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Cyclization of $Ph_2Si(NHNHMe)_2$. I. Reaction with *n*-BuLi/Ph_2SiCl_2 and Ring Rearrangement

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ABSTRACT

*Reaction of $Ph_2Si(NHNHMe)_2$ (**1**) with n-BuLi and Ph_2SiCl_2 in 1:2:1 ratio afforded 54% of 1,2,4-triaza-1-methyl-4-methylamino-3,3, 5, 5-tetraphenyl-3,5-disilacylopentane (**2**). In the presence of a catalytic amount of n-BuLi, **2** is rearranged to two isomers, 1,2,4,5-tetraaza-1,4-dimethyl-3,3,6,6-tetraphenyl-3,6-disilacyclohexane (**3**) and 1,2,4,5-tetraaza-1,5-dimethyl-3,3,6,6-tetraphenyl-3,6-disilacyclohexane (**4**), with **3** as the major product. The crystal structure of **3** reveals a twist-boat conformation of the Si_2N_4 ring [$a = 10.691(4)$ Å, $b = 13.178(4)$ Å, $c = 17.812(3)$ Å, $\beta = 95.11(7)°$; monoclinic, $P2_1/n$; $Z = 4$], in which each–N(Me)–N(H)–unit contains one pyramidal (NH) and one planar (NMe) nitrogen.*

INTRODUCTION

Silylhydrazine chemistry has been well developed since the 1950s [1,2]. Several methods have been employed to prepare silylhydrazine compounds. These include the reactions of hydrazines with halosilanes [2], reactions of metallated hydrazines with halosilanes [3,4], catalytic dehydrocoupling of hydrazines with organosilanes [5], and others [6,7,8a]. Besides a number of uses in organic synthesis [8], this class of compounds has attracted increasing attention due to its potential applications as precursors of silicon nitride ceramics [9,10] and as a single-source precursor for silicon nitride thin films as dielectrics in microwave devices [11].

Apart from the systematic work by West [12] on the anionic rearrangements of silylhydrazines, and the investigation of the structural changes of silylhydrazines accompanying electron transfer oxidation by Bock et al. [13], there has been relatively little research on the chemical and structural behavior of these interesting molecules. Only a few molecular structures containing the Si–N–N–unit have been previously described in the literature [4b–d,5,7,11,14]. Following the successful synthesis of bis(2-methylhydrazino)diphenylsilane, **1**, by catalytic dehydrocoupling [5], we have studied its chemical reactivity with the object of synthesizing new ring compounds containing–Si–N–N–units in the structural core. The results of this study are reported in the present article.

RESULTS AND DISCUSSION

Synthesis of 1,2,4-Triaza-1-methyl-4-methylamino-3,3,5,5-tetraphenyl-3,5-disilacyclopentane, 2

Compound **2** was synthesized by the reaction of **1** with *n*-BuLi and Ph_2SiCl_2 in 1:2:1 molar ratios (Equation 1) and was obtained in 64% yield (by ^1H NMR). In addition to the main product **2**, the two isomers of the six-membered Si_2N_4 ring, namely, 1,2,4,5-tetraaza-1,4-dimethyl-3,3,6,6-tetraphenyl-3,6-disilacyclohexane, **3** (22%), and 1,2,4,5-tetraaza-1,5-dimethyl-3, 3, 6,6-tetraphenyl-3,6-disilacyclohexane, **4** (14%), were also obtained by adding a mixture of **1** and *n*-BuLi to a solution of dichlorodiphenylsilane, in a molar ratio 1:2:1.

Dedicated to Prof. Adrian Gibbs Brook on the occasion of his seventieth birthday.

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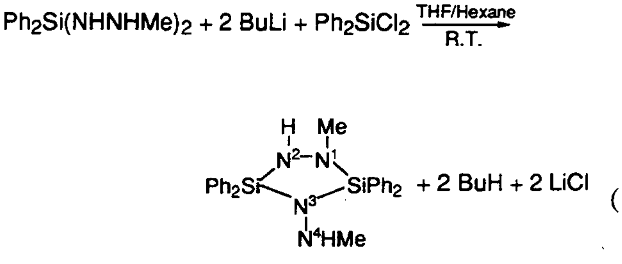


(1)

