Aluminium Hydrazides: Reactions of *tert*-Butylaluminium Chlorides with Dilithium Bis(trimethylsilyl)hydrazide – Formation of Iminoalanes and their Hydrazido Adducts

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Di(*tert*-butyl)aluminium chloride reacted with dilithium bis(trimethylsilyl)hydrazide with the formation of a novel $Al_2Li_2N_4$ cage compound **1**. Its structure consists of a dimeric iminoalane $RAl(\mu N{-}SiMe_3)_2AlR$ ($R = CMe_3$), which is bridged by a dilithiumhydrazido ligand with an intact N–N bond. A heterocubane-type molecule is formed, in which two vertices are occupied by Al atoms, two by Li atoms and four by N atoms. One face of the cube is strongly distorted due to the short N–N distance of the hydrazido group. A similar compound, **3**, was formed with bis(neopentyl)aluminium chloride but it was isolated in very poor yield. The reaction of $Li_2N_2(SiMe_3)_2$ with $(Me_3C)_2AlCl$ in the presence of THF yielded an adduct **2**, in which each Li atom of the cage is additionally coordinated via Li–Cl–Al bridges to the ligand $(Me_3Al)_2Cl{\cdot}THF$. The reaction of dilithium bis(trimethylsilyl)hydrazide with *tert*-butylaluminium dichloride gave, by complete cleavage of the hydrazido groups, a dimeric iminoalane $[Me_3C(THF)AlNSiMe_3]_2$ (**4**), in which each aluminium atom is coordinated to one THF molecule. Its uncoordinated, solvent-free form **5** was obtained by heating solid **4** to 150 °C in vacuo for several hours. It was identified as the tetrameric iminoalane $(Me_3CAlNSiMe_3)_4$, which does not adopt the expected heterocubane-type structure, but consists of three anellated four-membered Al_2N_2 heterocycles in a kind of ladder structure. Remarkably, the cage compound **1** could not be obtained by the reaction of an excess of lithium hydrazide with the iminoalanes **4** and **5**. All compounds **1**–**5** were characterized by crystal structure determinations.

Introduction

In contrast to boron hydrazides,[1] which form a well-established class of compounds, only a few hydrazides of the heavier group three elements aluminium to thallium are known in the literature.[2−7] Recently, such compounds have found an increased interest as they are potentially useful as precursors for the formation of ceramic materials like aluminium and gallium nitride.[4−6] Boron hydrazides are easily accessible by the reaction of boron halides with silylhydrazines and the release of chlorosilanes.[1] As recently shown by our group,[8] this method cannot be applied to the synthesis of aluminium hydrazides. Bis(trimethylsilyl)hydrazine, for instance, did not react with dialkylaluminium chlorides with the expected formation of trimethylchlorosilane, but a dismutation reaction occurred and adducts of the aluminium halides with the thermally unstable mono(trimethylsilyl)hydrazine were isolated. The few aluminium or gallium hydrazides known in the literature were obtained by four different methods: (i) the reaction of trialkyl derivatives with the N−H protons of hydrazines with the elimination of alkanes; (ii) the reaction of monolithiated hydrazides with the corresponding aluminium or gallium chlorides; (iii) the treatment of $LiAlH_4$ with hydrazines and the release of elemental hydrogen; (iv) the hydroalumination reaction of azobenzene with an arylaluminium dihydride, which gave elemental hydrogen and an iminoalane as by-products.[2−7] Recently, Nöth et al. synthesized a series of aluminium hydrazides using methods (ii) and (iii).[3] Beside the synthesis of monoaluminium derivatives, we are also very interested in the synthesis of polyaluminium or polygallium hydrazides which contain more than one aluminium or gallium atom coordinated to each N−N group, and we report here on the reaction of *tert*-butylaluminium chlorides with dilithium bis(trimethylsilyl)hydrazide.

Results and Discussion

Reaction of Dilithium Bis(trimethylsilyl)hydrazide with $(Me_3C)_2AlCl$

In order to obtain a hydrazido group attached to two aluminium atoms we treated the dilithium hydrazide $Li_2N_2(SiMe_3)_2$[9] with two equivalents of di(*tert*-butyl)aluminium chloride[10] in toluene at room temperature for 36 hours. A complicated mixture of products was formed, from which one component (**1**, Equation 1) was isolated in a pure form after a three-step recrystallization from toluene/*n*-hexane in a yield of 34%. The NMR spectroscopic characterization showed that the desired dialuminiumhydrazide could not have been formed. Three singlets were observed in the ^{1}H-NMR spectrum with equal intensities, of which two were in the characteristic range for trimethylsilyl groups (δ = 0.38 and −0.05). The other one was assigned to a *tert*-butyl group attached to an aluminium atom (δ = 1.42). Thus, a compound must have been formed which has two chemi-

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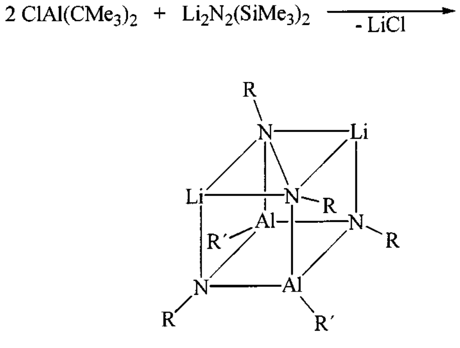
cally different $SiMe_3$ groups and one type of *tert*-butyl group. The constitution of **1** was established by an X-ray crystal structure determination.



