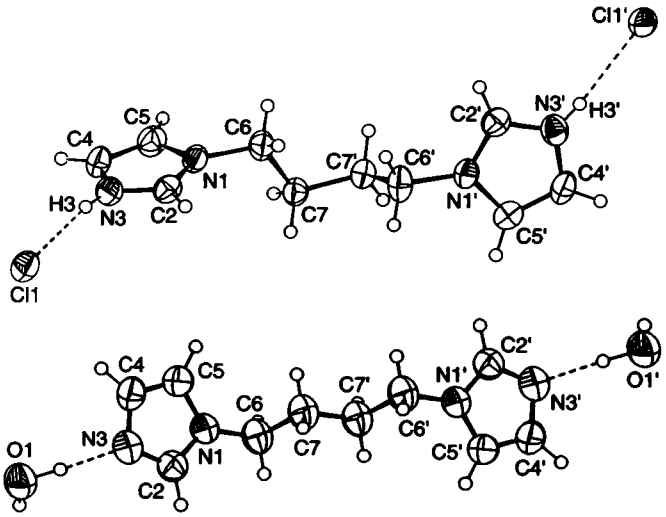
Crystal structures of 1,4-di(1-imidazolyl)butane dihydrochloride, $(C_{10}H_{16}N_4)Cl_2$, and 1,4-di(1-imidazolyl)butane dihydrate, $C_{10}H_{14}N_4 \cdot 2H_2O$

M. Królikowska and J. Garbarczyk*

Poznań University of Technology, Institute of Chemical Technology and Engineering, Pl. Skłodowskiej-Curie 2, 60-695 Poznań, Poland

Received March 14, 2004, accepted and available on-line January 13, 2005; CCDC nos. 1267/1308, 1267/1312



Abstract

$C_{10}H_{16}Cl_2N_4$, monoclinic, *C*12/*c*1 (no. 15), $a = 15.915(3)$ Å, $b = 8.230(2)$ Å, $c = 10.189(2)$ Å, $\beta = 103.98(3)°$, $V = 1295.0$ Å^3, $Z = 4$, $R_{gt}(F) = 0.038$, $wR_{ref}(F^2) = 0.113$, $T = 293$ K.

$C_{10}H_{18}N_4O_2$, orthorhombic, *Pbca* (no. 61), $a = 14.775(3)$ Å, $b = 5.222(1)$ Å, $c = 15.604(3)$ Å, $V = 1203.9$ Å^3, $Z = 4$, $R_{gt}(F) = 0.077$, $wR_{ref}(F^2) = 0.311$, $T = 293$ K.

Source of materials

The 1,4-di(1-imidazolyl)butane dihydrochloride (DIBHCl) was prepared by passing the HCl gas through a water solution of 1,4-di(1-imidazolyl)butane (DIB) [1], which was obtained according to the procedure described in [2]. The crystals of DIBHCl in the form of colorless needles were obtained by slow evaporation from methanol solution at room temperature.

Experimental details

The hydrogen atoms of C2, N3 or O1 atoms were refined freely, because of their participation on intermolecular hydrogen interactions. The remaining H atoms bonded to carbon atoms and less important in explanation of interactions in question were refined under restraints. Large *R* values and a small $N(hkl)_{gt}/N(param)$ ratio for the dihydrate are connected with the bad quality of crystals of this compound.