2

The identification of **2** is based on ^{1}H, ^{29}Si, and ^{15}N NMR, EI-MS, and elemental analysis. In the ^{1}H NMR spectrum, in addition to the phenyl resonances, there are four resonances from $\delta = 2.40$ to 3.50. The endocyclic N^2–H and N^1–Me resonances are shifted downfield relative to the exocyclic analogs, probably due to the ring current of the adjacent Ph groups. The exocyclic NHMe moiety exhibits coupling between the proton and the methyl group, as shown in Figure 1(f). The integral ratios of the various peaks correspond to the H, Me, and phenyl groups in the proposed structure. The connectivity between H and N atoms was confirmed by 2D ^{1}H NMR using a heteronuclear multiple quantum coherence (HMQC) pulse sequence through $^1J_{N-H}$. The ^{29}Si NMR showed two resonances, with one resonance (−16.38 ppm) having a stronger negative NOE effect than the other (−17.55 ppm). Mass spectroscopy showed a parent molecular ion ($m/e = 452$) corresponding to **2**. These facts unambiguously confirm the structure of **2**.

Compound **2** was isolated in 54% yield (96% isomeric purity) from the mixture in a toluene-hexane solution by controlled crystallization. Significant solubility differences allow the efficient separation of compounds **2**, **3**, and **4**. These three compounds are all soluble in toluene, whereas **2** is insoluble in hexane. **3** is soluble in warm hexane, but has low solubility in hexane at ambient temperature and is insoluble in pentane. **4** is soluble in hexane and pentane at ambient temperature. These compounds are colorless crystalline solids. They are mildly moisture-sensitive and can be handled in air for a short period.

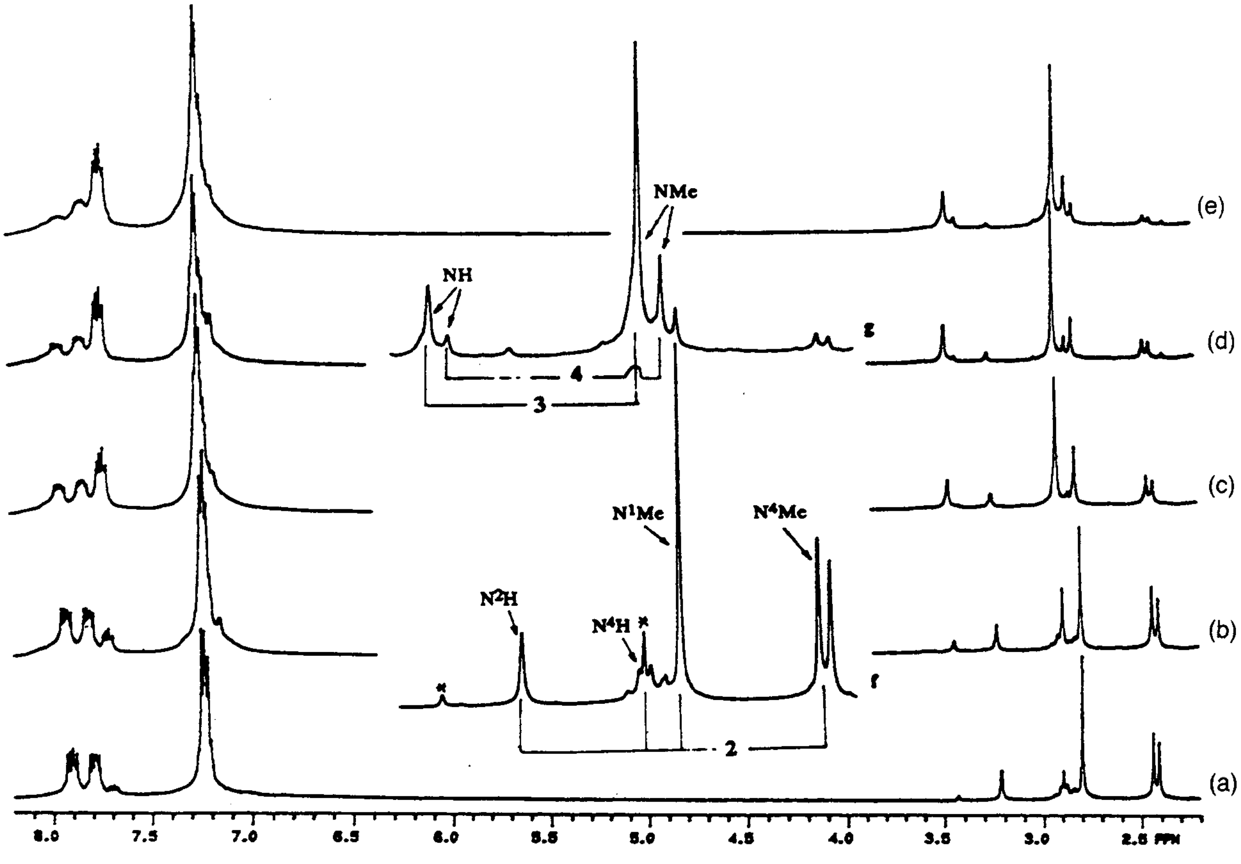
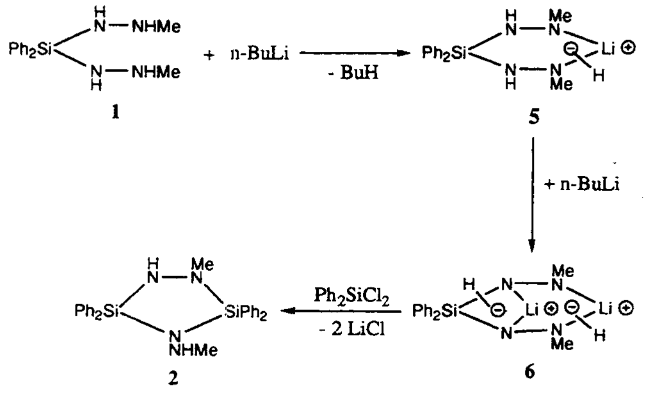
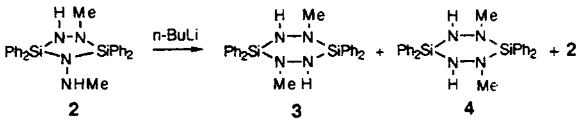


