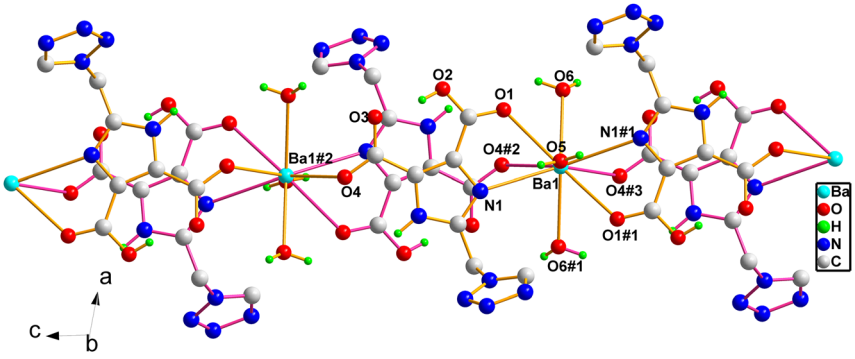
Wei Xie*, Rui Li and Chuan-Qi Wang

Crystal structure of *catena*-poly[triqua-bis(μ_2-4-carboxy-2-(1*H*-tetrazol-1-yl)-1*H*-imidazole-5-carboxylato-k^3*N*,*O*:*O*′)barium(II)] tetrahydrate, $C_{14}H_{14}BaN_{12}O_{15}$



https://doi.org/10.1515/ncrs-2020-0487
Received September 21, 2020; accepted October 16, 2020; published online November 6, 2020

Abstract

$C_{14}H_{14}BaN_{12}O_{15}$, monoclinic, *C*2/*c* (no. 15), a = 21.787(4) Å, b = 6.7594(11) Å, c = 18.143(3) Å, β = 102.456(2)°, V = 2609.0(8) Å^3, Z = 4, $R_{gt}(F)$ = 0.0209, $wR_{ref}(F^2)$ = 0.0567, T = 296(2) K.

CCDC no.: 2038892

A part of the polymeric title crystal structure is shown in the Figure (#1 = –*x*, *y*, 0.5–*z*; #2 = –*x*, –*y*, 1–*z*, #3 = *x*, –*y*, –0.5+*z*). Table 1 contains crystallographic data and Table 2 contains the list of the atoms including atomic coordinates and displacement parameters.

Table 1: Data collection and handling.

Crystal:	Yellow block
Size:	0.20 × 0.18 × 0.15 mm
Wavelength:	Mo *K*α radiation (0.71073 Å)
μ:	1.62 mm^{-1}
Diffractometer, scan mode:	Bruker D8 VENTURE PHOTON, φ and ω
θ_{max}, completeness:	28.4°, >99 %
$N(hkl)_{measured}$, $N(hkl)_{unique}$, R_{int}:	7939, 3174, 0.019
Criterion for I_{obs}, $N(hkl)_{gt}$:	I_{obs} > 2 $\sigma(I_{obs})$, 3033
$N(param)_{refined}$:	191
Programs:	SHELX [1], Bruker [2, 3]

Source of material

All chemicals were of AR grade and were used without purification. A mixture of $BaCl_2{\cdot}2H_2O$ (0.03 mmol), H_3tmidc systematic name: 2-(1*H*-tetrazol-1-yl)-1*H*-imidazole-4,5-dicarboxylic acid; (0.03 mmol), methanol (2 mL) and distilled water (2 mL) was sealed in a 25 mL Teflon lined stainless steel container and heated at 393 K for 72 h. After the mixture had been allowed to cool to room temperature at a rate of 5 K h^{-1}, light yellow crystals of $\{[Ba(H_2tmidc)_2(H_2O)_3]{\cdot}4H_2O\}_n$ suitable for X-ray analysis were obtained (yield 46.6%, based on H_3tmidc).

