Zhi-Yun Dong*, Fu-Gui Xi and Xing-Yu Xu

The crystal structure of 3,3'-((carbonylbis(azanediy))bis(ethane-2,1-diyl))bis(1-methyl-1*H*-benzo[*d*]imidazol-3-ium) tetrafluoroborate monohydrate, C₂₁H₂₈N₆O₃B₂F₈

Table 1: Data collection and handling.

Crystal:	Colorless block
Size:	0.25 × 0.15 × 0.07 mm
Wavelength:	Mo K α radiation (0.71073 Å)
μ :	0.14 mm ⁻¹
Diffractometer, scan mode:	φ and ω
$\theta_{\max}$, completeness:	28.1°, >99%
$N(hkl)_{\text{measured}}$, $N(hkl)_{\text{unique}}$, R_{int} :	32,623, 3127, 0.053
Criterion for I_{obs} , $N(hkl)_{\text{gt}}$:	$I_{\text{obs}} > 2 \sigma(I_{\text{obs}})$, 1897
$N(\text{param})_{\text{refined}}$:	183
Programs:	Bruker [1], SHELX [2–4], refinement [5], Olex2 [6]

Table 1 contains crystallographic data, and Table 2 contains the list of the atoms including atomic coordinates and displacement parameters.

Source of material

A solution of *N*-methylbenzimidazole 1.32 g (10 mmol) and 1,3-bis(2-chloroethyl)urea 0.74 g (4 mmol) in 10 mL ethanol was heated at 353 K for 38 h under N₂. After that the reaction mixture was cooled to room temperature, the white precipitate was collected and dissolved in 50 mL distilled water, and silver tetrafluoroborate 0.78 g (4 mmol) was added. In order to prevent the silver salt decomposing when exposed to light, the whole system was put in a dark place and stored at room temperature 5 h, removed the white AgCl precipitate, and finally evaporated the filtrate in vacuum. Colourless block crystals were grown from methanol by slow evaporation at ambient room temperature over a period of 2 weeks. Yield: 91%. ¹H NMR (500 MHz, DMSO-*d*₆): δ (ppm) 3.41–3.44 (m, 4H), 4.11 (s, 6H), 4.49 (t, J = 6.0 Hz, 4H), 6.85 (br, 2H), 7.64–7.68 (m, 4H), 8.01–8.07 (m, 4H), 10.04 (s, 2H); ¹³C NMR (125 MHz, DMSO-*d*₆): δ (ppm) 33.18, 38.43, 47.18, 113.40, 113.44, 126.26, 126.29, 130.92, 131.79, 143.18, 157.98.

<https://doi.org/10.1515/ncrs-2020-0646>

Received December 29, 2020; accepted January 14, 2021;
published online February 1, 2021

Abstract

C₂₁H₂₈N₆O₃B₂F₈, orthorhombic, *Pccn* (no. 56), $a = 17.7949(19)$ Å, $b = 9.3979(10)$ Å, $c = 15.3455(16)$ Å, $V = 2566.3(5)$ Å³, $Z = 4$, $R_{\text{gt}}(F) = 0.0567$, $wR_{\text{ref}}(F^2) = 0.1846$, $T = 296(2)$ K.

CCDC no.: 830078

*Corresponding author: Zhi-Yun Dong, Department of Chemistry, Xinzhou Teachers University, Xinzhou, Shanxi, 034000, P. R. China, E-mail: 16dongzhiyun@163.com. <https://orcid.org/0000-0001-5219-8045>

Fu-Gui Xi and Xing-Yu Xu, Department of Chemistry, Xinzhou Teachers University, Xinzhou, Shanxi, 034000, P. R. China

Table 2: Fractional atomic coordinates and isotropic or equivalent isotropic displacement parameters (Å²).

Atom	x	y	z	<i>U</i> _{iso} [*] / <i>U</i> _{eq}
O1	0.750000	1.250000	−0.46741 (13)	0.0587 (6)
N1	0.90084 (10)	1.0697 (2)	−0.35090 (12)	0.0560 (5)
N2	0.85795 (10)	0.99322 (19)	−0.47440 (12)	0.0454 (4)
N3	0.74597 (11)	1.1287 (2)	−0.59464 (12)	0.0501 (5)
H3	0.748929	1.134014	−0.650482	0.060*
C1	0.99436 (14)	1.2472 (3)	−0.4105 (2)	0.0704 (8)
H1	1.012511	1.280701	−0.357507	0.085*
C2	1.02111 (15)	1.2976 (3)	−0.4892 (3)	0.0844 (10)
H2	1.058155	1.367445	−0.489254	0.101*
C3	0.99404 (14)	1.2466 (4)	−0.5684 (2)	0.0793 (9)
H3A	1.014001	1.282641	−0.619869	0.095*
C4	0.93897 (13)	1.1450 (3)	−0.57293 (17)	0.0604 (7)
H4	0.920963	1.111445	−0.625981	0.072*
C5	0.91162 (11)	1.0951 (2)	−0.49427 (14)	0.0448 (5)
C6	0.93889 (12)	1.1439 (3)	−0.41512 (15)	0.0509 (6)
C7	0.85328 (12)	0.9825 (3)	−0.38898 (15)	0.0523 (6)
H7	0.820795	0.921533	−0.359617	0.063*
C8	0.81110 (13)	0.9160 (3)	−0.53636 (17)	0.0562 (6)
H8A	0.800703	0.821833	−0.513463	0.067*
H8B	0.838480	0.904656	−0.590577	0.067*
C9	0.73694 (13)	0.9916 (3)	−0.55477 (16)	0.0559 (6)
H9A	0.706696	0.932146	−0.592726	0.067*
H9B	0.709798	1.003015	−0.500399	0.067*
C10	0.750000	1.250000	−0.54705 (19)	0.0469 (7)
C11	0.91201 (18)	1.0821 (4)	−0.25698 (17)	0.0894 (11)
H11A	0.949626	1.152858	−0.245382	0.134*
H11B	0.865625	1.109509	−0.229797	0.134*
H11C	0.928134	0.992164	−0.233972	0.134*
B1	0.65409 (19)	0.8933 (5)	−0.2890 (2)	0.0760 (10)
F1	0.71942 (11)	0.9327 (3)	−0.24738 (12)	0.1138 (8)
F2	0.60551 (16)	1.0003 (3)	−0.2864 (2)	0.1612 (12)
F3	0.62177 (14)	0.7791 (3)	−0.24795 (14)	0.1301 (10)
F4	0.67229 (11)	0.8532 (2)	−0.37147 (11)	0.0955 (6)
O2 ^a	0.7355 (12)	1.231 (3)	−0.2835 (3)	0.105 (6)
H2A ^a	0.702052	1.278446	−0.301414	0.158*
H2B ^a	0.753411	1.237550	−0.332969	0.158*

