

Crystallographic Data for 1-3.

(1) Crystal data for $[\text{Ni}_8(\text{abo})_8(\text{Et}_2\text{O})_2(\text{MeOH})_2(\text{H}_2\text{O})]\cdot 2\text{Et}_2\text{O}$. $\text{C}_{130}\text{H}_{158}\text{N}_8\text{Ni}_8\text{O}_{23}$, $M = 2670.32$, orthorhombic, $a = 22.4470(6)$, $b = 55.1580(13)$, $c = 21.2010(5)$ Å, $\alpha = \beta = \gamma = 90^\circ$, $V = 26249.6(11)$ Å³, $T = 150$ K, space group $Fdd2$ (no. 43), $Z = 8$, 67387 reflections measured, 13364 unique ($R_{\text{int}} = 0.0565$) which were used in all calculations. The final $R(F_2)$ was 0.0551 (12122 data). Platon squeeze was applied to remove disordered Et_2O solvent molecules (2 per Ni_8 complex). The disordered ether in the centre of the wheel was treated as two components.

(2) Crystal data for $[\text{Ni}_6(\text{abo})_6(\text{aboH}_2)_2(\text{MeOH})_4]\cdot 2\text{Et}_2\text{O}$. $\text{C}_{124}\text{H}_{120}\text{N}_8\text{Ni}_6\text{O}_{22}$, $M = 2426.62$, triclinic, $a = 11.6358(4)$, $b = 16.1748(5)$, $c = 17.3392(6)$ Å, $\alpha = 112.085(2)^\circ$, $\beta = 105.280(2)^\circ$, $\gamma = 101.909(2)^\circ$, $V = 2741.43(18)$ Å³, $T = 150$ K, space group $P-1$ (no. 2), $Z = 1$, 32324 reflections measured, 14737 unique ($R_{\text{int}} = 0.0590$) which were used in all calculations. The final $R(F_2)$ was 0.0728 (8129 data).

(3) Crystal data for $[\text{NaNi}(\text{abo})(\text{aboH})(\text{MeOH})_3]$. $\text{C}_{31}\text{H}_{35}\text{N}_2\text{Na}_1\text{Ni}_1\text{O}_7$, $M = 629.33$, triclinic, $a = 13.3416(3)$, $b = 14.8361(3)$, $c = 15.1781(4)$ Å, $\alpha = 82.101(1)^\circ$, $\beta = 86.408(1)^\circ$, $\gamma = 82.228(1)^\circ$, $V = 2945.50(12)$ Å³, $T = 150$ K, space group $P-1$ (no. 2), $Z = 4$, 38427 reflections measured, 16043 unique ($R_{\text{int}} = 0.0390$) which were used in all calculations. The final $R(F_2)$ was 0.0474 (11490 data).

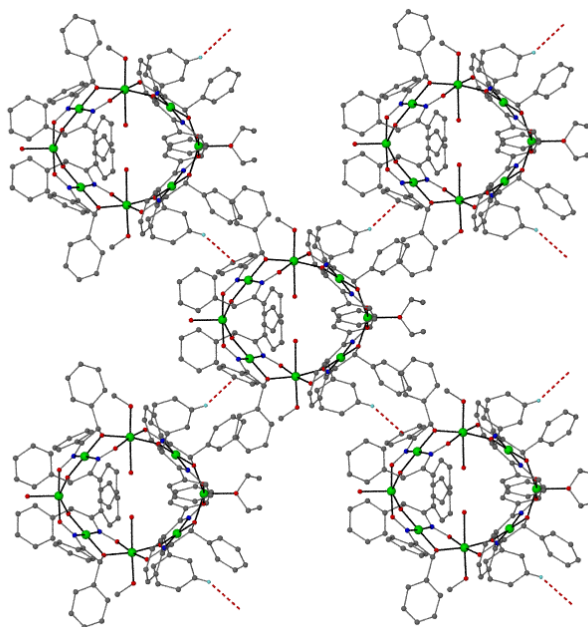


Fig. S1. Part of the (4,4)-grid layer in the ac plane made of H-bonded $[\text{Ni}_8]$ wheels. Most hydrogen and carbon atoms have been omitted for clarity.

