Crystal Structure of 1,2-Bis[*N,N'*-6-(3-pyridylmethylamido)pyridyl-2-carboxyamido]ethane

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Compound **1**, namely 1,2-bis[*N,N'*-6-(3-pyridylmethylamido)pyridyl-2-carboxyamido]ethane, crystallizes in the space group *C2/c*, with cell parameters $a = 13.669(6) \text{ \AA}$, $b = 17.763(7) \text{ \AA}$, $c = 11.013(4) \text{ \AA}$, $\beta = 108.857(16)^\circ$, $Z = 4$, and $V = 2530.5(17) \text{ \AA}^3$. The asymmetric unit comprises half of the title compound with the compound adopting a U-shaped conformation stabilized by intramolecular hydrogen bonding. Crystal packing is governed by N-H...N and C-H...O hydrogen bonds. The structure has been solved and refined to reach *R* and *wR* indices for all data of 0.0514 and 0.1366, respectively.

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Simple pyridyl functionalized amides have been commonly employed as organic linkers in the construction coordination polymers.¹⁻⁴ In this capacity, the pyridine nitrogen donor normally provides the metal coordinating sites, while amide NH moieties can bind counterions or provide for linker-linker interactions in the solid-state structures. In some instances, often with pre-organized hydrogen bonding domains, these can allow the coordination polymer to act as an anion binding host.⁵⁻⁸ In this paper, we present the crystal structure of a new amide compound designed with the combination of amide moieties and pyridine pendant arms (Fig. 1). In this approach, an ethyl unit is incorporated as the spacer between the two commonly used 2,6-pyridine dicarboxamide units. Compound **1**, namely 1,2-bis[*N,N'*-6-(3-pyridylmethylamido)pyridyl-2-carboxyamido]ethane, was obtained in moderate yield, approximately 55% from the reaction between a reported monoamide precursor, namely 2-6-[(3-pyridylmethylamino)carbonyl]-pyridine-2-carboxylic acid methyl ester⁴ (0.50 g, 1.8 mmol) and ethylenediamine (0.062 mL, 0.92 mmol) in toluene (4 mL), after recrystallization in methanol.

Crystals of the title compound were obtained by slow evaporation. The crystal and structure-refinement data for this

compound are summarized in Table 1. H atoms were positioned geometrically and allowed to ride on the parent C or N atoms, with C-H = 0.95 – 0.96 Å and N-H = 0.88 Å.

This compound crystallizes in the monoclinic space group *C2/c* with an asymmetric unit comprising half of the molecule. The extended structure has a U-shaped arrangement, with the hydrogen bond amide donors of the two 2,6-pyridine dicarboxamide moieties and both pendant pyridyl donors oriented in the same direction (Fig. 2). Such arrangements are typical of compounds incorporating 2,6-pyridine dicarboxamide cores,⁹ but the concomitant arrangement of both entities is coincidental. Intramolecular hydrogen bonding pre-organizes the NH-functionalities of the molecule into a central pocket

Fig. 1 Chemical diagram of the title compound.

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Table 1 Crystal data and experimental data of compound **1**

Chemical formula: C ₂₈ H ₂₆ N ₈ O ₄	
Molecular weight = 538.57	
<i>T</i> = 173(2)K	
Crystal system: monoclinic	space group: <i>C2/c</i>
<i>a</i> = 13.669(6) Å	
<i>b</i> = 17.763(7) Å	$\beta = 108.857(16)^\circ$
<i>c</i> = 11.013(4) Å	
<i>V</i> = 2530.5(17) Å ³	<i>Z</i> = 4
Radiation: Mo <i>K</i> α ($\lambda = 0.71073 \text{ \AA}$)	
$\mu(\text{Mo } K\alpha) = 10.03 \text{ mm}^{-1}$	<i>F</i> (0 0 0) = 1128
Crystal size = 0.35 × 0.06 × 0.02 mm	
No. of reflections collected = 15729	
No. of independent reflections = 2386	
θ range for data collection: 1.95 to 25.95°	
Data/Restraints/Parameters = 2386/0/181	
Goodness-of-fit on <i>F</i> ² = 1.034	
<i>R</i> indices [<i>I</i> > 2σ(<i>I</i>)] : <i>R</i> 1 = 0.0389, <i>wR</i> 2 = 0.1245	
<i>R</i> indices [all data] : <i>R</i> 1 = 0.0514, <i>wR</i> 2 = 0.1366	
Measurement: CrysAlisPro, Agilent Technologies	
Programs system: SHELXTL-97, SADABS	
Structure determination: SHELXS-97	
CCDC deposition number: 1567077

Fig. 2 ORTEP structure of compound **1** with ellipsoids shown at the 50% probability level.

Fig. 3 Packing of the structure, viewed down the *b*-axis, showing molecules connected by N-H...N and C-H...O hydrogen bonds (dashed lines).

(N4A...H4A = 2.703(3) Å; N-H...N angle = 107°) and (N2A...H2A = 2.647(3) Å, N-H...N angle = 109°), these parameters are typical.¹⁰ Intermolecular hydrogen bonding interactions between the CH of the central pyridine rings with the carbonyl oxygen atom of neighboring molecules was observed in the crystal packing (Fig. 3), arranged in a zigzag formation (N4-H4...N1 = 2.703(3) Å, C2-H2A...O2 = 3.273(3) Å and C12-H12...O1 = 3.181(3) Å). The crystal structure contains no solvate molecules, and thus no further significant hydrogen bonding interactions were observed in the structure. The bond lengths and angles for selected atoms are shown in Table 2, while the bond lengths of the weak hydrogen bonding are shown in Table 3.

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Table 2 Selected geometric parameters, bond lengths and angles (Å, °)

Bond	Bond length/Å	Bond	Bond angle/°
O(1)–C(6)	1.234(3)	C(5)–N(1)–C(1)	118.5(2)
N(1)–C(5)	1.338(3)	N(1)–C(1)–C(2)	122.9(2)
N(1)–C(1)	1.341(3)	N(1)–C(1)–C(13)	117.3(2)
C(1)–C(2)	1.391(3)	C(6)–N(2)–C(7)	123.5(2)
C(1)–C(13)	1.508(3)	C(3)–C(2)–C(1)	117.7(2)
O(2)–C(13)	1.230(3)	C(10)–N(3)–C(9)	116.3(2)

Table 3 Hydrogen-bond geometry (Å, °)

D–H...A	D–H	H...A	D...A	D–H...A
N(2)–H(2)–N(1)	0.88	2.23	2.647(3)	109
N(2)–H(2)–N(3)	0.88	2.47	3.242(3)	147
N(4)–H(4)–N(1)	0.88	2.31	2.703(3)	107
N(4)–H(4)–N(3)	0.88	2.25	3.030(3)	147
C(2)–H(2A)–O(2)	0.95	2.40	3.273(3)	153
C(12)–H(12)–O(1)	0.95	2.47	3.181(3)	132

Symmetry codes: (i) 1–*x*, *y*, 5/2–*z*, (ii) 1/2–*x*, 1/2–*y*, 2–*z*, (iii) 1/2–*x*, 3/2–*y*, 2–*z*.

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