Crystal Structures of Seven Terephthaldiamide Derivatives

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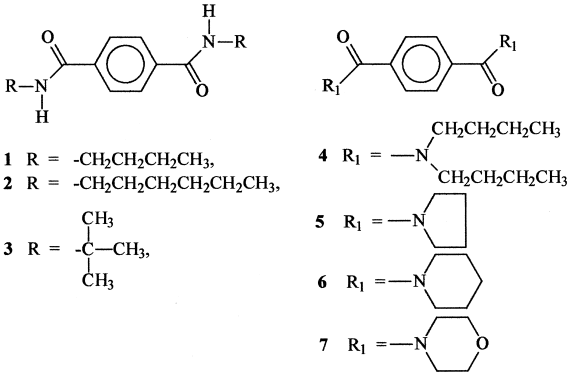
Terephthaldiamides, Crystal Structure, IR Spectra

N,N′-Dibutyl-terephthaldiamide (**1**), N,N′-dihexyl-terephthaldiamide (**2**), N,N′-di(*tert*-butyl)-terephthaldiamide (**3**), N,N,N′,N′-tetrabutyl-terephthaldiamide (**4**), 1,1′-terephthaloyl-bis-pyrrolidine (**5**), 1,1′-terephthaloyl-bis-piperidine (**6**), and 4,4′-terephthaloyl-bis-morpholine (**7**) have been synthesised and physicochemically characterised. The X-ray structure determinations reveal imposed inversion symmetry for compounds **1–6**; compound **3** has two independent molecules with inversion symmetry in the asymmetric unit. Compounds **1–3** form classical hydrogen bonds of the type N–H···O=C, leading to a ribbon-like arrangement of molecules (**1** and **2**) or a layer structure (**3**). Compound **3** also displays a very short C–H···O interaction, a type of hydrogen bond that is also observed in compounds **4–7**, which lack classical donors; thereby compounds **4–6** form layer structures and **7** a complex three-dimensional network.

Terephthalic acid and its derivatives are widely used organic compounds. Terephthalic acid is, for example, the basic component of a common polymer, PET. Since numerous terephthalic acid derivatives have found, or are expected to find, practical applications, reports in the literature have been largely confined to patent descriptions. This is also true for the compounds investigated in this contribution. *E.g.*, compound **1** has been described in early publications [1,2], but its physicochemical data have been limited to the melting point and a few IR bands [3]. Compound **2** was described, rather laconically, in the patent literature only [4]. Neither a preparative method nor physical or chemical data were given for this compound. Compound **3** has not been described earlier in the literature. Compounds **4–7** were described also very briefly in [5]. For compound **6** the ^{1}H NMR, and MS spectra were given [6]. The rotational barriers about the CO-N bond were calculated using ^{13}C NMR spectroscopy for compounds **5–7** [7]. Terephthaldiamides are generally and conveniently obtained from terephthalic acid chlorides and suitable amines. Terephthalic acid itself has been used less , largely because of its poor solubility in many solvents.



Results and Discussion

In order to obtain compounds **1**, **2**, **4–7** we proceeded differently, synthesizing suitable amides directly from dimethyl tetraphthalate and appro-

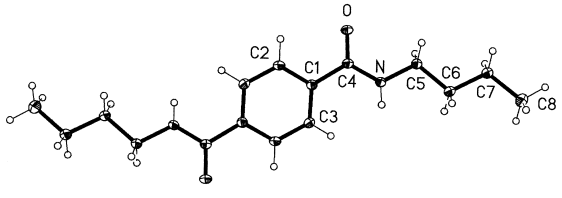


Fig. 1. The molecule of compound **1** in the crystal. Ellipsoids represent 50% probability levels. Only the asymmetric unit is numbered.

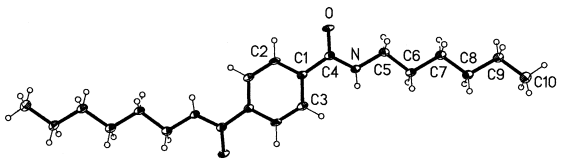


Fig. 2. The molecule of compound **2** in the crystal. Ellipsoids represent 50% probability levels. Only the asymmetric unit is numbered.

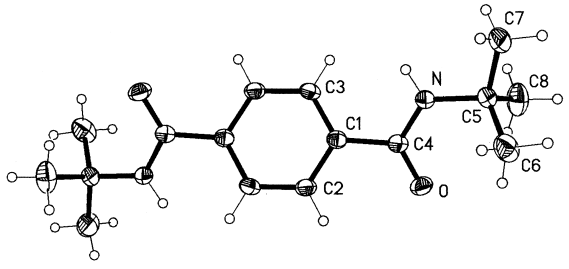


Fig. 3. The molecule of compound **3** in the crystal. Ellipsoids represent 50% probability levels. Only the asymmetric unit is numbered.