FIGURE 1 ^{1}H NMR spectra showing the rearrangement of **2** to **3** and **4** in the presence of 0.1 mol% *n*-BuLi at 30°C: (a) before the addition of *n*-BuLi; (b) 2 hours after the addition of *n*-BuLi; (c) 10 hours after the addition of *n*-BuLi; (d) 18 hours after the addition of *n*-BuLi; (e) 32 hours after the addition of *n*-BuLi; (f) expansion of (a) in the region of NH and NMe resonances; and (g) expansion of (e) in the region of NH and NMe resonances.



SCHEME 1



(2)

Inverse addition of diphenyldichlorosilane to the lithiohydrazide from **1** results in an increase of **3** and **4** at the expense of **2**. This is due to the rearrangement of **2** to **3** and **4** under basic conditions. We therefore undertook a study of the base catalyzed rearrangement of **2**, as described below.

Mechanism of Formation of 2

A tentative scheme for the formation of **2** is illustrated in Scheme 1. The first step is the reaction of one of the $N_\beta H$ in **1** with one equivalent of BuLi, leading to the formation of complex **5**. Although NMR data indicate that the proton on $N_\alpha H$ (bonded to the Si atom) is more acidic than that on $N_\beta H$ [15], the reaction of **1** with *n*-BuLi and Ph_2SiCl_2 in 1:1:1 ratio affords **4** as a dominant product [16], which indicates that the reaction of $N_\alpha H$ with the first equivalent of BuLi does not take place and **5** is a plausible product from the reaction of **1** with one equivalent of *n*-BuLi. In **5** and **6**, the lithium atoms are coordinated by two N atoms and the unionized hydrogen atoms may be attached to either N atom individually or, more likely, hydrogen bonded between two nitrogens. Therefore, in Scheme 1, the exact locations of the charges and the hydrogen atoms are left undetermined. Addition of a second mole of *n*-BuLi yields **6**. This is attributed to the tight hydrogen bonding and low acidity of the second $N_\beta H$, which is therefore more reluctant to react with *n*-BuLi than the proton of the $N_\alpha H$ moiety. When Ph_2SiCl_2 is added to **6**, **2** is formed by the elimination of two moles of LiCl.

Base Catalyzed Rearrangement of 2

The isomerization of **2** into **3** and **4** occurred in the presence of catalytic amounts of *n*-BuLi and finally reached an equilibrium among the three isomers (Equation 2).

