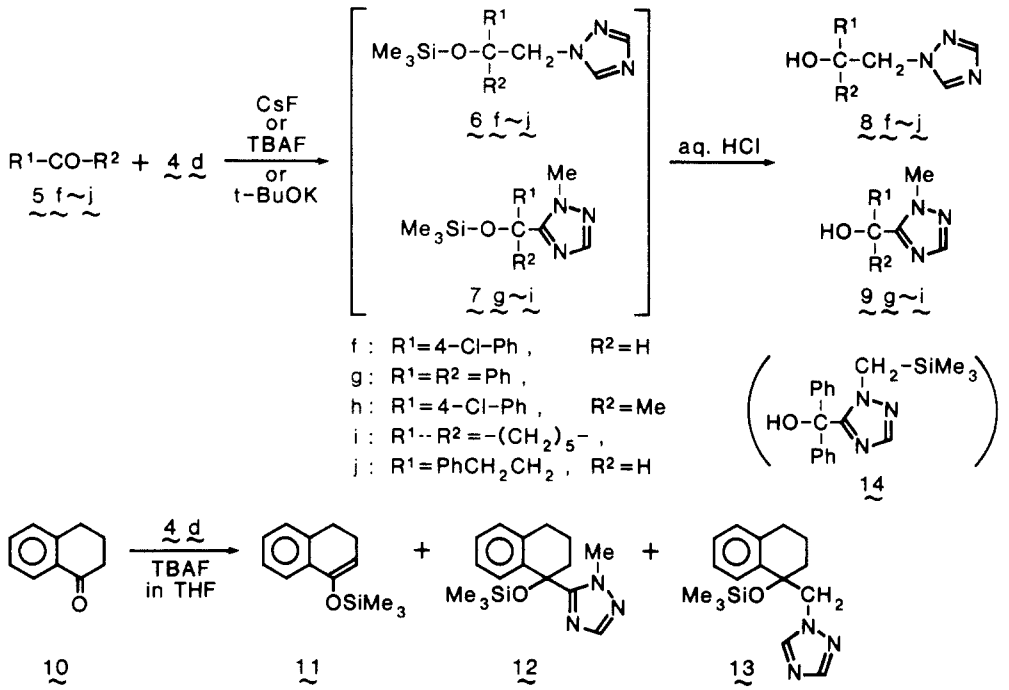
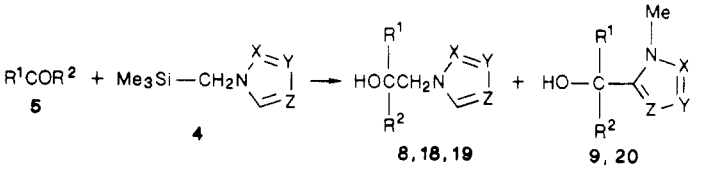