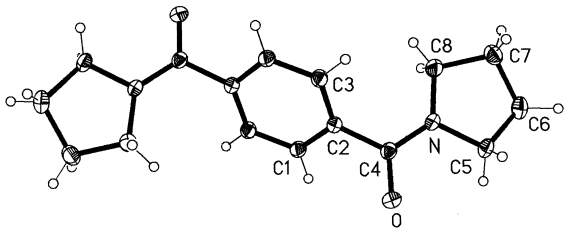


Fig. 5. The molecule of compound **5** in the crystal. Ellipsoids represent 50% probability levels. Only the asymmetric unit is numbered.

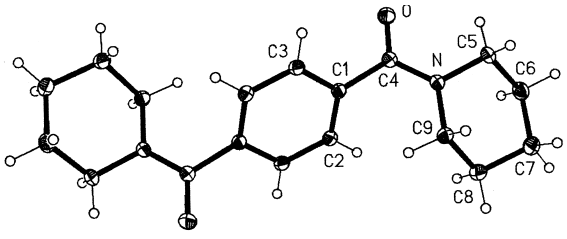


Fig. 6. The molecule of compound **6** in the crystal. Ellipsoids represent 50% probability levels. Only the asymmetric unit is numbered.

priate amines. Compound **3** was obtained from terephthaloyl chloride and *tert*-butylamine. Compound **2** is slightly soluble in DMSO, DMF, methanol, halogenated solvents and other common solvents. It is somewhat more soluble in pyridine. The other compounds are also poorly soluble, but more soluble than compound **2** (for example, their solubility in warm chloroform is sufficient to record NMR spectra). This is somewhat unexpected, since the presence of long hydrophobic chains of **2** should increase the solubility of compounds **1**–**4** in less polar solvents. If we look, however, at the corresponding crystal structures, hydrogen bonds between molecules can be seen, along with a typical mutual alignment of aliphatic hydrophobic chains. These structural effects are

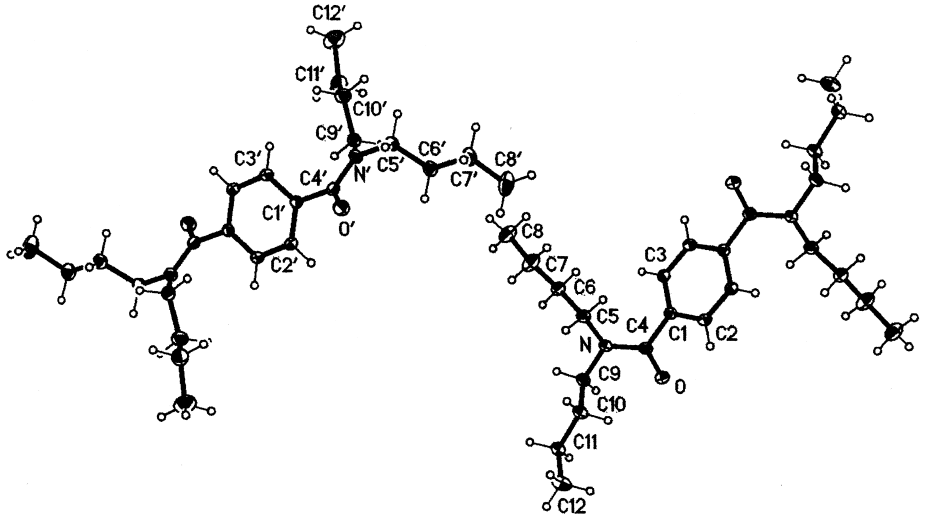


Fig. 4. The two independent molecules of compound **4** in the crystal. Ellipsoids represent 50% probability levels. Only the asymmetric unit is numbered.

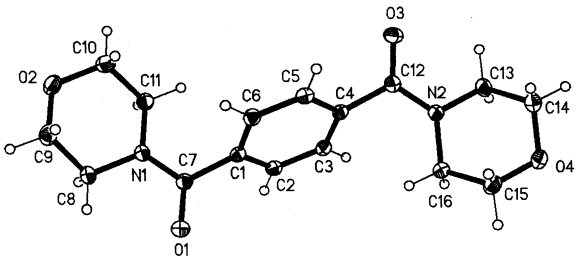


Fig. 7. The molecule of compound **7** in the crystal. Ellipsoids represent 50% probability levels.

probably the cause of the poor solubility of the compounds in both polar and non-polar solvents.