^aOccupancy: 0.5.

Comment

Ionic liquids are salts that can melt at temperatures below 100 °C and are typically composed of large and unsymmetrical organic cations and organic or inorganic anions [7]. ILs have been recognized as new green solvents for chemical reactions [8], extractions [9], catalysis [10], gas absorption [11], and promising candidate materials for electrolytes in electrochemical devices [12].

The asymmetric unit of the title compound contains one half of a bisbenzimidazolium diacation, one tetrafluoroborate anion and water molecules near to a two-fold axis with an occupancy of 0.5 (see the Figure), and lattice water was omitted for clarity. The bond lengths and angles are all in the expected ranges [13]. A carbon–oxygen double bonds exist in the compound, the C–O bond distance (C10–O1) is 1.222(4) Å, the angles of O1–C10–N3 and N3–C10–N3ⁱ (ⁱ = 1.5–x, 2.5–y, z) are 122.6(3)°, respectively. The counter tetrafluoroborate anions are located at the side of the benzimidazolium ring, the benzimidazolium group can make a strong interaction with tetrafluoroborate through (C–H)⁺–X[−] type ionic hydrogen bonding, where the ionic hydrogen-bonding distances are 2.496 Å [(C7–H7)⁺–F1], and 2.471 Å [(N3–H3)–F1ⁱⁱ; ⁱⁱ = 1.5–x, y, −0.5 + z]. There is also an intermolecular π–π stacking interaction (distance = 3.749 Å).

Author contributions: All the authors have accepted responsibility for the entire content of this submitted manuscript and approved submission.

Research funding: Scientific and Technological Innovation Programs of Higher Education Institutions in Shanxi (STIP).

Conflict of interest statement: The authors declare no conflicts of interest regarding this article.

Experimental details

The structures were solved by direct methods using SHELXT [3] and refined with SHELXL [4] following established refinement strategies [5] via OLEX-2 [6]. Carbon-bound H atoms were placed in calculated positions (C–H 0.95 Å) and were included in the refinement in the riding model approximation, with *U*_{iso}(H) set to 1.2 *U*_{eq}(C). All other hydrogen atoms bonded to heteroatoms were located on a difference Fourier map.

References

1. BRUKER. SAINT, APEX2 and SADABS; Bruker AXS Inc.: Madison, Wisconsin, USA, 2009.
2. Sheldrick G. M. A short history of *SHELX*. *Acta Crystallogr.* 2008, A64, 112–122.
3. Sheldrick G. M. SHELXT – integrated space-group and crystal-structure determination. *Acta Crystallogr.* 2015, A71, 3–8.
4. Sheldrick G. M. Crystal structure refinement with *SHELXL*. *Acta Crystallogr.* 2015, C71, 3–8.
5. Müller P. Practical suggestions for better crystal structures. *Crystallogr. Rev.* 2009, 15, 57–83.

6. Dolomanov O. V., Bourhis L. J., Gildea R. J., Howard J. A. K., Puschmann H. OLEX2: a complete structure solution, refinement and analysis program. *J. Appl. Crystallogr.* 2009, 42, 339–341.
7. Dong K., Liu X., Dong H., Zhang X., Zhang S. Multiscale studies on ionic liquids. *Chem. Rev.* 2017, 117, 6636–6695.
8. Clarke C. J., Tu W. C., Levers O., Brohl A., Hallett J. P. Green and sustainable solvents in chemical processes. *Chem. Rev.* 2018, 118, 747–800.
9. Toledo H., Ariel A. C., Maximo G. J., Costa M. C., Batista E. A. C., Meirelles A. J. A. Applications of ionic liquids in the food and bioproducts industries. *ACS Sustain. Chem. Eng.* 2016, 4, 5347–5369.
10. Salikolimi K., Sudhakar A. A., Ishida Y. Functional ionic liquid crystals. *Langmuir* 2020, 36, 11702–11731.
11. Yan S., Han F., Hou Q., Zhang S., Ai S. Recent advances in ionic liquid-mediated SO_2 capture. *Ind. Eng. Chem. Res.* 2019, 58, 13804–13818.
12. Zhao Y., Bostrom T. Application of ionic liquids in solar cells and batteries: a review. *Curr. Org. Chem.* 2015, 19, 556–566.
13. Kathuria L., Samuelson A. G. Chiral *N*-heterocyclic carbene-iridium complexes for asymmetric reduction of prochiral ketimines. *Polyhedron* 2021, 195, 114976.