

タイトル	Crystal Structure of Tris[(1-naphthoyloxy)ethyl]amine
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Crystal Structure of Tris[(1-naphthoyloxy)ethyl]amine

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Abstract — Structure of tris[(1-naphthoyloxy)ethyl]amine (**1**), C₃₉H₃₃NO₆, was determined by X-ray crystallographic analysis. It crystallizes in the space group *P31c* (#159) with cell parameters $a=b=14.3418(5)$ Å, $c=8.6968(2)$ Å, $\alpha=\beta=90^\circ$, $\gamma=120^\circ$, $Z=2$, and $V=1549.16(13)$ Å³. The molecule (**1**) has a tripod conformation with crystallographic C_3 symmetry. The crystal packing is consolidated by intermolecular C-H \cdots π interactions.

1. Introduction

During the last decades, macrocyclic and acyclic ligands have been designed to coordinate metal ions.^{1,2)} Polypod organic molecules containing donor atoms may be considered as non-cyclic cryptands³⁻⁷⁾ and indeed some of their complexes with alkali and alkali earth metal ions are more stable than the respective linear oligoether and cyclic crown ether complexes.⁵⁾ Recently, a number of fluoroionophores have been designed as ligands for metal cations. There are extensive investigations toward the characterization of photoinduced electron transfer (PET) fluoroionophores with naphthalene, anthracene, pyrene, and coumarin. Recently, it was found that the azacrown ethers act as electron donors in a typical exciplex-forming system that involves an appropriate electron acceptor. The addition of metal salts changed the fluorescence intensity of PET fluoroionophore. Moreover, the crystal structures of PET fluoroionophores such as 7,16-bis(1-naphthylmethyl)-1,4,10,13-tetraoxa-7,16-diazacyclooctadecane (**Np186Np**),⁸⁾ its HNCS salt (**Np186Np**·2HNCS),⁹⁾ lithium thiocyanate complex (**Np186Np**·2LiNCS),¹⁰⁾ potassium thiocyanate complex (**Np186Np**·KNCS),¹¹⁾ barium thiocyanate complex (**Np186Np**·Ba(NCS)₂·H₂O),¹¹⁾ zinc thiocyanate complex ((H₂·**Np186Np**)[Zn(NCS)₄])¹⁰⁾, 7,16-bis(1-pyrenylmethyl)-1,4,10,13-tetraoxa-7,16-diazacyclooctadecane, *N,N',N'',N'''*-tetrakis(1-naphthylmethyl)-1,4,8,11-tetraazacyclotetradecane,^{12,13)} *N,N'*-[9,10-anthracenediyl-bis(methylene)]bis[*N*-(2-ethoxyoxoethyl)]-glycine 1,1'-diethyl ester,¹⁴⁾ 1,4-bis(1-naphthylmethyl) piperazine,¹⁵⁾ 1,4-bis(9-anthracenylmethyl) piperazine,¹⁶⁾ *N9,N9,M10,M10*-tetraethyl-9,10-anthracenedimethanamine,¹⁷⁾ *N,N,N',N'*-tetraethyl-9,10-anthracenedimethanamine dihydrogen tetrachlorozincate,¹⁸⁾ 1,1'-[9,10-anthracenediyl-bis(methylene)]bis[4-methylpiperazine],¹⁸⁾

1,1'-[9,10-anthracenediylbis(methylene)]bis[4-(2-pyridinyl)piperazine],¹⁹⁾ *N*9, *N*9', *N*10, *N*10-tetrakis(2-pyridinylmethyl)-9,10-anthracenedimethanamine,¹⁸⁾ tetrachloro[μ -[*N*, *N*', *N*'', *N*''']tetrakis[(2-pyridinyl- κ *N*)methyl]-9,10-anthracenediamine]]di-zinc,¹⁸⁾ 9-(thiomorpholinomethyl)anthracene,²⁰⁾ and 9,10-bis(morpholinomethyl)anthracene²¹⁾ were elucidated by X-ray crystallographic analysis. We have reported tris[(1-naphthoyloxy)ethyl]amine (**1**) as a fluorescent pH indicator for Al³⁺.²¹⁾ We now report on the structure of tris[(1-naphthoyloxy)ethyl]amine (**1**) with the aim of contributing to a deeper understanding of PET systems.