**Table I. Reactions of 1-[(Trimethylsilyl)methyl]azoles 4 with Carbonyl Compounds 5**

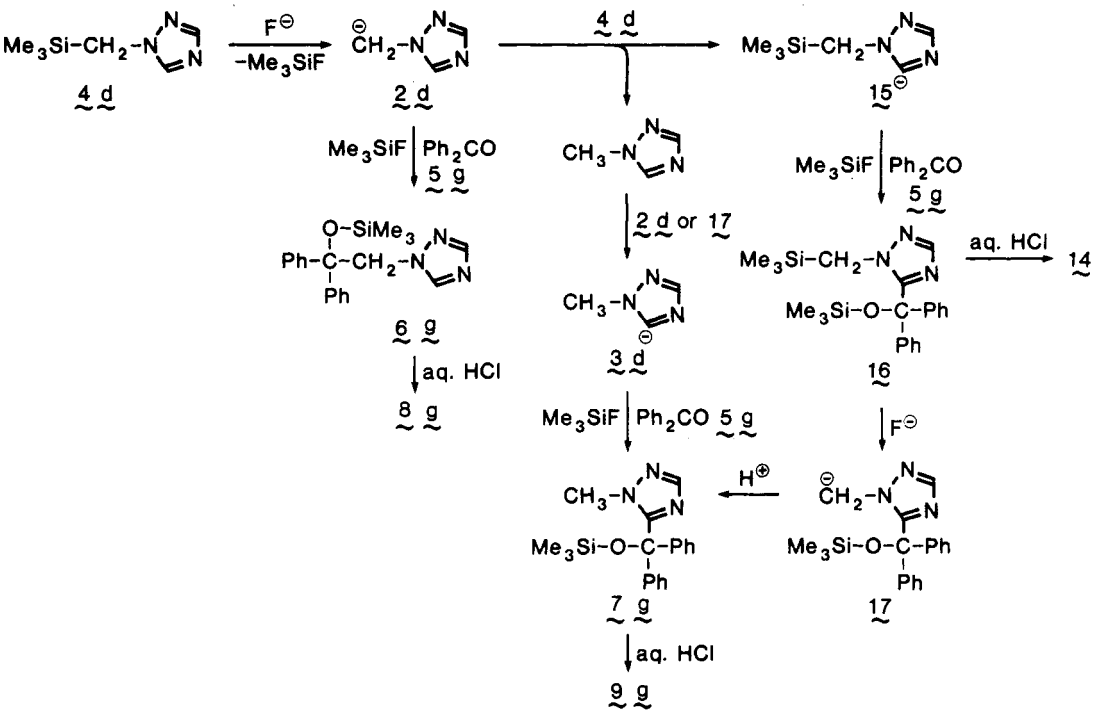



| entry | carbonyl compds | azoles | catalyst[b] | solvent/condns[c,i] | product[a] (yields, %)[d] mp, °C (recryst solv)[e] | | | |
|---|---|---|---|---|---|---|---|---|
| 1 | **5f** | **4d** | CsF | Dm, 4 h | **8f** (68) | 116–117 (A) | | |
| 2 | **5f** | **4d** | TBAF | T/refl, 2 h | **8f** (80) | | | |
| 3 | **5g** | **4d** | CsF | Dm, 4 h | **8g** (52) | 132–134 (A) | **9g** (28) | 175–177 (B) |
| 4 | **5g** | **4d** | TBAF | T/refl, 2 h | **8g** (21) | | **9g** (46) | |
| | | | | | | | **14** (4) | 165–167 (A) |
| 5[f] | **5h** | **4d** | CsF | Dm, 20 h | **8h** (44) | 90.5–91 (A) | | |
| 6[g] | **5h** | **4d** | TBAF | T/refl, 6 h | **8h** (8) | | **9h** (2) | 156–157 (A) |
| 7 | **5i** | 4d | CsF | Dm, 20 h | **8i** (4) | 99.5–100.5 (A) | | |
| 8 | **5i** | **4d** | TBAF | T/refl, 6 h | **8i** (3) | | **9i** (14) | 183–185 (A) |
| 9 | **5j** | **4d** | CsF | Dm, 20 h | **8j** (17) | 96.5–97.5 (A) | | |
| 10 | **5j** | **4d** | TBAF | T/refl, 6 h | **8j** (22) | | | |
| 11 | **5g** | **4d** | *t*-BuOK | T/−20 °C, 6 h | **8g** (61) | | **9g** (6) | |
| 12 | **5g** | **4d** | *t*-BuOK (18-crown-6)[h] | T/−20 °C, 4 h | **8g** (9) | | **9g** (81) | |
| 13 | **5f** | **4b** | TBAF | T/refl, 6 h | **18b** (22) | 128–129 (A) | | |
| 14 | **5f** | **4c** | TBAF | T/refl, 6 h | **18c** (35) | 174–176 (C) | | |
| 15 | **5g** | **4a** | *t*-BuOK | T/refl, 20 h | **19a** (40) | 70.5–71.5 (D) | | |
| 16 | **5g** | **4b** | *t*-BuOK | T/rt, 7 h | **19b** (52) | 161.5–163 (A) | | |
| 17 | **5g** | **4c** | *t*-BuOK | T/rt, 3 h | **19c** (69) | 199.5–200.5 (E) | | |
| 18 | **5g** | **4e** | *t*-BuOK | T/−20 °C, 1 h | | | **20e** (68) | 98–100 (F) |
| | | | | | | | **21e** (12) | 155.5–157.5 (G) |

[a] All products gave satisfactory elementary analyses (C, H, N, Cl ±0.4%). [b] 1.2 molar equiv of *t*-BuOK for **5g**. [c] Diglyme/100 °C = Dm, THF = T. [d] Isolated yield. [e] A = AcOEt/*i*-$Pr_2O$; B = AcOEt; C = MeOH/AcOEt; D = *i*-$Pr_2O$/petroleum ether; E = $CH_2Cl_2$; F = *i*-$Pr_2O$; G = $Et_2O$/petroleum ether. [f] **5h** recovered 26%. [g] **5h** recovered 36%. [h] 1.0 molar equiv of 18-crown-6 for *t*-BuOK. [i] rt = room temperature. refl = reflux.

