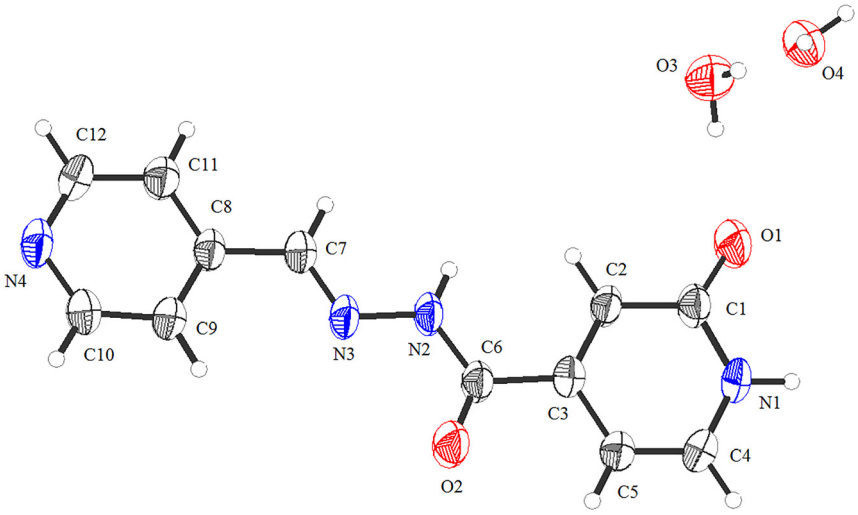
Zhao Zeng-Bing, Cheng Lan-Xing, Yang Shu-Cheng, Chai Yong-Li* and Tai Xi-Shi*

The crystal structure of (*E*)-4-(2-(pyridin-4-ylmethylene)hydrazine-1-carbonyl)pyridin-1-ium-2-olate dihydrate, $C_{12}H_{14}N_4O_4$



https://doi.org/10.1515/ncrs-2024-0276
Received July 1, 2024; accepted August 5, 2024;
published online August 14, 2024

Abstract

$C_{12}H_{14}N_4O_4$, triclinic, $P\bar{1}$ (no. 2), a = 6.79127(19) Å, b = 10.0328(3) Å, c = 10.0542(2) Å, α = 90.027(2)°, β = 97.124(2)°, γ = 101.983(3)°, V = 664.71(3) Å^3, Z = 2, $R_{gt}(F)$ = 0.0539, $wR_{ref}(F^2)$ = 0.1580, T = 292 K.

CCDC no.: 2375758

The molecular structure is shown in the figure. Table 1 contains crystallographic data and Table 2 contains the list of the atoms including atomic coordinates and displacement parameters.

***Corresponding authors: Chai Yong-Li**, Henan Academy of Sciences, Zhengzhou, 450046, P.R. China; and Henan Chemical Industry Research Institute Co., Zhengzhou, 450052, P.R. China E-mail: chaiyli@163.com, and **Tai Xi-Shi**, College of Chemistry and Chemical Engineering, Weifang University, Weifang, Shandong 261061, P. R. China, E-mail: taixs@wfu.edu.cn. https://orcid.org/0000-0002-0050-1900 (T. Xi-Shi)
Zhao Zeng-Bing, Cheng Lan-Xing and Yang Shu-Cheng, Henan Academy of Sciences, Zhengzhou, 450046, P.R. China; and Henan Chemical Industry Research Institute Co., Zhengzhou, 450052, P.R. China

Table 1: Data collection and handling.

Crystal:	Colourless block
Size:	0.14 × 0.12 × 0.10 mm
Wavelength:	Cu *K*α radiation (1.54184 Å)
μ:	0.90 mm^{-1}
Diffractometer, scan mode:	XtaLAB Synergy R, ω
θ_{max}, completeness:	73.2°, 99 %
$N(hkl)_{measured}$, $N(hkl)_{unique}$, R_{int}:	6,918, 2,553, 0.036
Criterion for I_{obs}, $N(hkl)_{gt}$:	$I_{obs} > 2\sigma(I_{obs})$, 2075
$N(param)_{refined}$:	197
Programs:	Bruker,[1] Olex2,[2] SHELX,[3] Diamond[4]

1 Source of materials

An amount of 0.107 g 4-pyridinecarboxaldehyde (1.0 mmol), 0.153 g 2-hydroxyisonicotinohydrazide (1.0 mmol), 0.040 g NaOH (1.0 mmol), and 0.1015 g magnesium chloride hexahydrate (0.50 mmol) were added together to the 18 ml ethanol-water solution (v:v = 2:1) and stirred to dissolve. Heating the above solution to 75 °C for reaction for 4 h. The colorless needle-like crystals were obtained from filtrate after 25 days.