Crystal structure determinations for compounds **1–7** (the molecules are shown in Figs. 1–7) reveal some notable similarities. Except for **7**, all the compounds crystallize with imposed inversion symmetry, either in space group P$\bar{1}$ or P2$_1$/c; compound **4** has two independent molecules in the asymmetric unit, both with inversion symmetry. Heterocyclic rings occur in conformations characteristic for the given ring type. The piperidine (in **6**) and morpholine (in **7**) rings adopt the usual chair conformation. The pyrrolidine group in **5** is partially twisted. Selected bond lengths and angles are given in Table 2. Hydrogen bonds are listed in Table 3.

The crystal packing of compounds **1** and **2** (Figs. 8, 9) is very similar. Both crystallize in the triclinic space group P$\bar{1}$. The molecules are aligned with their side chains parallel. There is only one independent classical hydrogen bond of the type N-H…O=C, but each molecule is involved in four such interactions with its translationally related neighbours, leading to two antiparallel chains of hydrogen bonds linking infinite chains of molecules parallel to the hydrogen bond direction (the *y* axis in **1**, the *x* axis in **2**; note the similarity in axis lengths). The alkyl chains of compound **2** are

Table 1. Crystallographic data and refinement details for compounds **1–7**.

Compound	**1**	**2**	**3**	**4**	**5**	**6**	**7**
Formula	$C_{16}H_{24}N_2O_2$	$C_{20}H_{32}N_2O_2$	$C_{16}H_{24}N_2O_2$	$C_{24}H_{40}N_2O_2$	$C_{16}H_{20}N_2O_2$	$C_{18}H_{24}N_2O_2$	$C_{16}H_{20}N_2O_2$
M_r	276.37	332.48	276.37	388.58	272.34	300.39	304.34
Habit	colourless tablet	colourless prism	colourless prism	colourless tablet	colourless prism	colourless lath	colourless tablet
Crystal size (mm)	0.36 × 0.15 × 0.05	0.44 × 0.10 × 0.07	0.4 × 0.24 × 0.20	0.34 × 0.24 × 0.11	0.38 × 0.17 × 0.15	0.45 × 0.20 × 0.13	0.4 × 0.35 × 0.2
Crystal system	triclinic	triclinic	monoclinic	triclinic	monoclinic	monoclinic	monoclinic
Space group	*P*(−1)	*P*(−1)	*P*2$_1$/*c*	*P*(−1)	*P*2$_1$/*c*	*P*2$_1$/*c*	*P*2$_1$/*c*
Cell constants:							
a (Å)	5.0624(10)	5.0698(15)	9.6892(16)	9.5630(8)	6.2736(6)	6.1815(6)	11.5375(8)
b (Å)	5.0820(10)	8.783(3)	8.8639(16)	10.6071(11)	9.7839(11)	12.5343(11)	9.5651(6)
c (Å)	14.764(3)	10.874(3)	9.9880(18)	11.9490(11)	11.5952(12)	10.0024(8)	13.9554(11)
α (°)	99.148(6)	97.103(7)	90	101.332(4)	90	90	90
β (°)	97.948(4)	102.399(6)	112.965(6)	90.263(4)	101.805(3)	97.780(4)	111.289(4)
γ (°)	92.636(4)	97.985(6)	90	100.755(4)	90	90	90
V (Å^3)	370.50(13)	462.4(2)	789.8(2)	1166.48(19)	696.66(13)	767.86(12)	1434.99(18)
Z	1	1	2	2	2	2	4
D_x (Mg m^{-3})	1.239	1.194	1.162	1.106	1.298	1.299	1.409
μ (mm^{-1})	0.08	0.08	0.08	0.07	0.09	0.09	0.10
F(000)	150	182	300	428	292	324	648
T (°C)	−140	−140	−140	−140	−140	−140	−140
$2\theta_{max}$	60	52.7	60	60	60	60	60
No. of reflections:							
measured	3749	4081	5502	13593	7863	8712	16274
independent	2078	1864	2282	6720	2034	2233	4204
R_{int}	0.120	0.156	0.035	0.023	0.027	0.031	0.032
Parameters	96	114	98	257	100	100	199
Restraints	0	0	0	0	5	0	0
$wR(F^2$, all refl.)	0.143	0.192	0.128	0.119	0.079	0.111	0.107
$R(F, >4\sigma(F))$	0.049	0.068	0.046	0.041	0.032	0.039	0.038
S	1.03	0.95	1.05	1.06	1.05	1.05	1.07
Max. $\Delta\varrho$ (e Å^{-3})	0.41	0.42	0.35	0.38	0.72	0.39	0.32

Table 2. Selected bond distances [Å] and angles [°] for compounds **1–7**.