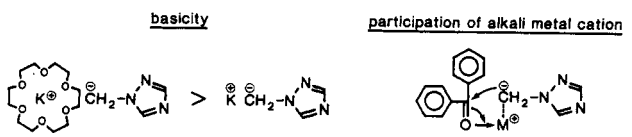
**Reaction of 1-[(Trimethylsilyl)methyl]azoles 4a–e with Carbonyl Compounds 5f–j.** 1-[(Trimethylsilyl)-methyl]-1,2,4-triazole (**4d**) reacted smoothly with carbonyl compounds **5f** and **5g** in the presence of a catalytic amount of cesium fluoride (CsF) or tetrabutylammonium fluoride (TBAF) to give 2-(1,2,4-triazol-1-yl)ethanols **8** with or without (1-methyl-1,2,4-triazol-5-yl)methanols **9** after acid-catalyzed hydrolysis (Scheme I). The results are summarized in Table I (entries 1–10). However, in the case of the enolizable carbonyl compounds **5h–j**, the product yields were comparatively low and the starting materials were recovered. This is due to a tendency to form enolate anions by proton transfer to **2d**. In fact, TBAF-catalyzed reaction of **4d** with α-tetralone **10** without subsequent hydrolysis afforded the corresponding silyl enol ether **11** (37%) with the accompanying silyl ethers **12** and **13** (25%

Scheme II



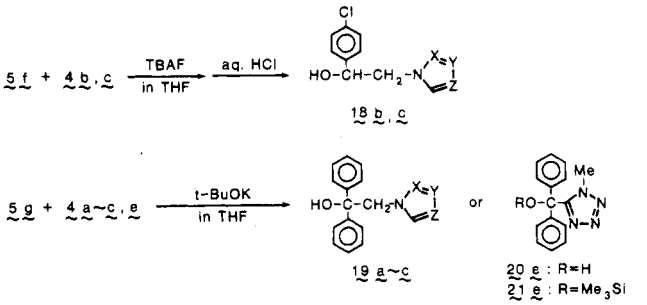
as the mixture, ratio **12**/**13** = 2.5). It should be noted that the CsF- and TBAF-catalyzed reaction of benzophenone (**5g**) with **4d** gave somewhat different product ratios (**8g**/**9g**). Namely, the reaction catalyzed by CsF yielded 52% **8g** and 28% **9g**, while that catalyzed by TBAF resulted in 21% **8g** and 46% **9g** with an accompanying 4% diphenylmethanol **14**. These findings can be explained by the process shown in Scheme **II**. The anion **2d** generated with CsF is assumed to react with **5g** to give **6g** as the main product. However, the naked anion **2d** generated with TBAF mainly reacts as a base to remove the proton at the 5-position of **4d** and give rise to the corresponding anion **15**. The anion **15** reacts with **5g** to give the trimethylsilyl ether **16** and subsequent hydrolysis gives **14**. On the other hand, **16** also undergoes desilylation under the reaction conditions to yield the 1-methyl-1,2,4-triazole **7g** via anion **17**, and subsequent hydrolysis of the 1-methyl derivative **7g** yields **9g**. Furthermore, the anion **3d** generated by proton abstraction at the 5-position of 1-methyl-1,2,4-triazole may react with **5g** to give **7g**.

The different product ratios may be due to the properties of the countercation of **2d**. Therefore, this reaction was examined from the standpoint of the influence of the countercation as follows. The reaction of **4d** with **5g** in the presence of a stoichiometric amount of potassium *tert*-butoxide (*t*-BuOK) in THF at −20 °C gave **8g** (61%) as the main product accompanied by **9g** (6%), whereas the above reaction in the presence of 18-crown-6 gave rise to **9g** (81%) as the main product accompanied by **8g** (9%) (entries 11, 12). The results suggest that the reactivity of **2d** generated from **4d** is related to the basicity and the participation of alkali metal cation in **2d**.

