

Supporting Information

Effect of N, S doping on graphene oxide aerogel for adsorption and photocatalytic reduction of carbon dioxide

Yuhan Liu, Jing Shang, Tong Zhu*

*SKL-ESPC & SEPKL-AERM, College of Environmental Sciences and Engineering,
and Center for Environment and Health, Peking University, Beijing, 100871, P. R.*

China

* Corresponding Author: shangjing@pku.edu.cn.

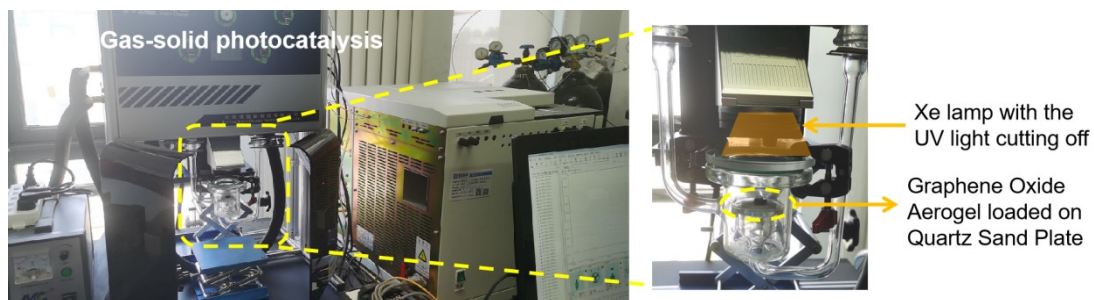


Fig.S1. Experimental setup of gas-solid CO₂ photocatalytic reduction.