	1	**2**	**3**	**4**	**5**	**6**	**7**
N–C(4)	1.3388(16)	1.331(3)	1.3426(15)	1.3571(12)	1.3391(11)	1.3496(11)	
N–C(5)	1.4578(16)	1.467(3)	1.4829(15)	1.4689(12)	1.4746(10)	1.4693(10)	
N–C(8)					1.4746(11)		
N–C(9)						1.4678(10)	
O-C(4)	1.2442(13)	1.246(3)	1.2371(13)	1.2350(11)	1.2399(10)	1.2356(10)	
C(1)–C(4)	1.4964(17)	1.493(3)	1.5000(16)	1.5036(12)		1.5069(11)	
C(1)–C(7)							1.5081(13)
C(2)–C(4)					1.5043(11)		
C(7)–O(1)							1.2297(12)
C(7)–N(1)							1.3517(13)
C(12)–O(3)							1.2326(12)
C(12)–N(2)							1.3524(12)
C(4)–N–C(5)	121.25(10)	121.2(2)	124.90(10)				
C(1)–C(2)–C(4)					117.37(7		
C(2)–C(1)–C(4)	118.60(10)	118.23(19)	117.61(10)	118.39(8)		121.75(7)	
C(3)–C(1)–C(4)	122.35(11)	123.0(2)	123.47(10)	122.16(8)		118.54(7)	
C(3)–C(2)–C(4)					122.81(7)		
O–C(4)–N	121.75(12)	121.7(2)	123.19(11)	121.85(8)	121.60(8	123.21(8)	
O–C(4)–C(1)	120.66(11)	120.8(2)	119.75(10)	119.01(8)		118.60(7)	
O–C(4)–C(2)					119.06(7)		
N–C(4)–C(1)	117.59(10)	117.44(19)	117.03(10)	119.11(8)		118.18(7)	
N–C(4)–C(2)					119.34(7)		
N–C(5)–C(6)			110.41(10)	114.16(8)	103.85(7)	109.98(7)	
N–C(5)–C(7)			106.48(10)				
N–C(5)–C(8)			110.21(10				
C(4)–N–C(9)				117.21(8)		126.09(7)	
C(4)–N–C(5)				124.71(8	119.57(7)	120.53(7)	
C(4)–N–C(8)					127.61(7)		
O(1)–C(7)–N(1)							122.89(9)
O(1)–C(7)–C(1)							119.46(9)
N(1)–C(7)–C(1)							117.62(8)

appreciably interdigitated, whereas in compound **1** only the terminal regions of the butyl chains are adjacent to each other.

The packing of compound **3** also involves just one independent classical hydrogen bond N-H…O=C, which is however accompanied by a very short 'weak' [8] hydrogen bond C3-H3…O (normalised [9] H…O only 2.19 Å) to the same oxygen acceptor. Again, each molecule is involved in four such bifurcated systems, but acts as a donor to two molecules and as an acceptor to two others. The hydrogen bonds assemble lines of molecules with overall direction parallel to the *y* axis to form layers parallel to the *yz* plane (Fig. 10). Note that the molecules of each line are appreciably angled to the *xy* plane, alternate lines in the opposite sense.

In the remaining structures, there are no classical hydrogen bond donors and thus only C-H…O interactions, generally from the aromatic hydrogens, are observed. Many of these are appreciably longer than their counterpart in compound **3**, but with maximum normalised H…O distances of under 2.6 Å may still be regarded as hydrogen bonds [8]. Each independent molecule of compound **4** forms a hydrogen bond C3-H3…O to the other molecule (Fig. 11); all hydrogen bonds are approximately parallel to the *z* axis, and the molecules form layers parallel to the *xz* plane.

In compound **5**, molecules related via the 2_1 screw axis in $P2_1/c$ are linked by a short interaction C3-H3…O (2.19 Å) to form layers parallel to the *yz* plane. A much longer interaction (H8A…O 2.56 Å) from a pyrrolidyl hydrogen atom to the same oxygen atom may play a supporting role, but is not included in Fig. 12.

Compound **6** exhibits just one hydrogen bond, namely C3-H3…O, which connects molecules via the glide and screw operators to form highly corrugated layers parallel to the *yz* plane (Fig. 13).

Compound **7** is the only one of the series that does not crystallize with imposed symmetry, and it

Table 3. Hydrogen bonds [Å and °] for compounds **1**–**7** [a].