Compound **3** was isolated from a hexane solution, and its ^1H NMR spectrum shows endocyclic NH (3.44 ppm) and NMe (2.89 ppm) resonances, in a 1:3 ratio. **3** exhibited a single ^{29}Si resonance at −19.92 ppm, very close to −19.89 ppm for the known analogue, 1,2,4,5-tetraaza-3,3,6,6-tetraphenyl-3,6-disilacyclohexane, **7** [5]. The connectivity between H and N atoms was also confirmed by 2D ^1H NMR using a HMQC pulse sequence. Compound **4** was recrystallized from a concentrated pentane solution and identified by its ^1H and ^{29}Si NMR spectra. In the ^1H NMR spectrum of **4**, both N–H and N–CH resonances are shifted upfield about 0.05 ppm relative to the corresponding peaks of **3**. The ^{29}Si NMR spectrum of **4** shows two resonances at −18.52 and −18.73 ppm. The −18.52 ppm resonance has almost twice the intensity of the −18.73 ppm resonance. This effect is due to a stronger NOE in the SiNH moiety than that in the SiNCH moiety. A comparison of the ^{29}Si resonances of **2**, **3**, and **4** reveals a dishielding effect on ^{29}Si chemical shifts associated with ring strain. This trend is in agreement with that found in most cyclic silane compounds [17].

The isomerization rate increased with increasing *n*-BuLi concentration, but no measurable effect on the proportion of isomers was observed at equilibrium by varying the amount of *n*-BuLi catalyst. However, the ratio of **3** to **4** was influenced by temperature, whereas **2** remained roughly constant (approximately 10%) at equilibrium. Lower temperature favors the formation of **3**. This is evident from the results shown in Figures 1 and 2, in which 69% **3** and 18% **4** were formed at 30 °C and 54% **3** and 36% **4** at 50 °C. In both cases, **3** is the dominant product.

The base catalyzed rearrangement of open-chain silylhydrazines was thoroughly investigated by West [12]. The proposed mechanism involves the deprotonation, anionic rearrangement, and reprotonation of the substrate. A pentacoordinate transition state is thought to occur in the rearrangement [18], and protonation and deprotonation are the rate-limiting steps [19]. This mechanism can also be applied to the present cyclic system and is illustrated in Scheme 2. Both steric and electronic effects of the adjacent NMe group disfavor the **4a** transition state. This accounts for **3** as the dominant product.

Structure of 3

The crystal structure of **3** was determined by X-ray crystallography, and an ORTEP diagram is

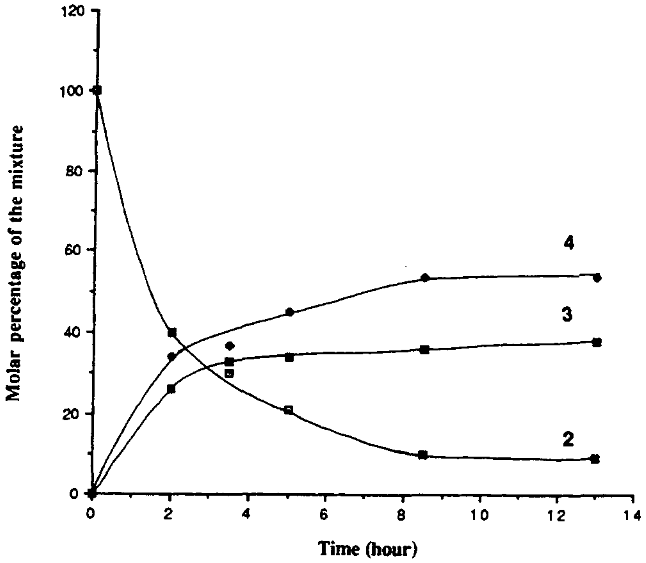


FIGURE 2 Catalytic rearrangement of **2** to **3** and **4** in the presence of 2.5% of BuLi at 50°C.

