X-ray Crystal Structures and Some Solution Structures of Lithium Amides with Intramolecular Complexation of Lithium by Tertiary Amide Groups

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Abstract: Lithium bis(*N*,*N*-dimethyl-2-aminoethyl)amide (**1a**) and lithium *N*-methyl-*N*-(3,6-diaza-3,6-dimethyl)heptylamide (**2**) are dimers in toluene, THF and in the crystalline state where they exhibit diamond-shaped Li_2N_2 cores. Tetracoordination of lithium is achieved by intramolecular binding of the amine nitrogen atoms. The dimers are broken down by hexamethylphosphoramide (HMPA), in the case of **1a** via **1a**$\cdot$HMPA, into solvent-separated ion pairs $RR'N^-$ $[Li(HMPA)_4]^+$ (R, R′ = $Me_2NCH_2CH_2$, $Me_2NCH_2CH_2$; $Me_2NCH_2CH_2(Me)NCH_2CH_2$, Me). For lithium *N*-methyl-*N*-(*N*′,*N*′-dimethyl-2-aminoethyl)amide (**3a**), seven ^6Li NMR signals are observed in toluene. In THF, **3a**$\cdot$2THF prevails, addition of HMPA leads to solvent-separated ion pairs through several intermediate species.

Keywords: coordination modes · ion pairs · HMPA · lithium amides · structure elucidation

Introduction

By complexation to lithium, Lewis bases such as R_2O, R_3N greatly influence the mode of aggregation and, as a consequence, the stabilities and reactivities of organolithium compounds (LiR) and lithium amides ($LiNR_2$). For $LiNR_2$ compounds, which are important reagents for the generation of enolates and their analogues from carbonyl compounds and related CH acids,[1] extensive work on solid-state[2, 8] and solution structures, much of the latter by the group of D.B. Collum,[3, 14] has established complexes, mostly $\{Li_m(NR_2)_m\cdot n(\text{Lewis base}), m = 1, 2, n = m, 2m\}$, that prevail in proportions that depend on R, Lewis base (which may be the solvent or an additive), temperature, and concentration. In some reactions, monomers were found to be most reactive,[4, 15] in other cases, however, full mechanistic understanding and rational selectivity control based on it, suffer from the possibility that several species may occur and that their rapid interconversion may prevent determination of individual contributions to the overall reactivity. In the LiR field, we have tackled this problem by studying organolithium compounds with *intramolecular* Lewis base $\{LiR'\text{-}(\text{Lewis base})_n\}$.[5] A single type of aggregate complexed intramolecularly according to the fixed ratio Li:Lewis base = 1:*n* is usually found for these RLi compounds in noncomplexing solvents. From the study of these species, relative energies and relative reactivities were obtained, which we consider representative of those of corresponding types of Lewis base complexed aggregates of ordinary LiR compounds.[6] The same aims have led us to apply the strategy of *intramolecular* Lewis base complexation to $LiNR_2$ compounds. We now report the X-ray crystal structures of lithium bis(*N*,*N*-dimethyl-2-aminoethyl)amide (**1a**)[7] and lithium *N*-methyl-*N*-(3,6-diaza-3,6-dimethyl)heptylamide (**2**)[7] and some solution structures of these lithium amides and their lower relative, lithium *N*-methyl-*N*-(*N*′,*N*′-dimethyl-2-aminoethyl)amide (**3a**). X-ray crystal structures are known of **1b**[8] and **3b**.[9] Compound **3b** and a few other intramolecularly Lewis base complexed lithium amides have been studied in the context of asymmetric induction.[9, 10]

Results and Discussion

Compounds **1a**, **2**, and **3a** were obtained by lithiation of the corresponding amine in pentane with BuLi at −78 °C. After stirring for 15 min at room temperature, the reaction mixture was evaporated to dryness after which the desired amount of solvent was added. Compound **1a** (Figure 1 top) crystallized from THF as the dimer **(1a)$_2$**. Each lithium atom of the diamond-shaped Li_2N_2 core is tetrahedrally surrounded by four nitrogen atoms, two of which are the amide nitrogen atoms (Li–N 2.02 Å), the other two (Li–N = 2.08 Å) are part

