Supplementary materials to

Confirming the Molecular Basis of the Solvent Extraction of Cadmium(II) using 2-pyridyl Oximes through a Synthetic Inorganic Chemistry Approach and a Proposal for more Efficient Extractants

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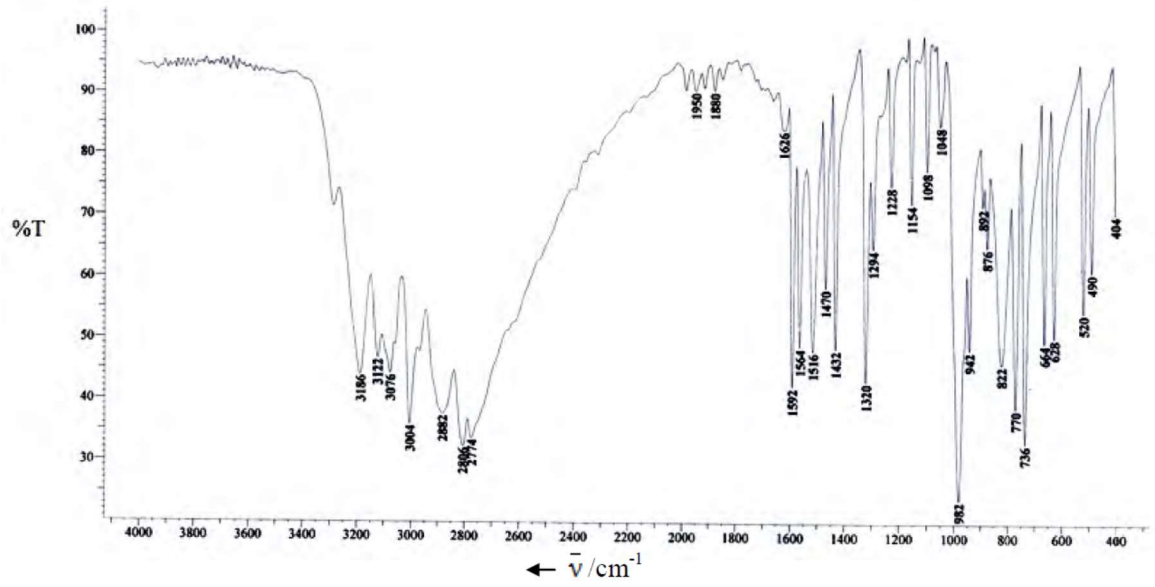

Figure S1. The FT-IR spectrum (KBr/cm^{-1}) of free paoH.

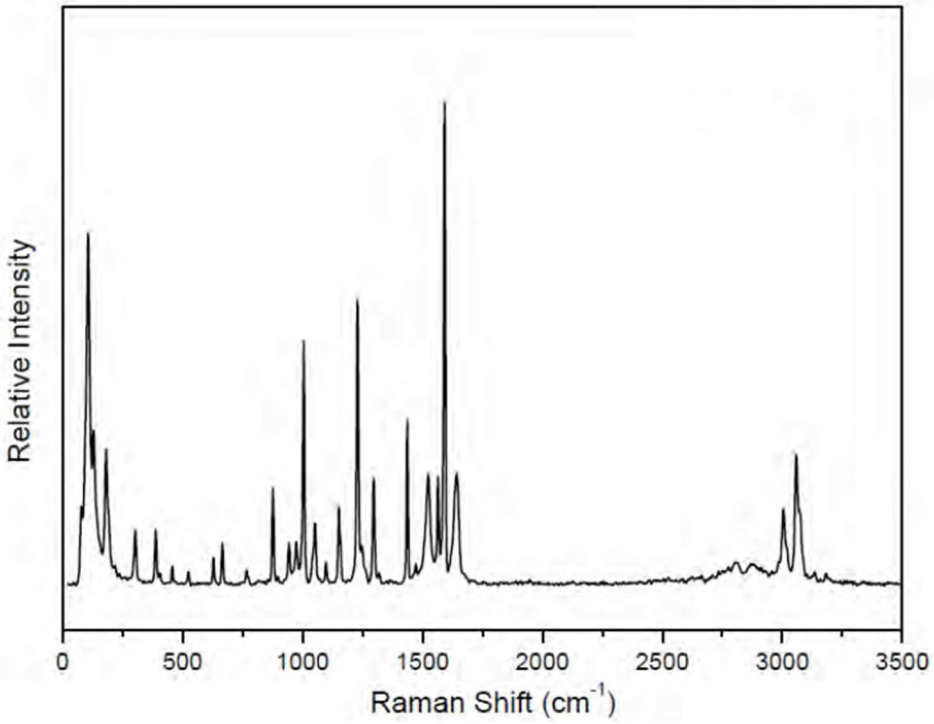

Figure S2. The FT-Raman spectrum of free paoH.

