

SUPPORTING INFORMATION

Three-Dimensional Graphene-Based Porous Adsorbents for Postcombustion CO₂ Capture

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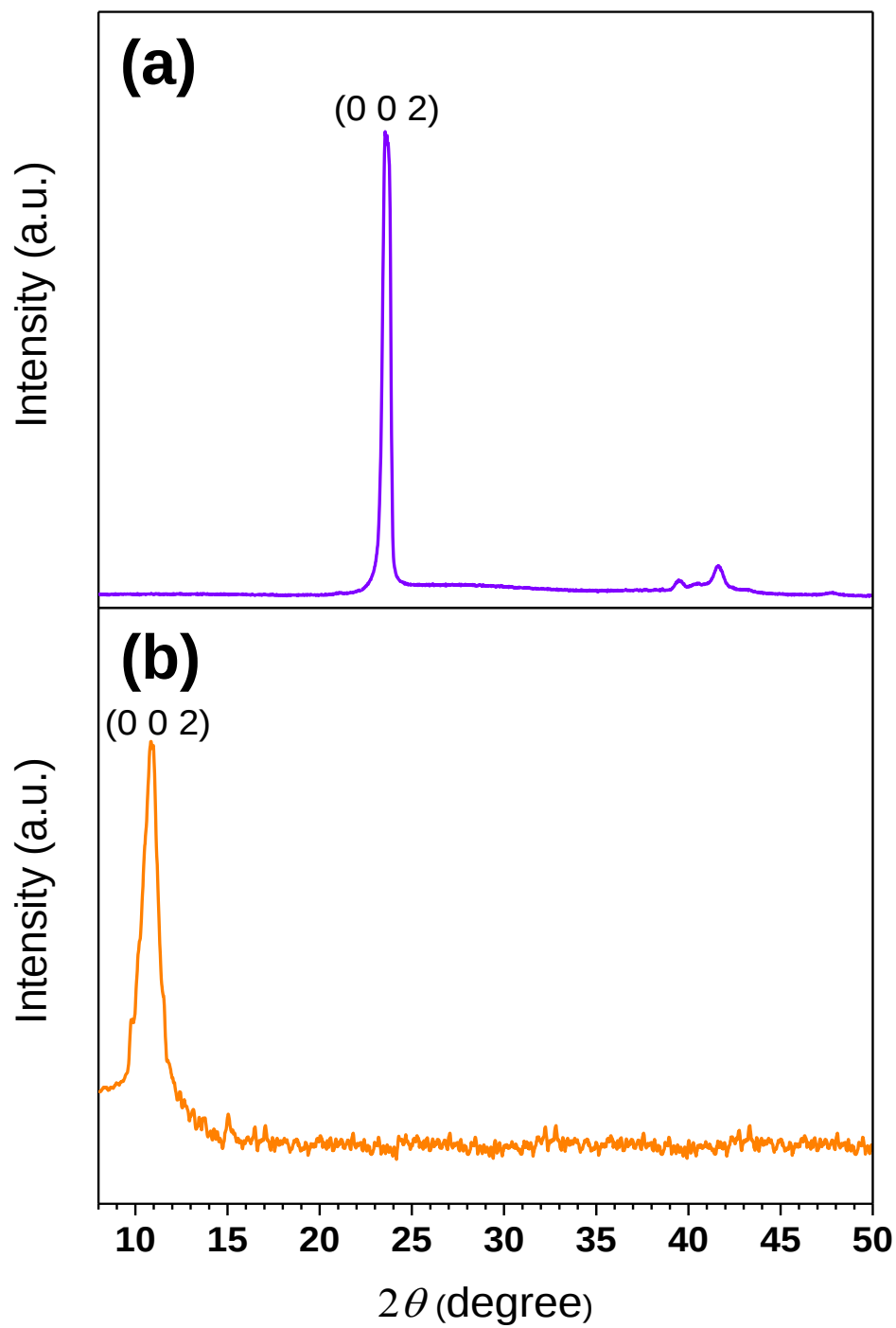


Figure S1. XRD patterns of (a) natural graphite and (b) GO.

