

Supporting materials

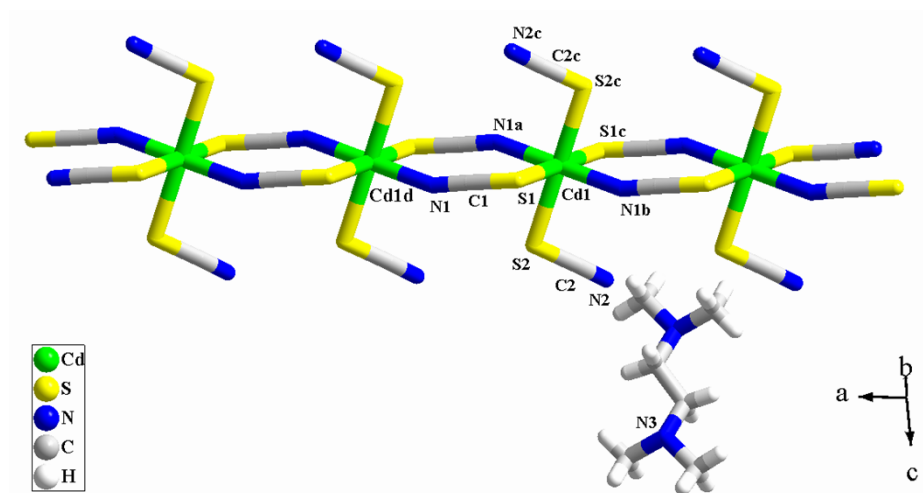


Fig. S1 1-D linear single-chain structure with $\text{H}_2(\text{tmen})^{2+}$ as templating agent in **1**.

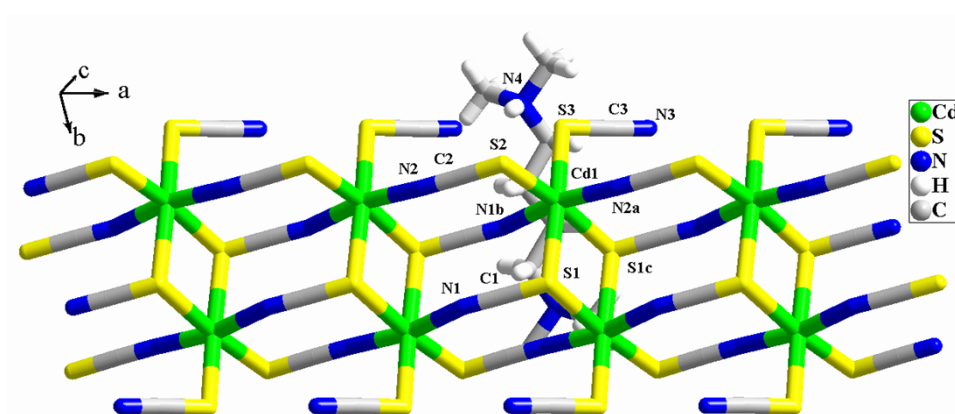


Fig. S2 1-D linear double-chain structure with $\text{H}_2(\text{tmiba})^{2+}$ as templating agent in **2**.

