Electronic Supplementary Information

One-Pot Facile Synthesis of Mesoporous Carbon and N-doped Carbon for CO_2 Capture by a Novel Melting-Assisted Solvent-Free Method

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Section I: Small angle XRD pattern of NMC-250D8H

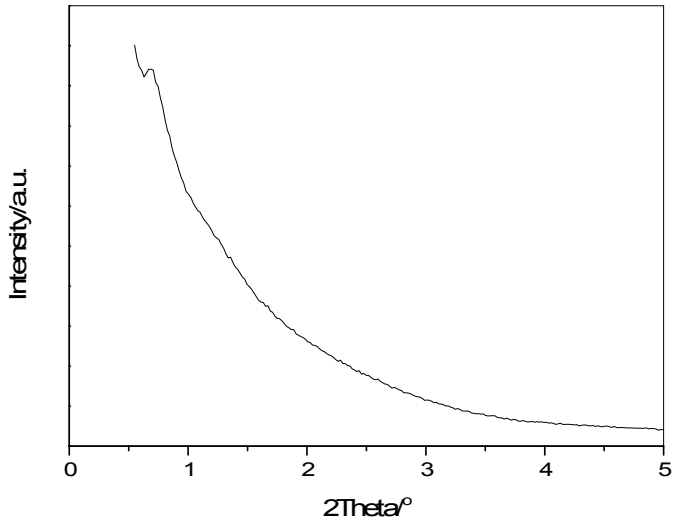

Fig.S1 Small angle XRD pattern of NMC-250D8H

Compared with that of MC samples, the small angle XRD pattern of NMC-250D8H sample does not show any diffraction peaks, indicating the mesostructure of NMC-250D8H is disordered.

Section II: Calculation of adsorption heat of CO_2 on MC-250D8H and NMC-250D8H

The enthalpy of CO_2 adsorption on MC-250D8H and NMC-250D8H was calculated using a modified version of the Clausius-Clapeyron equation as shown in Eq.(1):

$$\ln(\frac{p_1}{p_2}) = \Delta H_{ads} \times \frac{T_2 - T_1}{R \times T_1 \times T_2} \quad (1)$$

where ΔH_{ads} is the adsorption heat at specific loading (kJ/mol), *T* is the temperature (K), *p* is the pressure (bar), and *R* is the universal gas constant.

The isotherms of CO_2 at 0 and 25 ^{o}C on MC-250D8H and NMC-

250D8H (Fig.S2) were used to calculate the heat of adsorption. First, the Dual-Site Langmuir (DSL) model as defined by Eq. (2) was used to fit the isotherms:

$$n = q_{sat,A} \times \frac{b_A \times p}{1 + b_A \times p} + q_{sat,B} \times \frac{b_B \times p}{1 + b_B \times p} \quad (2)$$

where n is the adsorbed amount, q_{sat} is the saturation loading for site A and B, b is the Langmuir parameter associated with site A and B, and p is the pressure. The fitted parameters are displayed in Tal.S1.

Then, at a given adsorption amount, the equilibrium pressure at 0 and 25 ^{o}C can be solved out from the obtained fitted DSL equations. Finally, heats of adsorption can be obtained by adding these pressure values into Equ.(1). The calculated adsorption heats of CO_2 on MC-250D8H and NMC-250D8H at different adsorption amounts are listed in Tal.S2.

