

Supplementary Information

Double-site Langmuir (DL) isotherm model

In order to better describe the adsorption behavior and adsorption characteristics of the aerogel for CO₂ (Ar, N₂), the isotherm curves were fitted with the double-site Langmuir (DL) isotherm model (Eq. 1) and the Langmuir-Freundlich (LF) isotherm model (Eq. 2), respectively.

$$q = \frac{q_c \times k_c \times p}{1 + k_c \times p} + \frac{q_i \times k_i \times p}{1 + k_i \times p} \quad (1)$$

where q_c (mmol/g), q_i (mmol/g), k_c (kPa⁻¹), and k_i (kPa⁻¹) are the parameters of the double-site Langmuir model (q_c and q_i are the saturation capacities of sites c and i; k_c and k_i are the corresponding adsorption equilibrium constants reflecting the affinity coefficients of sites c and i; The subscripts c and i are on behalf of CO₂ and N₂, P corresponds to an equilibrium pressure.

Langmuir-Freundlich (LF) isotherm model

$$q = \frac{q_i \times k_i \times p^n}{1 + k_i \times p^n} \quad (2)$$

where q_i (mmol/g), k_i (kPa⁻¹), and n are the parameters of the Langmuir-Freundlich model (q_i is the saturation capacity; k_i is the corresponding adsorption equilibrium constant reflecting the affinity coefficient; n represents the deviation from the ideal homogeneous surface), P corresponds to an equilibrium pressure.

Ideal adsorbed solution theory (IAST)

The ideal adsorption solution theory (IAST) was used to predict binary adsorption isotherms using the adsorption isotherms of pure CO₂, and N₂. The

selectivity of the gas mixture was calculated. The DSL-IAST model was used to predict the selectivity of the adsorbent for a certain molar fraction of CO₂ and N₂ gas mixture.

$$S = \frac{X_i/X_j}{Y_i/Y_j} \quad (3)$$

Where x and y are the mole fraction of adsorbate in the adsorption phase and gas phase respectively. The subscripts i and j are on behalf of CO₂ and N₂.

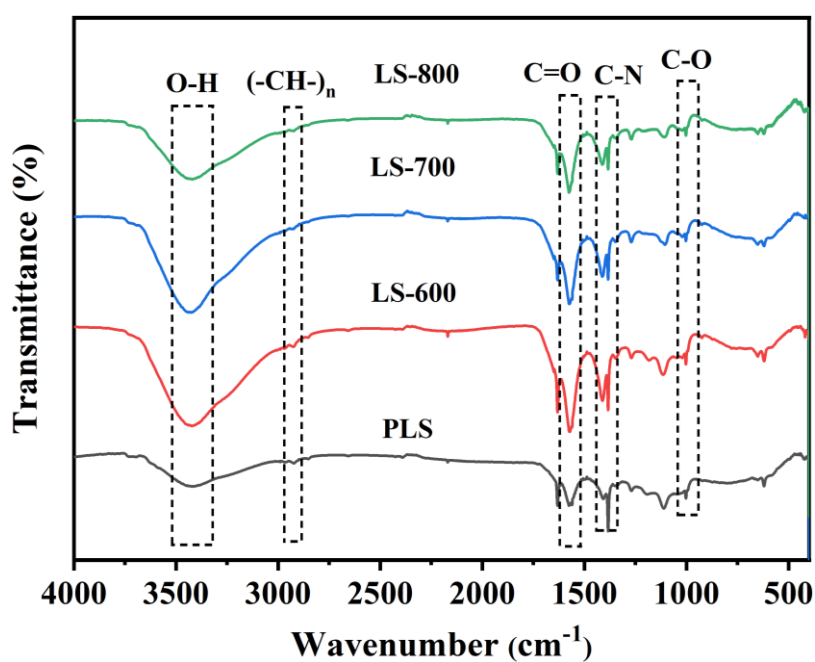


Fig. S1. FT-IR spectra of PLS, LS-600, LS-700 and LS-800.