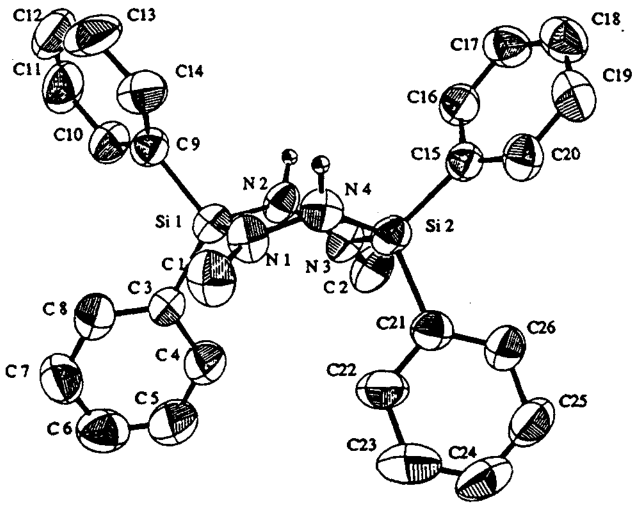
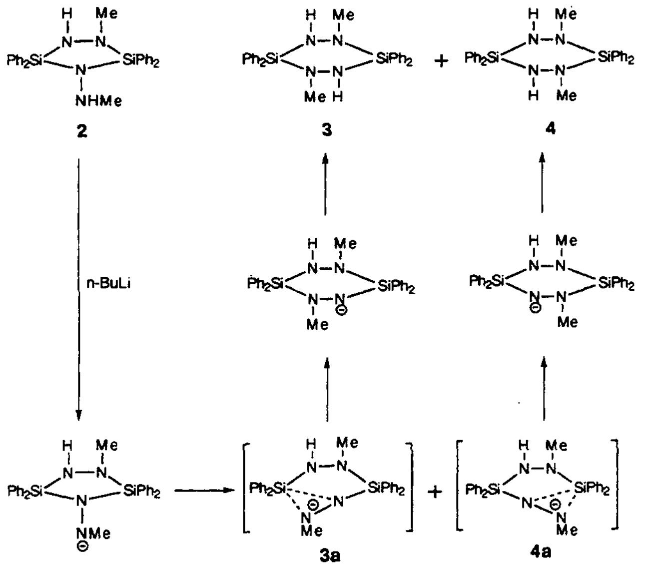


FIGURE 3 ORTEP view of **3**, showing the atom numbering scheme.

shown in Figure 3. The important bond lengths and bond angles are listed in Table 1.

The average bond lengths of Si–N (1.719(4) Å) and N–N (1.436(6) Å) are comparable to those of the known analogous tetraazadisilacyclohexane, **7** (Si–N: 1.731(4) Å; N–N: 1.455(5) Å) [5]. Similar to **7**, two geometries about nitrogens are found in the –N(Me)–N(H)–units, i.e., planar at NMe (sum of angles: N1: 359.9 and N3: 360°, respectively) and pyramidal at NH (sum of angles: N2: 350 and N4: 353.5°, respectively). However, the conformation varies dramatically between **3** and **7**. Whereas **3** adopts a twist-boat conformation, **7** adopts a conformation close to an ideal chair. The replacement of the H on the planar N of **7** by a Me group leads to the flattening of the adjacent pyramidal N of **3**, accompanied by ring flipping from the chair of **7** to the twist-boat of **3**. The conformation of 1,2,4,5-tetraaza-3,3,6,6-tetrafluoro-1,2,4,5-tetrakis(*tert*-butyldimethylsilyl)-3,6-disilacyclohexane is also a twist-boat, but in this case, all four nitrogens are close to planar (sum of angles: 357.6°) [4b]. The driving force of the conformational preference will be influenced by intramolecular steric and electronic effects and intermolecular lattice forces. It is premature to attempt a detailed discussion of this question, but it is clear that a simple interpretation based on, e.g., an N–N equatorial-axial preference is insufficient in these cases [13a].



SCHEME 2

TABLE 1 Selected Bond Distances (Å) and Angles (Degrees) for Compound **3**

Si1–N1	1.725(4)	Si2–N3	1.722(4)
Si1–N2	1.714(4)	Si2–N4	1.721(4)
Si1–C3	1.861(5)	Si2–C15	1.854(5)
Si1–C9	1.867(5)	Si2–C21	1.869(5)
N1–N4	1.428(7)	N2–N3	1.443(5)
N1–C1	1.449(6)	N3–C2	1.443(6)
N1–Si1–N2	105.98(21)	Si1–N1–N4	118.2(3)
N1–Si1–C3	106.75(21)	Si1–N1–C1	127.3(4)
N1–Si1–C9	115.40(21)	N4–N1–C1	114.4(4)
N2–Si1–C3	114.11(22)	Si1–N2–N3	115.2(3)
N2–Si1–C9	105.40(21)	Si1–N2–H	122.6(4)
C3–Si1–C9	109.39(23)	N3–N2–H	112.7(4)
N3–Si2–N4	105.28(21)	Si2–N3–N2	117.8(3)
N3–Si2–C15	115.52(21)	Si2–N3–C2	127.0(3)
N3–Si2–C21	105.37(19)	N2–N3–C2	114.1(3)
N4–Si2–C15	104.98(22)	Si2–N4–N1	117.7(3)
N4–Si2–C21	113.37(23)	Si2–N4–H	124.4(4)
C15–Si2–C21	112.32(23)	N1–N4–H	111.4(4)