Experimental details

Hydrogen atoms on carbon atoms were positioned geometrically and refined as riding atoms, with C–H = 0.97 (CH_2) or 0.93 Å (aromatic). Hydrogen atoms of the non-deprotonated carboxylic acid groups of H_2tmidc^- were

***Corresponding author**: **Wei Xie**, Yangzhou Polytechnic Institute, 225127, Yangzhou, Jiangsu, P. R. China, E-mail: xieweilirui@163.com. https://orcid.org/0000-0002-1105-3461
Rui Li, Yangzhou Polytechnic Institute, 225127, Yangzhou, Jiangsu, P. R. China, E-mail: 3947962@qq.com
Chuan-Qi Wang, School of Chemical Engineering, Henan Technical Institute, 450042, Zhengzhou, Henan, P. R. China, E-mail: 1519863150@qq.com

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

Atom	x	y	z	U_{iso}*/U_{eq}
Ba1	0.000000	−0.03302(2)	0.250000	0.02661(6)
O1	0.08744(7)	−0.1912(3)	0.37305(8)	0.0410(4)
O2	0.12099(6)	−0.2841(3)	0.49134(8)	0.0397(3)
H2	0.104121	−0.323273	0.524886	0.059*
O3	0.07947(7)	−0.3248(2)	0.60551(8)	0.0371(3)
O4	−0.01132(7)	−0.2772(2)	0.64014(7)	0.0338(3)
O5	0.000000	−0.4287(4)	0.250000	0.142(2)
H5	0.006590	−0.508505	0.216330	0.213*
O6	0.12169(10)	0.1028(3)	0.27319(11)	0.0639(5)
H6C	0.134012	0.198995	0.249878	0.096*
H6D	0.136022	0.102475	0.320638	0.096*
N1	−0.03704(7)	−0.1765(2)	0.38795(8)	0.0226(3)
N2	−0.07360(7)	−0.2189(2)	0.49158(8)	0.0223(3)
H2A	−0.100054	−0.225143	0.523520	0.027*
N3	−0.17954(7)	−0.3304(3)	0.33880(9)	0.0324(3)
N4	−0.20982(9)	−0.4553(3)	0.37632(14)	0.0466(5)
N5	−0.22477(10)	−0.6081(4)	0.33334(16)	0.0595(6)
N6	−0.20451(11)	−0.5848(4)	0.26813(16)	0.0638(7)
C1	0.07690(8)	−0.2324(3)	0.43464(10)	0.0261(3)
C2	0.01188(7)	−0.2230(2)	0.44692(9)	0.0199(3)
C3	−0.01022(7)	−0.2490(2)	0.51200(9)	0.0203(3)
C4	0.02102(9)	−0.2872(3)	0.59196(9)	0.0252(3)
C5	−0.08748(8)	−0.1770(3)	0.41696(9)	0.0225(3)
C6	−0.15296(8)	−0.1453(3)	0.37311(11)	0.0306(4)
H6A	−0.178765	−0.095112	0.406334	0.037*
H6B	−0.152818	−0.047620	0.333971	0.037*
C7	−0.17625(12)	−0.4118(5)	0.27337(15)	0.0506(6)
H7	−0.157137	−0.356011	0.237177	0.061*
O7	0.17338(8)	0.1956(3)	0.43363(11)	0.0605(5)
H7A	0.191034	0.305220	0.428304	0.091*
H7B	0.202284	0.110870	0.448274	0.091*
O8	0.22961(10)	0.5705(3)	0.44047(16)	0.0749(7)
H8A	0.237056	0.653122	0.408187	0.112*
H8B	0.201696	0.618432	0.461567	0.112*

refined as riding atoms, with O–H = 0.82 Å. Hydrogen atoms on nitrogen atoms and hydrogen atoms of the water molecules were located in a difference Fourier map and the N–H and O–H distances were constrained to 0.9 and 0.85 Å, respectively. Hydrogen atoms were refined with U_{iso}(H) = 1.2 U_{eq}(C, N) or 1.5 U_{eq}(O).