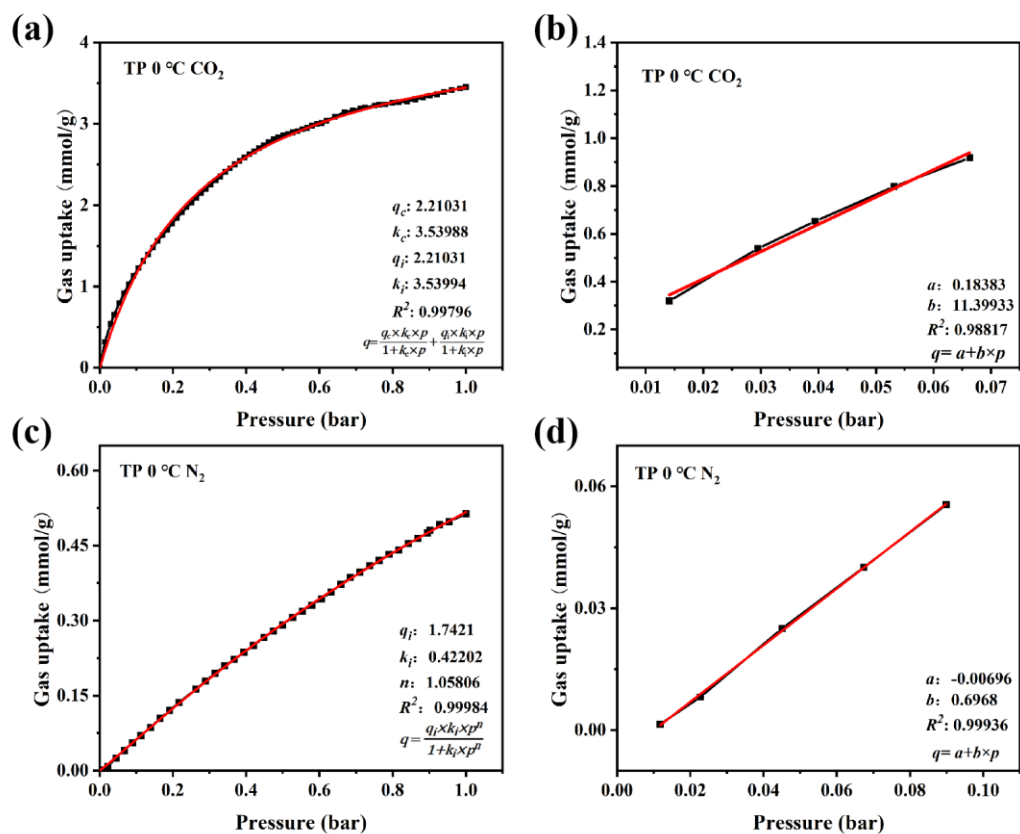


Fig. S2. Non-linear curve fitting and initial slope calculation for TP gas adsorption at 0°C (linear curve fitting at low pressure).