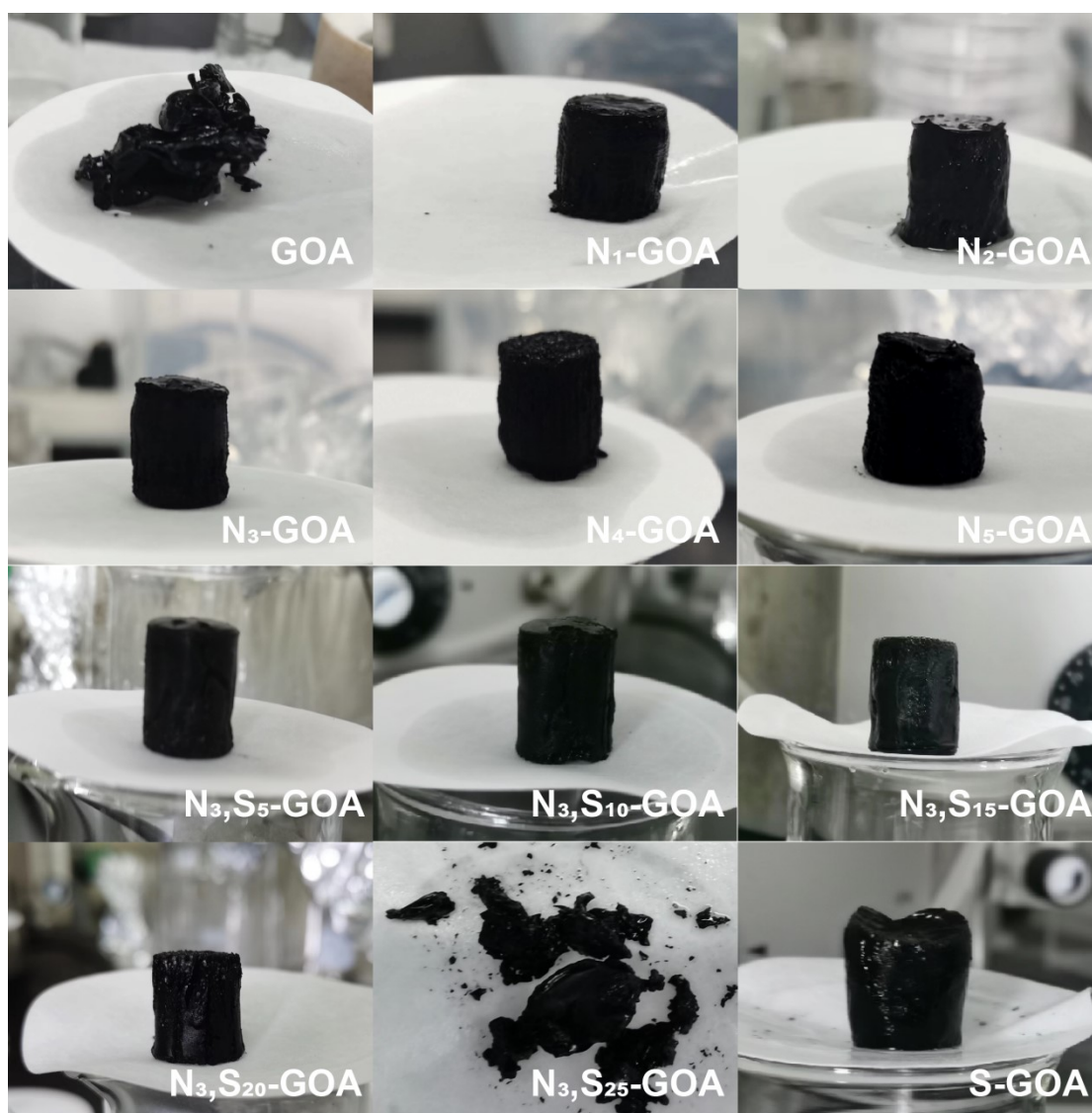


Fig.S2. The appearance of GOA obtained by different doping.

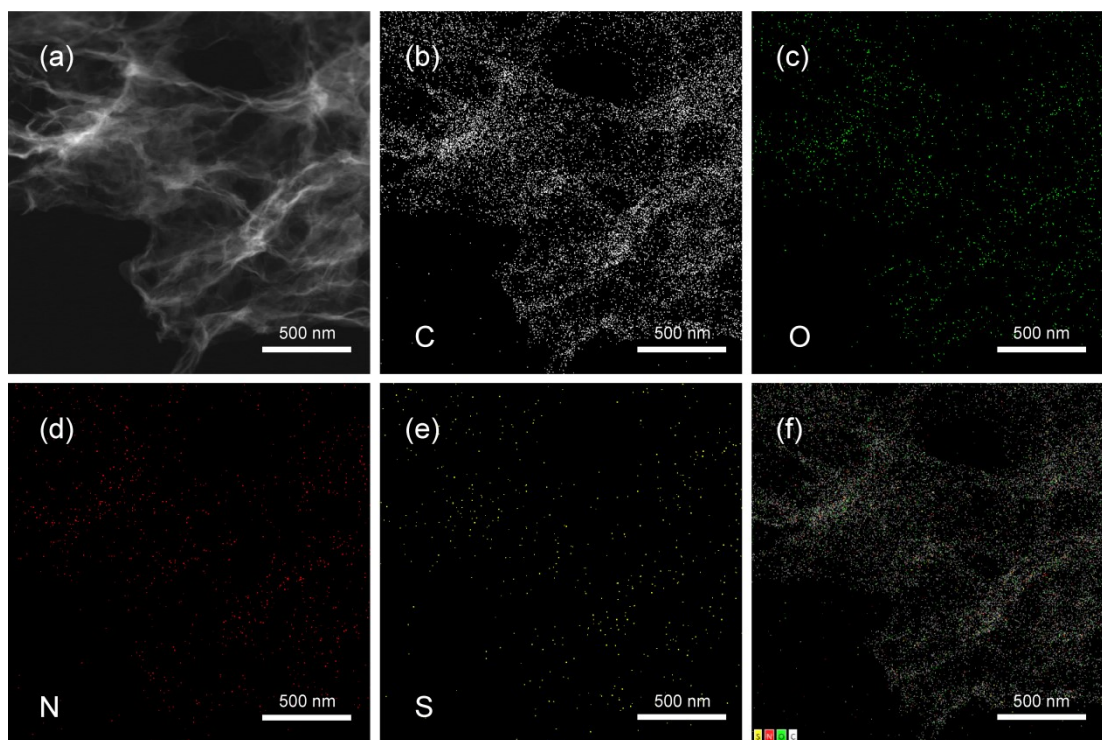


Fig. S3 The TEM image (a) of N,S-GOA with its corresponding EDS elemental mappings of carbon (b), oxygen (c), nitrogen (d) and sulfur (e).

