Ring Chain Transformations; X:[1] Synthesis of Condensed (ω-Aminoalkyl)imidazoles by Ring Chain Transformation

Jörg Bohrisch,[a] Michael Pätzel,[a] Jürgen Liebscher,*[a] Gerhard Maas[b]

[a] Fachbereich Chemie, Humboldt-Universität Berlin, Hessische Str. 1–2, D-1040 Berlin Germany
[b] Fachbereich Chemie, Universität Kaiserslautern, Erwin-Schrödinger-Str., D-6750 Kaiserslautern, Germany

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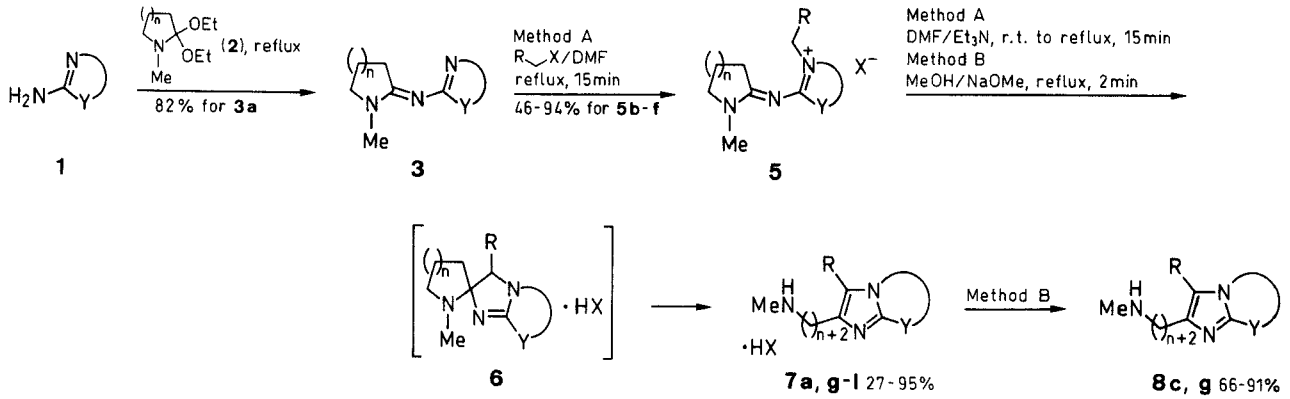
Dedicated to Professor Dr. Johannes C. Jochims on the occasion of his 60th birthday

2-Aminoazaheterocycles **1** react with lactam acetals (2,2-diethoxy-1-methylpyrrolidine, -1-methylpiperidine and -hexahydro-1-methyl-1*H*-azepine, **2**) affording 2-lactamimino azaheterocycles **3**. Reaction of the latter with CH-acidic methyl halides **4** allows a ring chain transformation via *N*-alkylation products **5** resulting in novel 2-[ω-(methylammonio)alkyl]imidazo[2,1-*b*]thiazole, -[1,2-*a*]pyridine and -[1,2-*a*]pyrimidine halides **7** and the corresponding 2-[ω-(methylamino)alkyl] compounds **8**.

Ring chain transformation is a useful principle for the synthesis of different types of (ω-aminoalkyl)heterocycles,[2–5] which are interesting heteroanalogues of histamine. Bridged, 1,3-dicarbonyl or imide heteroanalogues served as starting materials which, for example, were reacted with reagents possessing both, electrophilic and nucleophilic properties.[4,6]

Continuing our studies we tried now to apply doubly bridged heteroanalogues of imides, i.e. lactamimino heterocycles **3**, in order to synthesize condensed (ω-aminoalkyl)heterocycles, which might be of interest as analogues of biologically active compounds.[7,8]

The starting lactamimine derivatives **3** were prepared by reaction of 2-aminoazaheterocycles **1** with lactam acetals **2**. The products **3**, which often appear as oils were usually used in subsequent reactions without further purification. Attempts to use the lactamimino-substituted heterocycles **3** as bielectrophilic C–N–C building blocks in a reaction with hydrazines failed, because the starting aminoazaheterocycles **1** were formed by cleavage of the exocyclic C–N bond rather than the lactamimine ring. We further tried to apply the lactamimino heterocycles as an N–C–N–C building block for an imidazole ring by reacting with CH-acidic methyl halides **4** in the presence of a base (Method A). This procedure usually results in the formation of hydrohalides **7** of condensed [ω-(methylamino)-alkyl]imidazoles **8**. In some cases **(5b,c,d,e,f)**, only *N*-methylation products **5** were obtained, which resist further cyclization to the corresponding imidazole products **7** or **8** even with stronger bases and at prolonged heating. Probably, the success of the cyclization is