$2\ ClAl(CMe_3)_2 + Li_2N_2(SiMe_3)_2 \xrightarrow[-\ LiCl]{}$

(1)

1 (R = $SiMe_3$; R′ = CMe_3)

The molecular structure of **1** is depicted in Figure 1. A remarkable cage compound was formed, which comprises a four-membered Al_2N_2 heterocycle with each aluminium atom further attached to one *tert*-butyl group and each nitrogen atom to a trimethylsilyl group. Each aluminium atom becomes coordinatively saturated by an interaction with one nitrogen atom of a hydrazido dianion, which bridges the iminoalane heterocycle and has an intact N−N bond. The cage is completed by two lithium atoms, each of which is coordinated by both nitrogen atoms of the hydrazido ligand and one nitrogen atom of the iminoalane moiety. Thus, the structure resembles that of a heterocubane type molecule although it is strongly distorted due to the N−N single bond across one of its faces. The Al_2N_2 heterocycle is not planar, but folded across the N−N axis by 31.2°. Owing to a coordination number of four at the aluminium atoms the Al−N bond lengths (189.2 pm on average) are longer than in some dimeric iminoalanes known from the literature with three-coordinate Al and N atoms (180 to 182 pm),[11] and also longer than the bond lengths calculated by ab-initio methods for $(HAl{-}NH)_2$.[12] A remarkably short transannular Al−Al distance of 248.5(2) pm is observed, which is shorter than in dimeric trimethylaluminium with a 2e-3c AlCAl bond (262 pm)[13] or in dialanes(4) with Al−Al single bonds (264 to 275 pm).[14] Similar short transannular distances have been observed before in other dimeric iminoalanes[11] and in many other heterocyclic main-group compounds.

This iminoalane heterocycle is bridged by a hydrazido group, where each nitrogen atom is attached to one aluminium atom. The Al−N distances (201.1 pm) are longer than those of the Al_2N_2 heterocycle and are in the typical range for dative N→Al bonds.[15] The N−N single bond length [158.9(5) pm] is longer than those usually observed in hydrazine derivatives[8][16] or in the related dimetalated hydrazide $Li_2N_2(SiMe_3)_2$,[9] which has a side-on coordination of the N−N group by both lithium atoms similar to **1**. The Li−N distances are significantly different, those to the imino nitrogen atoms and to one atom of the hydrazido group are short (198.4 pm on average), whilst the distance to the second hydrazido nitrogen is lengthened (215.5 pm).

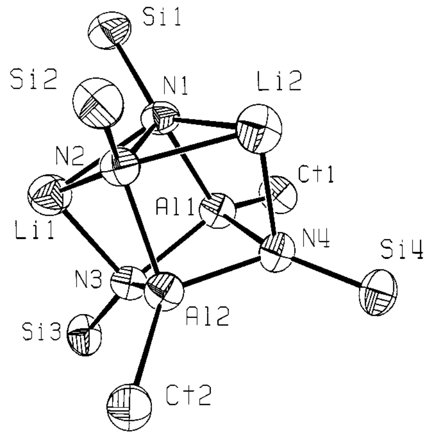


Figure 1. Molecular structure and numbering scheme of compound **1**; thermal ellipsoids are drawn at the 40% probability level (methyl groups omitted for clarity); selected bond lengths [pm] and angles [°]: Al1−N3 187.6(4), Al1−N4 191.4(4), Al2−N3 189.4(4), Al2−N4 188.3(4), N3−Si3 169.9(4), N4−Si4 168.3(4), Al1−Ct1 200.4(5), Al2−Ct2 200.9(5), Al1−N1 201.3(4), Al2−N2 200.8(4), Li1−N1 215.3(10), Li1−N2 196.8(9), Li1−N3 197.3(10), Li2−N1 197.8(9), Li2−N2 215.6(10), Li2−N4 201.8(10), N1−N2 158.9(5), N1−Si1 173.6(4), N2−Si2 174.0(4); N3−Al1−N4 93.8(2), Al1−N3−Al2 82.5(2), N3−Al2−N4 94.2(2), Al1−N4−Al2 81.8(2), Al1−N3−Li1 93.6(3), Al2−N3−Li1 86.2(3), Al1−N4−Li2 85.8(3), Al2−N4−Li2 93.7(3), N1−Al1−N3 92.6(2), N1−Al1−N4 91.3(2), N2−Al2−N3 91.2(2), N2−Al2−N4 92.3(2), N1−Li1−N2 45.1(2), N1−Li1−N3 85.8(4), N2−Li1−N3 90.1(4), N1−Li2−N2 45.0(2), N1−Li2−N4 89.3(4), N2−Li2−N4 84.5(4), Li1−N1−Li2 129.5(4), Li1−N2−Li2 129.9(4), Li1−N1−N2 61.3(3), Li1−N2−N1 73.6(3), Li2−N1−N2 73.5(3), Li2−N2−N1 61.6(3)

At first sight the Li atoms seem to be coordinatively unsaturated, with short contacts to only three nitrogen atoms, but each one also interacts with one methyl group of a trimethylsilyl substituent (Li1−C11 251.0 pm and Li2−C21 248.1 pm). Similar Li−C interactions have been observed in methyllithium[17] and in some lithium bis(trimethylsilyl)methylaluminates.[18]

We have also carried out quantum chemical calculations of the model compound $Li_2(H_3CAlNSiH_3)_2(H_3SiNNSiH_3)$ (**1a**), in which the *tert*-butyl and trimethylsiyl groups of **1** were replaced by CH_3 and SiH_3, respectively. The optimization of the molecular structure of **1a** resulted in a very good agreement between the experimental and calculated structural parameters. Some selected distances and bond angles are summarized in the Experimental Section, where it can be seen that the method chosen is suitable for the description of compound **1**. The largest deviations were observed for the Li−N separations (up to 5 pm) and for the N−N distance of the hydrazido group (151.3 pm instead of 158.9 pm), which may be partially caused by the smaller CH_3 and SiH_3 substituents employed in the calculated molecule **1a**. As clearly shown by NBO analysis, the short Al−Al distance (**1a**: 250.7 pm) must not be interpreted in terms of an Al−Al bonding interaction, and no two- or three-center interactions between the aluminium atoms can be found. The Al−Al bond order is close to zero, and no significant additional interaction between Al orbitals is observed similar to the four-membered heterocycles with short transannular E-E contacts reported previously.