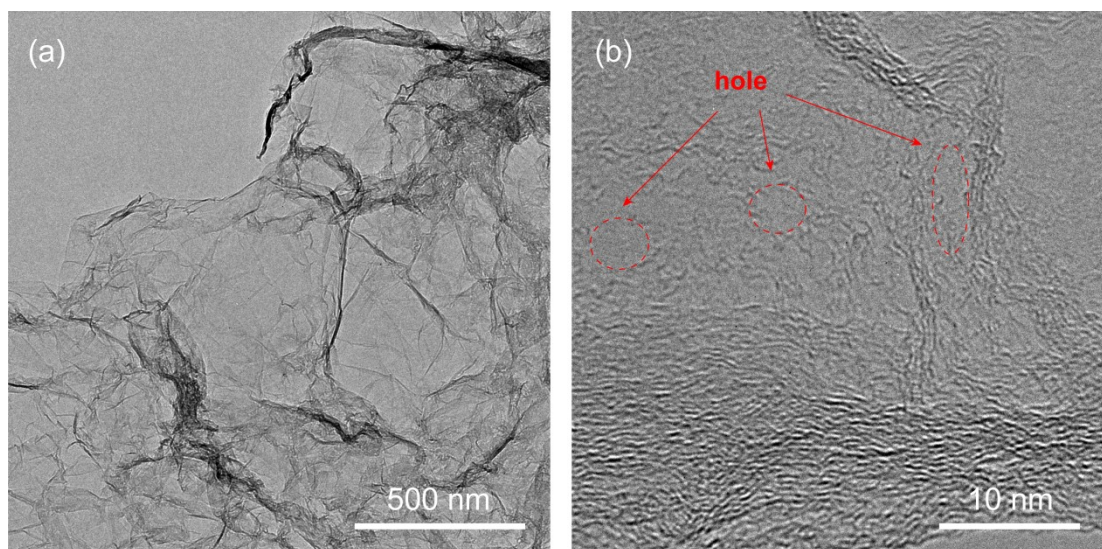


Fig. S4 Low-magnification (a) and high-resolution (b) TEM images of N,S-GOA.

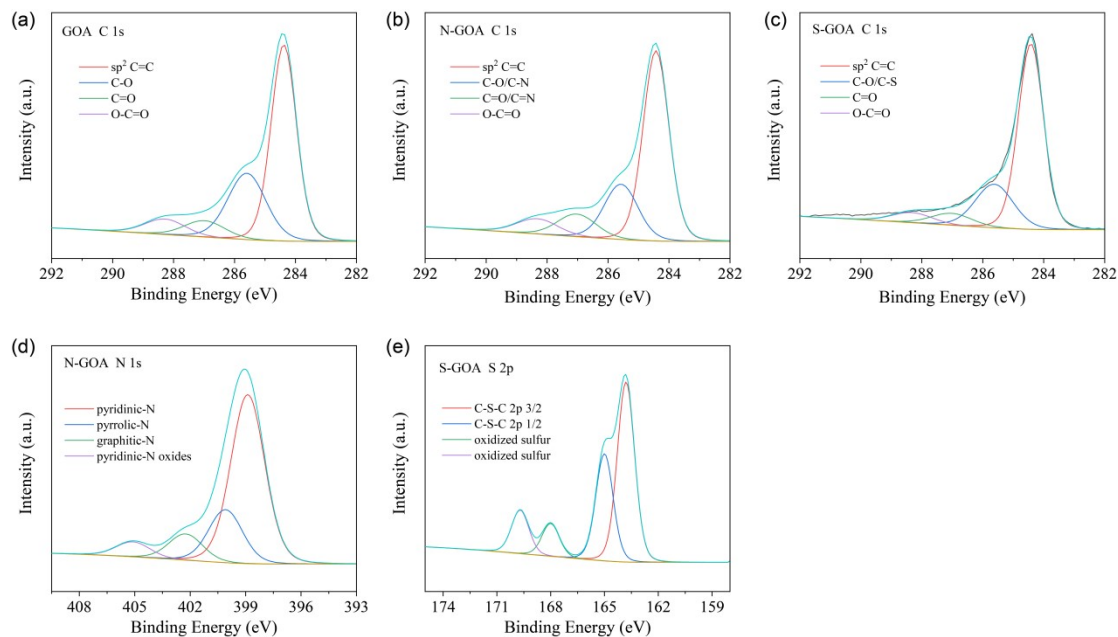


Fig. S5. XPS C 1s spectra of (a) GOA, (b) N-GOA, and (c) S-GOA. (d) XPS N 1s spectra of N-GOA. (e) XPS S 2p spectra of S-GOA.