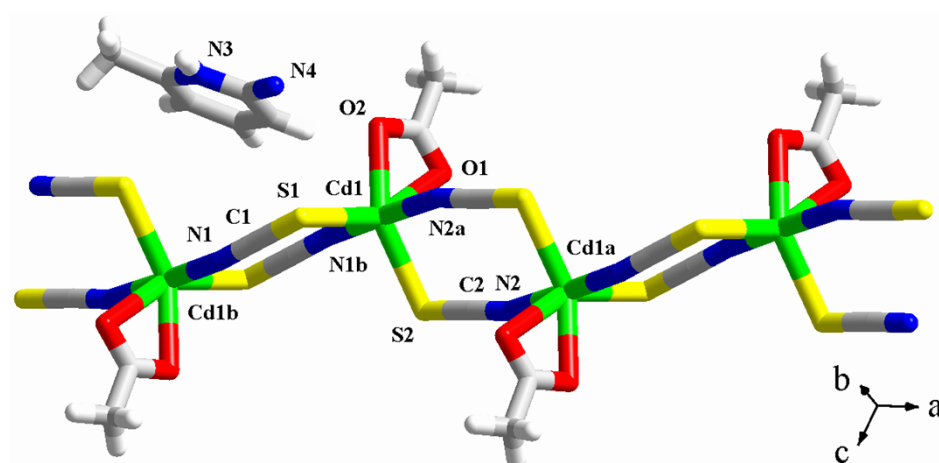


Fig. S3 1-D zigzag single-chain structure with H(amp)⁺ as templating agent in **4**.

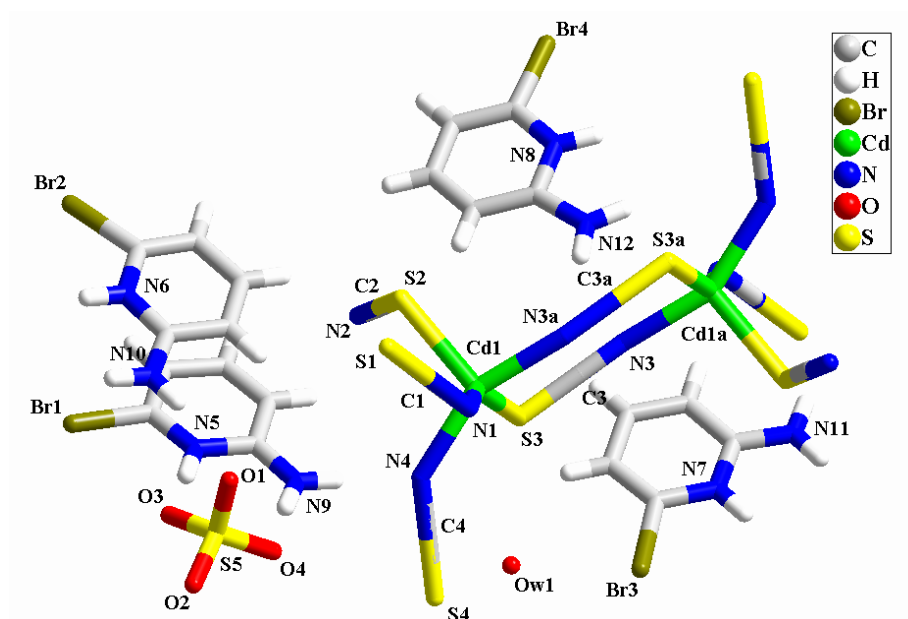


Fig. S4 Molecular structure of **5**.