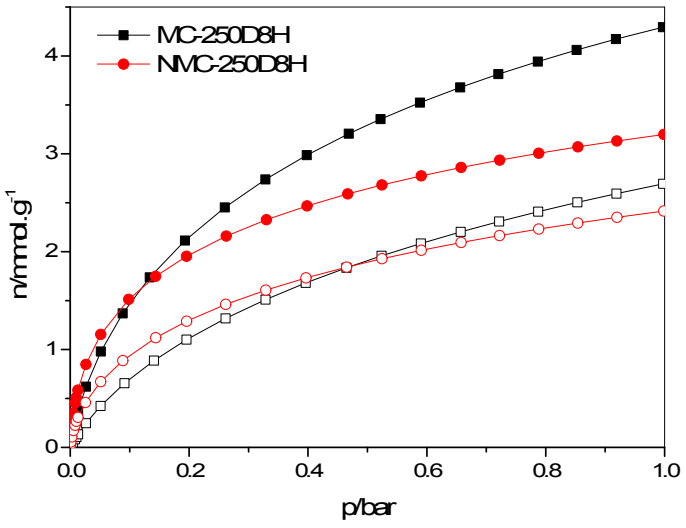

Fig.S2 CO_2 isotherms at 0 and 25 ^{o}C on MC-250D8H and NMC-250D8H

(Solid: 0 °C; open: 25 °C)

Tab.S1 Fitted parameters of Dual-Site Langmuir model for CO_2 isotherms on MC-250D8H and NMC-250D8H at 0 and 25 °C

Sample		$q_{sat,A}$	b_A	$q_{sat,B}$	b_B
MC-250D8H	0 °C	1.263	20.774	5.464	1.294
	25 °C	1.263	6.210	5.464	0.420
NMC-250D8H	0 °C	1.096	70.529	3.062	2.167
	25 °C	1.096	21.936	3.062	0.808

Tab.S2 The calculated adsorption heats of CO_2 on MC-250D8H and NMC-250D8H at different adsorption loadings

n	$\Delta H_{ads,MC-250D8H}$	$\Delta H_{ads,NMC-250D8H}$	n	$\Delta H_{ads,MC-250D8H}$	$\Delta H_{ads,MC-250D8H}$
mmol/g	kJ/mmol	kJ/mmol	mmol/g	kJ/mmol	kJ/mmol
0.01	31.17	32.17	2	27.13	30.89
0.2	31.02	32.08	2.2	27.01	30.81
0.4	30.78	31.96	2.4	26.94	30.75
0.6	30.42	31.83	2.6	26.88	30.70
0.8	29.90	31.68	2.8	26.84	30.66
1	29.23	31.52	3	26.81	30.63
1.2	28.54	31.37	3.2	26.79	30.60
1.4	27.97	31.22	3.4	26.77	30.58
1.6	27.57	31.09	3.6	26.75	30.56
1.8	27.30	30.98	3.8	26.74	30.54

Section III: Calculation of adsorption selectivity by Ideal Adsorption Solution Theory (IAST)

IAST has been widely used to predict the mixture adsorption equilibrium by using isotherms of pure gas (*Chem. Commun., 2014, 50, 6894*; *Energy Environ. Sci., 2011, 4, 2177; J. Mater. Chem., 2012, 22, 10274*). For the IAST application, the main requirement to be fulfilled is the availability of high quality adsorption data for each of the component, and excellent curve fitting model for such data (*Langmuir,*

2007, 23, 6431; Langmuir, 2008, 24, 8592).

In the current work, Dual-Site Langmuir (DSF) model was used to fit the pure CO_2 and N_2 isotherms because of its good fitting and simplicity as expressed by Eq.(2). The fitted equations for CO_2 and N_2 on MC-250D8H and NMC-250D8H are shown in Fig.S3 and Fig.S4, respectively.