Comment

It is well known that imidazole, tetrazole and their derivatives have been widely used as excellent building blocks for the preparation of complexes since they coordinate with most of metal ions with diverse coordination modes [4–10]. The N-heterocyclic carboxylic acid, 2-(1*H*-tetrazol-1-methyl)-1*H*-imidazole-4,5-dicarboxylic acid and the deprotonated anions are excellent ligands as there are potential N-donors and O-donors. Researchers have reported two transition metal complexes based on H_3tmidc and investigated their crystal structures and biological activities [11, 12]. In order to further enrich the number of complexes based on this ligand, we selected H_3tmidc as ligand to react with $BaCl_2{\cdot}2H_2O$ and obtained a new complex $\{[Ba(H_2tmidc)_2(H_2O)_3]{\cdot}4H_2O\}_n$.

There is one half Ba(II) ion, one H_2tmidc^- anion ligand, one and a half coordinated water molecules and two solvent water molecules in each asymmetric unit. The Ba1 ion is nine-coordinated by two N atoms from two H_2tmidc^- anions and seven O atoms from four H_2tmidc^- anions and three water molecules leading to a distorted BaN_2O_7 environment. The Ba–N bond length is 2.9541(14) Å, while the Ba–O distances span from 2.675(3) to 2.8669(14) Å, all of which are comparable to those observed for the other Ba(II) complexes based on N-heterocyclic carboxylic acids [13, 14]. Ba(II) ions are linked by H_2tmidc^- anion ligands into one-dimensional chains that run along the *c* axis. The intrachain Ba1–Ba1#2 distance is 9.0825(15) Å. There are O–H···O intramolecular hydrogen bonds between carboxyl and carboxylate groups, and O–H···O, O–H···N and N–H···O inter molecular hydrogen bonds involving carboxylate groups, imidazole ring, tetrazole ring, coordination water molecules and solvent water molecules. In addition, there are π–π stacking interactions between imidazole rings of adjacent chains with a centroid-centroid distance of 3.5699(7) Å, which is in the range for common π–π interactions [15–17]. Adjacent chains are linked by the aforementioned hydrogen bonds and π–π interactions, to a three-dimensional architecture in the solid state.

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

Research funding: We are very grateful for the financial assistance provided by Qinglan Project of Jiangsu Province (2020).

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

References

1. Sheldrick G. M. Crystal structure refinement with SHELXL. *Acta Crystallogr.* 2015, *C71*, 3–8.
2. Bruker. *SADABS*. Bruker AXS Inc.: Madison, Wisconsin, USA, 2000.
3. Bruker. *APEX3 and SAINT*. Bruker AXS Inc.: Madison, Wisconsin, USA, 2016.
4. Guo X.-Y., Chen J.-H., Meng X. R. Crystal structure of *catena*-poly[aqua[(μ_2-4,5-dicarboxylato-2-(2-carboxylatophenyl)imidazol-1-ido-κ^4N,O, O′: N′)] (μ2-4,4′-bipyridine κ^2N:N′)-N:) dicopper(II)], $C_{22}H_{14}Cu_2N_4O_7$. *Z. Kristallogr. NCS* 2020, *235*, 771–773.