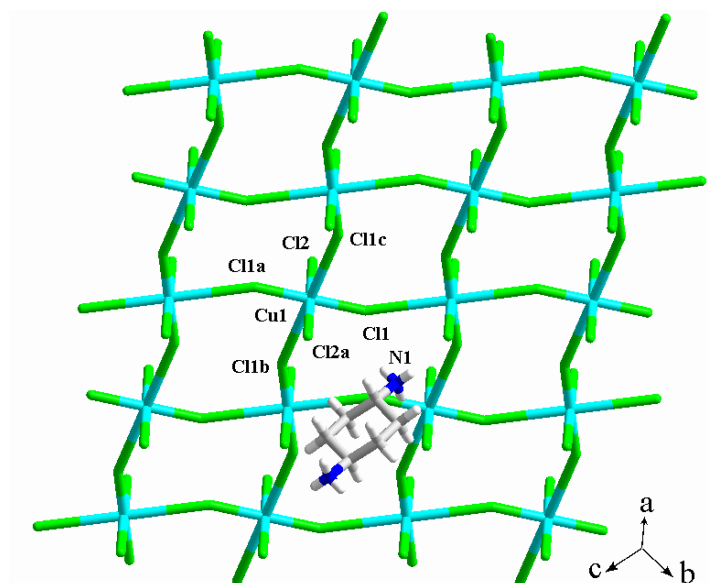
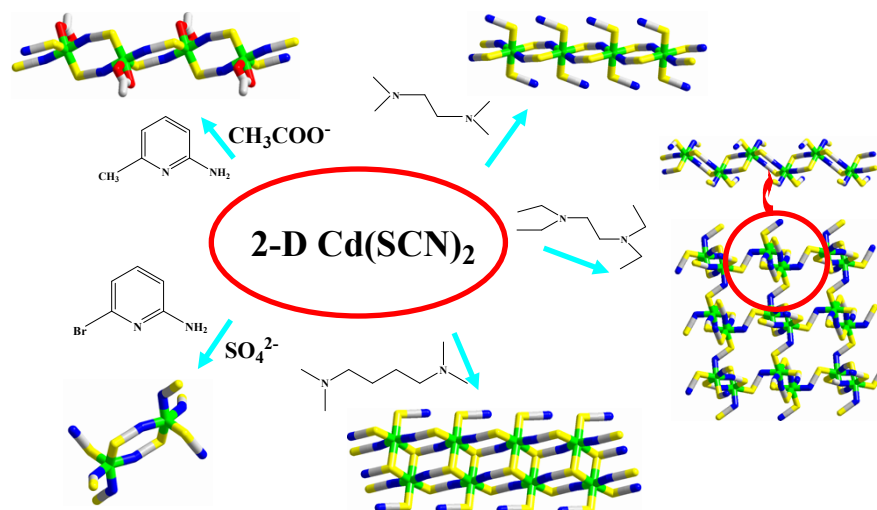
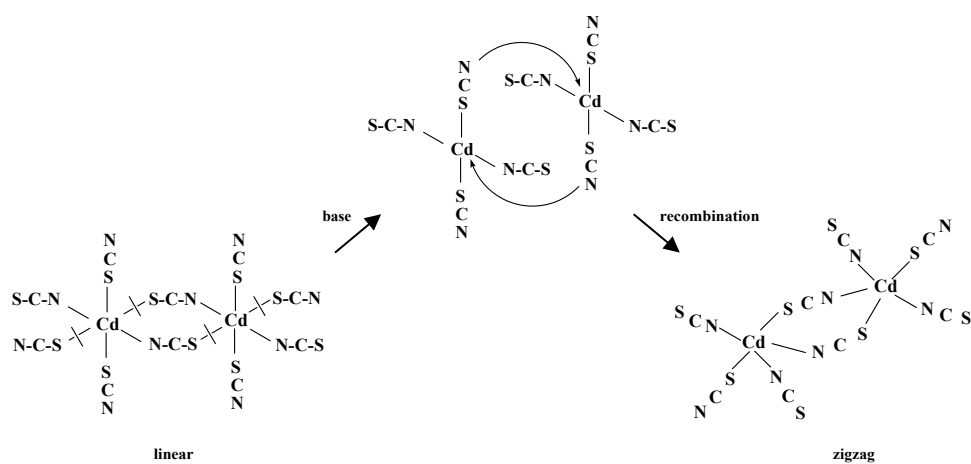


Fig. S5 2-D perovskite single-layer network with H₂(cha)²⁺ as templating agent in **6**.