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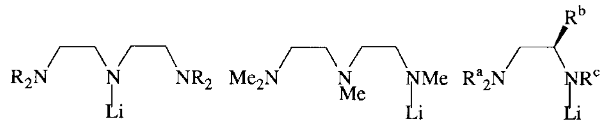


1 a: R = Me
b: R = Et

2

3 a: $R^a = R^c = Me$, $R^b = H$
b: $R^a_2 = (CH_2)_5$, $R^b = Ph$, $R^c = neo$-Pent

of the dimethylamino groups of chelating *N,N*-dimethyl-2-aminoethyl ligands of the amide nitrogen atoms, one from each monomer unit. A comparison with X-ray crystal structures of other $LiNR_2$ dimers, in particular with that of **1b**,[8] revealed no pecularities.

Compound **2** (Figure 1 bottom) crystallized from pentane as the dimer $\mathbf{2}_2$. Again, each lithium atom of the Li_2N_2 core is tetrahedrally surrounded by four nitrogen atomss, two of which are the amide nitrogen atoms (Li–N 2.00 Å), the other two the 5,8-nitrogen atoms of a 2,5,8-triaza-5,8-dimethyl-2-nonyl group that together with the bis-chelated lithium atom forms a bicyclo[3.3.0] ring system (Li–N(5) 2.19 Å, Li–N(8) 2.14 Å). The Li_2N_2 core is 25° out of plane, presumably due to the steric demands of six methyl groups located on the same face of the dimer.

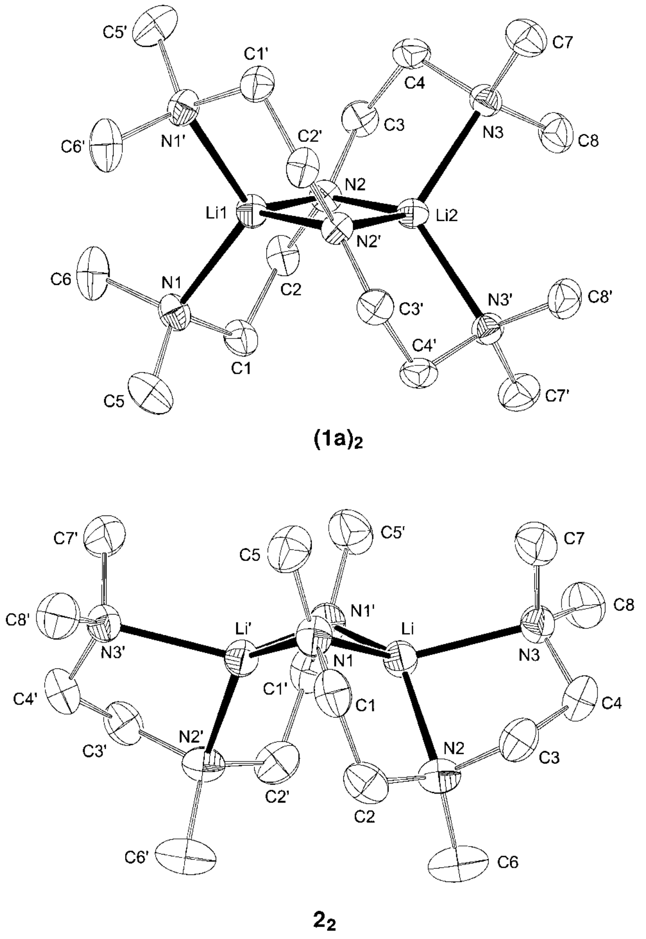
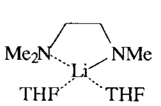