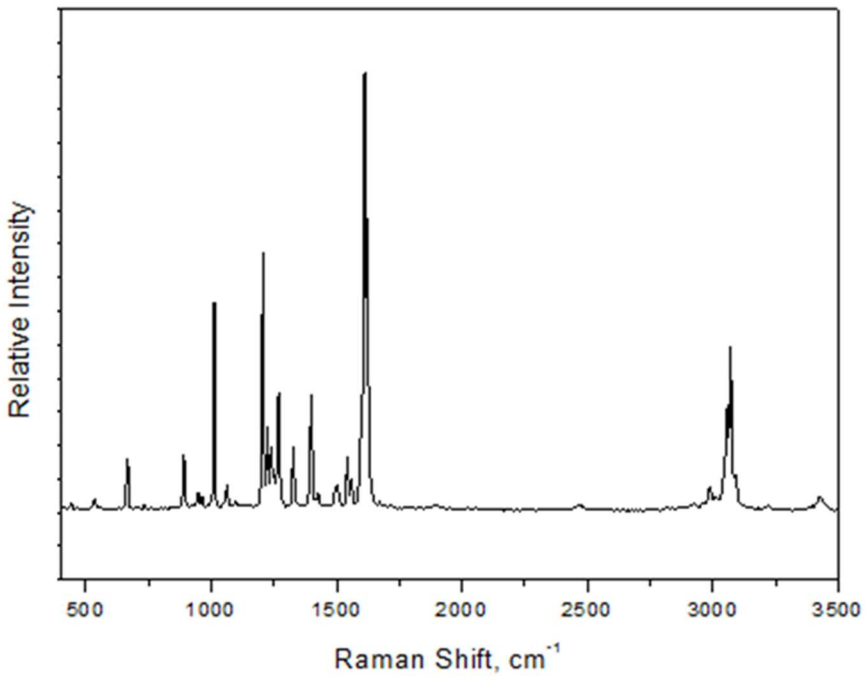

Figure S3. The FT-Raman spectrum of $\{[CdI_2(4paoH)_2]\}_n$ (**3**).

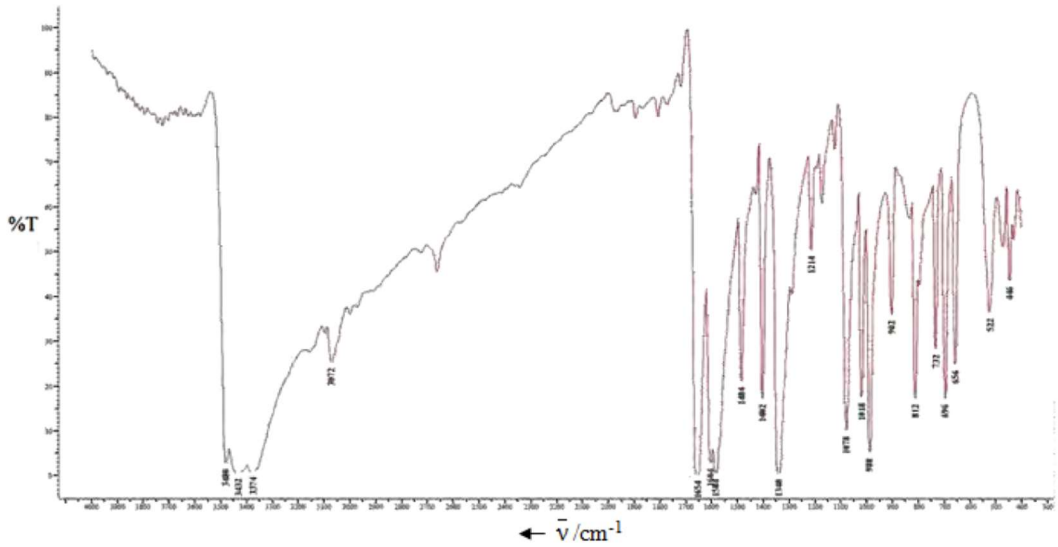

Figure S4. The FT-IR spectrum (KBr/cm^{-1}) of compound $[CdI_2(LH_4)]$ (**5a**).