	D–H···A	d(D–H)	d(H...A)	d(D...A)	<(DHA)
1[b]	N–H(0)...O#1	1.01	1.97	2.9632(14)	169
2[c]	N–H(0)...O#1	1.01	1.97	2.970(3)	169
3[d]	N–H(0)...O#1	1.01	2.04	3.0063(14)	160
	C(3)–H(3)...O#1	1.08	2.19	3.1053(15)	141
4[e]	C(3′)–H(3′)...O#1	1.08	2.43	3.2371(12)	131
	C(3)–H(3)...O′#2	1.08	2.47	3.2443(12)	128
5[f]	C(3)–H(3)...O#1	1.08	2.19	3.2644(11)	174
	C(8)–H(8A)...O#1	1.08	2.56	3.3922(12)	134
6[g]	C(3)–H(3)...O#1	1.08	2.49	3.3712(11)	138
7[h]	C(3)–H(3)...O(1)#1	1.08	2.46	3.3509(12)	139
	C(2)–H(2)...O(2)#2	1.08	2.49	3.4893(13)	153
	C(16)–H(16B)...O(2)#3	1.08	2.51	3.5259(13)	156
	C(6)–H(6)...O(3)#4	1.08	2.41	3.3365(12)	143
	C(13)–H(13A)...O(3)#5	1.08	2.53	3.4308(13)	141
	C(5)–H(5)...O(4)#6	1.08	2.48	3.3840(13)	141
	C(11)–H(11B)...O(4)#7	1.08	2.36	3.2386(13)	137

[a] N–H bond lengths normalised to 1.01 and C-H to 1.08 Å [9];
[b] symmetry transformation used to generate equivalent atoms: #1 x, y–1, z;
[c] symmetry transformation used to generate equivalent atoms: #1 x+1, y, z;
[d] symmetry transformation used to generate equivalent atoms: #1 x,–y+1/2,z–1/2;
[e] symmetry transformations used to generate equivalent atoms: #1 x,y+1,z–1; #2 x,y–1,z;
[f] symmetry transformation used to generate equivalent atoms: #1 –x+1,y–1/2,–z+1/2;
[g] symmetry transformation used to generate equivalent atoms: #1 x,–y+1/2,z+1/2;
[h] symmetry transformations used to generate equivalent atoms: #1 –x,–y+1,–z+1; #2 –x,y–1/2, –z+1/2; #3 x,–y+3/2, z+1/2; #4 –x+1,–y+1,–z+1; #5 –x+1,–y,–z+1; #6 -x+1,y+1/2,–z+3/2; #7 x,–y+1/2,z–1/2.

also bears more potential hydrogen bond acceptors in the morpholino oxygen atoms. Seven C-H...O hydrogen bonds are observed and the overall structure is a complex three-dimensional network. However, two regions can be easily interpreted (Figs. 14a, 14b); there are layers of molecules at $z \approx 0$ connected by three hydrogen bonds involving the carbonyl oxygen atoms as acceptors (numbers 1, 4 and 5 in the Table), and also layers at $x \approx 1/4$ involving hydrogen bonds to the morpholino oxygen atoms (numbers 3 and 7). The other two hydrogen bonds link these layers.

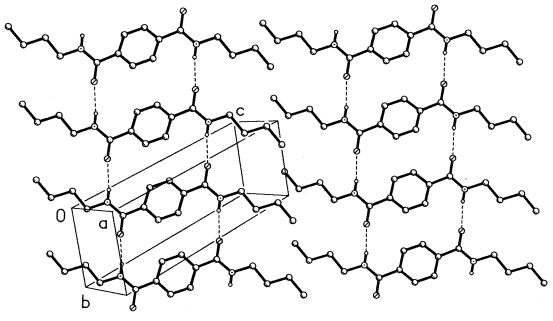


Fig. 8. Packing diagram for **1**. Hydrogen atoms not involved in hydrogen bonding are omitted. Hydrogen bonds are indicated by dashed lines. Radii are arbitrary.

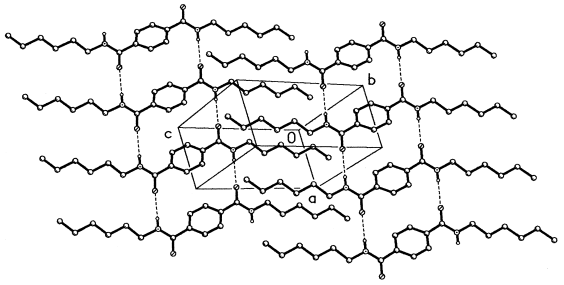


Fig. 9. Packing diagram for **2**. Hydrogen atoms not involved in hydrogen bonding are omitted. Hydrogen bonds are indicated by dashed lines. Radii are arbitrary.