Scheme

3,5–8	Y	n	R	X
a		1	4-BrC_6H_4CO	Br
b		3		Br
c		1	4-BrC_6H_4CO	Br
d		1	4-$O_2NC_6H_4$	Br
e		1		Br
f		3	4-BrC_6H_4CO	ClO_4

3,5–8	Y	n	R	X
g		1	4-BrC_6H_4CO	Br
h		1	CN	Cl
i		1	4-$O_2NC_6H_4CO$	Br
j		2	4-$O_2NC_6H_4CO$	Br
k		3	CN	Cl
l		1	4-BrC_6H_4CO	Br

Table 1. Interatomic Distances (d) Indicating Hydrogen Bonds[a]

	d[Å]		d[Å]
Br1 ... O1[I]	3.545(15)	N2 ... O2	2.894(24)
Br2 ... O2[II]	3.240(15)	N3 ... N4[IV]	3.182(20)
Br2 ... O3[III]	3.336(18)	N4 ... O3	2.766(22)
Br2 ... N4	3.233(18)	O2 ... O3[III]	2.631(24)

[a] Symmetry code: No index: x, y, z; I: x, y − 1, z; II: 1/2 − x, y − 1/2, 1/2 + z; III: 1/2 − x, 1/2 + y, 1/2 + z; IV: 1/2 − x, 1/2 + y, z − 1/2.

influenced by the nature of the electron-withdrawing substituent R and by ring size of the lactamimine ring.

The mechanism of formation of the (ω-aminoalkyl)-imidazoles **7** and **8** (Scheme) should resemble the classical mechanism of the comparable known synthesis of condensed imidazoles lacking the aminoalkyl substituent from non-bridged C–N–C–N building blocks and CH-acidic methyl halides.[9] The deprotonated CH_2 group of the *N*-alkylation product **5** attacks the lactam imine carbon atom giving a spiro intermediate **6**, which opens the previous lactamimine ring (ring chain transformation). An explanation of the failure of the cyclization of some *N*-alkylated compounds **5** might be given by the assumption that isomers of **5** are formed, derived from alkylation of the exocyclic N atom. However, NOE difference ^{1}H NMR investigations of compound **5d** revealed, that the RCH_2 group is in neighborhood of the NCH group of the thiazole ring thus proving alkylation of the ring N atom.

The structure of products **3, 5, 7,** and **8** was confirmed by spectroscopic methods (Table 2 and 3). The differentiation between the isomeric structures **5, 6** and **7** is possible by NMR (CH_2 signals are only observed in **5**) and mass spectroscopy (see previous publications[2,5]). The spiro compounds **6** are ruled out by ^{13}C NMR (no sp^3-NCN). In the case of compound **7l** (Method A) a byproduct was obtained whose analytical data did not match with structure **7** or **8**. X-ray crystal analysis (Figure, Table 1) revealed that unexpectedly this compound is the dihydrate **7l** · $2H_2O$ of hydrobromide **7l**. In the crystal lattice, the bromine atom, the nitrogen atom N4, and the two water oxygen atoms are involved in a number of hydrogen bonds that build up a three-dimensional network.

The isolated hydrobromide **7l** can be transformed into the corresponding dihydrate **7l** · $2H_2O$ by heating in aqueous dimethylformamide while other solvent/water mixtures leave **7l** unaffected. It is also worth mentioning that compound **7l** and its dihydrate **7l** · $2H_2O$ give different ^{1}H NMR spectra in DMSO-d_6 as well as in DMSO-d_6/water. Thermoanalysis of the dihydrate shows one molecule of water being eliminated on melting at about 107 °C while the second water unit sticks to the molecule until general decomposition takes place (above 205 °C). These results suggest that the dihydrate **7l** · $2H_2O$ is not a simple hydrate only existing in the crystal lattice but is a kind of supramolecular complex.