2. Experimental

2.1. Analytical Instruments

Elemental analyses were performed by Perkin Elmer PE2400 series II CHNS/O analyzer. Melting points were obtained with a Yanagimoto Micro Melting Point Apparatus and are uncorrected. NMR spectra were measured on a JEOL JNM-500 Model spectrometer in CDCl₃; the chemical shifts are expressed by an δ unit using TMS as an internal standard. IR spectra were recorded on a Hitachi Model 270-30 infrared spectrophotometer. UV spectra were measured using a Shimadzu Model UV-2200 spectrophotometer. Fluorescence spectra were measured with a Hitachi Model F-4500 spectrofluorometer. Mass spectra were measured with a JEOL O1SG-2 spectrometer.

2.2. Preparation of Tris[(1-naphthoyloxy)ethyl]amine (**1**)

A THF-Et₃N mixture solution (2 + 2 cm³) of triethanolamine (149 mg, 1.0 mmol) and 1-naphthoyl chloride (953 mg, 5.0 mmol) was refluxed for 6 hr. Cooling the mixture solution afforded the precipitate. The precipitate was filtered and washed with water (5 cm³). Recrystallization from ethyl acetate gave analytically pure sample (489 mg, 80%) with the following physical and spectroscopic properties. colorless crystals, mp 73-74°C, ¹H NMR (500 MHz, CDCl₃) δ =3.22 (3H, *t*, *J*=6.2 Hz), 4.57 (3H, *t*, *J*=6.2 Hz), 7.30 (3H, *dd*, *J*=8.2, 7.3 Hz), 7.48 (3H, *ddd*, *J*=8.6, 7.0, 1.2 Hz), 7.55 (3H, *ddd*, *J*=8.5, 7.0, 1.2 Hz), 7.80 (3H, *d*, *J*=8.6 Hz), 7.90 (3H, *d*, *J*=8.2 Hz), 8.11 (3H *d*, *J*=7.3 Hz), and 8.89 (3H, *d*, *J*=8.5 Hz). ¹³C NMR (125.7 MHz CDCl₃) δ =53.6 (3C), 63.1 (3C), 124.4 (3C), 125.8 (3C), 126.1 (3C), 126.9 (3C), 127.7 (3C), 128.5 (3C), 130.2 (3C), 131.4 (3C), 133.3 (3C), 133.8 (3C), and 167.4 (3C). IR (KBr) ν 774, 920, 1012, 1136, 1196, 1242, 1276, 1358, 1442, 1502, 1590, 1700, 2824, and 2952 cm⁻¹, UV (CH₃OH) λ =211 (ϵ =10000), 216 (10000), 233 (5800 sh), 297 (1700), and 324 (700 sh). FAB-MS (M+H) 612. E.A. Calcd for C₃₉H₃₃NO₆: C, 76.58; H, 5.44; N, 2.29. Found: C, 76.47; H, 5.45; N, 2.33.

2.3. Data Collection

A colorless prism crystal of $C_{39}H_{33}NO_6$ having approximate dimensions of 0.60 x 0.50 x 0.50 mm was mounted on a glass fiber. All measurements were made on Rigaku RAXIS RAPID imaging plate area detector with graphite monochromated Mo- $K\alpha$ radiation.

Cell constants and an orientation matrix for data collection, obtained from a least-squares refinement using the setting angles of 23183 carefully centered reflections in the range $5.80 < 2\theta < 55.00^\circ$ corresponded to a primitive trigonal cell (Laue class: $\bar{3}1m$) with dimensions: $a=14.3418(5)$ Å, $c=8.6968(2)$ Å, $V=1549.16(13)$ Å³. For $Z=2$ and F.W.=611.67, the calculated density is 1.311 g/cm³. Based on the systematic absences of $hh2-hl: l \neq 2n$ packing considerations, a statistical analysis of intensity distribution, and the successful solution and refinement of the structure, the space group was determined to be $P31c$ (#159).