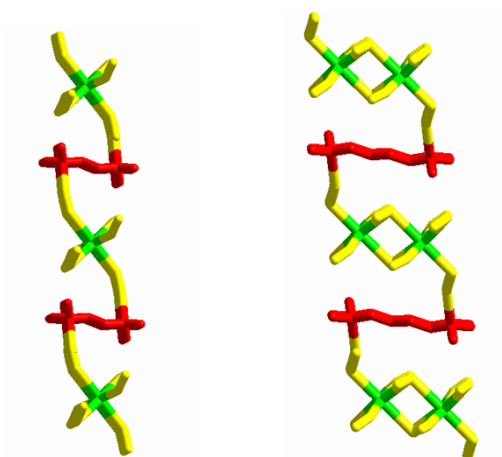


(a)



(b)

Scheme S1 Plots showing formation of thiocyanatocadmate frameworks in **1-5** (a) and recombination from linear chain to zigzag chain (b).



(a)

(b)

Fig. S6 Two supramolecular networks in **1** and **2**, showing how organic cation to control width of inorganic anion chain.

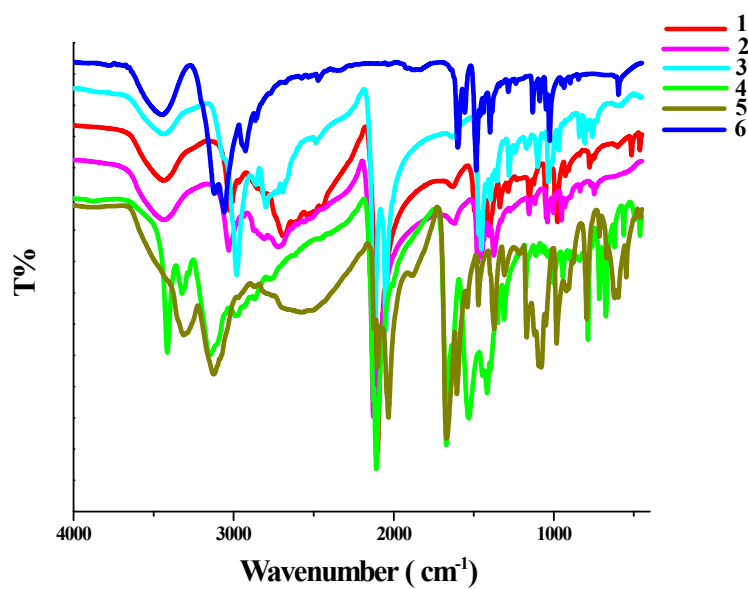


Fig. S7 IR spectra of 1-6.

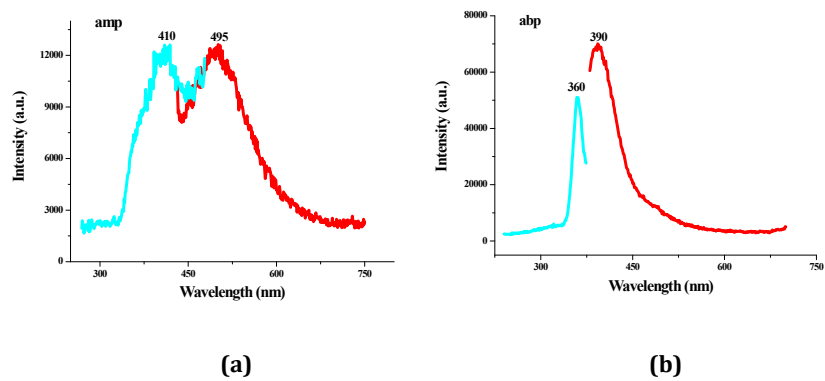


Fig. S8 Photoluminescence spectra of amp (a) and abp (b).

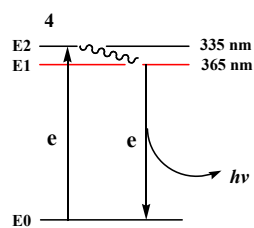


Fig. S9 Diagram of energy level of **4**.