Donor solvents like THF or diethyl ether had a strong influence on the course of this reaction (Equation 1). In diethyl ether no reaction was observed at all over a period of several days at room temperature, and in boiling ether decomposition occurred and only traces of the ether adduct $(Me_3C)_2AlCl{\cdot}OEt_2$ were isolated after recrystallization. Similar observations were made in THF as a solvent. Treatment of dilithium bis(trimethylsilyl)hydrazide with THF gave, after thorough evaporation of the obtained solid, a solvate which, from the integration of its ^1H NMR spectrum, contained half a molecule of THF for each formula unit of the hydrazide. The reaction of this solvate with di(*tert*-butyl)aluminium chloride in hexane gave another cage compound **2** in low yield and a complicated mixture of unknown products (Equation 2). The molecular structure of **2** in the solid state (Figure 2) can be derived from the cage of **1**, but each lithium atom is additionally coordinated via Li−Cl−Al bridges to a THF adduct of the starting chloroalane. The structural parameters of the cage are very similar to those of **1** and are not discussed here in detail. The chloro bridges are almost linear (Cl1 173.9°; Cl2 167.5°), which is in accordance with the steric shielding of the Li atom by the substituents of the cage and the high ionic character of the Li−Cl interaction. Li−Cl (240.1 pm) and Al−Cl distances (219.3 pm) are in the expected range.[19]

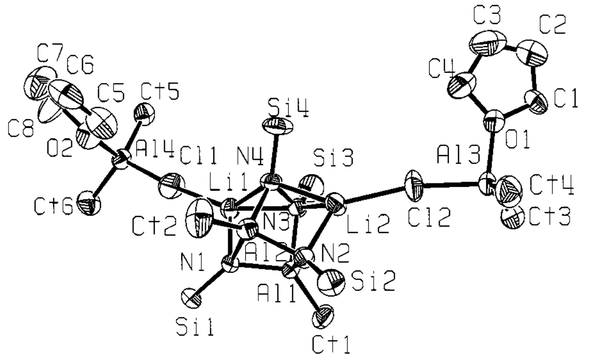


Figure 2. Molecular structure and numbering scheme of compound **2**; thermal ellipsoids are drawn at the 40% probability level (methyl groups omitted for clarity); selected bond lengths [pm] and angles [°]: Al1−N1 187.9(2), Al1−N2 188.8(3), Al2−N1 188.1(3), Al2−N2 188.5(3), N1−Si1 169.6(2), N2−Si2 168.9(2), Al1−Ct1 200.1(3), Al2−Ct2 201.7(4), Al1−N3 199.5(2), Al2−N4 200.0(3), Li1−N1 199.5(6), Li1−N3 212.4(5), Li1−N4 201.6(6), Li2−N2 198.8(5), Li2−N3 203.2(6), Li2−N4 216.4(6), N3−N4 156.1(4), N3−Si3 173.2(2), N4−Si4 172.3(3), Li1−Cl1 237.9(5), Li2−Cl2 242.2(5), Al3−Cl2 219.4(1), Al4−Cl1 219.3(1), Al3−O1 190.6(2), Al4−O2 189.3(3); N1−Al1−N2 93.5(1), Al1−N1−Al2 82.1(1), N1−Al2−N2 93.5(1), Al1−N2−Al2 81.7(1), Al1−N1−Li1 92.0(2), Al2−N1−Li1 86.8(2), Al1−N2−Li2 86.6(2), Al2−N2−Li2 93.6(2), N1−Al1−N3 92.9(1), N2−Al1−N3 92.6(1), N1−Al2−N4 92.6(1), N2−Al2−N4 92.4(1), N3−Li1−N4 44.2(1), N1−Li1−N3 85.9(2), N1−Li1−N4 88.8(2), N3−Li2−N4 43.5(1), N2−Li2−N3 88.6(2), N2−Li2−N4 84.9(2), Li1−N3−Li2 131.0(2), Li1−N4−Li2 129.5(2), Li1−N3−N4 64.3(2), Li1−N4−N3 71.6(2), Li2−N3−N4 72.7(2), Li2−N4−N3 63.7(2), Li1−Cl1−Al4 173.9(1), Li2−Cl2−Al3 167.5(1)

The reaction of dimethylaluminium chloride or di(*tert*-butyl)aluminium bromide with $Li_2N_2(SiMe_3)_2$ gave only mixtures of unknown products, from which no single component could be isolated by recrystallization. A further cage compound **3**) was obtained from the reaction with di(neopentyl)aluminium chloride, and was isolated in a very poor, but reproducible, yield of only 10% after recrystallization from diethyl ether/hexane (Equation 3). Compound **3** decomposes slowly in the reaction mixture, so that the reaction must be stopped after six hours at room temperature. The molecular structure of **3** (Figure 3) is quite similar to that of **1** and need not be discussed in detail. One of the Li atoms (Li2) is coordinated to an ether molecule. As a result the cage is slightly more distorted and the Li−N bonds to Li2 are longer (204.0 and 216.1 pm) than those to Li1 (197.5 and 210.2 pm).