2.4. Data Reduction

Of the 1197 reflections that were collected, 1197 were unique ($R_{int}=0.000$); equivalent reflections were merged. No decay correction was applied. The linear absorption coefficient, μ , for Mo- $K\alpha$ radiation is 0.881 cm⁻¹. An empirical absorption correction was applied which resulted in transmission factors ranging from 0.777 to 0.957. The data were corrected for Lorentz and polarization effects.

2.5. Structure Solution and Refinement

The structure was solved by direct methods²²⁾ and expanded using Fourier techniques.²³⁾ Some non-hydrogen atoms were refined anisotropically, while the rest were refined isotropically. Hydrogen atoms were refined using the riding model. The final cycle of full-matrix least-squares refinement ($\sum w(F_0^2 - F_c^2)^2$ where w =least-squares weights) on F^2 was based on 1197 observed reflections and 135 variable parameters and converged (largest parameter shift was 0.00 times its esd) with unweighted and weighted agreement factors of: $R_1 = \sum \|F_0\| - |F_c| / \sum |F_0| = 0.0310$, $wR_2 = [\sum (w(F_0^2 - F_c^2)^2) / \sum w(F_0^2)^2]^{1/2} = 0.1062$. The standard deviation of an observation of unit weight⁴ was 1.12. Unit weights were used. The maximum and minimum peaks on the final difference Fourier map corresponded to 0.20 and -0.29 e⁻/Å³, respectively.

Neutral atom scattering factors were taken from Cromer and Waber.²⁴⁾ Anomalous dispersion effects were included in F_{calc} .²⁵⁾ The values for $\Delta f'$ and $\Delta f''$ were those of Creagh and McAuley.²⁶⁾ The values for the mass attenuation coefficients are those of Creagh and Hubbell.²⁷⁾ All calculations were performed using the *CrystalStructure*²⁸⁾ crystallographic software package except for refinement, which was performed using *SHELXL97*.²⁹⁾ Fractional atomic coordinates

and equivalent isotropic displacement parameters were shown in Table 1. Anisotropic displacement parameters were shown in Table 2.

Table 1. Fractional atomic coordinates, equivalent isotropic displacement parameters ($B_{\text{iso}}/B_{\text{eq}}$)

atom	x	y	z	B_{eq}	occ
O1	0.93258(14)	0.59436(16)	1.2385(2)	3.61(3)	
O2	0.77692(14)	0.52220(14)	1.10884(18)	3.32(3)	
N1	0.6667	0.3333	0.9080(3)	2.71(5)	1/3
C1	0.77154(18)	0.56949(17)	1.3635(2)	2.70(3)	
C2	0.6647(2)	0.4910(2)	1.3710(2)	3.11(4)	
C3	0.5972(2)	0.4893(2)	1.4902(3)	3.60(4)	
C4	0.6376(2)	0.5670(2)	1.6001(2)	3.61(4)	
C5	0.7469(2)	0.6494(2)	1.5977(2)	3.30(4)	
C6	0.7892(2)	0.7309(2)	1.7118(3)	4.09(5)	
C7	0.8922(2)	0.8116(2)	1.7069(3)	4.63(6)	
C8	0.9610(2)	0.8159(2)	1.5876(3)	4.25(5)	
C9	0.9241(2)	0.73768(19)	1.4771(3)	3.35(4)	
C10	0.8167(2)	0.65172(19)	1.4788(2)	2.86(3)	
C11	0.83780(19)	0.56573(17)	1.2338(2)	2.84(3)	
C12	0.8282(2)	0.5052(2)	0.9760(2)	3.46(4)	
C13	0.7388(2)	0.4448(2)	0.8615(2)	3.17(4)	

$$B_{\text{eq}}=8/3\pi^2(U_{11}(aa^*)^2+U_{22}(bb^*)^2+U_{33}(cc^*)^2+2U_{12}(aa^*bb^*)\cos \gamma+2U_{13}(aa^*cc^*)\cos \beta+2U_{23}(bb^*cc^*)\cos \alpha)$$

Table 2. Anisotropic displacement parameters (\AA^2)