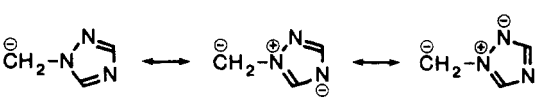



Reactions of the other 1-[(trimethylsilyl)methyl]azoles **4a,b,c,e** with **5f** and **5g** were examined in comparison with that of **4d**. The reaction of **4a** with **5f** in the presence of CsF or TBAF catalyst resulted in recovery of the starting material. However, compounds **4b** and **4c** reacted with **5f** in the presence of TBAF, though with low reactivity to give the azolylethanols **18b** (22%) and **18c** (35%), respectively (entries 13, 14). For further comparison with the reactivity of **4d** (entry 11), *t*-BuOK-induced reactions of **4a,b,c,e** with **5g** were examined (entries 15–18). Compounds **4a–c** reacted with **5g** to give the azolylethanols **19a** (40%), **19b** (52%), and **19c** (69%) as the sole products, respectively. The reaction of **5g** with **4e**, which has the most acidic ring proton, in the presence of *t*-BuOK gave (1-methyl-1,2,3,4-tetrazol-5-yl)methanol (**20e**, 68%) and its trimethylsilyl ether (**21e**, 12%) (Scheme III). Comparison of the above reactions in **4a–e** indicates that the reactivities of **4a–e** depend on the formation of the corresponding carbanions **2a–e** which undergoes dipole stabilization with the participation of nitrogen atom. Dipole stabilization is generally thought to play an important role in the formation of a $\alpha$-heteroatom carbanion.[15]

Scheme III



dipole stabilized carbanion



## Experimental Section

Melting points were determined on a Büchi apparatus and are uncorrected. $^1$H NMR spectra were recorded on a Varian T-60

(15) Beak, P.; Reitz, D. B. *Chem. Rev.* 1978, *78*, 275.

or EM-390 instrument with $Me_4Si$ as an internal standard. A Hitachi 260-10 spectrophotometer was used to obtain IR spectra. Chromatography was performed on 230-400-mesh silica gel.

**1-[(Trimethylsilyl)methyl]pyrrole (4a)** was prepared from pyrrole in 38% yield according to the procedure of Ashby:[12] bp 88.5-89 °C (37 mm) [lit.[12] bp 84 °C (30 mm)].

**1-[(Trimethylsilyl)methyl]imidazole (4c)** was prepared from imidazole in 41% yield according to the procedure of Barcza:[13] bp 110-111 °C (7 mm) [lit.[13] bp 65 °C (0.5 mm)].

**Preparation of 1-[(Trimethylsilyl)methyl]pyrazole (4b).** A suspension of pyrazole (10 g, 147 mmol), (chloromethyl)trimethylsilane (19.8 g, 161 mmol), powdered $K_2CO_3$ (24.4 g, 177 mmol), and dry $Me_2SO$ (200 mL) was stirred at 25 °C for 62 h. The resulting mixture was poured into ice-water and extracted with $Et_2O$. The organic layer was washed with water, dried ($Na_2SO_4$), and evaporated. The residue was distilled under reduced pressure to give 11.5 g of **4b** (51%): bp 118-119 °C (126 mm); $^1H$ NMR ($CDCl_3$) $\delta$ 0.11 (s, 9 H, $Me_3$), 3.71 (s, 2 H, $CH_2$), 6.08-6.21 (m, 1 H, 4-position of pyrazole ring), 7.14-7.26 (m, 1 H, 3-position), 7.33-7.46 (m, 1 H, 5-position); IR (neat) 3100, 2950, 2890, 1510, 1440, 1410, 1390, 1245, 1125, 1085, 1045, 960, 915, 850, 745, 700 $cm^{-1}$. Anal. Calcd for $C_7H_{14}N_2Si$: C, 54.49; H, 9.15; N, 18.16. Found: C, 54.20; H, 9.03; N, 17.95.