5. Li X.-F., Yang Y.-Q., Li Y.-X., Yang H.-X., Zhao W.-F., Meng X. R. Synthesis, crystal structure, and BSA binding studies of new Co(II) and Ni(II) complexes of 2-(hydroxymethyl)-1*H*-imidazole-4,5-dicarboxylate. *Inorg. Chim. Acta.* 2020, *505*, 119469.
6. Cheng D., Liu Y.-J., Cheng F.-R., Yang H.-X., Meng X. R. Synthesis, structure and fluorescence properties of a Zn(II) coordination polymer based on 2-(hydroxymethyl)-1*H*-imidazole-4,5-dicarboxylate. *J. Chem. Res.* 2018, *42*, 490–493.
7. Vukadinovic Y., Burkhardt L., Papcke A., Miletic A., Fritsch L., Altenburger B., Schoch R., Neuba A., Lochbrunner S., Bauer M. When Donors turn into acceptors: ground and excited state properties of Fe^{II} complexes with amine-substituted tridentate bisimidazole-2-ylidene pyridine ligands. *Inorg. Chem.* 2020, *59*, 8762–8774.
8. Li S.-S., Zhao X.-Y., Huang Q.-Y., Meng X. R. Crystal structure of bis(acetato-*O*\k)bis{2-((1*H*-tetrazol-1-yl)methyl)-1*H*-benzo[*d*]imidazole-\k{*N*}zinc(II), $C_{22}H_{22}N_{12}O_4Zn$. *Z. Kristallogr. NCS* 2018, *233*, 693–695.
9. Liu B., Zhao D., Li T., Meng X.-R. Syntheses, structures, and fluorescent properties of two new Zn(II) coordination polymers containing 2-((benzoimidazolyl)methyl)-1*H*-tetrazole. *J. Coord. Chem.* 2013, *66*, 139–151.
10. Wurzenberger M. H. H., Lommel M., Gruhne M. S., Szimhardt N., Stierstorfer J. Refinement of copper(II) azide with 1-alkyl-5*H*-tetrazoles: adaptable energetic complexes. *Angew. Chem. Int. Ed.* 2020, *59*, 1–5.
11. Guo X.-Y., Zhang J.-D., Li Y.-Y., Li X.-J., Meng X.-R. Synthesis, structure, and BSA binding studies of a new Co(II) complex based on 2-(1*H*-tetrazol-1-methyl)-1*H*-imidazole-4,5-dicarboxylic acid. *Inorg. Chem. Commun.* 2020, *119*, 108055.
12. Yan H.-Y., Li Y.-X., Yang H.-X., Li X.-J. Synthesis, molecular structure and BSA-binding properties of a new binuclear Cd(II) complex based on 2-(1*H*-tetrazol-1-methyl)-1*H*-imidazole-4,5-dicarboxylic acid. *Z. Naturforsch.* 2020, *75b*, 537–544.
13. Cai S.-L., Zheng S.-R., Wen Z.-Z., Fan J., Zhang W.-G. Construction of Ba(II) coordination polymers based on imidazole-based dicarboxylate ligands: structural diversity tuned by alcohol solvents. *Cryst. Growth Des.* 2012, *12*, 3575–3582.
14. Chen G., Lan H. H., Li Z. X., Li D. J., Peng G. J., Cai S. L., Zheng S. R., Zhang W. G. Syntheses, structures, and luminescent properties of two alkaline earth metal coordination polymers from hydroxymethyl imidazole dicarboxylate. *Russ. J. Coord. Chem.* 2018, *44*, 792–799.
15. Yang Y.-Q., Su C.-F., Zhang J.-D., Yang H.-X., Zhang G.-Y., Meng X.-R. Construction of Cd(II) complexes based on 2-(1*H*-imidazol-1-methyl)-1*H*-benzimidazole and 1,4-benzenedicarboxylate. *J. Coord. Chem.* 2016, *69*, 3762–3775.
16. Li X. F., Cheng D., Meng X. R., Yang H. X. Effects of two benzenedicarboxylic acids on the Cd^{II} construction of coordination polymers incorporating a flexible *N*-donor ligand. *Acta Crystallogr.* 2019, *C75*, 643–649.
17. Wei X., Li J. H., Huang Q. Y., Meng X. R. Two new isostructural mercury(II) complexes involving the *N*-heterocyclic 1-[(benzotriazol-1-yl)methyl]-1*H*-1,3-imidazole ligand: syntheses, structures and properties. *Acta Crystallogr.* 2017, *C73*, 314–318.