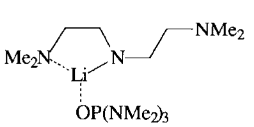
Figure 1. Displacement ellipsoid plots of $(\mathbf{1a})_2$ and $\mathbf{2}_2$ showing 50% probability. Hydrogen atoms have been omitted for clarity. Selected bond lengths [Å], angles [°] and torsion angles [°]: $(\mathbf{1a})_2$: N1–Li1 2.082(2), N2–Li1 2.013(2), N2–Li2 2.025(2), N3–Li2 2.083(2); N1-Li1-N2 90.69(4), N1-Li1-N1′ 118.04(10), N1-Li1-N2′ 126.15(4), N2-Li1-N1′ 126.15(4), N2-Li1-N2′ 108.44(10), N1′-Li1-N2′ 90.69(4), N2-Li2-N3 89.85(4), N2-Li2-N2′ 107.54(14), N2-Li2-N3′ 128.69(4), N3-Li2-N2′ 128.69(4), N3-Li2-N3′ 116.45(15), Li2-N2-Li1-N2′ 0.00(4). $\mathbf{2}_2$: N1–Li 2.004(2), N1–Li′ 2.003(3), N2–Li 2.192(2), N3–Li 2.144(3); N1-Li-N2 85.28(7), N1-Li-N3 124.06(10), N1-Li-N1′ 100.43(11), N2-Li-N3 85.34(9), N2-Li-N1′ 141.48(10), N3-Li-N1′ 119.85(9), Li′-N1-Li-N1′ 25.21(8).

Cryoscopy in THF on in situ generated **1a**–**3a** (ca. 0.12 M) indicated that at −108 °C, dimers prevail in the cases of **1a** (degree of association (*n*): 2.0 ± 0.5) and **2** (*n*: 2.1 ± 0.2), while monomer is the main component of solutions of **3a** (*n*: 1.3 ± 0.2). It is pointed out that for detectability reasons we had to operate at the limits of solubility of **1a**–**3a**. On occasions, **1a** crystallized from the solutions before THF started to freeze and after each measurement heterogenity was observed. Unfortunately, we have not yet succeeded in synthesizing the $^6Li^{15}NR_2$ isotopomers of **1a**–**3a** in yields providing sufficient amounts of material for more accurate distinctions between monomers, dimers, and higher oligomers by NMR spectroscopy.[11]

^{13}C and ^6Li NMR spectra recorded between −80 and −116 °C (Table 1) are in accord with the presence of $(\mathbf{1a})_2$ and $\mathbf{2}_2$, respectively: in toluene as well as in THF a single ^6Li signal is recorded for [^{6}Li]**1a** and for [^{6}Li]**2**, respectively, while two $^{13}CH_2$ and two $^{13}CH_3$ signals are consistent with the symmetry of $(\mathbf{1a})_2$ and the presence of $Li\cdots N(Me_2)$ bonds that are long-lived on the ^{13}C NMR time scale. Similarly, four $^{13}CH_2$ and four $^{13}CH_3$ signals are indicative of $\mathbf{2}_2$. By contrast, seven ^6Li signals in a solution of [^{6}Li]**3a** in toluene indicate the presence of several species. On addition of THF to this solution, the ^{13}C NMR spectrum changed until exactly two equivalents of THF per lithium had been added. The resulting spectrum, consisting of two $^{13}CH_2$ and three $^{13}CH_3$ signals, was identical with that of **3a** in neat THF. Together with the results of cryoscopy, the stoichiometric effect of THF indicates **3a**·2THF as the species predominating in THF and in THF/toluene. Its monomeric structure is in accord with the usual tetracoordination of lithium and is in analogy with **3b** which was proven to be present as a monomer in THF by NMR spectroscopy of [^{6}Li,^{15}N]**3b**.[9] Only one broad signal appeared in the ^6Li NMR spectrum of THF (containing) solutions of [^{6}Li]**3a**.