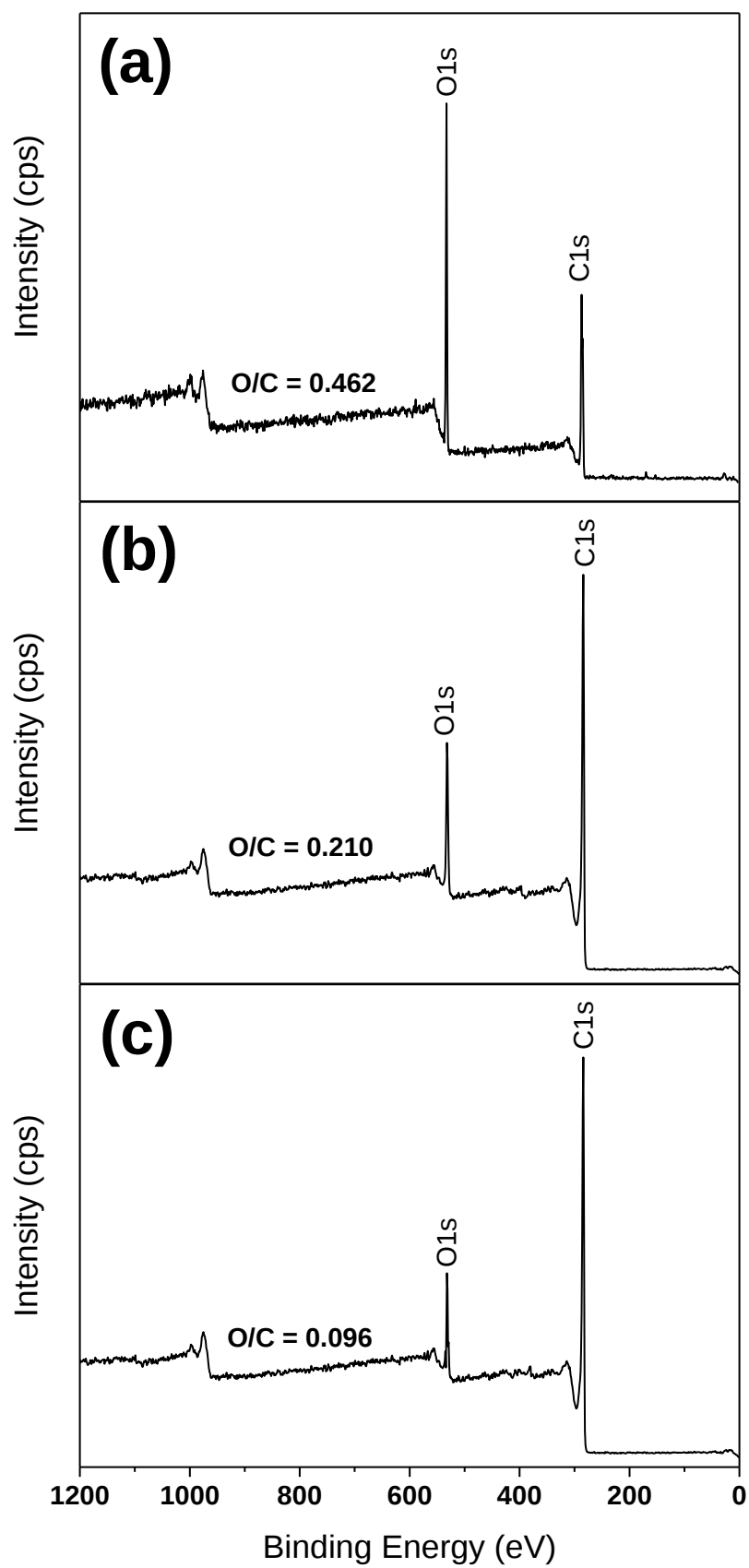


Figure S2. XPS survey scan spectra of (a) GO, (b) RGO and (c) a-RGO-950.

