Supporting materials

Table S1 The C-O distances of the acylhydrazide ring moiety in the title compounds.

	1		**2**		**3**		**4**	**5**	
	left	**right**	**I**	**II**	**left**	**right**		**I**	**II**
d(C-O$^-$) (Å)	1.274(4)	1.265(4)	1.295(7)	1.296(7)	1.290(4)	1.286(5)	1.294(2)	1.344(3)	1.340(3)
d(C=O) (Å)	1.238(4)	1.247(4)	1.237(7)	1.279(7)	1.244(4)	1.333(4)	1.258(2)	1.239(3)	1.260 (3)

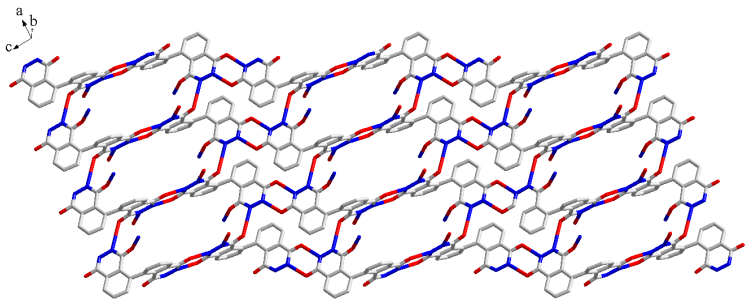


Fig. S1 A layer network of compound **8**, showing the difference from compound **3**.

Note for compound **8**: Fig. S1 illustrates a similar sheet network to that of compound **3**. The difference is (i) two acylamino groups outer isomerized into hydroxylimino groups, and they are involved in the formation of intermolecular hydrogen bonds; (ii) due to the absence of the water molecule, N or O forms the hydrogen bond to the other O or N from the neighboring layers, self-assembling compound **8** into a 3D supramolecular network.

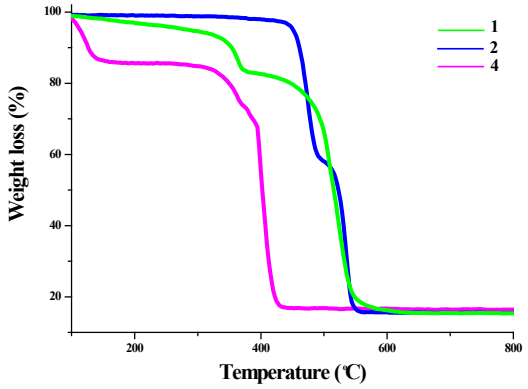


Fig. S2 The TG curve of compounds **1**, **2** and **4**.

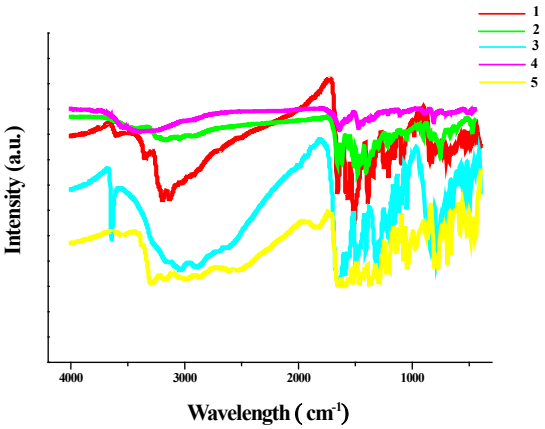
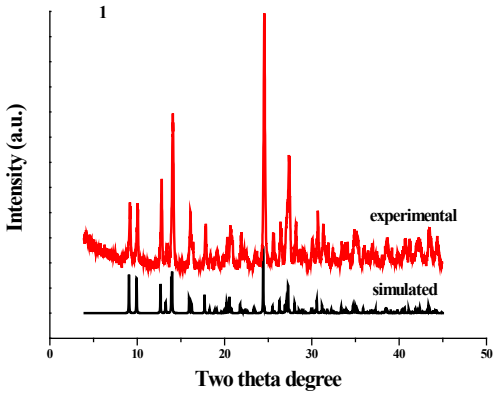
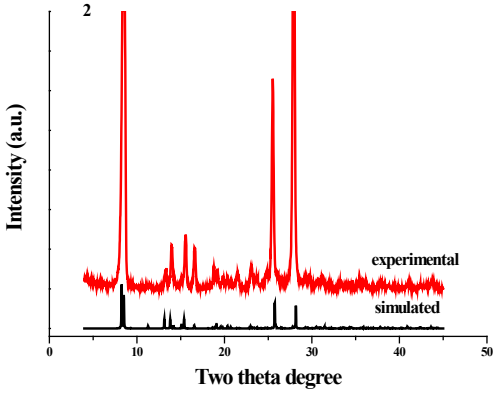


Fig. S3 The IR spectra of the title compounds.