$$2\ ClAl(CH_2CMe_3)_2 + Li_2N_2(SiMe_3)_2 \xrightarrow[-\ LiCl]{Et_2O} \qquad (3)$$

$$Li_2(Al\text{-}CH_2CMe_3)_2(Me_3SiN\text{-}NSiMe_3)(N\text{-}SiMe_3)_2 \cdot OEt_2$$

3

Reaction of Dilithium Bis(trimethylsilyl)hydrazide with $(Me_3C)AlCl_2$

The mechanism of the formation of the cage compounds is not well understood. We supposed that a dismutation reaction might play an important role although, in this case, the corresponding trialkylaluminium compound should be formed. However, these trialkylaluminium derivatives could not be unambiguously identified in the NMR spectra of the reaction mixtures. Nevertheless, *tert*-butylaluminium dichloride as a possible intermediate in a dismutation reaction seemed to be an appropriate starting compound for the synthesis of such derivatives in a higher yield. The imido group may be formed by a disproportionation of $[N_2(SiMe_3)_2]^{2-}$, which could yield dilithium or dialuminium trimethylsilylamide and a reactive nitrene or diimine intermediate which could be consumed by the formation of unknown secondary products.

Treatment of $Me_3C{-}AlCl_2$ with $Li_2N_2(SiMe_3)_2$ in an equimolar ratio without a coordinating solvent gave only decomposition products. Only with the THF solvate of the Li compound and reaction at low temperature was a pure product isolated after recrystallization in a yield of 31%. This compound was subsequently identified as the THF adduct of the corresponding dimeric iminoalane (**4**, Equation 4). Once again, the N−N bond of the hydrazido group was cleaved by the reaction with a chloroalane, and only half of the nitrogen atoms were transferred to the product. The other components of the reaction mixture, which should give more insight into the mechanism of the reaction and the fate of the missing NR groups, could neither be isolated nor identified by NMR spectroscopy. As expected, singlets of equal intensity were observed in the NMR spectra of **4** for the CMe_3 and $SiMe_3$ groups, and broad multiplets were detected for the protons of the coordinated THF molecules. The crystal structure of **4** (Figure 4) reveals two, crystallographically independent, planar Al_2N_2 heterocycles. One is

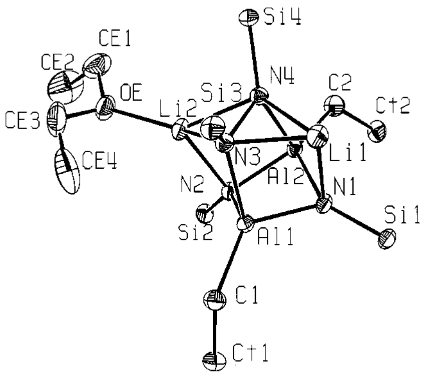
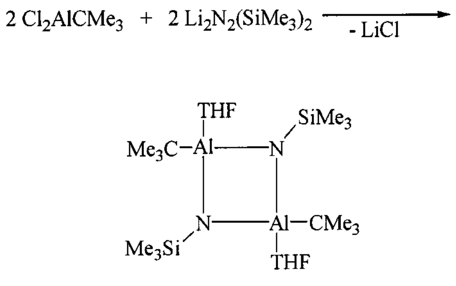


Figure 3. Molecular structure and numbering scheme of compound **3**; thermal ellipsoids are drawn at the 40% probability level (methyl groups omitted for clarity); selected bond lengths [pm] and angles [°]: Al1−N1 188.7(2), Al1−N2 187.2(2), Al2−N1 188.5(2), Al2−N2 188.9(2), N1−Si1 169.0(2), N2−Si2 169.8(2), Al1−C1 198.4(2), Al2−C2 197.9(2), Al1−N3 203.3(2), Al2−N4 200.9(2), Li1−N1 197.7(4), Li1−N3 210.2(4), Li1−N4 197.3(4), Li2−N2 203.9(4), Li2−N3 204.1(3), Li2−N4 216.1(4), N3−N4 157.1(2), N3−Si3 173.2(2), N4−Si4 174.9(2), Li2−OE 196.5(3); N1−Al1−N2 93.18(7), Al1−N1−Al2 82.46(6), N1−Al2−N2 92.67(7), Al1−N2−Al2 82.75(6), Al1−N1−Li1 91.5(1), Al2−N1−Li1 85.9(1), Al1−N2−Li2 88.4(1), Al2−N2−Li2 92.1(1), N1−Al1−N3 92.42(7), N2−Al1−N3 91.95(7), N1−Al2−N4 92.47(7), N2−Al2−N4 92.91(7), N3−Li1−N4 45.20(9), N1−Li1−N3 87.9(1), N1−Li1−N4 90.8(2), N3−Li2−N4 43.78(8), N2−Li2−N3 87.1(1), N2−Li2−N4 84.5(1), Li1−N3−Li2 129.4(2), Li1−N4−Li2 129.9(2), Li1−N3−N4 63.1(1), Li1−N4−N3 71.7(1), Li2−N3−N4 72.2(1), Li2−N4−N3 64.1(1)

located on a crystallographic inversion center, and the other shows a maximum deviation of any atom from the plane of 1 pm. Each Al atom is further coordinated to one *tert*-butyl group and one THF molecule in a distorted tetrahedral environment, while the nitrogen atoms are attached to only one silyl group besides the aluminium atoms of the ring. The nitrogen atoms are almost ideally planar-coordinated with the sum of the angles between 359.4 and 360.0°. The heterocycles deviate slightly from an ideal square geometry and have angles of 87.3° at the nitrogen atom and 92.7° at the aluminium atom. The Al−N distances (183.4 pm on average) are shorter than in the cage compounds described above, which may be due to less steric crowding in the molecules. They are similar to the solvent free iminoalanes reported in the literature.[11] The transannular Al−Al contacts are very small (252.7 pm). As predicted by theory (see above), they do not indicate any bonding interactions between the metal atoms.