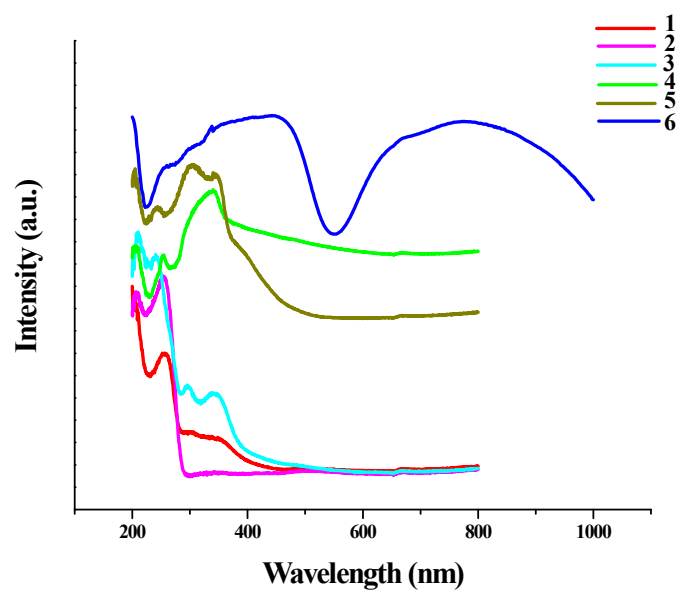
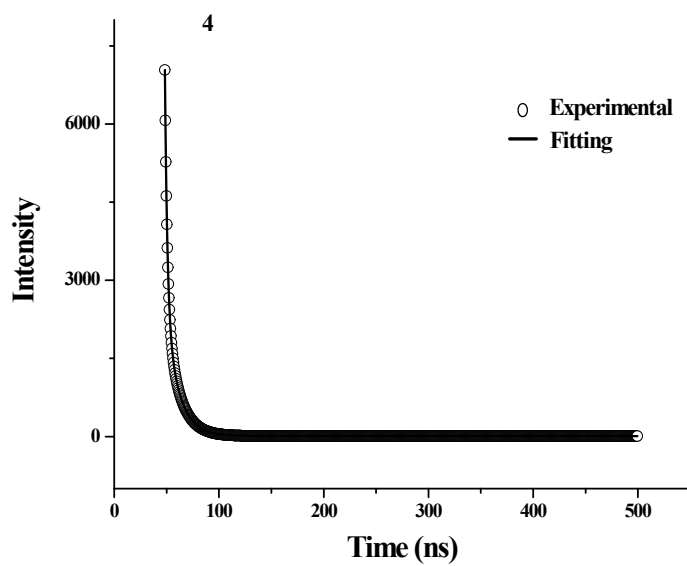


Fig. S10 Solid-state UV-vis spectra of title compounds.



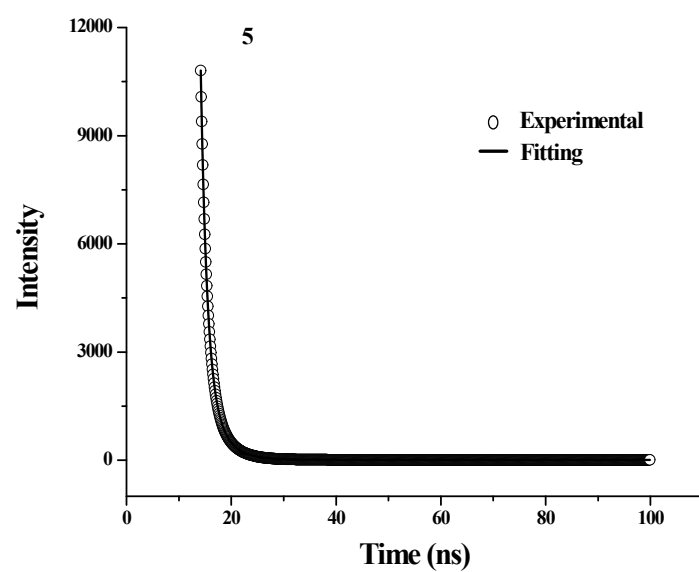


Fig. S11 Decay curves of **4** and **5**.