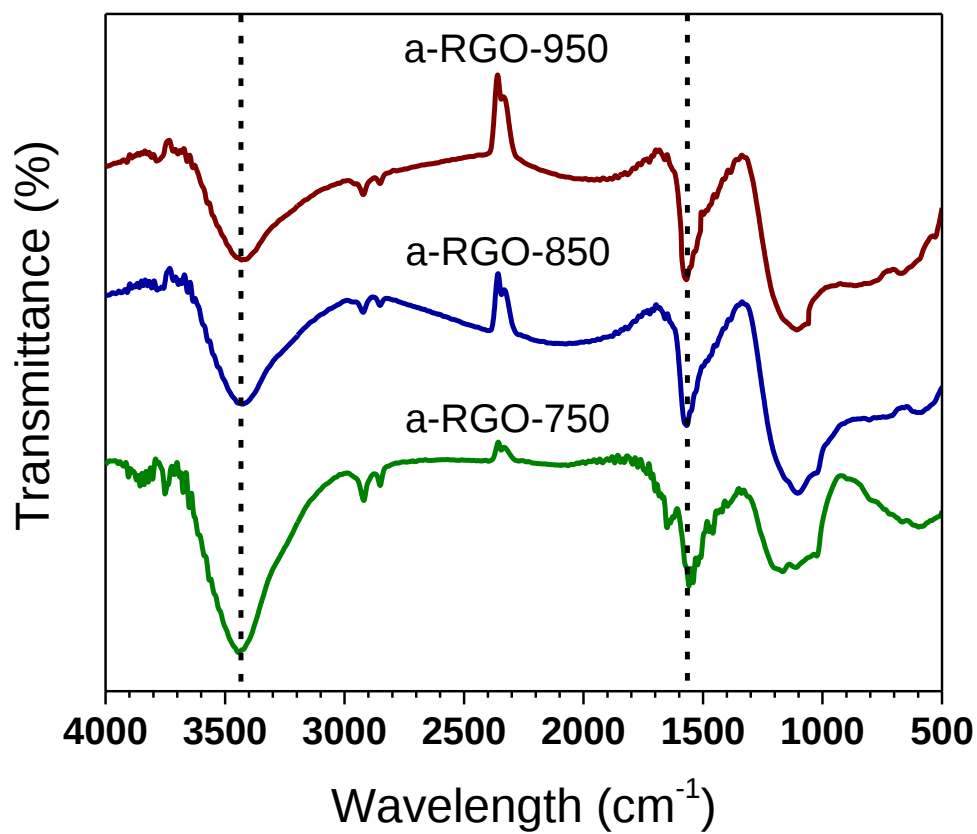


Figure S3. FTIR spectra of the a-RGO-*X* samples.