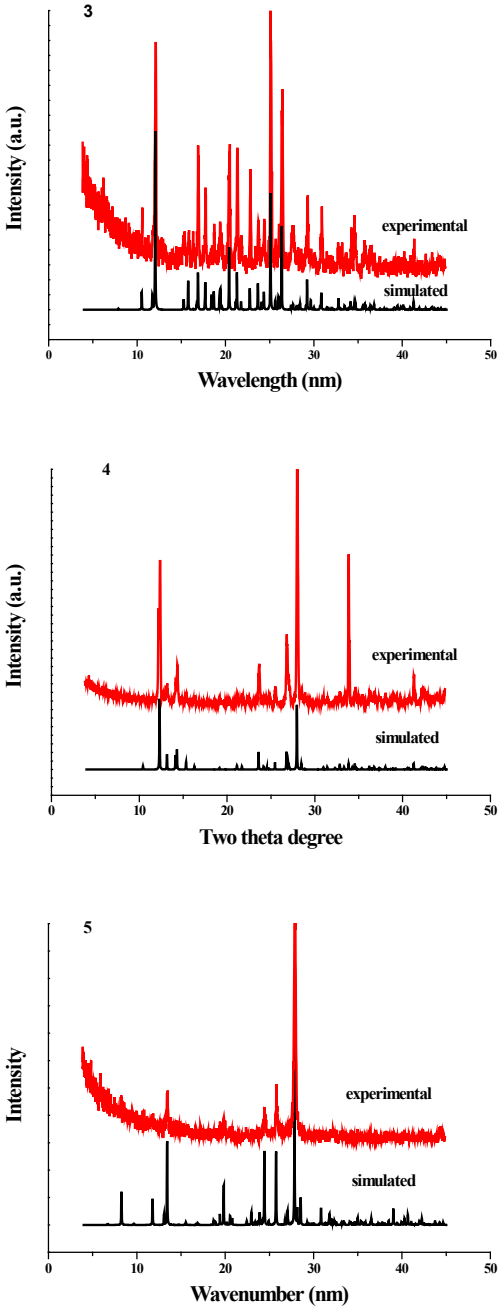


Fig. S4 The experimental (black) and simulated (red) powder XRD patterns for the title compounds.

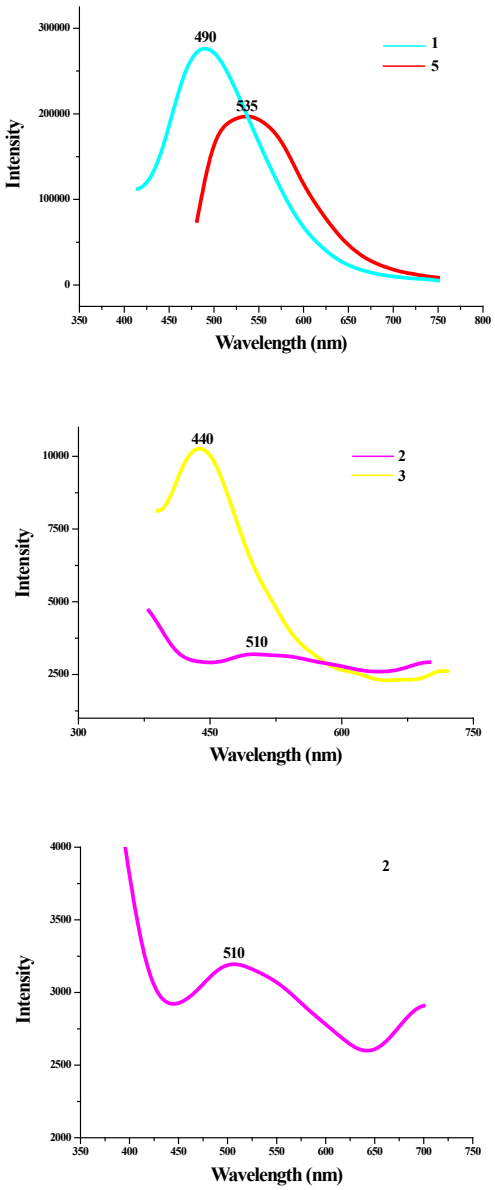
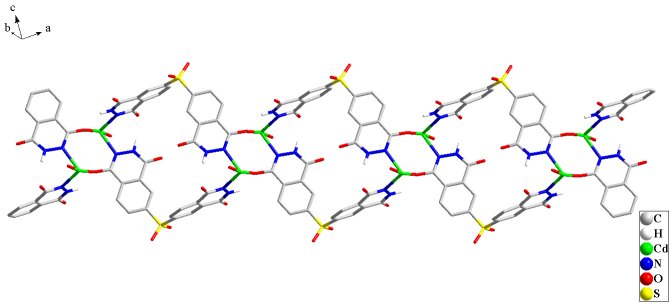


Fig. S5 The solid-state photoluminescence spectra of the title compounds.



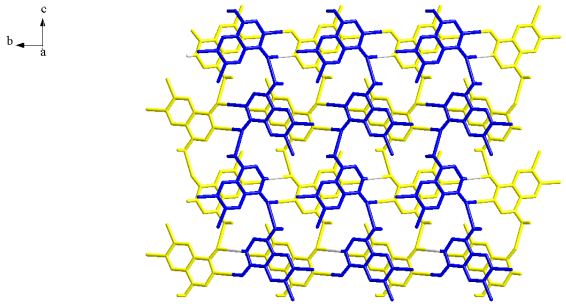


Fig. S6 The 1D chain structure of [Cd(sdpth)(phen)(H_2O)]·H_2O (above) and the 2D double-layer network of [dcpth] (below).

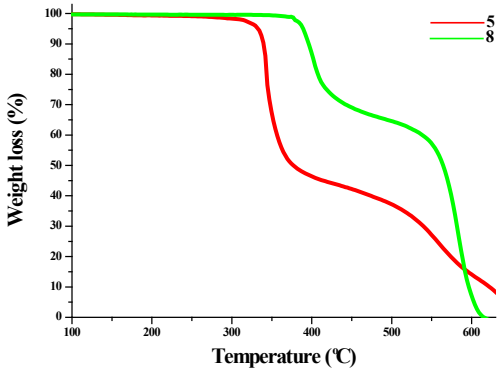


Fig. S7 The TG curves of compounds **5** and **8**, showing that the temperatures for the onset of decomposition for acylhydrazide molecules are *ca*. 300 ºC for **5** and *ca*. 370 ºC for **8**, respectively.