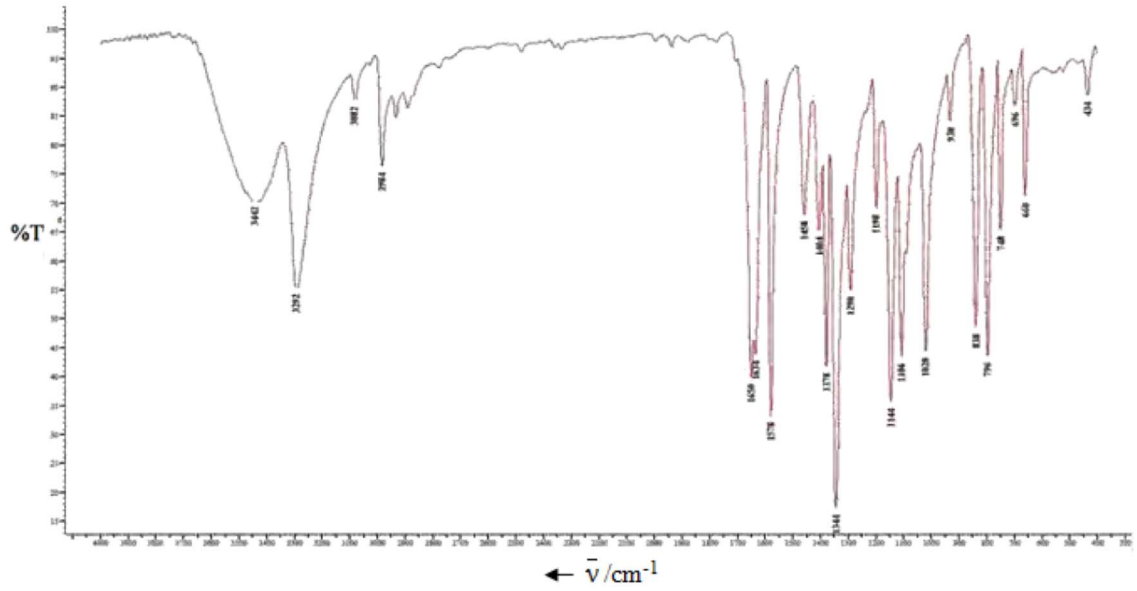

Figure S5. The FT-IR spectrum (KBr/cm^{-1}) of complex [$CdI_2(L'H)_2$] (**5**).

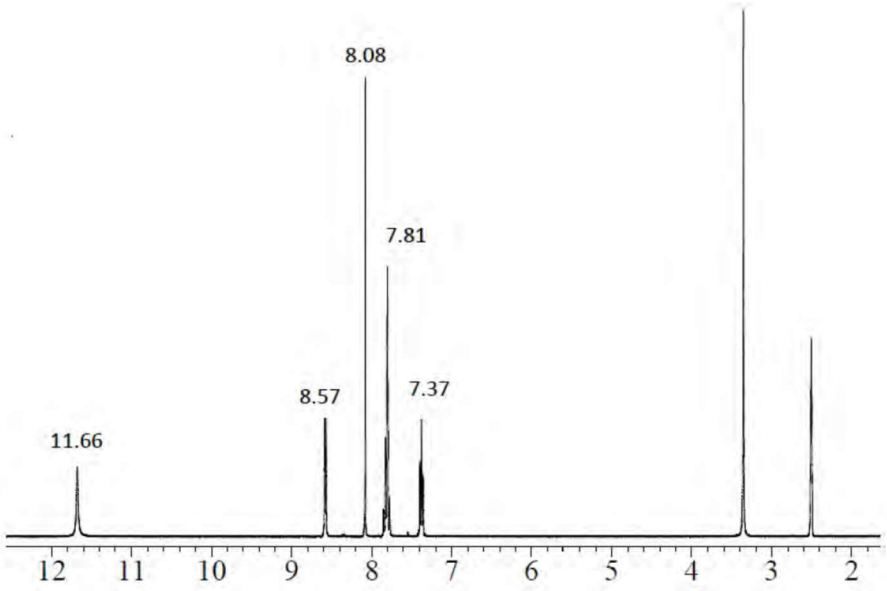

Figure S6. The ^{1}H NMR spectrum (δ/ppm) of the free ligand 2paoH in d_6-DMSO.