Discussion

The aim of the present analysis of 1,4-di(1-imidazolyl)butane dihydrochloride was defining the influence of protonation of imidazole ring on conformation of the molecule. For this purpose crystallographic data obtained for the salt (DIBHCl) and the neutral molecule 1,4-di(1-imidazolyl)butane have been compared. It was found that protonation of the nitrogen atoms of imidazole ring causes very significant changes in conformation of DIBHCl molecule (figure, top) in comparison to DIB one (figure, bottom). In the neutral DIB molecule the imidazole rings have *trans*-(*ap*)-position in respect to C7—C7′ bond (∠C6–C7–C7′–C6′ = 180.0°). However, after protonation the rings in question have *gauche*-(*sc*)-conformation (∠C6–C7–C7′–C6′ = 79.9(3)°) due to strong interactions between N–H$^+$ of the imidazolium ions and chloride anions, d(N3···Cl) = 3.035(3) Å and ∠N3–H3···Cl = 169.21(3)° (figure, top). The molecules of DIBHCl as well as DIB lie in special positions, but possess in the center of the molecule a two-fold rotation axis and a centre of symmetry, respectively. Contrary to DIBHCl, the DIB crystallizes with two molecules of water, which in the crystal lattice create chains joined by hydrogen bonds, parallel to the *b*-axis (d(O···O) = 2.892(3) Å and ∠O–H1W···O($-x+\frac{3}{2}, y+\frac{1}{2}, z$) = 165.91(3)°). There are also intermolecular hydrogen bonds between water and nitrogens from imidazole ring with d(O···N3) = 2.828(3) Å and ∠O–H1W···N3 = 173.98(2)°.

1. 1,4-Di(1-imidazolyl)butane dihydrochloride, $(C_{10}H_{16}N_4)Cl_2$

Table 1. Data collection and handling.

Crystal:	colorless needle, size 0.04 × 0.10 × 0.45 mm
Wavelength:	Cu K_α radiation (1.54178 Å)
μ:	43.49 cm^{-1}
Diffractometer, scan mode:	Kuma KM-4, $\theta/2\theta$
$2\theta_{max}$:	159.52°
$N(hkl)_{measured}$, $N(hkl)_{unique}$:	1239, 1177
Criterion for I_{obs}, $N(hkl)_{gt}$:	$I_{obs} > 2\,\sigma(I_{obs})$, 1004
$N(param)_{refined}$:	82
Programs:	SHELXS-97 [3], SHELXL-97 [4]

Table 2. Atomic coordinates and displacement parameters (in Å^2).

Atom	Site	x	y	z	U_{iso}
H(2)	8*f*	0.055(2)	0.159(3)	−0.396(3)	0.066(7)
H(3)	8*f*	0.152(2)	0.222(3)	−0.201(3)	0.070(8)
H(4)	8*f*	0.2577	0.4069	−0.2261	0.070
H(5)	8*f*	0.2248	0.4609	−0.4813	0.087
H(6A)	8*f*	0.0528	0.1931	−0.6473	0.080
H(6B)	8*f*	0.1254	0.2999	−0.6809	0.080
H(7A)	8*f*	0.0506	0.5360	−0.6432	0.062
H(7B)	8*f*	−0.0269	0.4197	−0.6226	0.058

* Correspondence author (e-mail: jozef.garbarczyk@put.poznan.pl)

Table 3. Atomic coordinates and displacement parameters (in $Å^2$).

Atom	Site	x	y	z	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
Cl(1)	8*f*	0.12018(3)	0.14025(6)	–0.00815(5)	0.0466(3)	0.0622(4)	0.0571(4)	0.0017(2)	0.0067(2)	0.0124(2)
N(1)	8*f*	0.12535(9)	0.3062(2)	–0.4844(2)	0.0410(8)	0.0490(8)	0.0443(8)	0.0055(6)	0.0017(6)	0.0010(6)
C(2)	8*f*	0.1011(1)	0.2265(2)	–0.3872(2)	0.0445(9)	0.0447(8)	0.056(1)	0.0028(7)	0.0085(8)	0.0007(8)
N(3)	8*f*	0.1551(1)	0.2622(2)	–0.2710(2)	0.0541(9)	0.0542(9)	0.047(1)	0.0108(7)	0.0086(7)	0.0044(7)
C(4)	8*f*	0.2156(1)	0.3683(2)	–0.2935(2)	0.0423(9)	0.072(1)	0.053(1)	0.0016(8)	–0.0036(8)	–0.0080(9)
C(5)	8*f*	0.1974(1)	0.3969(3)	–0.4272(2)	0.046(1)	0.065(1)	0.059(1)	–0.0074(8)	0.0094(8)	0.0001(9)
C(6)	8*f*	0.0824(1)	0.2953(3)	–0.6292(2)	0.058(1)	0.061(1)	0.044(1)	0.0167(9)	–0.0030(8)	–0.0066(8)
C(7)	8*f*	0.0183(1)	0.4316(2)	–0.6733(2)	0.0442(9)	0.0451(9)	0.043(1)	0.0007(7)	0.0033(7)	–0.0030(7)