2 Experimental details

The hydrogen atoms were positioned geometrically (C–H = 0.95 Å, N–H = 0.88–0.92 Å, O–H = 0.84–0.88 Å). Their U_{iso} values were set to 1.2U_{eq} or 1.5U_{eq} of the parent atoms.

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

Atom	*x*	*y*	*z*	U_{iso}*/U_{eq}
O1	0.3846 (2)	1.08152 (11)	0.22115 (11)	0.0593 (4)
O2	0.1810 (2)	0.47535 (11)	0.22257 (11)	0.0606 (4)
N1	0.2602 (2)	0.93718 (13)	0.04358 (11)	0.0438 (3)
H1	0.261 (3)	1.012 (3)	−0.009 (2)	0.071 (6)*
N2	0.25766 (19)	0.60161 (12)	0.41552 (11)	0.0398 (3)
H2	0.289019	0.683036	0.454680	0.048*
N3	0.24204 (19)	0.48575 (12)	0.48979 (12)	0.0406 (3)
N4	0.2440 (2)	0.16240 (13)	0.87182 (13)	0.0479 (4)
C1	0.3224 (2)	0.96118 (15)	0.17688 (14)	0.0414 (4)
C2	0.3104 (2)	0.84265 (14)	0.25766 (13)	0.0404 (4)
H2A	0.351244	0.853117	0.351580	0.048*
C3	0.2414 (2)	0.71539 (14)	0.20160 (13)	0.0368 (3)
C4	0.1930 (3)	0.81138 (17)	−0.01329 (14)	0.0478 (4)
H4	0.152380	0.801706	−0.107266	0.057*
C5	0.1828 (2)	0.69877 (15)	0.06207 (14)	0.0435 (4)
H5	0.136984	0.610345	0.021663	0.052*
C6	0.2234 (2)	0.58664 (14)	0.28049 (14)	0.0390 (3)
C7	0.2815 (2)	0.50292 (15)	0.61604 (14)	0.0421 (4)
H7	0.319939	0.592497	0.654484	0.050*
C8	0.2678 (2)	0.38445 (15)	0.70292 (14)	0.0385 (3)
C9	0.2075 (2)	0.25197 (16)	0.65100 (14)	0.0431 (4)
H9	0.173563	0.234795	0.557155	0.052*
C10	0.1979 (3)	0.14599 (16)	0.73859 (15)	0.0478 (4)
H10	0.155995	0.055660	0.702265	0.057*
C11	0.3145 (3)	0.40263 (16)	0.84091 (15)	0.0455 (4)
H11	0.355558	0.491749	0.880265	0.055*
C12	0.3005 (3)	0.28940 (18)	0.92052 (15)	0.0484 (4)
H12	0.333080	0.303309	1.014830	0.058*
O3	0.7087 (3)	1.16425 (14)	0.41834 (12)	0.0663 (4)
H3A	0.610 (3)	1.137 (3)	0.359 (2)	0.099*
H3B	0.818 (3)	1.173 (3)	0.382 (3)	0.099*
O4	1.0819 (2)	1.20317 (12)	0.31213 (12)	0.0594 (4)
H4A	1.182 (3)	1.168 (3)	0.292 (3)	0.089*
H4B	1.127 (4)	1.2888 (18)	0.309 (3)	0.089*

3 Comment

The study of the structure and properties of acylhydrazone compounds and their complexes has always been one of the priorities of coordination chemistry.[5,6] Because they show excellent properties such as vibrio harveyi inhibition activity,[7] turn-on fluorescent chemosensor,[8] neuritogenic activities in Neuro-2a cells,[9] and magnetic properties.[10] Some acylhydrazone compounds and their complexes have been synthesized and structurally characterized in our study group.[11–14] The molecular structure of the title compound is shown in figure. Our original intention was to synthesize an acylhydrazone magnesium complex, and unfortunately, magnesium ions are not involved in coordination. In the title compound, the bond lengths of C7=N3 and C6=O2 are 1.2684(19) and 1.2218(18) Å, respectively, which are shorter to C6–N2 (1.3516(18) Å) and C1–O1 (1.2551(19) Å), showing that C7=N3 and C6=O2 are expected for double bonds. The dihedral angle between two pyridine rings is 3.72°, showing that the 2-hydroxy-isonicotinic acid pyridin-4-ylmethylene-hydrazide molecule is almost coplanar. The title compound molecules form 1D chained structure by intermolecular N(1)–H(1)···N(4) hydrogen bonds, and the 1D chains further form a three-dimensional network structure by hydrogen bonds.

