Title: Supplementary Video 1.

**Description:** In situ optical microscopy movie of ELM-11 particles during the introduction of CO<sub>2</sub> at 0.8 kPa s<sup>-1</sup> and ~297 K.

Title: Supplementary Video 2.

**Description:** The same video as Supplementary Video 1, but in each frame, the pixels that changed from the first frame (0 s) are coloured red.

Title: Supplementary Video 3.

**Description:** Schematic movement of the structural transition mechanism of  $CO_2$  gate adsorption on ELM-11.

Title: Supplementary Video 4.

**Description:** In situ optical microscopy movie of MIL-53(Al) particles during the introduction of CO<sub>2</sub> at 0.8 kPa s<sup>-1</sup> and ~297 K.

Title: Supplementary Video 5.

**Description:** The same video as Supplementary Video 4, but in each frame, the pixels that changed from the first frame (0 s) are coloured red.

Title: Supplementary Video 6.

**Description:** In situ optical microscopy movie of CuFB particles during the introduction of CO<sub>2</sub> at 0.8 kPa s<sup>-1</sup> and ~297 K.

Title: Supplementary Video 7.

**Description:** The same video as Supplementary Video 6, but in each frame, the pixels that changed from the first frame (0 s) are coloured red.