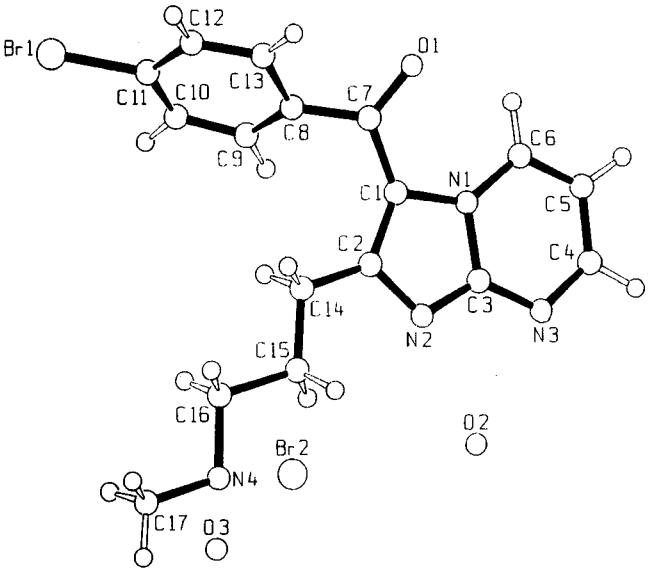


Figure. Plot (SCHAKAL) of **7l** · $2H_2O$ in the crystalline state. The hydrogen atoms at N4, Br2, O2, and O3 have not been localized.

The results described here demonstrate that the principle of ring chain transformation can also be successfully applied to the synthesis of condensed (ω-aminoalkyl)-heterocycles.

2-(1-Methyl-2-pyrrolidinylideneamino[-2-piperidinylideneamino or -hexahydro-1*H*-azepin-2-ylideneamino]) Substituted Thiazoles, Pyridines and Pyrimidine 3; General Procedure:
A mixture of 2-aminoazaheterocycle **1** (0.01 mol) and of lactam acetal **2** (0.01 mol) was heated until the evaporation of EtOH was complete. After cooling to r.t. products **3** either crystallized or remained oily. They could be purified by recrystallization or column chromatography, respectively (Table 2). Usually, they were used for Method A without further purification.

1-Alkyl-2-(1-methyl-2-pyrrolidinylideneamino[-2-piperidinylideneamino or -hexahydro-1*H*-azepin-2-ylideneamino]thiazolium Halides 5b–f and 2-[ω-(methylammonio)alkyl]imidazo[2,1-*b*]thiazole-[1,2-*a*]pyridine and -[1,2-*a*]pyrimidine Halides, 7a,g–l; General Procedure:
Method A: A solution of **3** (0.01 mol) and CH-acidic methyl halide **4** (0.01 mol) in DMF (15 mL) was heated under reflux for 15 min. After cooling to r.t. Et_3N (2 mL) was added and heating was continued for further 15 min. The product was precipitated by gradual dilution of the cold mixture with Et_2O. In some cases oily salts were obtained, which had to be washed with Et_2O several times until they crystallized upon scratching with a glass rod. Compound **5f** was isolated as a perchlorate by adding $HClO_4$ to the mixture (Table 2).

3-(4-Bromobenzoyl)-2-[3-(methylamino)propyl]imidazo[2,1-*b*]thiazole (8c) and -[1,2-7*a*]pyridine (8g); General Procedure:
Method B: *N*-Alkylation product **5** or cyclization product **7** obtained from method A was mixed with a solution of Na (0.34 g, 0.015 mol) in MeOH (20 mL). After short heating to reflux the product was precipitated from the cold mixture by dilution with ice-water. Product **8** was filtered and recrystallized (Table 2).

X-ray Structural Analysis of 7l · $2H_2O$:
Crystal Data: Orthorhombic space group $Pna2_1$; a = 24.795 (11), b = 9.855 (5), c = 8.186 (10) Å; $\alpha = \beta = \gamma = 90°$; D_{calc} = 1.628 g · cm^{-3}; Z = 4. *Data Collection:* Crystal size 0.14 × 0.18 × 0.45 mm; Enraf-Nonius CAD4 diffractometer, monochromatized Mo K_α radiation; 1633 independent reflections were measured, scan width (0.95 + 0.35 tanθ)°. An empirical (Fourier series) absorption