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

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

Research funding: Henan Academy of Sciences (220608035, 231508001, 242208025) and the National Natural Science Foundation of China (No. 21171132, https://doi.org/10.13039/501100001809).

References

1. Bruker: SAINT and SADABS; Bruker AXS Inc.: Madison, Wisconsin, USA, 2000.
2. 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.
3. Sheldrick, G. M. Crystal Structure Refinement with *SHELXL*. *Acta Crystallogr.* **2015**, *C71*, 3–8.
4. Brandenburg, K. *DIAMOND. Visual Crystal Structure Information System. Ver. 3.2*; Crystal Impact: Bonn, Germany, 2012.
5. Tai, X. S.; Wang, X. Synthesis, Structural Characterization and Antitumor Activity of a Ca(II) Coordination Polymer Based on 4-Formyl-1,3-Benzenedisulfonate-2-Furoic Acid Hydrazide Ligands. *Crystallogr. Rep.* **2017**, *62*, 242–245.
6. Kuriakose, D.; Prathapachandra Kurup, M. R. Supramolecular Frameworks Formed via Hydrogen Bonding and Non-covalent Interactions and Interaction Energy Calculations of Solvent Coordinated cis-Dioxomolybdenum(VI) Complexes Derived from ONO Donor Aroylhydrazone: Cytotoxicity Studies. *Inorg. Chim. Acta* **2020**, *505*, 119472.
7. Shao, Z. B.; Jiang, K. J.; Cao, L. G.; Wu, Y. Y.; Su, Z. Q.; Wang, Y. X.; Liu, S. H.; Wu, Y. R.; Liu, W. W. Synthesis of Emodin Acylhydrazone Derivatives and Determination of Vibrio Harveyi Inhibitory Activity. *Chem. Nat. Compd.* **2022**, *58*, 222–226.
8. Liu, Y. C.; Zhang, L. L.; Chen, L.; Liu, Z. X.; Liu, C. B.; Che, G. B. 2-Hydroxynaphthalene Based Acylhydrazone as a Turn-On Fluorescent Chemosensor for Al^{3+} Detection and Its Real Sample Applications. *Spectrochim. Acta A* **2021**, *248*, 119269.
9. Jiang, X.; Tang, G. Y.; Yang, J.; Ding, J. C.; Lin, H. W.; Xiang, X. L. Synthesis of Some New Acylhydrazone Compounds Containing the 1,2,4-Triazole Structure and Their Neuritogenic Activities in Neuro-2a Cells. *RSC Adv.* **2020**, *10*, 18927–18935.

10. Zhang, Y. M.; Zheng, F. W.; Chen, H. T.; Li, D. J.; Han, F. J.; Yang, L. Structure and Magnetic Properties of Tetranuclear Dysprosium Cluster Based on 2,6-Bis(2-((*E*)-(8-hydroxyquinolin-2-yl)methylene) hydrazine-1-Carbonyl)pyridine 1-oxide. *J. Cluster Sci.* **2021**, *32*, 737–742.
11. Wang, L. H.; Tai, H. W.; Song, G. Q.; Tai, X. S.: The Crystal Structure of Aqua-bis{2-bromo-6-((2-(2-phenylacetyl)hydrazineylidene)methyl) phenolato-κ^3N,O,O'-dimethylformamide-κ^1O-Erbium(III) Chloride-Dimethylformamide-Water (1/2/1), $C_{39}H_{49}N_7O_9Br_2ClEr$. *Z. Kristallogr. N. Cryst. Struct. 238* (**2023**) 437–439.
12. Wang, L. H.; Zhou, X. J.; Liu, L. L.; Zhang, A. L.; Tai, X. S. The Crystal Structure of (*E*)-2-(5-bromo-2-Hydroxybenzylidene)-*N*-Phenylhydrazine-1-Carboxamide Monohydrate, $C_{14}H_{14}BrN_3O_3$. *Z. Kristallogr. N. Cryst. Struct.* **2021**, *236*, 251–252.
13. Tai, X. S.; Wang, L. H.; Liu, L. L.; Zhou, X. J.: The Crystal Structure of (*E*)-*N*′-((2-hydroxynaphthalen-1-yl)methylene)-2-Phenylacetohydrazide, $C_{19}H_{16}N_2O_2$. *Z. Kristallogr. N. Cryst. Struct.* **2019**, *234*, 723–724.
14. Tai, X. S.; Li, P. F.: The Crystal Structure (*E*)-4-methyl-*N*′-(2-nitrobenzylidene)benzenesulfonohydrazide, $C_{14}H_{13}N_3O_4S$. *Z. Kristallogr. N. Cryst. Struct.* **2017**, *232*, 1031–1032.