Atom	U^{11}	U^{22}	U^{33}	U^{12}	U^{13}	U^{23}
O1	0.0385(9)	0.0437(9)	0.0493(9)	0.0164(7)	0.0008(7)	-0.0035(7)
O2	0.0397(8)	0.0487(9)	0.0356(8)	0.0204(7)	0.0006(6)	-0.0080(7)
N1	0.0381(10)	0.0381(10)	0.0267(13)	0.0190(5)	0.0000	0.0000
C1	0.0379(11)	0.0335(11)	0.0352(10)	0.0209(9)	-0.0026(9)	0.0004(8)
C2	0.0413(12)	0.0385(11)	0.0402(11)	0.0213(10)	-0.0005(10)	0.0002(9)
C3	0.0401(12)	0.0537(14)	0.0447(12)	0.0248(11)	0.0029(10)	0.0029(10)
C4	0.0529(14)	0.0631(16)	0.0360(11)	0.0400(13)	0.0032(10)	0.0034(10)
C5	0.0584(15)	0.0478(13)	0.0353(10)	0.0388(12)	-0.0071(10)	-0.0013(10)
C6	0.0732(19)	0.0647(17)	0.0407(13)	0.0518(16)	-0.0119(12)	-0.0128(12)
C7	0.085(2)	0.0581(17)	0.0532(15)	0.0509(17)	-0.0265(15)	-0.0232(12)
C8	0.0614(16)	0.0393(13)	0.0613(17)	0.0256(12)	-0.0208(13)	-0.0114(11)
C9	0.0501(13)	0.0350(11)	0.0452(12)	0.0236(11)	-0.0098(10)	-0.0036(9)
C10	0.0486(12)	0.0364(11)	0.0343(10)	0.0291(10)	-0.0059(9)	0.0001(8)
C11	0.0379(11)	0.0278(10)	0.0396(11)	0.0145(9)	-0.0013(9)	-0.0012(8)
C12	0.0394(12)	0.0445(13)	0.0395(11)	0.0148(10)	0.0071(9)	-0.0039(9)

The general temperature factor expression:

$$\exp(-2\pi^2(a^*U_{11}h^2+b^*U_{22}k^2+c^*U_{33}l^2+2a^*b^*U_{12}hk+2a^*c^*U_{13}hl+2b^*c^*U_{23}kl))$$

3. Result & Discussion

Compound (**1**) was synthesized by the esterification of triethanolamine with 1-naphthoyl chloride (Fig. 1).²¹⁾ The purity of **1** was ascertained by ¹H, ¹³C NMR spectroscopic data, and elemental analysis.

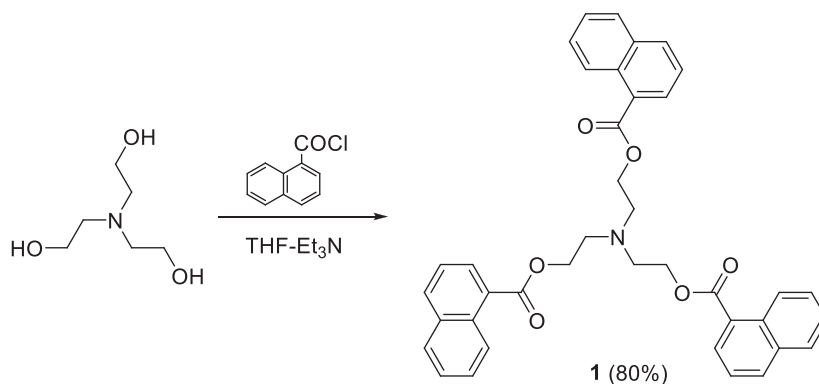


Fig. 1. Synthesis of title compound (**1**).