The solvent free compound **5** was obtained by heating the THF adduct to 150 °C under vacuum for several hours (Equation 5). Compound **5** is almost insoluble in noncoordinating solvents like pentane or toluene and was purified by extraction with boiling toluene in a Soxhlet apparatus. Therefore, we were not able to determine its molar mass in solution by cryoscopy. Two singlets were observed in the ^1H NMR spectrum for the $SiMe_3$ and CMe_3 groups with equal intensity, which remained unchanged upon cooling solutions in toluene to −90 °C. In the solid state, **5** reveals a unique molecular structure of a tetrameric iminoalane (Figure 5), which comprises three annelated planar four-membered heterocycles. Usually, iminoalanes give cage molecules with up to eight formula units,[20] while very large substituents give dimers or trimers with four- or six-membered heterocycles.[11] No cage compound is formed here, instead a kind of a ladder structure results with angles between the normals of the planes of 63.7°. Different Al−N bond lengths are observed. The shortest one is found between the coordinatively unsaturated atoms Al2 and N2 (180.2 pm), intermediate are the distances between the tri- and tetracoordinated atoms (188.8 pm) and the longest is the separation between the tetracoordinated atoms (Al1−N1 200.6 and Al1−N1′ 193.8 pm). The Al−Al distances are longer than described before (Al1−Al2 262.4 pm, Al1−Al1′ 278.3 pm). As expected, the N−Si bond lengths depend on the coordination number of the nitrogen atom, a short bond is observed to N2 (172.5 pm) and a longer one to N1 (177.4 pm). A similar structure has been observed in a tetraaluminium tetraoxo compound.[21]



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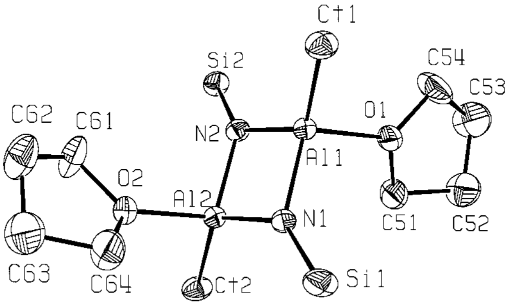
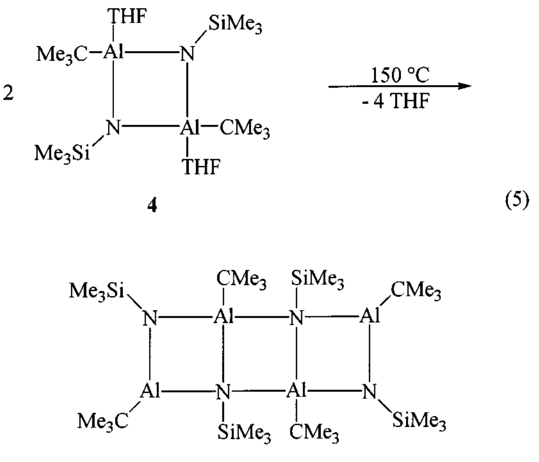


Figure 4. Molecular structure and numbering scheme of compound **4**; thermal ellipsoids are drawn at the 40% probability level (methyl groups omitted for clarity); selected bond lengths [pm] and angles [°]: Al1−N1 184.5(2), Al2−N1 183.0(2), Al1−N2 183.0(2), Al2−N2 183.8(2), Al1−O1 192.8(2), Al1−Ct1 200.5(3), Al2−O2 192.9(2), Al2−Ct2 200.5(3), N1−Si1 167.7(2), N2−Si2 168.5(2); Al1−N1−Al2 87.34(8), Al1−N2−Al2 87.51(8), N1−Al1−N2 92.45(9), N1−Al2−N2 92.67(9), Al1−N1−Si1 133.9(1), Al2−N1−Si1 138.7(1), Al1−N2−Si2 137.1(1), Al2−N2−Si2 134.8(1)

In agreement with the NMR spectroscopic data described above, the structure of **5** in solution may be quite different from that in the solid state. A dimeric iminoalane may be formed by dissociation, or a heterocubane or larger aggregate may exist as the most stable form. Remarkably, the reaction according to Equation 5 is irreversible, and treatment of **5** with a large excess of THF did not yield the THF adduct **4**. Also, the formation of the cage compound **1** was not achieved by the reaction of the solvent-free iminoalane **5** with $Li_2N_2(SiMe_3)_2$ under various conditions,



5

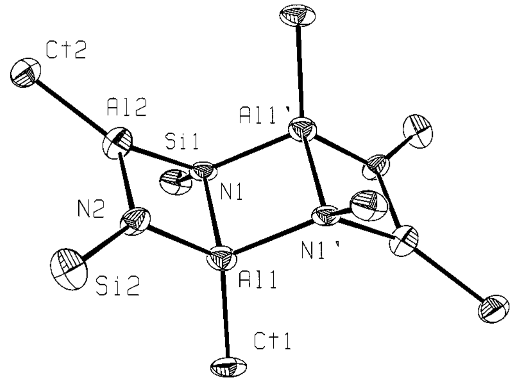


Figure 5. Molecular structure and numbering scheme of compound **5**; thermal ellipsoids are drawn at the 40% probability level (methyl groups omitted for clarity); selected bond lengths [pm] and angles [°]: Al1–N1 200.6(4), Al1–N1′ 193.8(3), Al1–N2 186.6(4), Al1–Ct1 200.8(5), Al2–N1 191.0(4), Al2–N2 180.2(4), Al2–Ct2 196.2(4), N1–Si1 177.4(4), N2–Si2 172.5(3); N1–Al1–N1′ 90.3(1), N1–Al1–N2 89.7(2), Al1–N1–Al1′ 89.7(1), Al1–N1–Al2 84.1(1), Al1′–N1–Al2 117.2(2), N1–Al2–N2 94.8(2), Al1–N2–Al2 91.3(2); Al1′ and N1′ generated by $-x + 1$, $-y$, $-z + 1$

and even the THF adduct did not react with an excess of the lithium hydrazide to form **1**. Obviously, the mechanism of the formation of such cages is very complicated and seems to be determined by several factors, which must remain unclear as long as we have no information concerning the constitution of the by-products