Table 2. Lactamimino Heterocycles **3**, *N*-Alkylated Lactamimino Heterocycles **5** and Condensed (ω-Aminoalkyl)imidazoles **7** and **8**

Compound	Yield (%) Method	mp (°C)	Molecular Formula[a]	^{1}H NMR[b] δ, J (Hz)
3a	82	84–86 (cyclohexane)	$C_8H_{13}N_3S$ (183.3)	2.03 (quint, 2H, J = 7, 4-CH_2), 2.82 (t, 2H, J = 7, 3-CH_2), 2.90 (s, 3H, NCH_3), 3.40 (t, 2H, J = 7, SCH_2), 3.42 (t, 2H, J = 7, CH_2NMe), 4.19 (t, 2H, J = 7, NCH_2S)
5b	55/A	192–195 (AcOH/Et_2O)	$C_{15}H_{21}BrN_4O_3S$ (417.4)	1.70 (m, 6H, 4-CH_2, 5-CH_2, 6-CH_2), 2.98 (m, 2H, 3-CH_2), 3.29 (s, 3H, NCH_3), 3.60 (t, 2H, J = 6, SCH_2), 3.78 (m, 2H, CH_2NMe), 4.06 (t, 2H, J = 6, N^+CH_2), 4.90 (s, 2H, N^+CH_2), 6.95 (d, 1H, J = 5, CH_{furyl}), 7.71 (d, 1H, J = 5, CH_{furyl})
5c	78/A	244–246 (AcOH)	$C_{16}H_{17}Br_2N_3OS$ (459.2)	2.13 (quint, 2H, J = 7, 4-CH_2), 2.89 (s, 3H, NCH_3), 3.05 (t, 2H, J = 7, 3-CH_2), 3.75 (t, 2H, J = 7, 5-CH_2), 5.98 (s, 2H, N^+CH_2), 7.50 (d, 1H, J = 5, SCH=), 7.85 (d, 1H, J = 5, N^+CH=), 7.85 (d, 2H, J = 7, CH_{arom}), 8.06 (d, 2H, J = 7, CH_{arom})
5d	94/A	240–245 (AcOH)	$C_{15}H_{17}BrO_2S$ (397.3)	2.28 (quint, 2H, J = 7, 4-CH_2), 3.06 (m, 2H, 3-CH_2), 3.25 (s, 3H, NCH_3), 3.75 (t, 2H, J = 7, 5-CH_2), 5.62 (s, 2H, N^+CH_2), 7.53 (d, 1H, J = 5, SCH=), 7.65[c] (d, 2H, J = 8, CH_{arom}), 8.08[c] (d, 1H, J = 5, N^+CH=), 8.31 (d, 2H, J = 8, CH_{arom})
5e	95/A	198–200 (AcOH/Et_2O)	$C_{13}H_{15}BrN_4O_3$ (387.3)	2.47 (quint, 2H, J = 7, 4-CH_2), 3.06 (t, 2H, J = 7, 3-CH_2), 3.16 (s, 3H, NCH_3), 3.62 (t, 2H, J = 7, 5-CH_2), 5.59 (s, 2H, N^+CH_2), 6.94 (d, 1H, J = 6, CH_{furyl}), 7.40 (d, 1H, J = 6, SCH_2), 7.65 (d, 1H, J = 6, CH_{furyl}), 7.92 (d, 1H, J = 6, N^+CH_2)
5f	46/A	180–182 (EtOH)	$C_{18}H_{21}BrClN_3O_5S$ (506.8)	1.71 (m, 6H, 4-CH_2, 5-CH_2, 6-CH_2), 2.94 (m, 2H, 3-CH_2), 3.04 (s, 3H, NCH_3), 3.70 (m, 2H, 7-CH_2), 5.78 (s, 2H, N^+CH_2), 7.32 (d, 1H, J = 5, N^+CH=), 7.85 (d, 2H, J = 7, CH_{arom}), 7.98 (d, 2H, J = 7, CH_{arom})
7a	80/A	118–121 (AcOH)	$C_{16}H_{19}Br_2N_3OS$ (461.3)	1.81 (quint, 2H, J = 7, β-CH_2), 2.33 (t, 2H, J = 7, α-CH_2), 2.53 (s, 3H, NCH_3), 2.75 (m, 2H, γ-CH_2), 3.94 (t, 2H, J = 7, SCH_2), 4.30 (t, 2H, J = 7, NCH_2), 7.59 (d, 2H, J = 9, CH_{arom}), 7.78 (d, 2H, J = 9, CH_{arom})
7g	95/A	167–169 (MeCN)	$C_{18}H_{19}Br_2N_3O$ (453.2)	2.19 (quint, 2H, J = 7, β-CH_2), 2.68 (t, 2H, J = 7, α-CH_2), 2.76 (s, 3H, NCH_3), 3.11 (t, 2H, J = 7, γ-CH_2), 7.13 (t, 1H, J = 6, CH_{pyr}), 7.56 (m, 1H, CH_{pyr}), 7.56 (d, 2H, J = 8, CH_{arom}), 7.69 (d, 2H, J = 8, CH_{arom}), 7.71 (t, 1H, J = 6, CH_{pyr}), 9.36 (d, 1H, J = 6, CH_{pyr})