Experimental Section

The proton NMR spectra were recorded with a Varian Unity INOVA-300 spectrometer, chemical shifts are in ppm ($CDCl_3$/TMS). The IR spectra were recorded with the Perkin Elmer Spectrum One FT-IR spectrometer in KBr pellets. Melting points are uncorrected.

X-ray structure determinations: Colorless single crystals were obtained by slow evaporation from: **1,**

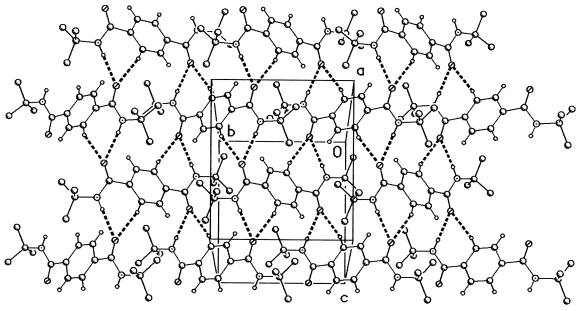


Fig. 10. Packing diagram for **3**. Hydrogen atoms not involved in hydrogen bonding are omitted. Hydrogen bonds are indicated by dashed lines. Radii are arbitrary.

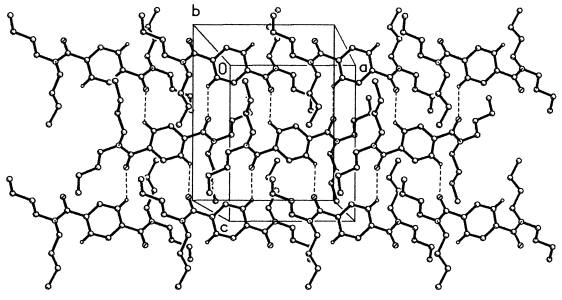


Fig. 11. Packing diagram for **4**. Hydrogen atoms not involved in hydrogen bonding are omitted. Hydrogen bonds are indicated by dashed lines. Radii are arbitrary.

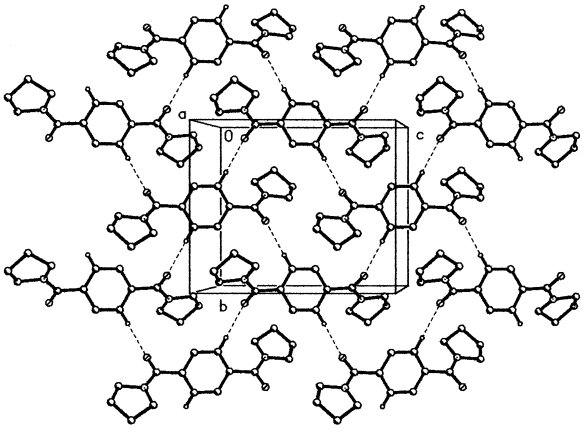


Fig. 12. Packing diagram for **5**. Hydrogen atoms not involved in hydrogen bonding are omitted. Hydrogen bonds are indicated by dashed lines. Radii are arbitrary.

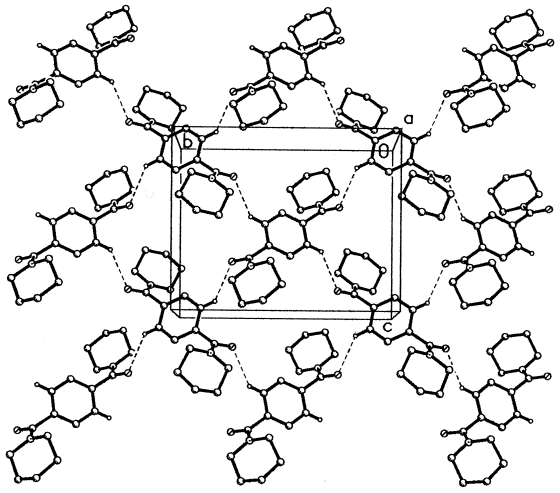


Fig. 13. Packing diagram for **6**. Hydrogen atoms not involved in hydrogen bonding are omitted. Hydrogen bonds are indicated by dashed lines. Radii are arbitrary.