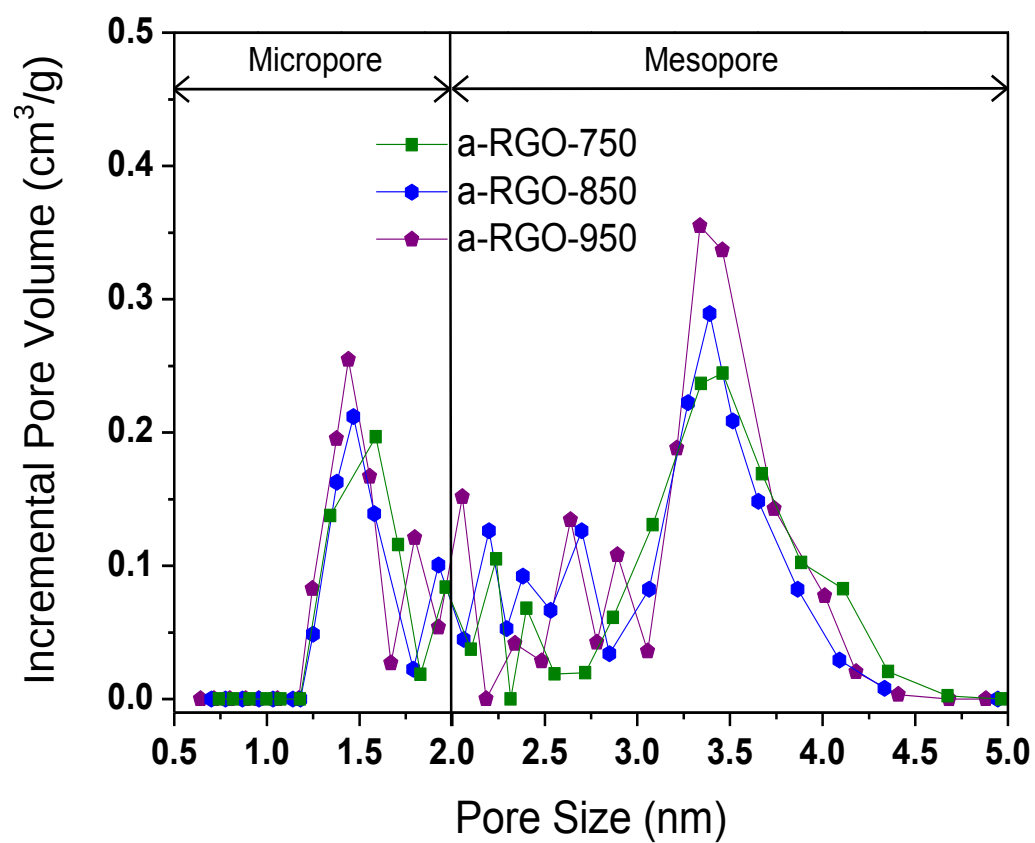


Figure S4. NLDFIT pore size distributions of the a-RGO-*X* materials.

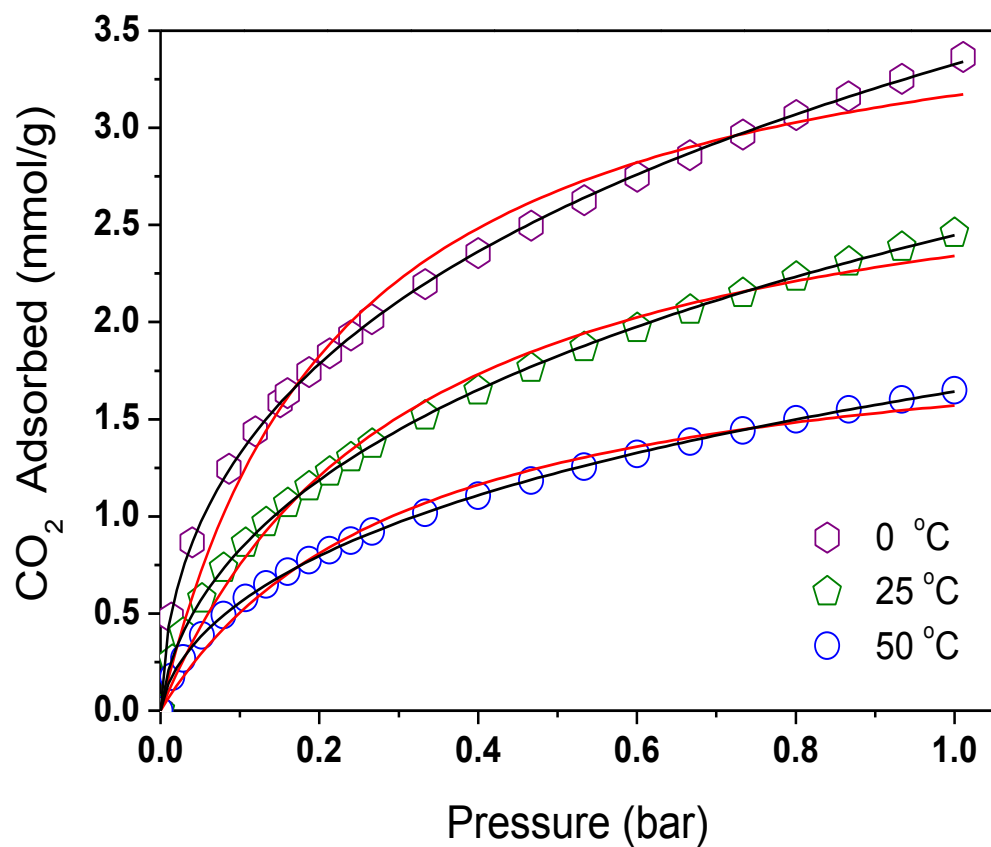


Figure S5. Effect of temperature on CO₂ adsorption by a-RGO-950 and nonlinear fit of the Langmuir (—) and Toth (—) isotherm models to the experimental equilibrium data.