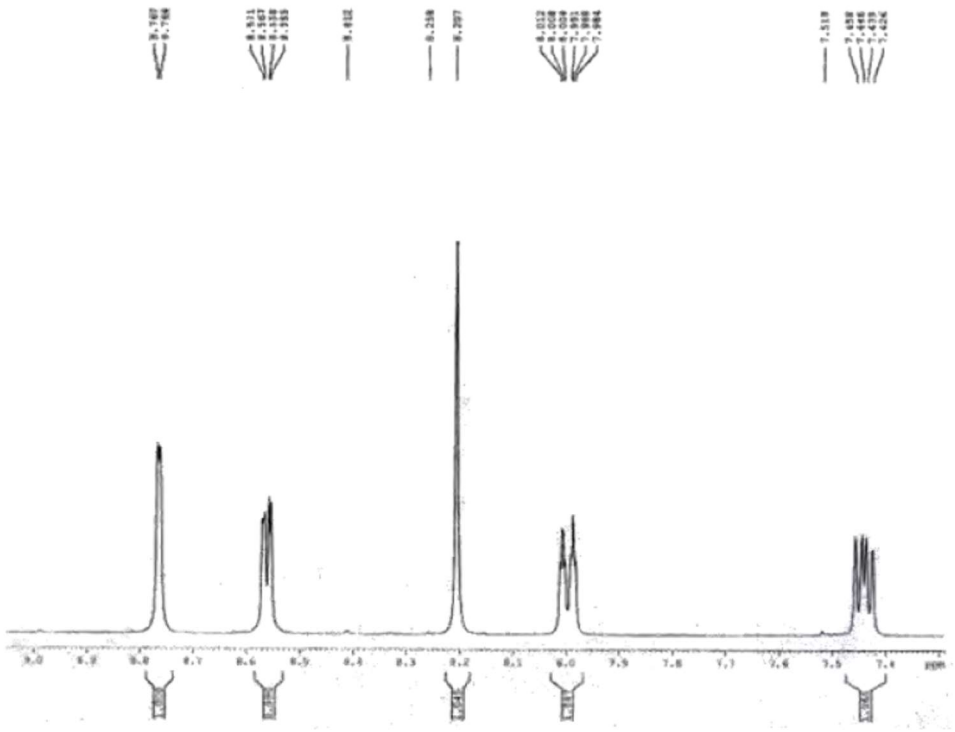

Figure S7. The ^{1}H NMR spectrum (δ/ppm) of complex $\{[CdI_2(3paoH)]\}_n$ (**2**)$_x$ in d_6-DMSO in the 9.0-7.33 ppm region.

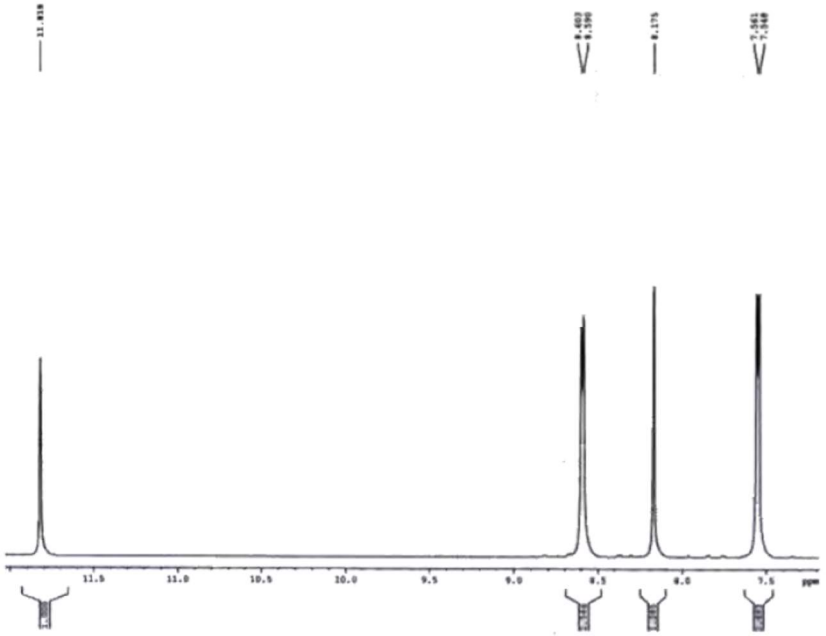

Figure S8. The ^{1}H NMR spectrum (δ/ppm) of compound $\{[CdI_2(4paoH)_2]\}_n$ (**3**) in d_6-DMSO in the 12.0-7.3 ppm region.