EXPERIMENTAL

General Considerations

All reactions were carried out under an atmosphere of dry argon. The solvents used were dried over Na/benzophenone and distilled prior to use. Bis(2-methylhydrazino)diphenylsilane, **1**, was prepared by the catalytic dehydrocoupling of diphenylsilane with methylhydrazine as described elsewhere. Other chemicals were used as received from the Aldrich Chemical Co. Milwaukee, WI. All glassware was heated over 120°C and evacuated and purged with dry argon several times before use. ^1H (^{29}Si, ^{15}N) NMR spectra were recorded on a Varian XL-200 (300, 500) FT-NMR spectrometer at 20°C with internal TMS (^1H, ^{29}Si) and external CH_3NO_2 (^{15}N) as standards. Benzene-d_6 was used as solvent. Low resolution mass spectra were obtained on a Du Pont 21-492b spectrometer with ionizing energy 70 eV. IR spectra were recorded on a Bruker IFS-88 spectrometer with a resolution of 4 cm^{-1} under dry N_2 purge, with a mercury cadmium telluride detector. Elemental analyses were performed by Galbraith Laboratories Inc., Knoxville, TN.

Reaction of $Ph_2Si(NHNHMe)_2$ with n-BuLi and Ph_2SiCl_2 and Five-Membered Ring Formation

A solution of 1.12 g (4.12 mmol) of **1** in 20.0 mL of THF/Hexane (5:1) under dry argon was cooled to −30°C, and 3.2 mL (8.24 mmol) of *n*-BuLi (2.5 M in hexane) was added dropwise with stirring. The pale yellow solution was allowed to warm to room temperature and stirred for 20 minutes. It was then added dropwise into a solution containing 0.86 mL (4.12 mmol) of Ph_2SiCl_2 in 4 mL of THF/Hexane (5:1). After having been stirred for 1 hour, the solvents (THF/Hexane) were removed under reduced pressure and the residue was redissolved in Toluene/Hexane (10/1) to precipitate LiCl, which was removed by filtration through celite. After workup, the product was found to be **2** (64%), **3** (22%), and **4** (14%) by ^1H NMR spectroscopy. Compound **2** was isolated in 54% yield by crystallization from the mixture in Toluene/Hexane (1/2) at ambient temperature. Further separation of **3** from **4** in the residue was not performed. Analytical data for **2**: ^1H NMR (200 MHz): δ 2.42 (d, $^3J_{HNCH}$ = 4.2 Hz, 3H, N^4CH_3), 2.79 (s, 3H, N^1CH_3), 2.89 (q, $^3J_{HNCH}$ = 4.2 Hz, 1H, N^4H), 3.23 (s, 1H, N^2H), 7.20 (m, 12H, Ph_2Si), 7.86 (m, 8H, Ph_2Si). ^{29}Si NMR (59.6 MHz): δ −16.38 (SiNH), −17.55 ($SiNCH_3$). $^{15}N\{^1H\}$ NMR (HMQC, 499.84 MHz, $^1J_{N-H}$ = 90 Hz): δ −307.02 ($N^2\{H\}$), −326.68 ($N^4\{H\}$). EI-MS (*m/e*, abundance, fragmentation): 452(100) [$M^{\cdot+}$], 407(5) [$M^{\cdot+}$ − HNNHMe], 378(27) [$M^{\cdot+}$ − HNNHMe − NMe], 331(38) [$M^{\cdot+}$ − SiPh − MeH], 302(27) [$M^{\cdot+}$ − HNNMe − SiPh], 226(12) [$M^{\cdot+}$ − HNNMe − $SiPh_2$]. IR (neat, γ cm^{-1}): 3433 (br), 3315 (br), (N–H); 994s, 934s (Si–N). Mp: 146–148°C (uncalibrated).

Anal. calcd for $C_{26}H_{28}N_4Si_2$ (**2**): C, 68.98; H, 6.23; N, 12.37. Found: C, 68.53; H, 6.28; N, 12.23.