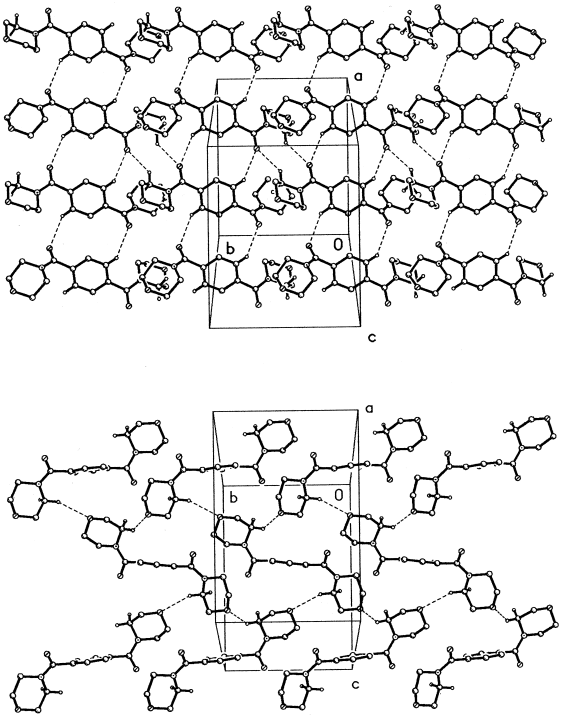


Fig. 14a,b. Packing diagrams for **7**. Hydrogen atoms not involved in hydrogen bonding are omitted. Hydrogen bonds are indicated by dashed lines. Radii are arbitrary.

4 and **7** – methanol, **2** and **3** – DMF, **5** and **6** – ethyl acetate. Crystallographic, experimental and computational details are given in Table 1. Scattering factors are from *International Tables for Crystallography (Vol.C)*. Data collection, cell refinement and data reduction: Bruker SMART 1000 CCD system [10]. Program used to solve structures: SHELXS86 [11]. Program used to refine structures: SHELXL97 [12]. Molecular graphics: Siemens XP [13]. Refinement details : H atoms bonded to nitrogen were refined freely, methyl H using rigid groups, other H using a riding model. The pyrrolidine ring of compound **5** is disordered over two positions; the minor sites for C6 and C7 are 10% occupied. Complete crystallographic data have been deposited at the Cambridge Crystallographic Data Centre under the numbers CCDC-181608 for **1**, CCDC-181609 for **2** CCDC-181610 for **3**, CCDC-181611 for **4**, CCDC-181612 for **5**, CCDC-181613 for **6** and CCDC-181614 for **7**. Copies can be obtained free of charge from CCDC, 12 Union Road, Cambridge CB2 1EZ, U. K. (Fax: Int.+1223-336-033; E-mail: deposit@ccdc.cam.ac.uk).

General procedure for synthesis of terephthaldiamides from dimethyl terephthalate.

A mixture of 50 mmol of dimethyl terephthalate and 0.5 mol of amine was refluxed for 2 h and left overnight. The precipitated products were filtered, washed with water and with cold methanol. Yield: 80–90%.

1: M.p. 229–232 °C (lit[4]. 228–229 °C) – IR (KBr): ν = 3316 (N–H), 3064, 3030, 2957, 2940, 2910, 2871, 1626 (C=O), 1543, 1499, 1472, 1463, 1353, 1314, 1293, 1258, 1159, 1135, 1017, 970, 911, 861, 847, 805, 734, 652, 538, 492 cm^{-1}. – ^{1}H NMR (300.05 MHz, $CDCl_3$): δ = 7.79 (s, 4H, Ar*H*), 6.18 (t, 2H, N*H*), 3.46 (q, 4H, NH–*CH*$_2$), 1.61 (qu, 4H, *CH*$_2$), 1.44 (sex, 4H, *CH*$_2$–CH$_3$), 0.96 (t, 6H, *CH*$_3$). – $C_{16}H_{24}N_2O_2$: calcd. C 69.52, H 8.76, N 10.14; found C 69.27, H 8.84, N 10.21.

2: M.p. 220–221 °C–IR (KBr): ν = 3322 (N-H), 3062, 3025, 2956, 2939, 2924, 2869, 2855, 1627 (C=O), 1539, 1496, 1472, 1464, 1374, 1348, 1317, 1299, 1275, 1231, 1158, 1133, 1115, 1069, 1053, 1038, 1017, 971, 892, 862, 851, 800, 734, 646, 541, 505 cm^{-1}. – ^{1}H NMR (300.05 MHz, pyridine-d_5): δ = 8.71 (s, 4H, Ar*H*), 8.71 (t, 2H, N*H*), 3.61 (q, 4H, NH-*CH*$_2$), 1.65 (qu, 4H, *CH*$_2$), 1.28 (qu, 4H, *CH*$_2$), 1.1–1.2 (m, 8H, *CH*$_2$), 0.75 (t, 6H, *CH*$_3$). – $C_{20}H_{32}N_2O_2$: calcd. C 72.24, H 9.71, N 8.43; found C 72.15, H 9.75, N 8.11.