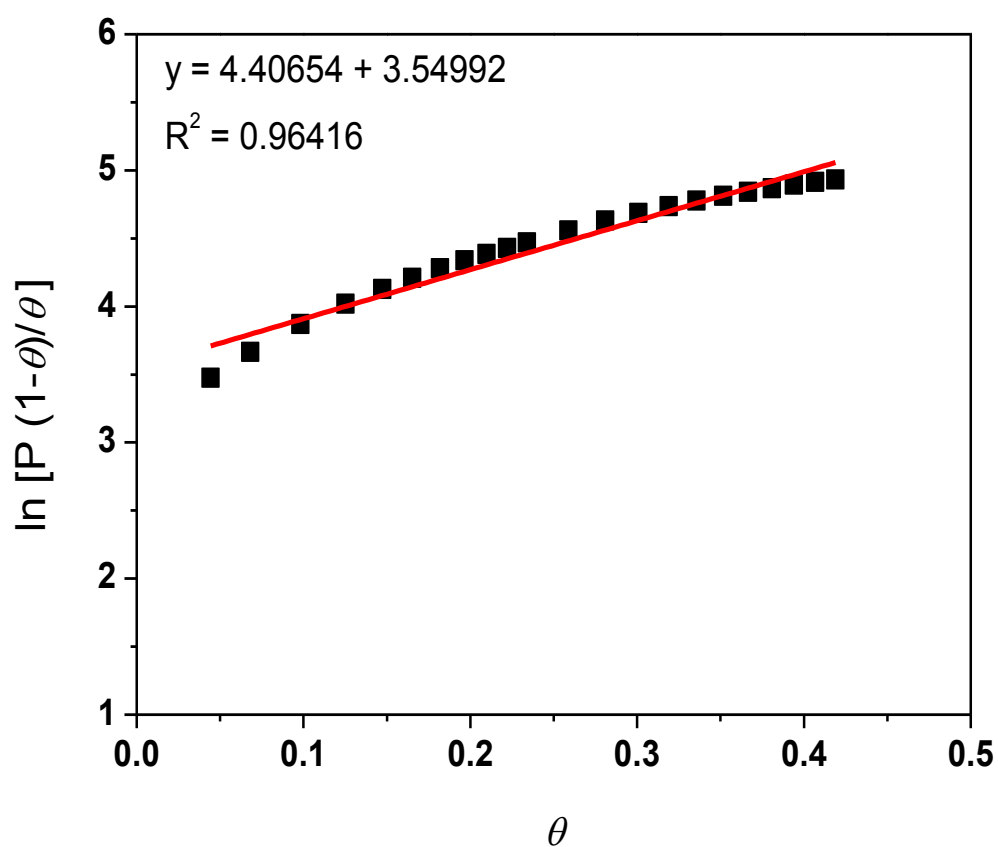


Figure S6. Fowler-Guggenheim plot for CO₂ adsorption on a-RGO-950 at 25 °C.