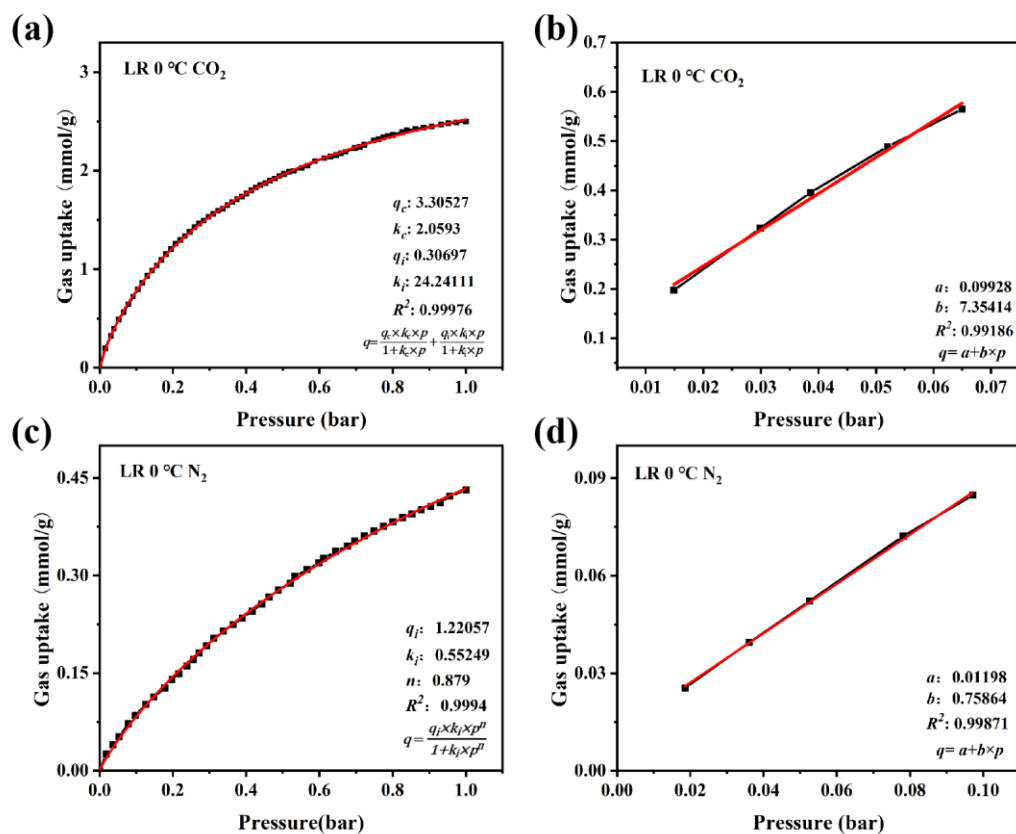


Fig. S3. Non-linear curve fitting and initial slope calculation for LR gas adsorption at 0°C (linear curve fitting at low pressure).

