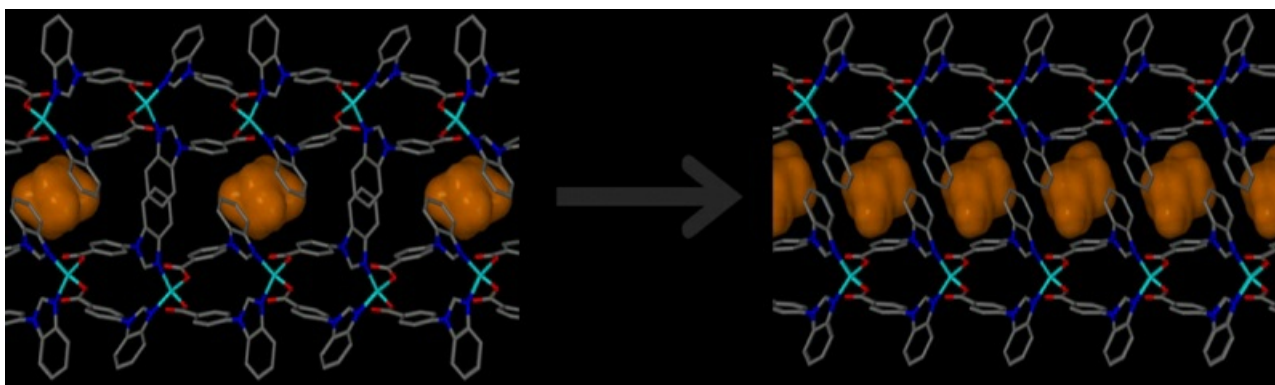


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A two-dimensional (2-D) coordination polymer $\{[\text{Zn}(\text{L})_4 \cdot \text{DMF} \cdot \text{H}_2\text{O}]_n\}$, (1), prepared by the reaction of 4-(1H-benzo[d]imidazol-1-yl)benzoic acid with $\text{Zn}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ under solvothermal conditions, can adapt its structure during supercritical carbon dioxide (CO_2) guest-exchange and CO_2 sorption. The 2-D layers of the coordination polymer are associated by weak interactions that enable the framework to undergo guest-induced expansion and contraction. At high pressure, additional space is created between adjacent layers resulting in a significant increase in the CO_2 sorption capacity (Figure 1). Moreover, the structural changes and gas uptake by the host occur preferentially for CO_2 at 25 °C while nitrogen (N_2) and methane (CH_4) are not adsorbed under the same conditions. Pressure differential scanning calorimetry (PDSC) and in situ single-crystal X-ray diffraction (SCD) experiments under gas loading were carried out to obtain insight into the dynamic behaviour of the framework.

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