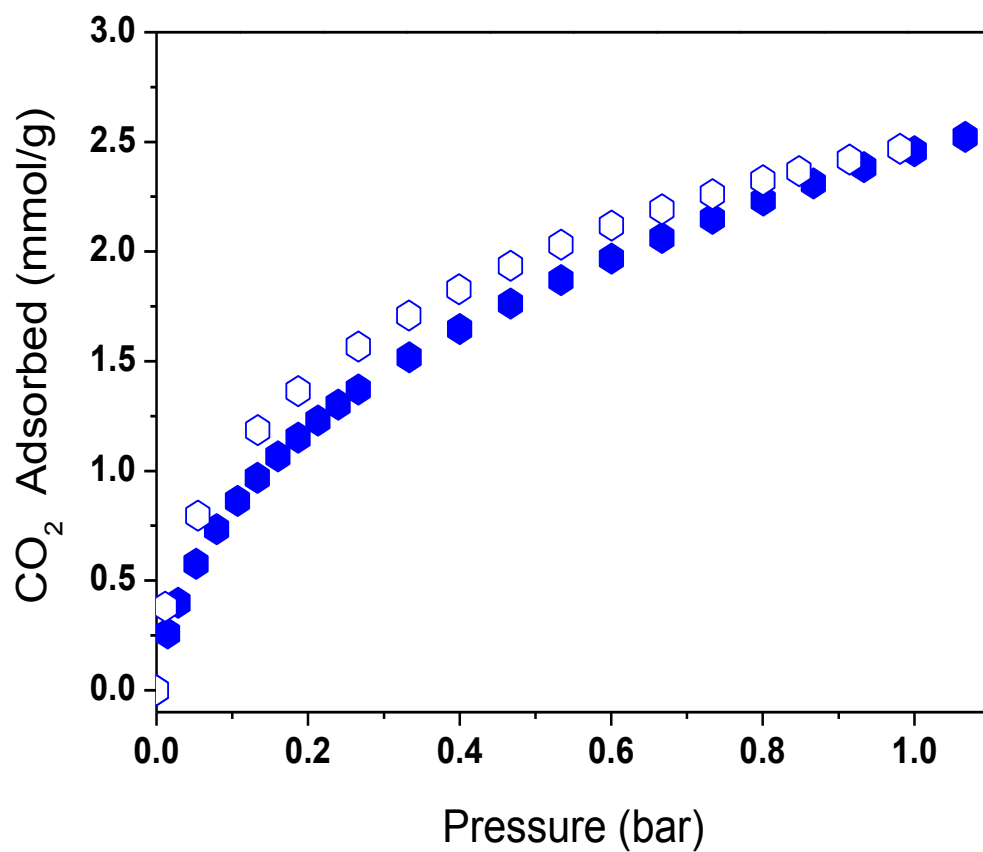


Figure S7. CO₂ adsorption/desorption isotherms of a-RGO-950 at 25 °C (solid symbols denote adsorption and open symbols denote desorption).

Table S1. Surface elemental composition of GO, RGO and the a-RGO-X samples as determined from XPS measurements.

Sample	O content (at. %)	C species distribution (at.%)		
		285.0 eV	287.1 eV	288.7 eV
GO	31.6	29.8	33.5	5.1
RGO	17.4	65.1	12.6	4.9
a-RGO-750	11.7	77.9	7.1	3.3
a-RGO-850	9.5	81.2	6.6	2.7
a-RGO-950	8.8	83.4	5.7	2.1

Table S2. Comparison of the zero-coverage isosteric heat of CO₂ adsorption for a-RGO-950 with other major types of solid adsorbents.

Adsorbent	$-Q_{0,st}$ (kJ mol ⁻¹)	Reference
<i>Zeolites</i>		
KCHA ^a	52.2	Ridha and Webley. ¹
K-BEA ^b	59.6	Yang <i>et al.</i> ²
Na-A ^c	48.1	Zukal <i>et al.</i> ³
Na-X ^d	42.5	Li <i>et al.</i> ⁴
<i>MOFs</i>		
Cu-BTtri-mmen ^e	96	McDonald <i>et al.</i> ⁵
TBA@bio-MOF-1 ^f	55	An and Rosi. ⁶
ZIF-68 ^g	25.1	Liu <i>et al.</i> ⁷
Li@CNT@Cu ₃ (BTC) ₂ ^h	36.8	Xiang <i>et al.</i> ⁸
<i>Activated carbons</i>		
Ammonia-modified activated carbon	73.2	Shafeeyan <i>et al.</i> ⁹
Activated carbon from oil palm shell	31.5	Hoseinzadeh Hesas <i>et al.</i> ¹⁰
Pitch-based activated carbon beads	23.2	Shen <i>et al.</i> ¹¹
N-doped activated carbon	58	Yang <i>et al.</i> ¹²
a-RGO-950	27.4	This study