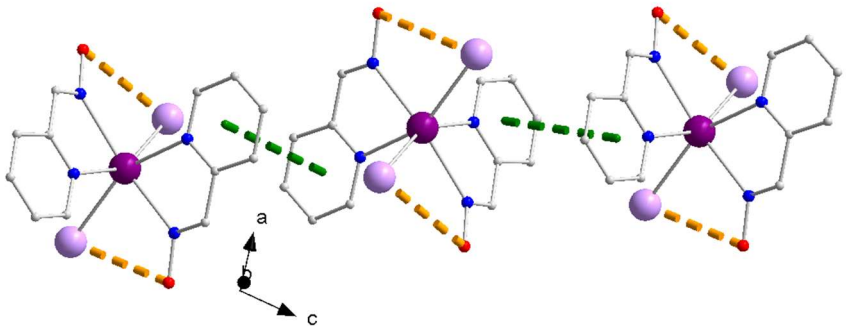

Figure S9. A portion of one chain, parallel to the crystallographic direction [101], in the crystal structure of [$CdI_2(2paoH)_2$] (**1**). The thick dashed orange and green lines represent the intramolecular O1-H(O1)···I1′ and O1′-H(O1′)…I1 hydrogen bonds [(′)= *-x*, *y*, *-z*+1/2] and the π-π interactions, respectively.

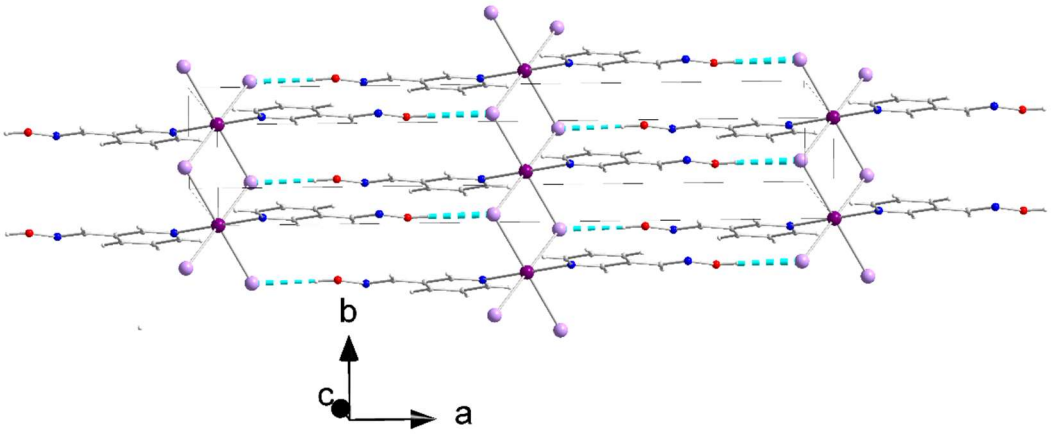

Figure S10. Layers of chains parallel to the (110) plane in the crystal structure of {[$CdI_2(3paoH)_2$]}$_n$ (**2**). The layers are formed through O-H···I hydrogen bonds (thick dashed cyan lines); see text for more details.