3a·2THF



1a·HMPA

On addition of up to two equivalents of HMPA to a solution of **1a** in THF, an additional set of ^{13}C NMR signals appeared, one half of which resembled those of $(\mathbf{1a})_2$, while the chemical shifts ($\delta = 60.03, 48.30, 45.82$) of the remaining signals were very close to those of the free $CH_2CH_2NMe_2$ groups of the solvent-separated ion pair $(Me_2NCH_2CH_2)_2N^-[Li(hmpa)_4]^+$ (vide infra). Complexation to lithium of a single molecule of HMPA was indicated by a doublet in the ^6Li NMR spectrum of [^{6}Li]**1a** ($^2J(^{31}P,^6Li) = 3$ Hz). The monomeric structure **1a**·HMPA was assigned to the new species on analogy with the findings for **3b** in toluene/HMPA.[9]

Table 1. ^{13}C[a] and ^{6}Li[b] Chemical shifts of the species formed by **1a**, **2**, and **3a** at −80 °C.

	$(\mathbf{1a})_2$[c,d]	**1a** · HMPA[e]	$(Me_2NCH_2CH_2)_2N^-[Li(hmpa)_4]^+$ [f]
^{13}C	64.16, 58.78, 48.11, 41.95	64.80, 60.03, 57.80, 49.00, 48.30, 45.82, 43.6	60.22, 48.51, 46.00
^{6}Li	(−95 °C) 0.960, s	(−114 °C) −0.690, d, $^2J(^{31}P,^{6}Li) = 3$ Hz	(−116 °C) −0.776, s
	$\mathbf{2}_2$[c,h]		$Me_2NCH_2CH_2(Me)NCH_2CH_2(Me)N^-[Li(hmpa)_4]^+$ [f]
^{13}C	62.90, 60.38, 58.07, 49.95, 48.51, 47.50, 45.62, 44.39		58.67 (2 ×), 57.14, 50.82, 46.78, 46.60, 43.44
^{6}Li	(−95 °C) 0.806, s		(−80 °C) −0.814, s
	3a · 2THF[c]		$Me_2NCH_2CH_2(Me)N^-[Li(hmpa)_4]^+$ [f]
^{13}C	62.96, 60.31, 47.63, 47.50, 41.00		60.25, 50.86, 46.21, 37.75
^{6}Li	(−100 °C) 0.530, bs		(−100 °C) −0.719, s

[a] δ (ppm, relative to TMS). [b] δ (ppm, relative to LiBr). [c] THF, 1 M.[g] [d] ^{13}C NMR chemical shifts in $[D_8]$toluene are lower by $\delta = 0.6–0.9$. [e] THF + 1.1 equivalents HMPA/Li, 1 M.[g] [f] THF + 20 equivalents HMPA/Li, 0.06 M.[g] [g] Lock $[D_8]$THF. [h] ^{13}C NMR chemical shifts in $[D_8]$toluene are lower by $\delta = 0.3–1.1$.

With more than two equivalents of HMPA, a gradual increase occurred of a three signal ^{13}C NMR pattern. Since the chemical shift of the ^{6}Li signal of $[^{6}Li]$**1a** ($\delta(^{6}Li, -116\,°C) = -0.776$) observed under these conditions is in the range typical for lithium ions tetracoordinated by HMPA,[12] the new set of signals was assigned to the solvent-separated ion pair $(Me_2NCH_2CH_2)_2N^-[Li(hmpa)_4]^+$ (solvent-separated ion pair). With four equivalents of HMPA, only the presumed **1a** · HMPA and solvent-separated ion pair were present in the ratio 1:1. Thus, the deaggregation of $(\mathbf{1a})_2$ by HMPA to solvent-separated ion pairs takes place in a stepwise manner via **1a** · HMPA.