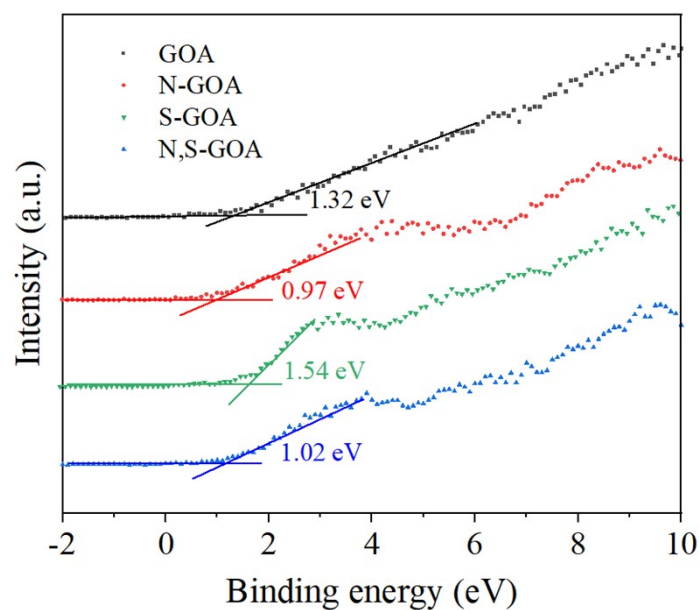


Fig. S6. XPS valence band potentials of GOA, N-GOA, S-GOA, and N,S-GOA. After eliminating the measurement error by the formula (1) with the work function (Φ) of XPS analyzer and vacuum level to be 4.39 and 4.44 eV (vs. NHE),¹ the calculated valence band (VB) positions (vs. NHE) of GOA, N-GOA, S-GOA, and N,S-GOA are 1.27, 0.92, 1.49, and 0.97 eV, respectively.

$$VB \text{ (vs. NHE)} = \phi + VB_{XPS} - 4.44 \text{ eV} \quad (1)$$

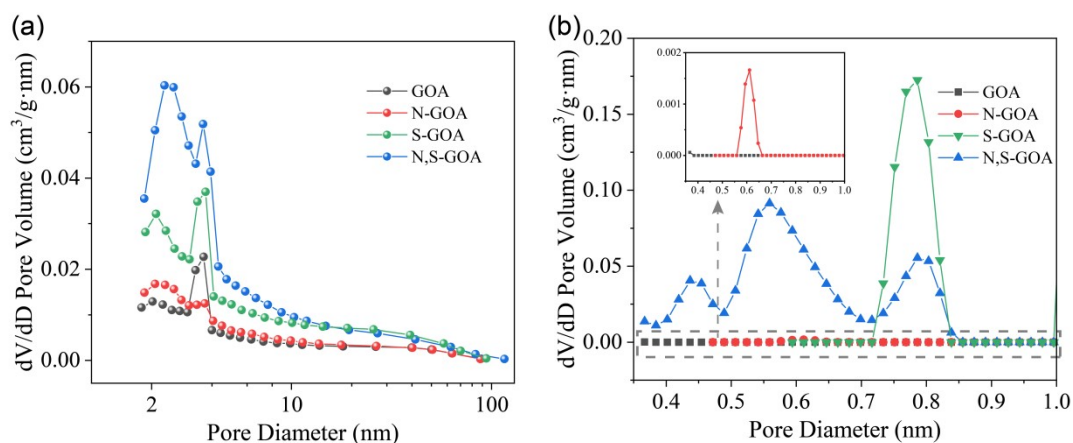


Fig. S7. (a) Pore volume distribution of GOA, N-GOA, S-GOA, and N,S-GOA by (a) N_2 adsorption-desorption test and (b) CO_2 adsorption-desorption test (inset is an enlarged view of the GOA and N-GOA curves).

Table S1. Contents of major elements of samples based on XPS analyses.

Sample	C (at.%)	O (at.%)	O/C ratio (%)	N (at.%)	S (at.%)
GOA	87.21	12.43	14.25	0.36	/
N-GOA	84.56	9.28	10.97	6.16	/
S-GOA	88.26	9.25	10.48	0.25	2.24
N,S-GOA	85.01	7.66	9.01	5.97	1.36

Table S2. Assigned C 1s, N 1s, and S 2p peaks of samples based on XPS analyses.