**1-[(Trimethylsilyl)methyl]-1,2,4-triazole (4d)** was prepared in a similar manner. 1,2,4-Triazole (12 g, 174 mmol) was converted into 20.7 g of **4d** (77%): bp 102-103 °C (15 mm); $^1H$ NMR ($CCl_4$) $\delta$ 0.11 (s, 9 H, $Me_3$), 3.67 (s, 2 H, $CH_2$), 7.62 (s, 1 H, 3-position of 1,2,4-triazole ring), 7.76 (s, 1 H, 5-position); IR (neat) 3100, 2950, 2890, 1500, 1410, 1345, 1290, 1270, 1250, 1210, 1140, 1010, 950, 850, 760, 700, 680 $cm^{-1}$. Anal. Calcd for $C_6H_{13}N_3Si$: C, 46.41; H, 8.44; N, 27.06. Found: C, 46.00; H, 8.47; N, 26.90.

**1-[(Trimethylsilyl)methyl]tetrazole (4e) and 2-[(trimethylsilyl)methyl]tetrazole** were prepared in a similar manner. Tetrazole (6 g, 86 mmol) gave **4e** (3.35 g, 25%) and 2-[(trimethylsilyl)methyl]tetrazole (1.97 g, 15%), which were separated from each other by flash chromatography using AcOEt-benzene (1:4) as an eluent. **4e**: mp 56-57.5 °C; $^1H$ NMR ($CDCl_3$) $\delta$ 0.17 (s, 9 H, $Me_3$), 3.94 (s, 2 H, $CH_2$), 8.50 (s, 1 H, 5-position of tetrazole ring); IR (Nujol) 3090, 1485, 1425, 1270, 1255, 1240, 1180, 1120, 1100, 975, 900, 870, 850, 775, 710 $cm^{-1}$. Anal. Calcd for $C_5H_{12}N_4Si$: C, 38.43; H, 7.74; N, 35.85. Found: C, 38.16; H, 7.71; N, 35.64. 2-[(Trimethylsilyl)methyl]tetrazole: bp 85-86 °C (17 mm); $^1H$ NMR ($CDCl_3$) $\delta$ 0.18 (s, 9 H, $Me_3$), 4.22 (s, 2 H, $CH_2$), 8.43 (s, 1 H, 5-position of tetrazole ring); IR (neat) 3135, 2950, 2895, 1445, 1410, 1350, 1275, 1250, 1170, 1130, 1090, 1020, 1000, 850, 765, 740, 700 $cm^{-1}$. Anal. Calcd for $C_5H_{12}N_4Si$: C, 38.43; H, 7.74; N, 35.85. Found: C, 37.91; H, 7.58; N, 35.55.

**General Procedure for CsF-Catalyzed Reaction of 1-[(Trimethylsilyl)methyl]azole 4 with Carbonyl Compounds.** To a solution of carbonyl compound (500 mg, 2.7-5.1 mmol) and **4** (1.2 molar equiv/mol of carbonyl compound) in dry diglyme (1.9 mL/mmol of carbonyl compound) under nitrogen atmosphere at room temperature was added powdered CsF (0.1 molar equiv/mol of carbonyl compound, dried 150 °C under vacuum). The suspension was stirred at 100 °C for the period shown in Table I. After cooling to room temperature, 6 N HCl (2 mL) was added and the mixture was stirred for 6 h. The reaction mixture was poured into 5% NaOH and extracted with $CH_2Cl_2$. The organic layer was washed with water, dried ($Na_2SO_4$), and evaporated. The residue was purified by flash chromatography and the results are summarized in Table I (entries 1, 3, 5, 7, 9).

**General Procedure for TBAF-Catalyzed Reaction of 4 with Carbonyl Compounds.** To a mixture of carbonyl compound (500 mg, 2.7-5.1 mmol) and **4** (1.2 molar equiv/mol of carbonyl compound) in dry THF (1.9 mL/mmol of carbonyl compound) under nitrogen atmosphere at room temperature was added anhydrous TBAF (0.1 molar equiv/mol of carbonyl compound, 1 M in THF). The mixture was refluxed for the period shown in Table I. The reaction mixture was worked up in a similar manner to that described above, and purified by flash chromatography to give **8** with or without **9**, as shown in Table I (entries 2, 4, 6, 8, 10, 13 14).