7h	54/A	237–240 (*i*-PrOH)	$C_{12}H_{14}Cl_2N_4$ (285.2)	2.12 (quint, 2H, 2H, J = 7, β-CH_2), 2.59 (s, 3H, NCH_3), 2.97 (m, 4H, α-CH_2, γ-CH_2), 7.69 (d, 1H, J = 10, CH_{pyr}), 7.89 (d, 1H, J = 10, CH_{pyr}), 8.90 (s, 1H, CH_{pyr})
7i	80/A	207–210 (MeCN)	$C_{18}H_{17}BrClN_4O_3$ (453.8)	1.91 (quint, 2H, J = 7, β-CH_2, 2.38 (t, J = 7, 2H, α-CH_2, 2.52 (s, 3H, NCH_3), 2.77 (t, 2H, J = 7, γ-CH_2), 7.90 (m, 2H, CH_{pyr}), 8.00 (d, 2H, J = 9, CH_{arom}), 8.50 (d, 2H, J = 9, CH_{arom}), 9.53 (s, 1H, CH_{pyr})
7j	93/A	213–215 (MeCN)	$C_{19}H_{19}BrClN_4O_3$ (467.8)	1.50 (m, 4H, β-CH_2, γ-CH_2), 2.32 (t, 2H, J = 7, α-CH_2), 2.53 (s, 3H, NCH_3), 2.75 (t, 2H, J = 7, δ-CH_2), 7.89 (m, 2H, CH_{pyr}), 8.01 (d, 2H, J = 8, CH_{arom}), 8.50 (d, 2H, J = 8, CH_{arom}), 9.56 (s, 1H, CH_{pyr})
7k	61/A	178–179 (*i*-PrOH)	$C_{14}H_{18}Cl_2N_4$ (313.3)	1.62 (m, 6H, β-CH_2, γ-CH_2, δ-CH_2), 2.54 (s, 3H, NCH_3), 2.85 (t, 4H, J = 7, α-CH_2, ε-CH_2), 7.65 (d, 1H, J = 10, CH_{pyr}), 7.85 (d, 1H, J = 10, CH_{pyr}), 8.89 (m, 1H, CH_{pyr})
7l	27/A	230–235 (EtOH)	$C_{17}H_{18}Br_2N_4O$ (454.2)	2.05 (m, 2H, β-CH_2), 2.61 (s, 3H, NCH_3), 3.08 (t, 2H, J = 7, α-CH_2), 3.37 (m, 2H, γ-CH_2), 7.31 (dd, 1H, J_1 = 7, J_2 = 4, CH_{pyrim}), 7.84 (d, 2H, J = 8, CH_{arom}), 8.36 (d, 2H, J = 8, CH_{arom}), 8.69 (dd, 1H, J_1 = 4, J_2 = 2, CH_{pyrim}), 8.79 (vb, 1H, NH), 9.26 (dd, 1H, J_1 = 6, J_2 = 2, CH_{pyrim})
7l·2H$_2$O	33/A	97–103 ($CHCl_3$)	$C_{17}H_{18}Br_2N_4O \cdot 2H_2O$) (490.2)	1.98 (m, 2H, β-CH_2), 2.57 (s, 3H, NCH_3), 2.81 (m, 4H, α-CH_2, γ-CH_2), 7.44 (dd, 1H, J_1 = 7, J_2 = 4, CH_{pyrim}), 7.72 (d, 2H, J = 8, CH_{arom}), 7.89 (d, 2H, J = 8, CH_{arom}), 8.53 (vb, 1H, NH), 8.89 (dd, 1H, J_1 = 4, J_2 = 2, CH_{pyrim}), 9.59 (dd, 1H, J_1 = 6, J_2 = 2, CH_{pyrim})
8c	66/B[d]	157–160 (H_2O)	$C_{16}H_{16}BrN_3OS$ (378.3)	1.59 (quint, 2H, J = 7, β-CH_2), 2.14 (s, 3H, NCH_3), 2.31 (m, 4H, α-CH_2, γ-CH_2), 7.26 (d, 1H, J = 5, CH_{thiaz}), 7.56 (d, 2H, J = 9, CH_{arom}), 7.81 (d, 2H, J = 9, CH_{arom}), 7.94 (d, 1H, J = 5, CH_{thiaz}), 8.26 (s, 1H, NH)
8g	91/B[e]	75–77 (cyclohexane)	$C_{18}H_{18}BrN_3O$ (372.3)	1.15 (br s, 1H, NH), 1.78 (quint, 2H, J = 7, β-CH_2), 2.32 (s, 3H, NCH_3), 2.39 (t, 2H, J = 7, α-CH_2), 2.51 (t, 2H, J = 7, γ-CH_2), 7.04 (t, 1H, J = 6, CH_{pyr}), 7.49 (t, 1H, J = 6, CH_{pyr}), 7.56 (d, 2H, J = 9, CH_{arom}), 7.66 (d, 2H, J = 9, CH_{arom}), 7.69 (d, 1H, J = 6, CH_{pyr}), 9.40 (d, 1H, J = 6, CH_{pyr})