When the same reaction was carried out with "inverse" addition of Ph_2SiCl_2 to the lithiohydrazide, the product was shown by NMR spectroscopy to be 48% **2**, 30% **3**, and 22% **4**.

Catalytic Rearrangement of **2** *to* **3** *and* **4**

The following is an example with 0.1% of *n*-butyllithium as catalyst, where 0.5 mL of *n*-butyllithium solution (2.5 × 10^{-3} M in C_6D_6, diluted from the commercial 2.5 M in hexane), was added to 0.565 g (1.25 mmol) of **2** in toluene at 30°C. Stirring was continued for 60 hours. After workup, the product was found to be 69% **3**, 18% **4**, and 13% **2**. After removal of **2**, **3** was isolated in 56% yield from the mixture by crystallization in hexane solution at 4°C. Crystals of **3** suitable for X-ray crystallography were grown by cooling a warm hexane solution slowly to ambient temperature. Analytical data for **3**: ^1H NMR (200 MHz): δ 2.89 (s, 6H, NCH), 3.44 (s, 2H, NH), 7.20 (m, 12H, Ph), 7.70 (m, 8H, Ph). ^{29}Si NMR (56.9 MHz): −19.92. $^{15}N\{^1H\}$ NMR (HMQC, 499.84 MHz): δ −311.82 (N{H}). EI-MS (*m/e*, abundance, fragmentation): 452(100) [$M^{\cdot+}$], 407(4) [$M^{\cdot+}$ − HNNHMe], 378(19) [$M^{\cdot+}$ − HNNHMe − NMe], 331(26) [$M^{\cdot+}$ − SiPh − MeH], 302(22) [$M^{\cdot+}$ − HNNHMe − SiPh], 226(12) [$M^{\cdot+}$ − HNNMe − $SiPh_2$]. IR (neat, γ cm^{-1}): 3331s, 3302s (N–H); 1018vs, 994s (Si–N). Mp: 149–150°C.

Anal. calcd for $C_{26}H_{28}N_4Si_2$ (**3**): C, 68.98; H, 6.23. Found: C, 68.58; H, 6.32.

When the base catalyzed rearrangement of **2** was carried out with 2.5% of *n*-butyllithium at 50°C, the product was found to be 10% **2**, 54% **3**, and 36% **4**. After the separation of **2** and **3** from **4**, 15% of **4** was isolated by recrystallization from a concentrated pentane solution at 4°C. Analytical data for **4**: ^1H NMR (200 MHz): δ 2.83 (s, 6H, NCH), 3.39 (s, 2H, NH), 7.20 (m, 12H, Ph), 7.82 (m, 8H, Ph). ^{29}Si NMR (56.9 MHz): −18.52 (SiNH), −18.73 ($SiNCH_3$). IR (neat, γ cm^{-1}): 3373s, 3321s (N–H); 1022s, 998s (Si–N). Mp: 102–104°C.

The progress of the rearrangement was followed by NMR spectroscopy. A typical experiment is as follows. A sample of **2** (0.0119 g; 0.0263 mmol) was placed in a clean, dry sample tube and dissolved in C_6D_6 (0.4 mL). *n*-Butyllithium (0.26 mL; 2.5 × 10^{-3} M in C_6D_6) was injected into the solution. After mixing, the sample tube was sealed and then placed in an oil bath at the required temperature. The progress of the rearrangement was monitored by ^1H NMR spectroscopy.

Crystal Structure Measurement

A crystal of **3** was sealed in a thin-walled capillary under argon and mounted on a Rigaku diffracto-

TABLE 2 Summary of the Crystal Data Collection for **3**

Formula	$Si_2N_4C_{26}H_{28}$
Fw	452.70
Cryst size, mm	0.40 × 0.25 × 0.23
Cryst syst	monoclinic
Space group	$P2_1/n$
a, Å	10.691(4)
b, Å	13.178(4)
c, Å	17.812(3)
β, deg	95.11(7)
V, Å^3	2499.5(13)
Z	4
d_{calcd}, Mg · m^{-3}	1.203
Temp, °C	20
Radiation	graphite monochromator, Mo(γ = 1.54056 Å)
μ, mm^{-1}	1.43
2θ range, deg	20–35
Scan type	$2\theta/\theta$
No. of reflcts collected	2924
No. of unique reflcts	2724
R	0.054
R_w	0.051
GoF	2.66