**General Procedure for *t*-BuOK-Induced Reaction of 1-[(Trimethylsilyl)methyl]azole 4 with Benzophenone (5g).** *t*-BuOK (148 mg, 1.3 mmol) was added to a solution of **5g** (200 mg, 1.1 mmol) and **4** (1.2 molar equiv/mol of **5g**) in dry THF (2 mL) and the mixture was stirred under the conditions described in Table I. The mixture was poured into ice-water and extracted with $CH_2Cl_2$. The organic layer was washed with water, dried ($Na_2SO_4$), and evaporated. The residue was chromatographed on silica gel to give the products listed in Table I (entries 11, 12, 15-18).

**TBAF-Catalyzed Reaction of 1-[(Trimethylsilyl)methyl]-1,2,4-triazole (4d) with α-Tetralone (10).** To a solution of **10** (470 mg, 3.2 mmol) and **4d** (1 g, 6.4 mmol) in dry THF (10 mL) was added anhydrous TBAF (0.3 mL, 1 M in THF) under nitrogen atmosphere at room temperature, and the mixture was refluxed for 2 h. The reaction mixture was evaporated to remove THF and chromatographed on silica gel. The fractions eluted with benzene-hexane (1:1) gave **11** (257 mg, 37%): $^1H$ NMR ($CCl_4$) $\delta$ 0.24 (s, 9 H, $Me_3$), 2.06-2.91 (m, 4 H, -$(CH_2)_2$-), 5.05 (t, 1 H, $J$ = 4.2 Hz, CH=), 6.88-7.54 (m, 4 H, Ar H); IR (neat) 1640 $cm^{-1}$. The fractions eluted with benzene gave α-tetralone (137 mg, 29%). The fraction eluted with benzene-AcOEt (10:1) afforded a mixture of **12** and **13** (272 mg, 12:13 = 2.5:1 by $^1H$ NMR spectrum, 25%). **12**: $^1H$ NMR ($CCl_4$) $\delta$ 0.0 (s, 9 H, $Me_3$), 1.57-2.95 (m, 6 H, -$(CH_2)_3$-), 3.80 (s, 3 H, Me), 7.02-7.43 (m, 4 H, Ar H), 7.48 (s, 1 H, 3-position of 1,2,4-triazole ring). **13**: $^1H$ NMR ($CCl_4$) $\delta$ 0.0 (s, 9 H, $Me_3$), 1.57-2.95 (m, 6 H, -$(CH_2)_3$-), 4.13 (d, 1 H, $J$ = 14.4 Hz, NCH*H*), 4.40 (d, 1 H, $J$ = 14.4 Hz, NC*H*H), 7.02-7.43 (m, 4 H, Ar H), 7.70 (s, 1 H, 3-position of 1,2,4-triazole ring), 8.05 (s, 1 H, 5-position of 1,2,4-triazole ring).

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**Registry No.** **4a**, 5833-50-1; **4b**, 92525-04-7; **4c**, 39579-48-1; **4d**, 103817-03-4; **4e**, 103817-04-5; **5f**, 104-88-1; **5g**, 119-61-9; **5h**, 99-91-2; **5i**, 108-94-1; **5j**, 104-53-0; **8f**, 62881-59-8; **8g**, 76674-04-9; **8h**, 79983-74-7; **8i**, 103817-05-6; **8j**, 103817-06-7; **9g**, 103817-08-9; **9h**, 103817-15-8; **9i**, 103817-16-9; **10**, 529-34-0; **11**, 38858-72-9; **12**, 103817-13-6; **13**, 103817-14-7; **18b**, 103817-07-8; **18c**, 24155-45-1; **19a**, 103817-09-0; **19b**, 103817-10-3; **19c**, 65570-68-5; **20e**, 33452-23-2; **21e**, 103817-12-5; CsF, 13400-13-0; TBAF, 429-41-4; *t*-BuOK, 865-47-4; pyrazole, 288-13-1; 1,2,4-triazole, 288-88-0; (chloromethyl)trimethylsilane, 288-94-8; tetrazole, 288-94-8; 2-[(trimethylsilyl)methyl]tetrazole, 103817-11-4.

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## Synthesis of Some Macrocyclic Compounds Containing 2,6-Bis(*N*-alkylamino)phenol Units

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Many macrocyclic compounds showing cation-selective behavior[2,3] are commonly prepared either by metal template condensation reactions[4] or by high-dilution techniques.[5] Structural, magnetic, spectroscopic, and redox

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