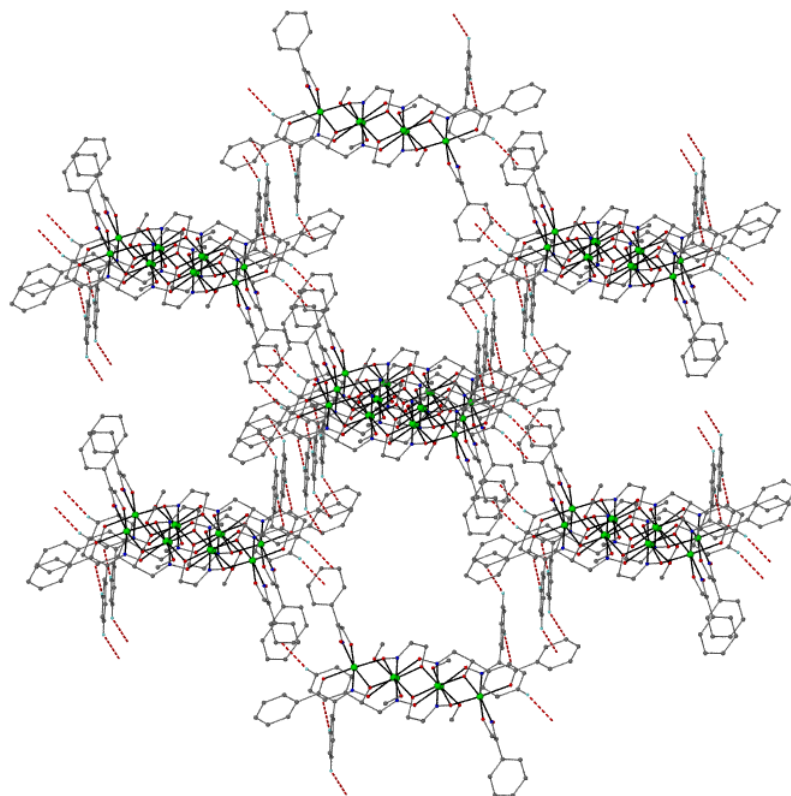


Fig. S2. The **pcu** network made of H-bonded [Ni₆] wheels. Most hydrogen and carbon atoms have been omitted for clarity.

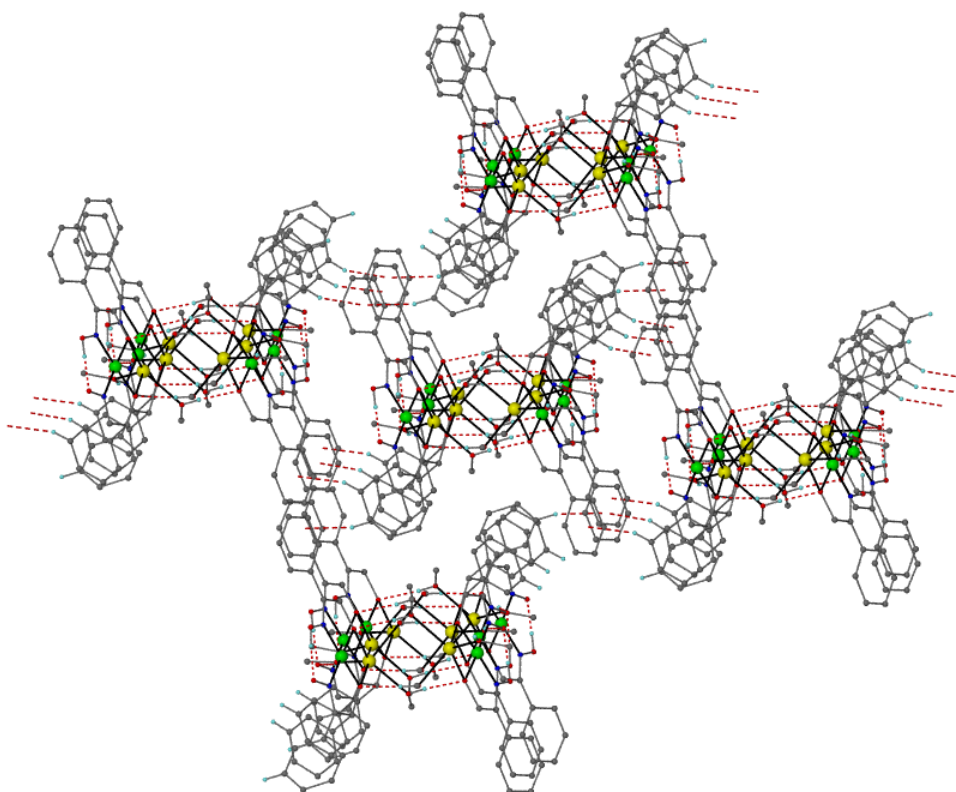


Fig. S3. The **pcu** rod-packing of the H-bonded tapes of (3)₂. Most hydrogen and carbon atoms have been omitted for clarity