	GOA	N-GOA	S-GOA	N,S-GOA
C=C (284.4 eV)	56.68	59.32	63.33	62.23
sp ² carbon bonded with heteroatoms (285.6 eV)	28.69	22.44	22.82	24.27
C=O/C=N (287.0 eV)	7.57	10.61	7.17	8.06
O-C=O (288.3 eV)	7.16	7.63	6.68	5.44
pyridinic-N (398.1-399.3 eV)	/	64.18	/	48.22
pyrrolic-N (399.8-401.2 eV)	/	20.15	/	35.04
graphitic-N (401.1-402.7 eV)	/	9.97	/	12.06
pyridinic-N oxides (402-406 eV)	/	5.70	/	4.68
Thiophenic(C-S-C) (163.8- 165.2 eV)	/	/	72.05	79.04
Oxidized sulfur (167.4- 170.0 eV)	/	/	27.95	20.96

Table S3. Surface area and CO₂ uptake capacity for the samples.

Sample	S _{BET} ^a (m ² /g)	S _L ^b (m ² /g)	PV ^c (cm ³ /g)	Quantity Adsorbed CO ₂ ^d (cm ³ /g STP)
GOA	79	189	0.246	1.04
N-GOA	88	210	0.256	2.72
S-GOA	172	402	0.507	56.55
N,S-GOA	257	575	0.568	20.62

^aSurface area calculated from N₂ adsorption isotherms at 77.5 K using BET equation.

^bSurface area calculated from N₂ adsorption isotherms at 77.5 K using Langmuir equation. ^cPore volume calculated from nitrogen isotherm at $p/p_0=0.986$, 77.5 K.

^dQuantity Adsorbed CO₂ determined volumetrically using a Micromeritics TriStar II Plus analyzer at 1.00 bar and 273.15 K.

Table S4. Comparison of CO₂ capture capacity on various nanoamaterials.

Nanoamaterials	Exp. Cond.	Capacity (mmol/g)	Ref.
S-GOA	273.15 K, 1 bar	2.52	This work
MgO/GO	60 °C, 1 bar	2.79	2
Mo/g-C ₃ N ₄	273.15 K, 1 bar	0.07	3
N,O-codoped microporous carbon	25 °C, 1 atm	3.05	4
Triethylamine/rice husk silica	600 °C, 1 atm	0.75	5
70T-MM-550	75 °C, 1 atm	3.43	6
HPSA	25 °C, 1 bar	2.07	7
Zn ₂ (TRZ) ₂ (Fuma)	298 K, 101 kPa	2.78	8

References

- 1 Y. Shang, Y. Wang, C. Lv, F. Jing, T. Liu, W. Li, S. Liu and G. Chen, *Chem. Eng. J.* 2022, **431**, 133898.
- 2 C. A. Gunathilake, G. G. T. A. Ranathunge, R. S. Dassanayake, S. D. Illesinghe, A. S. Manchanda, C. S. Kalpage, R. M. G. Rajapakse and D. G. G. P. Karunaratne, *Environ. Sci.-Nano*, 2020, **7**, 1225-1239.
- 3 S. L. Huang, H. Yi, L. H. Zhang, Z. Y. Jin, Y. J. Long, Y. Y. Zhang, Q. F. Liao, J. Na, H. Z. Cui, S. C. Ruan, Y. Yamauchi, T. Wakihara, Y. V. Kaneti and Y. J. Zeng, *J. Hazard. Mater.*, 2020, **393**, 122324.
- 4 X. Li, G. Y. Ma, X. R. Chen, L. Y. Cheng, C. Fang, H. P. Li, L. Wang, J. Ding, H. Wan and G. F. Guan, *J. Environ. Chem. Eng.*, 2022, **10**, 108526.
- 5 F. Zarei and P. Keshavarz, *Environ. Sci. Pollut. R.*, 2023, **30**, 19278-19291.
- 6 N. Mahinpey and D. Karami, *Catal. Today*, 2022, **404**, 237-243.
- 7 D. Peltzer, J. Múnera and L. Cornaglia, *J. Environ. Chem. Eng.*, 2019, **7**, 102927.
- 8 C. G. Zhou, S. A. Yu, K. Ma, B. Liang, S. Y. Tang, C. J. Liu and H. R. Yue, *Chem. Eng. J.*, 2021, **413**, 127675.