**Supplementary Information**

**High and selective CO\textsubscript{2} uptake in a nitrogen-rich pillar-layered metal organic framework**

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**Material synthesis**: Co(Imda) (4, 4’-bpy) was synthesized referring to the method of Li et al.\textsuperscript{13} A mixture of NaOH (1.50 mmol) in H\textsubscript{2}O (5.0 ml), 4, 5-imidazole dicarboxylic acid (1.0 mmol) and 4, 4’-bipyridyl (1.0 mmol) was added to an aqueous solution (5.0 ml) of Co(NO\textsubscript{3})\textsubscript{2}•6H\textsubscript{2}O (1.5 mmol) and stirred. The resulting solution was kept statically under autogenous pressure at 180 °C for 3 days in a teflon-lined autoclave. The resultant solid was isolated by filtration and washed with deionized water.

**Characterization methods**: Powder X-ray powder diffraction (XRD) measurement was conducted using CuK\textalpha\ (\lambda=1.54 Å) radiation on a Rigaku diffractometer. The N\textsubscript{2} adsorption-desorption measurements were carried out on a BEL adsorption instrument (BELsorp(II)-Max) at 77 K. The surface areas of the samples were calculated using the Brunauer–Emmett–Teller (BET) method. Thermogravimetric analysis (TGA) was carried out on a SCINCO thermal gravimeter S-1000. Scanning electron microscopy (SEM) was carried out using a Hitachi S-4200 instrument to observe particle morphologies.

**CO\textsubscript{2} or N\textsubscript{2} adsorption-desorption measurements**: Equilibrium CO\textsubscript{2} and N\textsubscript{2} adsorption isotherms at 298 K were obtained using a BEL adsorption instrument (BELsorp(II)-mini) using ultra high purity gases (U-Sung, 99.999%). The samples were pretreated under high vacuum at 150 °C overnight before the measurement.
The TGA unit was used to perform the cyclic CO₂ adsorption-desorption runs. High purity CO₂ (99.999%, U-Sung) and 15% CO₂ (85% N₂ as balance gas, U-Sung) were used as adsorption gases, while argon (Ultra high purity, 99.999%, U-Sung) was used as purge gas in the desorption process. The feed gas flow rate was maintained to 30 mL/min using a mass flow controller. Both adsorption and desorption experiments were carried out at 298 K.

**CO₂/N₂ selectivity calculation:** CO₂/N₂ selectivity was calculated using Ideal Adsorbed Solution Theory (IAST) method [19,20]. The absolute CO₂ loading was fitted with a dual–site Langmuir model, while the N₂ loading was fitted with a single–site Langmuir model. Both adjusted R² values of fitting exceed 0.9998. The single–site Langmuir model can be defined as,

\[
q = q_{sat}bp/(1+bp)
\]

The dual–site Langmuir model can be defined as,

\[
q = q_A+q_B = q_{sat,A}b_Ap/(1+b_Ap) + q_{sat,B}b_Bp/(1+b_Bp)
\]

Where, \(q\) is molar loading of adsorbate; \(q_{sat}\) is saturation loading; \(b\) is parameter in the pure component Langmuir adsorption isotherm, A and B is referring to two different sites. The IAST selectivity for the CO₂:N₂ (0.15:0.85) gas mixture was calculated using following equation,

\[
S = (q_1/q_2)/(p_1/p_2)
\]

Where, \(S\) is the selectivity factor, \(q_1\) and \(q_2\) represent the adsorbed amount of CO₂ and N₂, and \(p_1\) and \(p_2\) represents the partial pressure of CO₂ and N₂.
**Fig. S1:** A comparison of XRD patterns of 1) Co(Imda) (4, 4’-bpy), 2) CoO, and 3) Co(OH)$_2$. 
**Fig. S2:** CO$_2$ and N$_2$ adsorption isotherms were fitted by dual-site Langmuir model and single-site Langmuir model respectively for IAST CO$_2$/N$_2$ selectivity calculation of Co(Imda) (4, 4’-bpy).