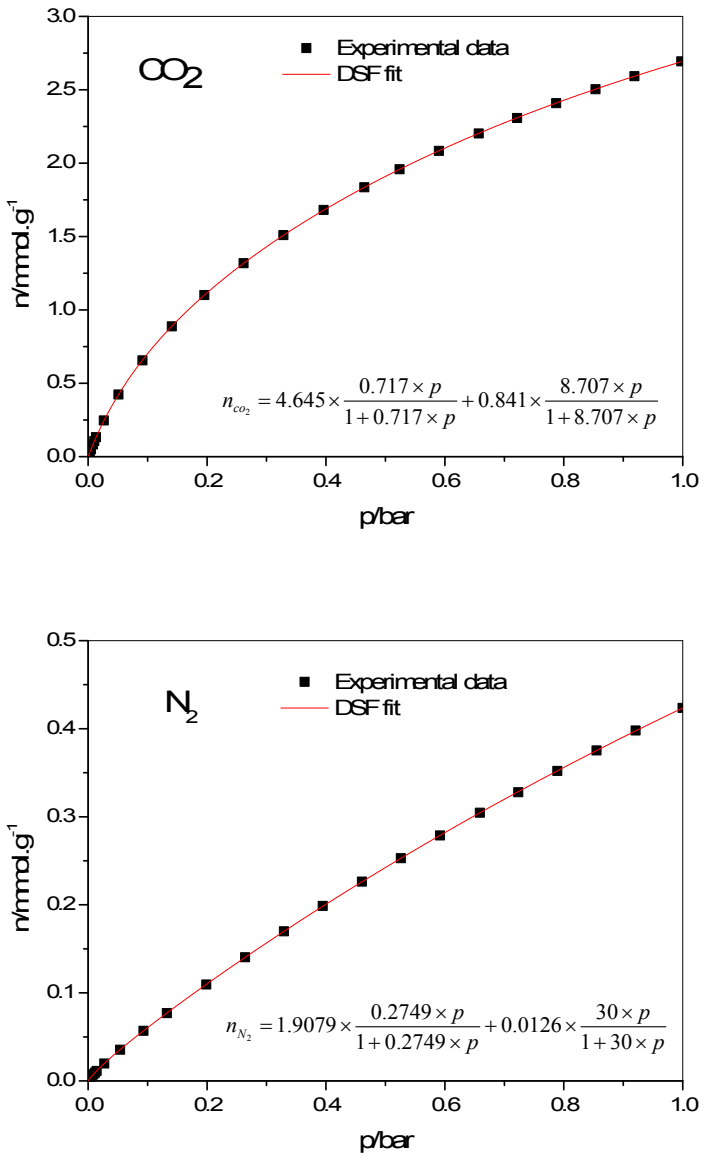

Fig.S3 CO_2 and N_2 isotherms for MC-250D8H at 298 K (black circles) and corresponding DSF fits (solid line)