^a Potassium chabazite

^b Potassium-exchanged zeolite beta

^c Sodium form of zeolite A

^d Sodium form of zeolite X

^e H₃[(Cu₄Cl)₃(BTtri)₈(N,N'-dimethylethylenediamine)₁₂]

^f Zn₈(adenine)₄(biphenyl-4,4'-dicarboxylate)₆O₃.tetrabutylammonium

^g Zeolitic imidazolate framework-68

^h Li-doped carbon nanotube-impregnated Cu₃(1,3,5-benzenetriscarboxylate)₂

■ REFERENCES

- (1) Ridha, F.N.; Webley, P.A. Anomalous Henry's law behavior of nitrogen and carbon dioxide adsorption on alkali-exchanged chabazite zeolites. *Sep. Purif. Technol.* **2009**, *67*, 336-343.
- (2) Yang, S.-T.; Kim, J.; Ahn, W.-S. CO₂ adsorption over ion-exchanged zeolite beta with alkali and alkaline earth metal ions. *Micropor. Mesopor. Mat.* **2010**, *135*, 90-94.
- (3) Zukal, A.; Areat, C.O.; Delgado, M.R.; Nachtigall, P.; Pulido, A.; Mayerova, J.; Cejka, J. Combined volumetric, infrared spectroscopic and theoretical investigation of CO₂ adsorption on Na-A zeolite. *Micropor. Mesopor. Mat.* **2011**, *146*, 97-105.
- (4) Li, Y.; Yi, H.; Tang, X.; Li, F.; Yuan, Q. Adsorption separation of CO₂/CH₄ gas mixture on the commercial zeolites at atmospheric pressure. *Chem. Eng. J.* **2013**, *229*, 50-56.
- (5) McDonald, T.M.; D'Alessandro, D.M.; Krishna, R.; Long, J.R. Enhanced carbon dioxide capture upon incorporation of N,N'-dimethylethylenediamine in the metal-organic framework CuBTtri. *Chem. Sci.* **2011**, *2*, 2022-2028.
- (6) An, J.; Rosi, N.L. Tuning MOF CO₂ adsorption properties via cation exchange. *J. Am. Chem. Soc.* **2010**, *132*, 5578-5579.
- (7) Liu, Y.; Kasik, A.; Linneen, N.; Liu, J.; Lin, Y.S. Adsorption and diffusion of carbon dioxide on ZIF-68. *Chem. Eng. Sci.* **2014**, *118*, 32-40.
- (8) Xiang, Z.; Hu, Z.; Cao, D.; Yang, W.; Lu, J.; Han, B.; Wang, W. Metal organic frameworks with incorporated carbon nanotubes: improving carbon dioxide and methane storage capacities by lithium doping. *Angew. Chem. Int. Ed.* **2011**, *50*, 491-494.

- (9) Shafeeyan, M.S.; Wan Daud, W.M.A.; Shamiri, A.; Aghamohammadi, N. Adsorption equilibrium of carbon dioxide on ammonia-modified activated carbon. *Chem. Eng. Res. Des.* **2015**, *104*, 42-52.
- (10) Hoseinzadeh Hesas, R.; Arami-Niya, A.; Wan Daud, W.M.A.; Sahu, J.N. Microwave-assisted production of activated carbons from oil palm shell in the presence of CO₂ or N₂ for CO₂ adsorption. *J. Ind. Eng. Chem.* **2015**, *24*, 196-205.
- (11) Shen, C.; Grande, C.A.; Li, P.; Yu, J.; Rodrigues, A.E. Adsorption equilibria and kinetics of CO₂ and N₂ on activated carbon beads. *Chem. Eng. J.* **2010**, *160*, 398-407.
- (12) Yang, Z.; Xia, Y.; Zhu, Y. Preparation and gases storage capacities of N-doped porous activated carbon materials derived from mesoporous polymer. *Mater. Chem. Phys.* **2013**, *141*, 318-323.