By contrast, no changes were observed upon addition of up to two equivalents of HMPA to $\mathbf{2}_2$. With more than two equivalents of HMPA, a gradual increase of solvent-separated ion pair $Me_2NCH_2CH_2(Me)NCH_2CH_2(Me)N^-[Li(hmpa)_4]^+$ ($\delta(^{6}Li, -80\,°C) = -0.814$) was evidenced. Thus, it appears that contrary to $(\mathbf{1a})_2$, $\mathbf{2}_2$ is transformed by HMPA into solvent-separated ion pairs directly. The reason for the different behavior may be that $\mathbf{2}_2$ can deaggregate into two monomers (which then immediately form solvent-separated ion pairs with HMPA) without loss of $Li \cdots N(Me_2)$ bonding, while formation of monomer from $(\mathbf{1a})_2$ is more difficult, requiring, in addition to cleavage of the Li_2N_2 core, breaking of two $Li \cdots N(Me_2)$ bonds. Binding of HMPA (under formation of **1a** · HMPA) probably assists in the latter process. On addition of one equivalent of HMPA to $[^{6}Li]$**3a** in THF, six ^{6}Li signals appeared in the ^{6}Li NMR spectrum. Upon further addition, the number of signals decreased until only the signal of the solvent-separated ion pair, $Me_2NCH_2CH_2(Me)N^-[Li(hmpa)_4]^+$ ($\delta(^{6}Li, -100\,°C) = -0.719$), was left.

Conclusion

Neat THF, effecting di-etherates $(THF \cdot Li)_2(NR_2)_2$ is the strongest complexation medium for dimeric lithium amides.[13] Even the least hindered trialkylamines are relatively poor ligands for lithium amide dimers and chelation by *N,N,N′,N′*-tetramethylethylenediamine (TMEDA), 1,2-dimethoxyethane (DME), or related compounds occurs only with monomeric $LiNR_2$ compounds which are azaphilic, whereas dimers are oxaphilic.[14] In the lithium amide dimers $(\mathbf{1a})_2$ and $\mathbf{2}_2$, multiple intramolecular/intraaggregate complexation, favored by entropy and abating adverse steric demands of intermolecularly complexing trialkylamines, conspicuously alters these patterns: even a change of solvent from toluene to neat THF does not affect the $Li \cdots N(Me_n)$ bonds of $(\mathbf{1a})_2$ and $\mathbf{2}_2$! The notion that very similar species prevail in THF and in hydrocarbons such as toluene is supported by the finding that changing the solvent from pentane to THF hardly affects the kinetically controlled *E*/*Z* selectivity of lithium enolate formation from 3-pentanone by **1a** (ca. 20 °C, 15/85 and 12/88, respectively).[15] So far, only HMPA was found to deaggregate $(\mathbf{1a})_2$ and $\mathbf{2}_2$, thereby leading to solvent-separated ion pairs, $RR'N^-[Li(hmpa)_4]^+$ (R, R′ = $Me_2NCH_2CH_2$, $Me_2NCH_2CH_2$ (*E*/*Z* lithium 2-penten-3-olate: 7/93);[15] $Me_2NCH_2CH_2(Me)NCH_2CH_2$, Me) as the most stable states. The different ways in which $(\mathbf{1a})_2$ and $\mathbf{2}_2$ are converted by HMPA into solvent-separated ion pairs may prefigure different kinetic behaviour of the isomeric amides **1a** and **2** with reaction partners. For **3a**, in which only a single dimethylamino group provides chelation, behavior more akin to that of ordinary $LiNR_2$/Lewis base[13] is demonstrated by the conversion, upon addition of THF, of the unknown species present in toluene (*E*/*Z* lithium 2-penten-3-olate (pentane): 13/87)[15] into monochelated monomer **3a** · 2THF (*E*/*Z* lithium 2-penten-3-olate: 44/56).[15] The good solubility of $Li \cdots N(Me_n)$ stabilized/activated $(\mathbf{1a})_2$ and $\mathbf{2}_2$ in a (higher boiling) arene may be of promise for large-scale (industrial) syntheses, when avoidance of dangerous ethers is desirable.

Experimental Section

General: All reactions were run under an argon atmosphere in oven-dried glassware. Pentane and THF were freshly distilled from $LiAlH_4$, THF was stored over sodium wire. $[D_8]$THF was distilled from sodium/potassium alloy in sealed glassware, HMPA was distilled from KOH and stored over molecular sieves (4 Å). Solvents and reagents were manipulated with gas-tight syringes. ^{13}C, ^{6}Li, and ^{31}P NMR spectra were recorded with a Bruker MSL 400 at 100.62, 58.88, and 162.00 MHz, respectively. $[^{6}Li]$BuLi was prepared according to literature methods[16] and purified by short path distillation at 5×10^{-4} mbar. The colorless crystals were isolated after several crystallizations from a concentrated solution (>4 M) at −15 °C.