2. 1,4-Di(1-imidazolyl)butane dihydrate, $C_{10}H_{14}N_4 \cdot 2H_2O$

Table 4. Data collection and handling.

Crystal:	colorless plate, size 0.04 × 0.25 × 0.35 mm
Wavelength:	Cu K_α radiation (1.54178 Å)
μ:	7.35 cm^{-1}
Diffractometer, scan mode:	Kuma KM-4, $\theta/2\theta$
$2\theta_{max}$:	161.36°
$N(hkl)_{measured}$, $N(hkl)_{unique}$:	1254, 1253
Criterion for I_{obs}, $N(hkl)_{gt}$:	$I_{obs} > 2\,\sigma(I_{obs})$, 594
$N(param)_{refined}$:	86
Programs:	SHELXS-97 [3], SHELXL-97 [4]

Table 5. Atomic coordinates and displacement parameters (in $Å^2$).

Atom	Site	x	y	z	U_{iso}
H(2)	8*c*	0.946(3)	–0.186(9)	0.768(3)	0.07(1)
H(4)	8*c*	0.8051	0.4632	0.7940	0.085
H(5)	8*c*	0.8276	0.3644	0.6263	0.068
H(6A)	8*c*	0.8696	–0.1163	0.5595	0.080
H(6B)	8*c*	0.9488	–0.2442	0.6092	0.080
H(7A)	8*c*	0.9615	0.2278	0.5265	0.080
H(7B)	8*c*	1.0414	0.0989	0.5766	0.080
H(1W)	8*c*	0.821(5)	0.43(1)	0.410(5)	0.15(3)
H(2W)	8*c*	0.757(7)	0.65(2)	0.459(5)	0.16(3)

Table 6. Atomic coordinates and displacement parameters (in $Å^2$).

Atom	Site	x	y	z	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
N(1)	8*c*	0.8949(2)	0.0464(6)	0.6729(2)	0.063(2)	0.062(2)	0.049(2)	0.002(2)	0.003(2)	0.000(2)
C(2)	8*c*	0.9127(3)	–0.028(1)	0.7533(3)	0.062(3)	0.069(3)	0.054(2)	0.006(2)	–0.000(2)	0.002(2)
N(3)	8*c*	0.8795(3)	0.1287(8)	0.8112(2)	0.069(2)	0.074(2)	0.058(2)	–0.003(2)	0.005(2)	–0.001(2)
C(4)	8*c*	0.8370(3)	0.3143(9)	0.7643(3)	0.067(3)	0.072(3)	0.070(3)	0.006(2)	0.016(2)	–0.001(2)
C(5)	8*c*	0.8458(3)	0.2686(8)	0.6791(3)	0.063(3)	0.067(3)	0.068(3)	0.007(2)	0.009(2)	0.009(2)
C(6)	8*c*	0.9217(3)	–0.0830(9)	0.5944(3)	0.076(3)	0.077(3)	0.052(2)	–0.006(2)	0.003(2)	–0.007(2)
C(7)	8*c*	0.9890(3)	0.0676(9)	0.5417(3)	0.075(3)	0.075(3)	0.054(3)	–0.004(2)	0.006(2)	–0.005(2)
O(1)	8*c*	0.7921(2)	0.4455(7)	0.4705(2)	0.096(3)	0.083(2)	0.061(2)	–0.003(2)	0.005(2)	0.003(2)

Acknowledgments. This work was partly supported by the Poznań University of Technology (grant no. DS/32/114/2004). The authors thank Dr. A. Skrzypczak for providing compounds.

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