As reported in a previous paper,²¹⁾ the amine (**1**) revealed a broad emission band around 440 nm that is typical of a structureless emission of a naphthoyl excimer and a naphthalene-amine exciplex. When the metal salts (Na⁺, K⁺, Ca²⁺, Ba²⁺, Al³⁺, Ni²⁺, Cu²⁺, Zn²⁺, Cd²⁺, and Hg²⁺) were added (2.5x10³ molar equivalent), the relative emission intensity (I_{complex}/I_1), being used as a measure of the molecular recognition, changed from 0.04 to 5 depending on the nature of metal cations. The intensity ratio depends on ligand metal cations and decreases in the following order: Al³⁺ (5.3) > Zn²⁺ (3.4) > Ni²⁺ (1.6) > Ca²⁺, Cd²⁺ (1.2) > Na⁺, K⁺, Ba²⁺, Hg²⁺ (0.9) > Cu²⁺ (0.04). The amine (**1**) had a high fluorescence switch-on ability as an Al³⁺ sensing fluoroionophore.

With the aim of the contributing to a deeper understanding of the complexation behavior of **1**, the crystal structure of **1** was elucidated by X-ray analysis. Single crystals of **1** for X-ray analysis were grown by slow evaporation of a chloroform solution at room temperature.

The crystal structure of **1** was shown in Fig. 2. The selected geometric parameters were shown in Table 3. Compound (**1**) adopts a tripod conformation with crystallographic C₃ symmetry. Intramolecular C-H···O interactions are observed in **1**. The H2···O1 is 2.722 Å (symmetry code: (i) 1-y, x-y, z), which is close to this type interaction (2.5–2.7 Å). The two dihedral angles between the least-squares naphthalene plane A (defined by C1/C2/C3/C4/C5/C6/C7/C8/C9/C10) and the naphthalene plane B (defined by C1ⁱ/C2ⁱ/C3ⁱ/C4ⁱ/C5ⁱ/C6ⁱ/C7ⁱ/C8ⁱ/C9ⁱ/C10ⁱ) and between plane A

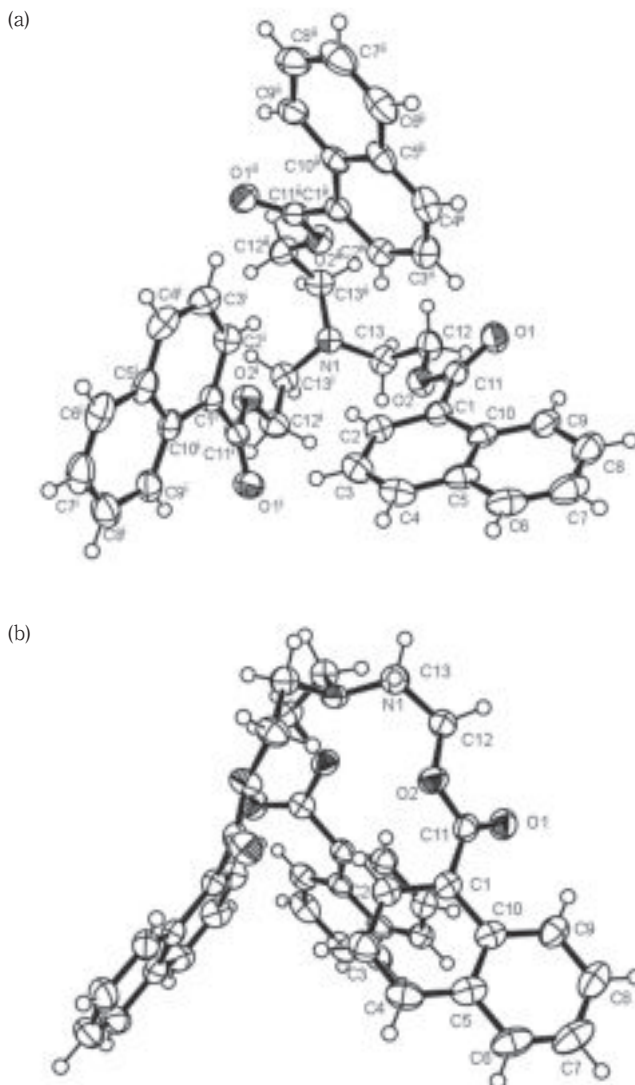


Fig. 2. ORTEP drawings ((a) top view and (b) side view) of **1** showing 50% probability displacement ellipsoids. Symmetry codes: (i) $1-y, x-y, z$, (ii) $1-x+y, 1-x, z$.

and the naphthalene plane C (defined by $C1^{ii}/C2^{ii}/C3^{ii}/C4^{ii}/C5^{ii}/C6^{ii}/C7^{ii}/C8^{ii}/C9^{ii}/C10^{ii}$) are $92.0(1)^\circ$. The conformation of the triethanolamine moiety is: $N1(t)C13(g)C12(t)O2(t)C11$, where *t* and *g* denote *trans* and *gauche* form, respectively.

