SUPPLEMENTARY MATERIAL

Tuning Packing, Structural Flexibility and Porosity in 2D Metal-Organic Frameworks by Metal Node Choice

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1. Additional Structural Information for $\beta$-ML1 and ML1 ($M = \text{Co(II)}$ or Ni(II))

**Figure S1.** a) A view of the coordination sphere of the Co(II) and Ni(II) centres of (a) $\beta$-CoL1 and (b) $\beta$-NiL1 respectively. The hydrogen atoms of the water ligand could not be located in the difference map.

**Figure S2.** a) A view of the coordination sphere of the Co(II) centre of CoL1 highlighting the perpendicular relationship between the metal centres of two separate 2-D layers; b) the zig-zag conformation of the Co(II) metal centres viewed along the $b$ axis (identical for NiL1).
## 2. Unit Cell Parameters for the Solvent Exchanged Forms of NiL1

Table S1. Unit cell parameters for the various solvent exchanged forms of NiL1.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Treatment</th>
<th>$a$ (Å)</th>
<th>$b$ (Å)</th>
<th>$c$ (Å)</th>
<th>$V$ (Å$^3$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NiL1·$xS$</td>
<td>as-synthesised</td>
<td>11.2054(2)</td>
<td>20.1664(4)</td>
<td>13.3065(3)</td>
<td>3006.90(11)</td>
</tr>
<tr>
<td>NiL1·EtOH</td>
<td>ethanol exchanged</td>
<td>10.6799(2)</td>
<td>20.3654(3)</td>
<td>13.2980(3)</td>
<td>2892.32(9)</td>
</tr>
<tr>
<td>NiL1·MeOH</td>
<td>methanol exchanged</td>
<td>11.409(2)</td>
<td>20.322(4)</td>
<td>13.065(3)</td>
<td>3029.2(11)</td>
</tr>
<tr>
<td>NiL1·Tol</td>
<td>toluene exchanged</td>
<td>10.933(2)</td>
<td>20.875(4)</td>
<td>11.989(2)</td>
<td>2736.2(10)</td>
</tr>
</tbody>
</table>
3. Powder X-ray Diffraction

Figure S3. a) PXRD patterns of a) $\beta$-CoL1 simulated; b) $\beta$-CoL1 as-synthesised; c) $\beta$-NiL1 simulated; d) $\beta$-NiL1 as-synthesised.

Figure S4. a) PXRD patterns of a) CoL1 simulated; b) CoL1 as-synthesised; c) NiL1 simulated; d) NiL1 as-synthesised.
**Figure S5.** a) PXRD patterns of a) NiL1 as-synthesised; b) NiL1·xCH₃OH heated to 100 °C; c) CoL1 treated at 260 °C; d) NiL1 treated at 260 °C.
4. Gas Adsorption

Figure S6. a) $\text{N}_2$ isotherm of NiL1-\textit{ac} measured at 77 K. Filled and empty symbols represent the adsorption and desorption processes respectively.

Figure S7. a) 293 K CO$_2$ and $\text{N}_2$ isotherms of NiL1-\textit{ac}. Filled and empty symbols represent the adsorption and desorption processes respectively.