3: M.p. 246–249 °C–IR (KBr): ν = 3302 (N-H), 3042, 2972, 2930, 2866, 1634 (C=O), 1543, 1502, 1475, 1453, 1392, 1365, 1324, 1295, 1256, 1231, 1220, 1120, 1033, 1020, 980, 930, 864, 762, 737, 656, 544 cm^{-1}. – ^{1}H NMR (300.05 MHz, $CDCl_3$): δ = 7.75 (s, 4H, Ar*H*), 5.95 (s, 2H, N*H*), 1.48 (s, 18H, *CH*$_3$). – $C_{16}H_{24}N_2O_2$: calcd. C 69.52, H 8.76, N 10.14; found C 69.34, H 8.73, N 10.07.

4: M.p. 79–80 °C (lit[5]. 79–80 °C) – IR (KBr): ν = 2971, 2952, 2932, 2870, 1621 (C=O), 1503, 1469, 1453, 1426, 1394, 1378, 1316, 1291, 1254, 1235, 1197, 1118, 1104, 1054, 1008, 984, 953, 899, 862, 796, 736, 596, 530, 464 cm^{-1}. – ^{1}H NMR (300.05 MHz, $CDCl_3$): δ = 7.38 (s, 4H, Ar*H*), 3.49 (t, 4H), 3.17 (t, 4H, N–*CH*$_2$), 1.65 (m, 4H, *CH*$_2$), 1.41–1.46 (m, 8H, *CH*$_2$), 1.7 (m, 4H, *CH*$_2$–CH$_3$), 0.95 (t, 6H, *CH*$_3$), 0.82 (t, 6H, *CH*$_3$). – $C_{24}H_{40}N_2O_2$: calcd. C 74.18, H 10.38, N 7.21; found C 74.46, H 10.49, N 7.21.

5: M.p. 218–219 °C (lit[5] 213 °C) – IR (KBr): ν = 3026, 2973, 2952, 2866, 1607 (C=O), 1517, 1429, 1339, 1281, 1250, 1226, 1207, 1180, 1156, 1113, 1032, 1022, 994, 932, 902, 864, 841, 729, 618, 576, 488 cm^{-1}. – ^{1}H NMR (300.05 MHz, $CDCl_3$): δ = 7.54 (s, 4H, Ar*H*), 3.65 (t, 4H, N–*CH*$_2$), 3.39 (t, 4H, N–*CH*$_2$), 1.97 (q, 4H, *CH*$_2$), 1.89 (q, 4H, *CH*$_2$). – $C_{16}H_{20}N_2O_2$: calcd. C 70.56, H 7.40, N 10.29; found C 70.21, H 7.39, N 10.26.

6: M.p. 211–213 °C (lit[5] 211–213 °C) – IR (KBr): ν = 3029, 2992, 2955, 2935, 2920, 2868, 2856, 1614 (C=O), 1507, 1486, 1465, 1440, 1392, 1366, 1349, 1286, 1272, 1233, 1173, 1123, 1114, 1105, 1064, 1023, 1001, 979, 953, 886, 853, 843, 821, 730, 676, 606, 537, 470 cm^{-1}. – ^{1}H NMR (300.05 MHz, $CDCl_3$): δ = 7.42 (s, 4H, Ar*H*), 3.71 (bs, 4H, N-*CH*$_2$), 3.32 (bs, 4H, N–*CH*$_2$), 1.68 (bs, 8H, *CH*$_2$), 1.50 (bs, 4H, *CH*$_2$). – $C_{18}H_{24}N_2O_2$: calcd. C 71.96, H 8.06, N 9.33; found C 71.60, H 8.03, N 9.25.

7: M.p. 239–242 °C (lit[5] 218–219 °C) – IR (KBr): ν = 2963, 2922, 2902, 2860, 1626 (C=O), 1509, 1433, 1396, 1365, 1332, 1302, 1271, 1164, 1115, 1067, 1026, 1009, 936, 894, 860, 838, 734, 615, 568 cm^{-1}. – ^{1}H NMR (300.05 MHz, $CDCl_3$): δ = 7.39 (s, 4H, Ar*H*), 3.71 (bs, 8H, O–*CH*$_2$), 3.56 (bs, 4H, N–*CH*$_2$), 3.39 (bs, 4H, N–*CH*$_2$). – $C_{16}H_{20}N_2O_4$: calcd. C 63.14, H 6.62, N 9.20; found C 63.07, H 6.60, N 9.18.

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