Some intermolecular $C-H \cdots \pi$ interactions are observed in **1** as shown in Fig. 3 and Table 4. The $H \cdots C$ distances (2.722, 2.779, 2.862, and 2.898 Å) are within the range associated with $C-H \cdots \pi$ interaction (2.8–3.1 Å).^{7–21)}

In conclusion, the crystal structure of tris[(1-naphthoyloxy)ethyl]amine (**1**) was elucidated by

Table 3. Selected geometric parameters (Å, °)

O1—C11	1.208(3)	O2—C11	1.337(2)
O2—C12	1.454(3)	N1—C13	1.461(2)
N1—C13 ⁱ	1.462(2)	N1—C13 ⁱⁱ	1.460(3)
C1—C2	1.376(2)	C1—C10	1.433(3)
C1—C11	1.493(3)	C2—C3	1.411(4)
C3—C4	1.358(3)	C4—C5	1.415(3)
C5—C6	1.418(3)	C5—C10	1.427(4)
C6—C7	1.347(4)	C7—C8	1.412(5)
C8—C9	1.367(3)	C9—C10	1.412(3)
C12—C13	1.508(3)		
C11—O2—C12	117.2(2)	C13—N1—C13 ⁱ	112.60(18)
C13—N1—C13 ⁱⁱ	112.68(17)	C13 ⁱ —N1—C13 ⁱⁱ	112.67(17)
C2—C1—C10	120.0(2)	C2—C1—C11	118.32(19)
C10—C1—C11	121.65(18)	C1—C2—C3	121.5(2)
C2—C3—C4	119.7(2)	C3—C4—C5	121.0(2)
C4—C5—C6	121.1(2)	C4—C5—C10	120.1(2)
C6—C5—C10	118.8(2)	C5—C6—C7	121.3(3)
C6—C7—C8	120.2(2)	C7—C8—C9	120.3(2)
C8—C9—C10	121.0(2)	C1—C10—C5	117.7(2)
C1—C10—C9	123.9(2)	C5—C10—C9	118.3(2)
O1—C11—O2	123.2(2)	O1—C11—C1	126.2(2)
O2—C11—C1	110.6(2)	O2—C12—C13	105.4(2)
N1—C13—C12	112.3(2)		
C11—O2—C12—C13	173.8(2)	C12—O2—C11—O1	2.7(3)
C12—O2—C11—C1	-175.4(2)	C13—N1—C13 ⁱ —C12 ⁱ	-85.0(2)
C13 ⁱ —N1—C13—C12	146.2(2)	C13—N1—C13 ⁱⁱ —C12 ⁱⁱ	146.2(2)
C13 ⁱⁱ —N1—C13—C12	-85.0(2)	C13 ⁱ —N1—C13 ⁱⁱ —C12 ⁱⁱ	-85.0(2)
C13 ⁱⁱ —N1—C13 ⁱ —C12 ⁱ	146.2(2)	C2—C1—C10—C5	-0.9(4)
C2—C1—C10—C9	-177.4(3)	C10—C1—C2—C3	0.3(4)
C2—C1—C11—O1	-147.5(2)	C2—C1—C11—O2	30.4(3)
C11—C1—C2—C3	180.0(2)	C10—C1—C11—O1	32.1(4)
C10—C1—C11—O2	-149.9(2)	C11—C1—C10—C5	179.4(2)
C11—C1—C10—C9	3.0(4)	C1—C2—C3—C4	0.5(5)
C2—C3—C4—C5	-0.5(5)	C3—C4—C5—C6	179.7(3)
C3—C4—C5—C10	-0.1(4)	C4—C5—C6—C7	-177.6(3)
C4—C5—C10—C1	0.9(4)	C4—C5—C10—C9	177.5(3)
C6—C5—C10—C1	-179.0(3)	C6—C5—C10—C9	-2.3(4)
C10—C5—C6—C7	2.2(5)	C5—C6—C7—C8	-0.7(6)
C6—C7—C8—C9	-0.7(6)	C7—C8—C9—C10	0.5(5)
C8—C9—C10—C1	177.4(3)	C8—C9—C10—C5	1.0(4)
O2—C12—C13—N1	-71.8(3)		