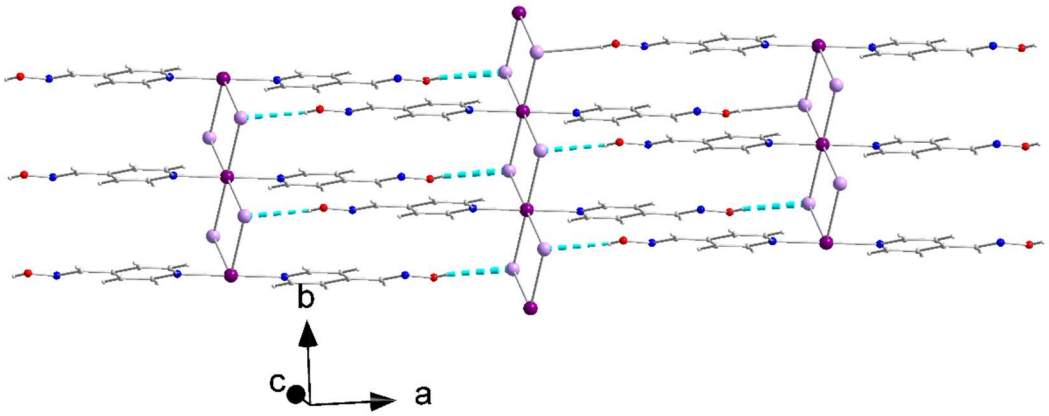

Figure S11. Layers of chains parallel to the (110) plane in the crystal structure of {[$CdI_2(4paoH)_2$]}$_n$ (**3**). The layers are formed through O-H···I hydrogen bonds (thick dashed cyan lines); see text for the metric parameters.

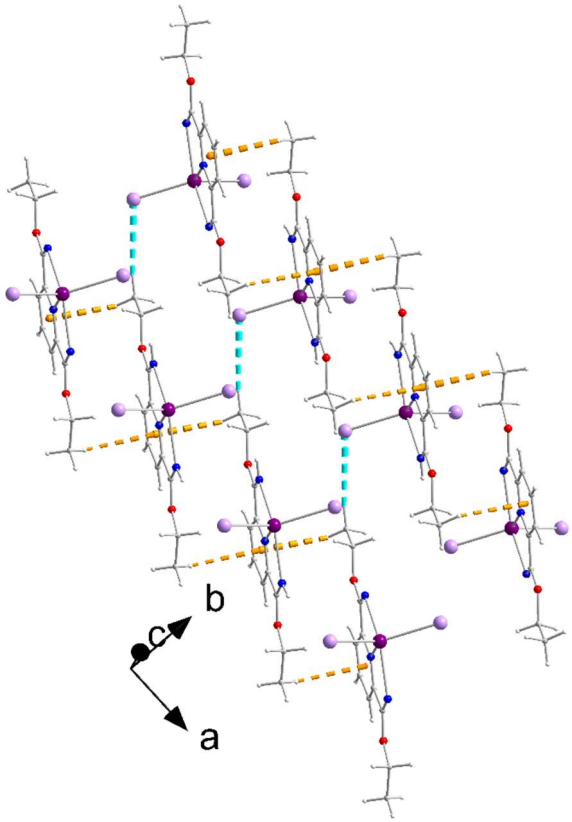

Figure S12. Formation of chains parallel to the *a* axis and layers parallel to the (001) crystallographic plane in the crystal structure of complex [CdI_2(L'H_2)] (**5**). The dashed thick orange line and the cyan lines represent the C(methyl)-H···π and C(methyl)-H···I interactions; see Table S2 for metric parameters.

Table S1. Hydrogen bonding interactions (Å, deg) in the crystal structure of [CdI_2(dapdoH_2)]·2EtOH (**4**·2EtOH).[a]

D—H···A	d(D···A)	d(D···H)	d(H···A)	<DHA	Symmetry Code of A
O1—H(O1)···O4	2.614 (4)	0.84	1.79	168	*x*,*y*,*z*
O2—H(O2)···O3	2.622 (3)	0.84	1.82	158	*x*,*y*,*z*
C4—H(C4)···I2	3.889 (3)	0.95	3.08	144	-*x*, -*y*+2, -*z*+2
C9—H_C(C9)···O2	3.440 (4)	0.98	2.54	152	-*x*+1, -*y*+1, -*z*+2

[a] For more details, see text. D= donor; A= acceptor.

Table S2. Hydrogen bonding interactions (Å, deg) in the crystal structure of [CdI_2(L'H_2)] (**5**).

D—H···A	d(D···A)	d(D···H)	d(H···A)	<DHA	Symmetry Code of A
C1—H_A(C1)···I1	4.143(6)	0.98	3.17	171	-*x*+1, -*y*+1, -*z*+1
C1—H_C(C1)···Cg1[a]	3.721(6)	0.98	2.95	136	*x*+1, *y*, *z*

[a] C_g1 is the centroid of the ring N2C4C5C6C5'C4' [(')= -*x*, -*y*+3/2, *z*]. D= donor; A= acceptor.