Preparation of 1a, 2 or 3a: To a solution of the corresponding amine (0.5 mmol) in pentane (5 mL) was added 1.0 equivalent of $[^{6}Li]$BuLi at −78 °C. After stirring for 15 min at room temperature, the reaction mixture was evaporated to dryness after which the desired solvent (0.5 mL) was added. Solutions prepared in this way proved to be perfectly stable for a year when stored at −80 °C.

X-ray structure analysis of $(\mathbf{1a})_2$: $C_{16}H_{40}Li_2N_6$, $M_r = 330.42$, monoclinic, *C*2/*c*, $a = 15.564(2)$, $b = 10.076(3)$, $c = 14.6643(13)$ Å, $\beta = 111.301(10)°$, $V =$

2142.6(7) Å^3, $Z=4$, $\rho_{calcd}=1.024$ g cm^{-3}, $T=150(2)$ K, $(\sin\theta/\lambda)_{max}=0.649$ Å^{-1}, colorless block $0.3\times0.5\times0.5$ mm, measured reflections: 8.145, unique reflections: 2444 ($R_{int}=0.059$), R values ($I>2\sigma(I)$): $R1=0.0395$, $wR2=0.0948$, all data: $R1=0.0558$, $wR2=0.1029$. $-0.17<\Delta\rho<0.19$ e Å^{-3}. Diffractometer: Enraf–Nonius CAD4T with rotating anode ($\lambda=0.71073$ Å). Absorption correction was not considered necessary. Structure solution with direct methods (SHELXS-96.)[17] Structure refinement with SHELXL-97[18] against F^2. 190 parameters, no restraints. Non-hydrogen atoms were refined with anisotropic temperature parameters, hydrogen atoms were refined freely with isotropic temperature parameters. Structure graphics and checking for higher symmetry were performed with the program PLATON.[19]

X-ray structure analysis of $\mathbf{2_2}$: $C_{16}H_{40}Li_2N_6$, $M_r=330.42$, monoclinic, $C2/c$, $a=21.998(3)$, $b=8.211(4)$, $c=15.815(3)$ Å, $\beta=130.232(18)^\circ$, $V=2180.8(12)$ Å^3, $Z=4$, $\rho_{calcd}=1.006$ g cm^{-3}, $T=150(2)$ K, $(\sin\ \theta/\lambda)_{max}=0.649$ Å^{-1}, colorless cube $0.5\times0.5\times0.5$ mm, measured reflections: 10100, unique reflections: 2500 ($R_{int}=0.039$), R values ($I>2\sigma(I)$): $R1=0.0394$, $wR2=0.1036$, all data: $R1=0.0472$, $wR2=0.1101$. $-0.24<\Delta\rho<0.19$ e Å^{-3}. Diffractometer: Enraf–Nonius CAD4T with rotating anode ($\lambda=0.71073$ Å). Structure solution with direct methods (SIR97).[20] Structure refinement with SHELXL-97[18] against F^2. 189 parameters, no restraints. Non-hydrogen atoms were refined with anisotropic temperature parameters, hydrogen atoms were refined freely with isotropic temperature parameters. Structure graphics, checking for higher symmetry and absorption correction (routine DELABS, 0.38–0.79% transmission) were performed with the program PLATON.[19]

Crystallographic data (excluding structure factors) for the structures reported in this paper have been deposited with the Cambridge Crystallographic Data Center as supplementary publication no. CCDC-101758 (structure $\mathbf{(1a)_2}$) and CCDC-101759 (structure $\mathbf{2_2}$). Copies of the data can be obtained free of charge on application to CCDC, 12 Union Road, Cambridge CB21EZ, UK (fax: (+44)1223-336-033; e-mail: deposit@ccdc.cam.ac.uk).

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