TABLE 3 Atomic Parameters *x*, *y*, *z*, and Beq[a] ESDs Refer to the Last Digit Printed

	x	*y*	*z*	*Beq*
Si1	0.95621(13)	0.97867(10)	0.22405(7)	4.09(7)
Si2	1.24080(14)	1.08507(9)	0.25600(7)	3.99(6)
N1	0.9937(4)	1.1047(3)	0.21236(21)	4.72(23)
N2	1.0965(4)	0.9158(3)	0.23958(23)	4.45(20)
N3	1.1923(4)	0.9688(3)	0.28650(19)	4.24(19)
N4	1.1182(4)	1.1291(3)	0.19553(25)	4.56(23)
C1	0.9111(5)	1.1915(4)	0.2154(3)	6.0(3)
C2	1.2529(5)	0.9077(4)	0.3464(3)	5.7(3)
C3	0.8565(5)	0.9728(3)	0.30441(25)	4.2(3)
C4	0.9097(5)	0.9773(4)	0.3783(3)	6.3(3)
C5	0.8363(7)	0.9766(5)	0.4385(3)	7.9(4)
C6	0.7083(8)	0.9719(5)	0.4272(4)	7.2(4)
C7	0.6520(6)	0.9676(5)	0.3550(4)	7.2(4)
C8	0.7273(6)	0.9680(4)	0.2947(3)	6.0(3)
C9	0.8725(5)	0.9160(4)	0.1400(3)	4.61(25)
C10	0.8122(5)	0.8229(4)	0.1462(3)	5.7(3)
C11	0.7490(6)	0.7782(5)	0.0826(4)	8.1(4)
C12	0.7445(7)	0.8270(8)	0.0125(5)	9.7(5)
C13	0.8040(7)	0.9170(6)	0.0072(3)	8.6(4)
C14	0.8685(5)	0.9618(4)	0.0694(3)	5.9(3)
C15	1.3778(4)	1.0824(4)	0.19908(24)	4.05(23)
C16	1.4092(5)	0.9926(4)	0.1626(3)	4.8(3)
C17	1.5044(6)	0.9899(5)	0.1152(3)	5.8(3)
C18	1.5713(5)	1.0758(5)	0.1019(3)	6.0(3)
C19	1.5438(5)	1.1656(4)	0.1381(3)	5.6(3)
C20	1.4489(5)	1.1687(4)	0.1852(3)	4.8(3)
C21	1.2706(5)	1.1636(3)	0.34312(25)	4.3(3)
C22	1.1694(5)	1.1986(4)	0.3789(3)	5.8(3)
C23	1.1863(7)	1.2529(4)	0.4461(3)	7.3(4)
C24	1.3050(8)	1.2720(4)	0.4771(3)	7.2(4)
C25	1.4087(6)	1.2397(5)	0.4437(3)	7.1(3)
C26	1.3901(6)	1.1857(4)	0.3759(3)	5.8(3)
HN2	1.130(5)	0.875(4)	0.202(3)	7.3(15)
HN4	1.122(5)	1.139(4)	0.146(3)	7.9(17)

[a] Beq is the mean of the principal axes of the thermal ellipsoid for atoms refined anisotropically. For hydrogen atoms, Beq = Biso.

meter, controlled by TEXRAY software [20]. Cell dimensions were obtained from 25 reflections with 2θ in the range 25.00–35.00°. The intensity data were collected using the $\theta/2\theta$ scan mode. The last least-squares cycle was calculated with 60 atoms, 298 parameters, and 2024 out of 2724 reflections. Weights based on counting-statics were used. The weight modifier K in KFo^2 is 0.000050. Standard intensities remained steady for the first 2700 reflections then dropped sharply, showing 25% decay. Merging *R* was 2.9% for 200 pairs of symmetry related reflections. The structure was solved by direct methods [21]. Hydrogens on phenyls were included in calculated positions, and hydrogens on nitrogens were found in a difference map. The structure was refined by full-matrix least squares with all nonhydrogens anisotropic. All computations were done using the NRCVAX system of crystallographic software [22].

Crystal data and collection parameters are listed in Table 2. Atomic coordinates and thermal parameters are listed in Table 3.

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SUPPLEMENTARY MATERIAL AVAILABLE

Complete tables of bond distances and bond angles, calculated hydrogen atom parameters, and anisotropic thermal parameters for **3** (3 pages) can be obtained by sending a request to John F. Harrod.

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