Electronic supplementary information (ESI)

## Amorphous nano-structured silicas for high-performance carbon dioxide confinement

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#### 1. Additional experimental section

#### 1.1. Raman spectroscopy

Raman spectra of adsorbed CO<sub>2</sub> were measured using an NRS-2100 spectrometer (JASCO) with a 514.5-nm laser (laser power 5 mW, exposure time 900 s  $\times$  2) and a resolution of 2 cm<sup>-1</sup>. Indene under inert atmosphere or an artificial diamond<sup>S1</sup> (Tokyo Progress System, Raman shift 1332 cm<sup>-1</sup>) was used for the wavenumber calibration. The spectra of CO<sub>2</sub>-condined adsorbents were recorded under ambient conditions, while the measurement of solid CO<sub>2</sub> was performed at -170 °C with a cooled cell (Linkam, model THMS600/LK-600PM/L-600A).

#### 1.2. Transmission electron microscopy (TEM)

TEM measurements were performed on a JEM-2010F (JEOL) equipped with a field emission gun (operated at 200 kV). The S-400 and S-800 samples were pretreared at 400 °C for 3 h in a stream of dry air (dew point ca. -40 °C) prior to the measurements.

#### 1.3. N<sub>2</sub> and Ar adsorption measurements

Surface areas and pore structures were characterized by measuring the  $N_2$  and Ar adsorption–desorption isotherms using a BELSORP-mini (BEL Japan) for  $N_2$  and an Autosorb-1-MPa analyzer (Quantachrome) for Ar, respectively. Before the measurements, the samples were treated at 400 °C for 3 h in a stream of dry air (dew point ca. –40 °C), and subsequently at 120 °C for 3 h under  $N_2$  flow on the BELSORP-mini apparatus and 200 °C for 3 h under vacuum (<10<sup>-5</sup> Pa) on the Quantachrome apparatus, respectively.

### 1.4. References

S1 S. A. Solin and A. K. Ramdas, *Phys. Rev. B*, 1970, 1, 1687.



**Fig. S1** Comparison of the Raman spectra of adsorbed CO<sub>2</sub> between S-Kan and zeolite 13X. For experimental details, see "**Notes and references** section§" and "**Additional experimental section** in ESI". Indene under inert atmosphere was used for the wavenumber calibration.



Fig. S2 Saito–Foley pore-size distributions of S-400 and S-800.



Fig. S3 Pore-size distributions of S-400 and S-800 determined by the non-local density functional theory (NL-DFT).



**Fig. S4** TEM images of S-400 (left) and S-800 (right). Note that the mean first-order-particle size was larger for S-800 than for S-400 due to the difference in calcination temperature that determines the extent of sintering.