[a] Satisfactory microanalyses obtained: C + 0.37, H + 0.18, N + 0.32.
[b] 80 MHz (DMSO-d_6/TMS): **5e**, **7a**, **7a–l**; ($CDCl_3$/TMS): **8c**
300 MHz (DMSO-d_6/TMS): **5b**, **5c**, **5d**, **5f**, **7g**; ($CDCl_3$/TMS): **3a**, **8g**
[c] Positive NOE signal after irradiation at δ = 5.62.
[d] Starting from **5c**.
[e] Starting from **7g**.

correction was applied; transmission ranged from 0.673 to 1.388. *Structure solution and refinement:* The structure was solved by direct methods and refined by a full-matrix-least-squares method. Hydrogen atoms were calculated as far as possible (except for NH, HBr, and H_2O) and included in the structure factor calculation. Carbon atoms C3, C8 were refined only isotropically. Refinement with 897 reflections [$I > 2\sigma(I)$] and 224 parameters converged at R = 0.0673, R_w = 0.0595; the residual electron density was ≤ 0.77. The absolute structure (i.e. the direction of the polar axis) was determined by comparison of the R values of the two enantiomorphic structures; the wrong structure gave R = 0.070, R_w = 0.063. Tables of positional and thermal parameters, the list of bond lengths and angles, and a list of the observed and calculated structure factors have been deposited.[10,11]

