Supporting Information

Modulating the porosity of N-doped carbon materials for

enhanced CO₂ capture and methane uptake

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	<u>CO₂ uptake (mmol/g)</u>		Reference
	1 bar	0.15 bar	
Sawdust-derived activated carbon	4.8	1.2	1
Petroleum pitch-derived activated carbon	4.55	~1.0	2
Activated carbon spheres	4.55	~1.1	3
Phenolic resin activated carbon spheres	4.5	~1.2	4
Fungi-derived activated carbon	3.5	~1.0	5
Chitosan-derived activated carbon	3.86	~1.1	6
Polypyrrole derived activated carbon	3.9	~1.0	7
Soya bean derived N-doped activated carbon	4.24	1.2	8
N-doped ZTCs	4.4	~1.0	9
Activated templated N-doped carbon	4.5	1.4	10
Polyaniline derived activated carbon	4.3	1.38	11
N-doped activated carbon monoliths	5.14	1.25	12
Activated hierarchical N-doped carbon	4.8	1.4	13
Activated N-doped carbon from algae	4.5	~1.1	14
Compactivated carbons from sawdust	5.8	2.0	15
Fern-derived activated carbon	5.67	~1.7	16
Compactivated carbons from polypyrrole	5.5	2.1	17
Clove-derived activated carbon	5.4	1.4	18
Potassium oxalate-activated carbon from date seed	5.0	1.8	19
Potassium phthalimide derived carbons	5.2	1.7	20

Table S1. CO_2 uptake capacity of various porous carbons at 25 °C and 0.15 bar or 1 bar (The data in the table is adapted from reference 29 in main manuscript).

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Sample	Density	65 bar (g g ⁻¹) (cm ³ cm ⁻³)		80 bar		100 bar		Reference
Sample	$(g \text{ cm}^{-3})$			$(g g^{-1})$	$(g g^{-1}) (cm^3 cm^{-3})$		$(\mathrm{cm}^3\mathrm{cm}^{-3})$	
DSM2800-1	0.65	0.25	230	0.27	248	0.29	266	This work
DSM4700-1	0.59	0.27	219	0.29	237	0.31	256	This work
DSU4800-1	0.41	0.35	197	0.39	217	0.42	239	This work
PPI-800-2	0.92	0.21	270	0.22	287	0.25	309	1
PPI-900-2	0.87	0.23	283	0.25	303	0.28	331	1
PPI-1000-2	0.83	0.25	288	0.27	311	0.29	338	1
CHCC2800	0.82	0.26	293	0.28	315	0.30	339	2
CHCC4700	0.75	0.27	282	0.29	306	0.32	334	2
CHCC4800	0.58	0.32	258	0.35	279	0.38	309	2
CNL4800	0.67	0.26	241	0.29	269	0.31	291	3
PPYCNL124	0.52	0.30	217	0.33	238	0.36	260	3
PPYCNL214	0.36	0.36	183	0.41	204	0.46	229	3
ACDS4800	0.69	0.25	243	0.27	262	0.29	282	3,4
PPYSD114	0.47	0.32	211	0.35	231	0.39	254	4
AX-21 carbon	0.487	0.30	203	0.33	222	0.35	238	5
HKUST-1	0.881	0.21	263	0.22	272	0.23	281	5
Ni-MOF-74	1.195	0.15	259	0.16	267	0.17	277	5
Al-soc-MOF-1	0.34	0.41	197	0.47	222			6
MOF-210	0.25	0.41	143	0.48	168			7
NU-1500-Al	0.498	0.29	200	0.31	216	0.34	237	8
NU-1501-Fe	0.299	0.40	168	0.46	193	0.52	218	8
NU-1501-Al	0.283	0.41	163	0.48	190	0.54	214	8
monoHKUST-1	1.06	0.17	261	0.18	278	0.18	275	9
monoUiO-66_D	1.05	0.14	210	0.17	245	0.20	296	10

Table S2. Methane uptake for the best performing N-doped activated carbons compared to selected benchmark MOFs and carbons reported in the literature. Volumetric uptake of powder

MOFs is calculated based on crystallographic density rather than packing density.

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Table S3: Methane uptake working capacity for N-doped activated carbons compared to selected benchmark MOFs and carbons reported in the literature.

Sample	65 bar		80 bar		100 bar		Reference
	$(g g^{-1}) (cm^3 cm^{-3})$		$(g g^{-1}) (cm^3 cm^{-3})$		$(g g^{-1}) (cm^3 cm^{-3})$		
DSM2800-1	0.17	159	0.19	177	0.21	195	This work
DSM4700-1	0.20	159	0.22	177	0.24	196	This work
DSU4800-1	0.27	151	0.31	171	0.34	193	This work
PPI-800-2	0.13	174	0.14	190	0.16	212	1
PPI-900-2	0.15	183	0.17	203	0.19	231	1
PPI-1000-2	0.17	199	0.19	222	0.21	249	1
CHCC2800	0.18	200	0.20	222	0.22	246	2
CHCC4700	0.20	210	0.22	234	0.25	262	2
CHCC4800	0.25	197	0.28	218	0.31	248	2
CNL4800	0.19	182	0.22	202	0.24	224	3
PPYCNL124	0.23	167	0.26	188	0.29	209	3
PPYCNL214	0.29	146	0.34	167	0.39	192	4
ACDS4800	0.18	171	0.20	189	0.22	209	3,4
PPYSD114	0.25	162	0.28	182	0.32	205	4
AX-21 carbon	0.23	155	0.26	174	0.28	190	5
HKUST-1	0.15	179	0.16	198	0.17	207	5
Ni-MOF-74	0.08	148	0.09	152	0.10	162	5
Al-soc-MOF-1	0.36	176	0.42	201			6
MOF-210	0.38	134	0.45	157			7
NU-1500-A1	0.24	165	0.26	181	0.29	202	8
NU-1501-Fe	0.36	151	0.42	176	0.48	201	8
NU-1501-A1	0.37	147	0.44	174	0.50	198	8
monoHKUST-1	0.12	184	0.13	201	0.13	198	9
monoUiO-66_D	0.11	167	0.14	202	0.17	253	10

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Supporting Figure 1. Photograph of the attempted activation of melamine and urea.



Supporting Figure 2. The N/C atomic ratio of activated carbons as a function of the activation temperature.



Supporting Figure 3. TGA curves of N-doped activated carbons prepared at KOH/ACDS ratio of 2 or 4 at 600, 700 or 800 °C and melamine/ACDS ratio of 1.



Supporting Figure 4. TGA curves of N-doped activated carbons prepared at 800 °C and KOH/ACDS ratio of 4 and various amounts of melamine or urea.



Supporting Figure 5. Powder XRD patterns of raw date seeds (Raw-DS) and air-carbonised dated seeds (ACDS).



Supporting Figure 6. Powder XRD patterns of N-doped carbons activated at KOH/ACDS ratio 2 or 4 with melamine or urea as dopants.



Supporting Figure 7. SEM images of selected N-doped activated carbons.



Supporting Figure 8. TEM images of selected N-doped activated carbons.



Supporting Figure 9. Excess CO₂ and methane uptake of N-doped carbons.