Experimental Section

General: All procedures were carried out under purified argon in dried solvents (THF, diethyl ether and toluene over Na/benzophenone; *n*-hexane and cyclopentane over $LiAlH_4$; pentafluorobenzene over molecular sieves). $Li_2N_2(SiMe_3)_2$,[9] di(*tert*-butyl)aluminium chloride[10] and *tert*-butyl aluminiumdichloride[18] (similar to the bromo derivative) were synthesized according to literature procedures.

Synthesis of 1: A suspension of dilithium bis(trimethylsilyl)hydrazide (0.788 g, 4.19 mmol) in 50 mL of toluene was treated at room temperature with a solution of di(*tert*-butyl)aluminium chloride (1.48 g, 8.38 mmol) in 50 mL of toluene. The mixture was stirred for 36 h, then filtered and evaporated to dryness. Colorless **1** was purified by threefold recrystallization from a toluene/hexane mixture. Yield: 0.38 g (34% based on hydrazide). – M. p. (argon, sealed capillary): 152°C (dec). – ^1H NMR (C_6D_6, 300 MHz): δ = 1.42 (s, 18 H, CMe_3), 0.38 (s, 18 H, $Al_2N{-}SiMe_3$), −0.05 [s, 18 H, $N_2(SiMe_3)_2$]. – ^{13}C NMR (C_6D_6, 75 MHz): δ = 31.2 (CMe_3), 7.7 and 2.8 ($N{-}SiMe_3$), AlC not detected. – IR (CsBr, paraffin): $\tilde{\nu}$ = 1302 vw, 1252 s, 1223 w $\delta(CH_3)$, 1182 vw, 1169 vw, 1152 vw, 1063 w, 997 m, 978 m, 937 m, 889 m $\nu(CC)$, $\nu(NN)$; 837 vs, 748 s, 723 m $\rho(CH_3)$; 658 w, 635 m, 619 w, 602 w $\nu(SiC)$, $\nu(SiN)$; 515 m, 457 w, 438 w $\nu(AlC)$, $\nu(AlN)$; 351 vw $\delta(SiC)$. – $C_{20}H_{54}Al_2Li_2N_4Si_4$ (530.9): calcd. Al 10.2; found Al 10.1. – Mol. mass: 522 (cryoscopically in benzene).

Synthesis of 2: Dilithium bis(trimethylsilyl)hydrazide (0.602 g, 3.20 mmol) was dissolved in THF. The solvent was then removed in vacuo and an NMR spectroscopic characterization showed that the adduct $Li_2N_2(SiMe_3)2{\cdot}0.5THF$ had formed. This compound was suspended in 50 mL of *n*-hexane and cooled to −30°C. A solution of di(*tert*-butyl)aluminium chloride (1.132 g, 6.41 mmol) in 50 mL of *n*-hexane was then added dropwise, and the mixture was stirred at room temperature for 16 h. After filtration and evaporation the product was purified by recrystallizing twice from *n*-hexane. Yield: 0.170 g (10% based on hydrazide). – M. p. (argon, sealed capillary): 133°C (dec). – ^1H NMR (C_6D_6, 133 MHz): δ = 3.61 (m, 8 H, $O{-}CH_2$ of THF), 1.46 (s, 18 H, CMe_3 of iminoalane), 1.19 (s, 36 H, CMe_3 of chloroalane ligand), 0.97 (m, 8 H, CH_2 of THF), 0.42 (s, 18 H, $Al_2N{-}SiMe_3$), 0.15 [s, br., 18 H, $N_2(SiMe_3)_2$]. – ^{13}C NMR (C_6D_6, 75 MHz): δ = 73.4 ($O{-}CH_2$ of THF), 31.5 (CMe_3 of iminoalane), 30.5 (CMe_3 of chloroalane), 24.6 (CH_2 of THF), 7.8 and 3.7 ($N{-}SiMe_3$); AlC not detected. – IR (CsBr, paraffin): $\tilde{\nu}$ = 1300 vw cm^{-1}, 1250 s δCH_3; 1182 w, 1138 vw, 1109 vw, 1040 m, 1003 m, 968 s, 934 s $\nu(CC)$, $\nu(CO)$, $\nu(NN)$; 883 m, 839 vs $\rho(CH_3)$; 814 s $\nu(C_3C)$; 754 s, 723 w $\rho(CH_3)$; 664 w $\nu_{as}(SiC_3)$; 638 m, 621 m, 598 m $\nu_s(SiC_3)$, $\nu(SiN)$; 546 w, 523 w, 463 w $\nu(AlC)$; 434 m, 403 m $\nu(AlCl)$; 382 vw, 361 w $\delta(SiC_3)$. – $C_{44}H_{106}Al_4Cl_2Li_2N_4O_2Si_4$ (1028.4): calcd. Al 10.5; found Al 10.3.