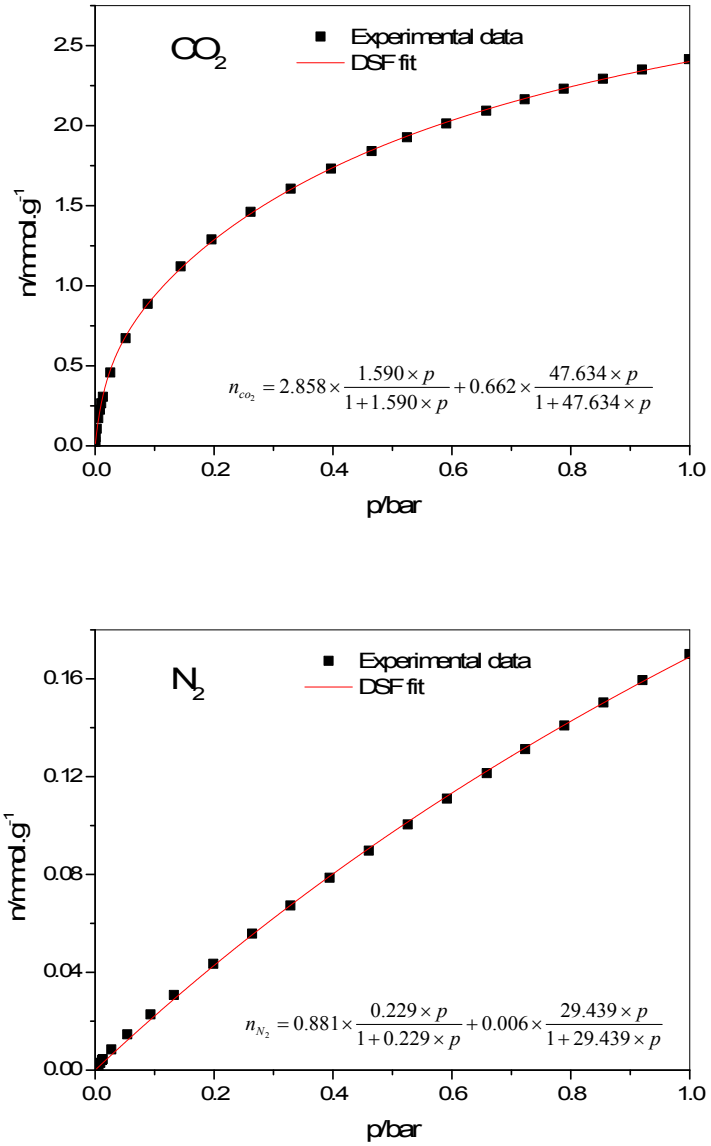

Fig.S4 CO_2 and N_2 isotherms for NMC-250D8H at 298 K (black circles) and corresponding DSF fits (solid line)

Then, the so-called spreading pressure as defined by Eq. (3) for CO_2 and N_2 in a mixture with fixed concentration and pressure can be obtained from the pure component adsorption isotherms, where *n* is the experimentally measured gas uptake as a function of pressure *p*, *A* is the available surface area (assumed to be the same for all gases), *i* and *j* represent different gases, π^o is the so-called spreading pressure, and P^o

is the equilibrium pressure of the single-component gas corresponding to π^o

$$\pi_i^o(P_i^o) = \frac{RT}{A}\int_{p=0}^{P_i^o}\frac{n_i^o(p)}{p}dp \quad (3)$$

At equilibrium, Eq.(4) need to be fulfilled, together with Eq.(5) and Eq.(6), adsorption selectivity then can be calculated by Eq.(7).

$$Py_i = P_i^o x_i \quad (4)$$

$$\sum x_i = 1 \quad (5)$$

$$\sum y_i = 1 \quad (6)$$

$$S_{CO_2,N_2} = \frac{x_{CO_2}/y_{CO_2}}{x_{N_2}/y_{N_2}} = \frac{x_{CO_2}}{y_{CO_2}} \times \frac{1-y_{CO_2}}{1-x_{CO_2}} \quad (7)$$

where x_i and y_i are the adsorbed and gas phase molar fractions of gas i, respectively.

At 298 K and 1 bar, the calculated equilibrium CO_2 molar fractions and selectivities for MC-250D8H and NMC-250D8H at different CO_2 concentrations in N_2 are listed in Tal.S3 and Tal.S4.

Tab.S3 The calculated equilibrium CO_2 molar fraction in adsorbed phase and corresponding CO_2/N_2 selectivity for MC-250D8H at different CO_2 concentrations in N_2

	10%CO_2	15%CO_2	20%CO_2	30%CO_2	40%CO_2	50%CO_2	60%CO_2	70%CO_2	80%CO_2
x_{CO2}	0.692	0.788	0.846	0.911	0.946	0.966	0.979	0.987	0.993
$S_{CO2,N2}$	20.22	21.06	21.97	23.88	26.28	28.41	31.08	32.54	35.46

Tab.S4 The calculated equilibrium CO_2 molar fraction in adsorbed phase and corresponding CO_2/N_2 selectivity for NMC-250D8H at different CO_2 concentrations in N_2

	10%CO_2	15%CO_2	20%CO_2	30%CO_2	40%CO_2	50%CO_2
x_{CO2}	0.958	0.977	0.985	0.993	0.996	0.998
$S_{CO2,N2}$	205.3	240.7	262.7	331	373.5	499