Symmetry codes: (i) 1-y, x-y, z, (ii) 1-x+y, 1-x, z.

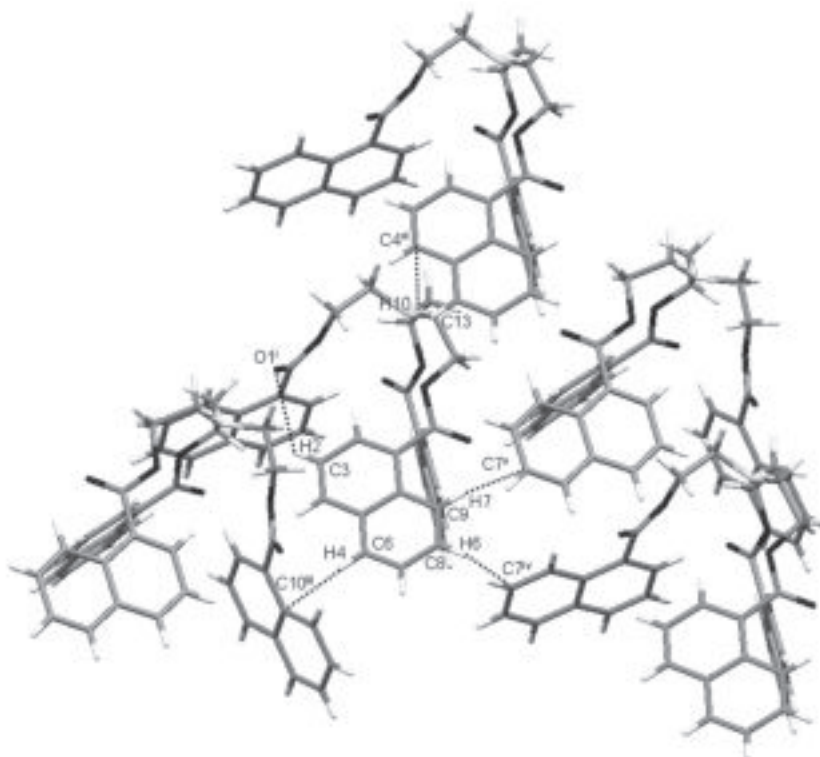


Fig. 3. Intermolecular C-H $\cdots\pi$ interactions of **1**. Symmetry codes: (i) 1-y, x-y, z, (iii) y, x, 1/2+z, (iv) z-y, 1+x-y, z, (v) y, x, 1/2+z, (vi) x, y, z-1.

Table 4. Hydrogen bond geometry (\AA , $^\circ$) of **1**

D	H	A	D-H	H \cdots A	D \cdots A	D-H \cdots A	Symmetry codes
C3	H2	O1 ⁱ	0.930	2.722	3.329(3)	124	(i) 1-y, x-y, z
C6	H4	C10 ⁱⁱⁱ	0.930	2.779	3.636(4)	154	(iii) y, x, 1/2+z
C8	H6	C7 ^{iv}	0.930	2.898	3.707(4)	146	(iv) z-y, 1+x-y, z
C9	H7	C7 ^v	0.930	2.862	3.520(4)	129	(v) y, x, 1/2+z
C13	H10	C4 ^{vi}	0.970	2.838	3.590(4)	135	(vi) x, y, z-1

X-ray crystallographic analysis. Intermolecular C-H $\cdots\pi$ interactions help stabilized the crystal packing.

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