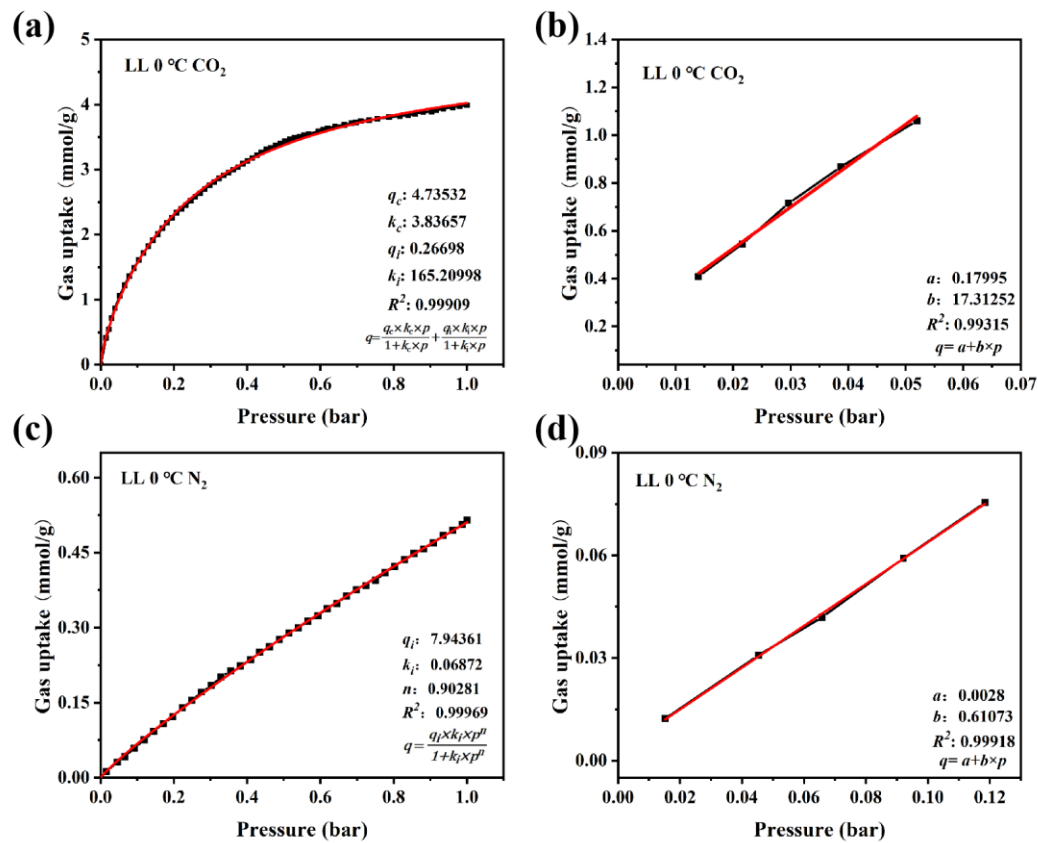


Fig. S4. Non-linear curve fitting and initial slope calculation for LL gas adsorption at 0°C (linear curve fitting at low pressure).

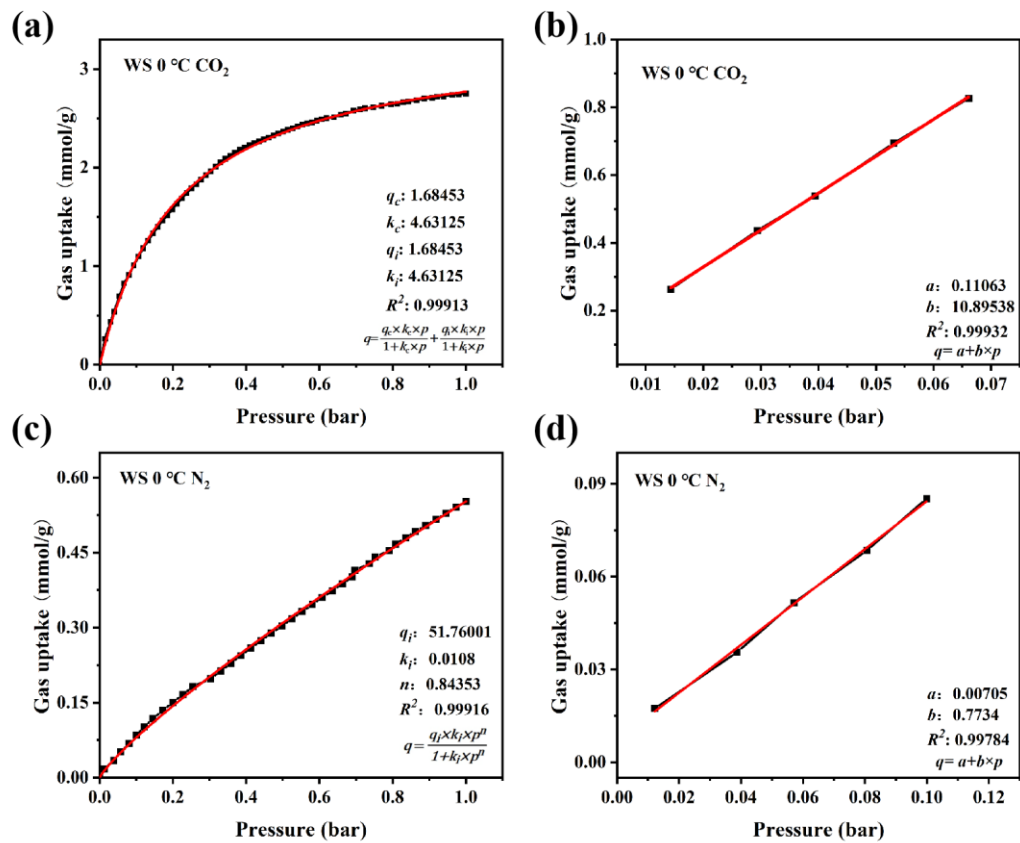


Fig. S5. Non-linear curve fitting and initial slope calculation for WS gas adsorption at 0°C (linear curve fitting at low pressure).