Table 3. ^{13}C NMR Data of compounds[a] **5**, **7** and **8**

Compound	δ
5d	18.7 (4-CH_2), 30.4 (3-CH_2), 32.9 (NCH_3), 51.1 (5-CH_2), 53.3 (N^+CH_2), 111.3 (CH_{thiaz}), 123.9 (CH_{arom}), 129.2 (CH_{arom}), 130.8 (CH_{arom}), 142.8 (C_{arom}), 147.5 (C_{arom}), 169.2 (C_{arom}), 170.7 (C_{amidin})
7a	24.7 (β-CH_2), 26.1 (α-CH_2), 32.5 (NCH_3), 35.7 (SCH_2), 47.7 (NCH_2), 47.9 (NCH_2), 47.9 (NCH_2), 126.3 (C_{arom}), 127.3 (C_{arom}), 130.5 (CH_{arom}), 131.9 (CH_{arom}), 137.8 (C_{arom}), 153.6 (C_{arom}), 155.1 (C_{arom}), 183.6 (C=O)
7h	24.3 (β-CH_2), 25.3 (α-CH_2), 32.4 (NCH_3), 47.8 (γ-CH_2), 96.1 (C_{arom}), 111.1 (CN), 117.9 (CH_{arom}), 121.8 (C_{arom}), 125.2 (CH_{arom}), 130.1 (CH_{arom}), 144.8 (C_{arom}), 156.5 (C_{arom})
7i	24.3 (β-CH_2), 26.9 (α-CH_2), 32.5 (NCH_3), 48.0 (γ-CH_2), 117.6 (CH_{arom}), 120.8 (C_{arom}), 122.2 (C_{arom}), 124.1 (CH_{arom}), 126.2 (CH_{arom}), 129.5 (CH_{arom}), 130.9 (CH_{arom}), 145.2 (C_{arom}), 145.7 (C_{arom}), 149.3 (C_{arom}), 155.8 (C_{arom}), 184.2 (C=O)
7j	25.1 (γ-CH_2), 25.4 (β-CH_2), 29.1 (α-CH_2), 32.5 (NCH_3), 48.2 (δ-CH_2), 117.5 (CH_{arom}), 120.8 (C_{arom}), 122.0 (C_{arom}), 124.0 (CH_{arom}), 126.2 (CH_{arom}), 129.5 (CH_{arom}), 130.9 (CH_{arom}), 145.4 (C_{arom}), 145.8 (C_{arom}), 149.3 (C_{arom}), 157.0 (C_{arom}), 184.3 (C=O)
7k	25.1 (γ-CH_2), 25.6 (β-CH_2), 27.6 (δ-CH_2), 27.9 (α-CH_2), 32.9 (NCH_3), 48.1 (ε-CH_2), 96.0 (C_{arom}), 111.3 (CN), 117.9 (CH_{arom}), 121.7 (C_{arom}), 125.0 (CH_{arom}), 130.0 (CH_{arom}), 144.7 (C_{arom}), 157.8 (C_{arom})
7l	20.3 (β-CH_2), 24.0 (α-CH_2), 32.5 (NCH_3), 47.7 (γ-CH_2), 110.2 (CH_{arom}), 127.1 (C_{arom}), 128.9 (C_{arom}), 131.4 (CH_{arom}), 132.7 (CH_{arom}), 134.1 (CH_{arom}), 136.7 (C_{arom}), 138.6 (C_{arom}), 146.0 (C_{arom}), 153.0 (CH_{arom}), 188.7 (C=O)
7l·2H$_2$O	24.4 (β-CH_2), 26.8 (α-CH_2), 32.5 (NCH_3), 48.0 (γ-CH_2), 111.4 (CH_{arom}), 119.1 (C_{arom}), 126.3 (C_{arom}), 130.7 (CH_{arom}), 132.1 (CH_{arom}), 136.7 (CH_{arom}), 138.5 (C_{arom}), 149.8 (C_{arom}), 154.2 (CH_{arom}), 155.6 (C_{arom}), 185.0 (C=O)
8c	27.6 (β-CH_2), 29.0 (α-CH_2), 36.0 (NCH_3), 51.0 (γ-CH_2), 115.4 (CH_{arom}), 121.6 (CH_{arom}), 123.6 (C_{arom}), 125.6 (C_{arom}), 130.3 (CH_{arom}), 131.8 (CH_{arom}), 138.5 (C_{arom}), 154.1 (C_{arom}), 157.2 (C_{arom}), 183.6 (C=O)
8g	26.8 (β-CH_2), 28.0 (α-CH_2), 36.3 (NCH_3), 51.4 (γ-CH_2), 114.4 (CH_{arom}), 116.8 (CH_{arom}), 120.7 (C_{arom}), 126.5 (C_{arom}), 128.4 (CH_{arom}), 129.3 (CH_{arom}), 129.9 (CH_{arom}), 131.8 (CH_{arom}), 139.1 (C_{arom}), 147.7 (C_{arom}), 157.0 (C_{arom}), 185.5 (C=O)

[a] For solvents see footnote [b] of Table 2.

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(11) Further information on the X-ray analysis of **7l·2H$_2$O** is available from Fachinformationszentrum Karlsruhe, Gesellschaft für wissenschaftlich-technische Information mbH, D-7514 Eggenstein-Leopoldshafen 2. Requests should indicate the depository number CSD 56611, the names of the authors, and the literature citation of this paper.