Synthesis of 3: A solution of dineopentylaluminium chloride (0.826 g, 4.04 mmol) in 25 mL of toluene was added at −40°C to a suspension of dilithium bis(trimethylsilyl)hydrazide (0.381 g, 2.02 mmol) in 25 mL of toluene. The mixture was allowed to warm slowly to room temperature. After stirring for 6 h LiCl was filtered off, and the solvent was removed in vacuo. The residue was recrystallized from diethyl ether/*n*-hexane. Yield: 0.130 g (10% based on hydrazide). – M. p. (argon, sealed capillary): 141°C (dec). – ^1H NMR (C_6D_6, 300 MHz): δ = 3.20 (q, 4 H, $O{-}CH_2$), 1.43 (s, 18 H, CMe_3), 0.98 (t, 6 H, CH_3 of diethyl ether), 0.75 (s, 4 H, $Al{-}CH_2$), 0.43 (s, 18 H, $Al_2N{-}SiMe_3$), 0.11 [s, 18 H, $N_2(SiMe_3)_2$]. – ^{13}C NMR (C_6D_6, 75 MHz): δ = 65.5 ($O{-}CH_2$), 36.0 (CMe_3), 14.8 (CH_3 of diethyl ether), 7.4 and 4.1 ($N{-}SiMe_3$); AlC not detected. – IR (CsBr, paraffin): $\tilde{\nu}$ = 1304 w cm^{-1}, 1250 s $\delta(CH_3)$; 1182 w 1167 w, 1152 w, 1125 m, 1096 m, 1065 m, 1005 m, 970 m, 934 s $\nu(CO)$, $\nu(CC)$, $\nu(NN)$; 889 s, 837 vs, 756 s, 721 m $\rho(CH_3)$; 667 m, 662 sh, 621 w, 604 w $\nu(SiC)$, $\nu(SiN)$; 563 w, 525 m, 463 m $\nu(AlC)$; 425 w $\nu(AlCl)$. – $C_{22}H_{58}Al_2Li_2N_4Si_4{\cdot}OC_4H_{10}$ (633.0): calcd. Al 8.5; found Al 8.2.

Synthesis of 4: Dilithium bis(trimethylsilyl)hydrazide (1.52 g, 8.07 mmol) was dissolved in THF and the solvent removed in vacuo to give a solid containing half a molecule of THF per formula unit of the hydrazide. This solid was treated with 50 mL of *n*-hexane and cooled to −50°C. *tert*-Butylaluminium dichloride (1.26 g, 8.07 mmol) in 50 mL of *n*-hexane was then added, and the mixture was slowly warmed to room temperature in 8 h. Colorless crystals of **4** were obtained after filtration, evaporation and recrystallization (twice) from a mixture of hexane and toluene. Yield: 1.21 g (31%). − M. p. (argon, sealed capillary): 255 °C (dec). − ^{1}H NMR (C_6D_6, 300 MHz): δ = 3.95 (m, 8 H, O−CH_2), 1.30 (s, 18 H, CMe_3), 1.23 (m, 8 H, CH_2 of THF), 0.30 ppm (s, 18 H, N−$SiMe_3$). − ^{13}C NMR (C_6D_6, 75 MHz): δ = 71.6 (O−CH_2), 31.6 (C*Me*$_3$), 25.0 (CH_2 of THF), 7.0 (N−$SiMe_3$), Al−C not detected. − IR (CsBr, paraffin): ṽ = 1298 w cm^{-1}, 1246 m, 1235 m δ(CH_3); 1181 w, 1074 m, 1017 vs, 934 w, 922 w ν(CC), ν(CO); 845 s ρ(CH_3); 822 s ν_{C3C}; 760 m, 754 m, 721 w δ(CH_3); 681 m, 658 m, 629 w ν(SiC), ν(SiN); 544 w 519 w, 492 w, 463 w, 434 m ν(AlC), ν(AlN), ν(AlO); 397 vw, 374 vw, 351 vw, 328 vw δ(C_3C), δ(SiC). − $C_{22}H_{52}Al_2N_2O_2Si_2$ (486.8): calcd. Al 11.1; found Al 11.1. − Mol. mass: 455 (cryoscopically in benzene).

Synthesis of 5: Compound **4** (1.0 g, 2.05 mmol) was heated in vacuo to 150 °C for 5 h. Product **5** was extracted from the almost insoluble residue by treating with boiling toluene. It can be recrystallized from toluene. Yield: 0.25 g (36%), colorless crystals, sparingly soluble in organic solvents. − M. p. (argon, sealed capillary): >400 °C. − ^{1}H NMR (C_6D_6, 300 MHz): δ = 1.31 (s, 9 H, CMe_3), 0.42 (s, 9 H, N−$SiMe_3$). − ^{13}C NMR (C_6D_6, 75 MHz): δ = 32.3 (C*Me*$_3$), 8.0 ($SiMe_3$), Al−C not detected. − IR (CsBr, paraffin): ṽ = 1312 w, 1252 vs δ(CH_3); 1167 w, 1155 w, 1086 w, 1030 w, 1003 m, 936 s ν(CC); 876 vs, 837 vs ρ(CH_3); 808 vs ν(C_3C); 754 vs, 725 s ρ(CH_3); 679 w, 621 vs ν(SiC), ν(SiN); 552 vs, 465 m ν(AlC). − $C_{28}H_{72}Al_4N_4Si_4$ (685.2): calcd. Al 15.8; found Al 15.7.

X-ray Crystal Structure Determinations: Single crystals were obtained upon cooling solutions of the compounds in hexane/toluene (**1** and **4**), hexane (**2**), hexane/diethyl ether (**3**) or toluene (**5**). Crystal data and structure refinement parameters are given in Table 1.[24] In compound **2**, the trimethylsilyl groups of Si3 and Si4 are disordered. The carbon atoms were refined with occupancy factors of 0.6 and 0.4. The THF carbon atoms C1 and C8 were refined on split positions with occupancy ratios of 0.5 to 0.5 and 0.4 to 0.6, respectively. In compound **4**, the THF molecules of the heterocycle in general position were disordered, and the atoms C11, C12, C14, C21 and C24 were refined in split positions. The molecule located on a crystallographic inversion center was completely disordered and refined in two positions with occupancy factors of 0.68 and 0.32.

Quantum Chemical Calculations: Standard ab initio molecular orbital calculations[25] were carried out with the GAUSSIAN 94 set of programs.[26] The structure of **1a** was optimized at the HF level by using the 6−31G* basis set. Population analysis was performed by employing the NBO program[27] within GAUSSIAN. Bond lengths [pm] and angles [°] of **1a** (average values, experimental values in brackets): Al−N(1,2) 201.6 (201.1), Al−N3 189.5 (188.5), Al−N4 190.2 (189.9), Li−N(3,4) 193.9 (199.6), Li−N(1,2) 198.3 and 219.3 (197.3 and 215.5), N−N 151.3 (158.9), Al−C 197.1 (200.7), N−Si(1,2) 172.8 (173.8), N−Si(3,4) 170.2 (169.1), Al···Al 250.7 (248.5), Li···Al 266.3 and 277.8 (265.9 and 282.8), Al−N−Al 82.7 (82.2), N−Al−N 93.0 (94.0), Li−N−Li 130.2 (129.7), N1−Li−N2 42.1 (45.1), Li−N−N 61.5 and 76.3 (61.5 and 73.6).

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Table 1. Crystal data, data collection parameters and structure refinement of compounds **1−5**[a]

	1	**2**	**3**	**4**	**5**
Formula	$C_{20}H_{54}Al_2Li_2N_4Si_4$	$C_{44}H_{106}Al_4Cl_2Li_2N_4O_2Si_4$	$C_{26}H_{68}Al_2Li_2N_4OSi_4$	$C_{22}H_{52}Al_2N_2O_2Si_2$	$C_{28}H_{72}Al_4N_4Si_4$
Crystal system	monoclinic	monoclinic	orthorhombic	monoclinic	triclinic
Space group	*C*2/c; No. 15[22]	$P2_1$/c; No. 14[22]	$P2_12_12_1$; Nr. 19[22]	$P2_1$/c; No. 14[22]	*P*−1; No. 2[22]
Z	8	4	4	6	1
Temperature [K]	293	213	213	213	213
$d_{calc.}$ [g/cm^3]	1.039	1.050	1.057	1.067	1.078
a [pm]	1943.2(4)	1618.9(1)	1200.2(2)	992.33(5)	1049.0(2)
b [pm]	1260.6(3)	1963.16(8)	1500.3(3)	1620.04(8)	1080.6(1)
c [pm]	2883.0(6)	2048.5(1)	2209.7(4)	2849.0(2)	1087.8(2)
α [°]	90	90	90	90	68.24(1)
β [°]	106.03(3)	91.845(7)	90	96.738(8)	78.81(2)
γ [°]	90	90	90	90	67.44(1)
V [10^{-30} m^3]	6788(3)	6507.0(6)	3979(1)	4548.4(5)	1055.6(3)
μ [mm^{-1}]	0.241	0.221[b]	0.217[b]	0.157[b]	0.246[b]
Crystal size [mm]	0.49 × 0.38 × 0.30	0.45 × 0.30 × 0.25	0.90 × 0.66 × 0.35	0.72 × 0.50 × 0.44	0.35 × 0.32 × 0.30
Diffractometer	AED-2	Stoe-IPDS	Stoe-IPDS	Stoe-IPDS	Stoe-IPDS
Radiation	Mo-K_α; graphite monochromator				
2Θ range [°]	$3.90 \le 2\Theta \le 45.94$	$4.14 \le 2\Theta \le 52.1$	$4.72 \le 2\Theta \le 52.02$	$4.84 \le 2\Theta \le 52.18$	$4.22 \le 2\Theta \le 52.24$
Index ranges	$-21 \le h \le 20$ $0 \le k \le 13$ $0 \le l \le 31$	$-20 \le h \le 19$ $-24 \le k \le 24$ $-25 \le l \le 25$	$-14 \le h \le 14$ $-18 \le k \le 18$ $-24 \le l \le 24$	$-11 \le h \le 11$ $-19 \le k \le 19$ $-35 \le l \le 34$	$-12 \le h \le 12$ $-12 \le k \le 12$ $-13 \le l \le 13$
Independent reflections	4710	12722	7355	8339	3795
Reflections $F > 4\sigma(F)$	3018	6960	6739	6306	2491
Parameters	307	657	372	563	193
$R = \Sigma\lVert F_o\rvert - \lvert F_c\rVert/\Sigma\lvert F_o\rvert$ ($F > 4\sigma(F)$)	0.0780	0.0569	0.0338	0.0582	0.0726
$wR^2 = \{\Sigma w(F_o^2 - F_c^2)^2/\Sigma w(F_o^2)^2\}^{1/2}$ (all data)	0.1107	0.1520	0.0982	0.1835	0.1259
Max./min. residual [10^{30} e/m^3]	0.34/−0.33	0.53/−0.38	0.49/−0.22	0.54/−0.64	0.85/−0.39

[a] Program: SHELXTL, SHELXL-93;[23] solutions by direct methods; full matrix refinement with all independent structure factors. − [b] Numerical absorption correction.

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