

## Electronic Supplementary Information for

### Electrostatically regulated ternary-doped carbon foams with exposed active sites as metal-free oxygen reduction electrocatalysts

Md. Selim Arif Sher Shah,<sup>a¶</sup> Jooyoung Lee,<sup>b</sup> Ali Rauf,<sup>a</sup> Jong Hyeok Park,<sup>c</sup> Byungkwon Lim,<sup>b</sup>  
and Pil J. Yoo<sup>a,d,\*</sup>

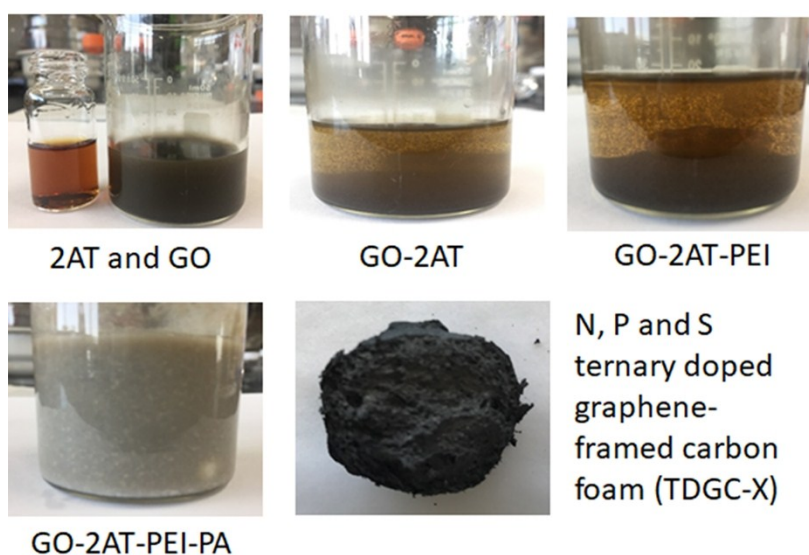
<sup>a</sup> School of Chemical Engineering, <sup>b</sup> School of Advanced Materials Science and Engineering, and <sup>d</sup> SKKU Advanced Institute of Nanotechnology (SAINT), Sungkyunkwan University (SKKU), Suwon 16419, Republic of Korea.

<sup>c</sup> Department of Chemical and Biomolecular Engineering, Yonsei University, Seoul 03722, Republic of Korea

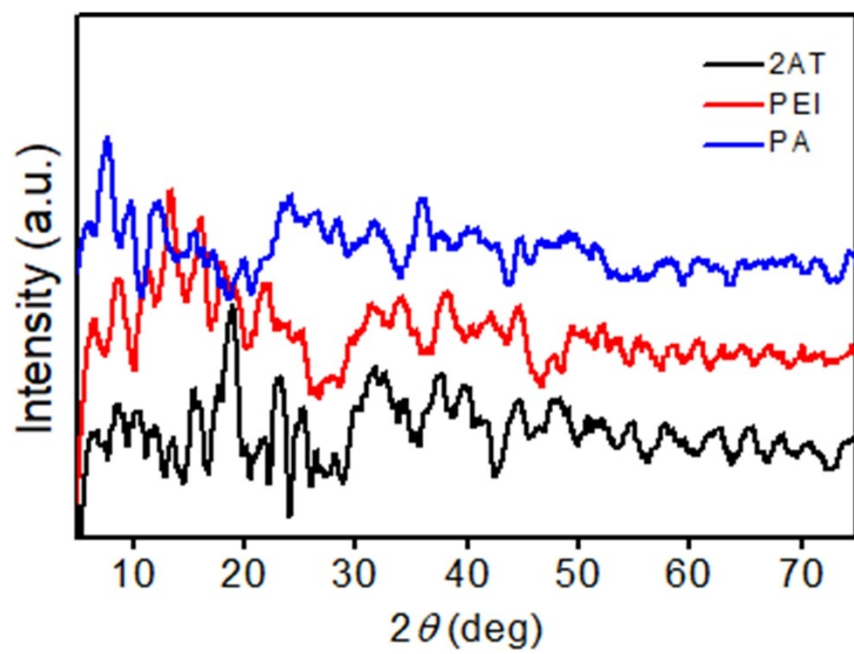
#### ¶ Present address

<sup>c</sup> Department of Chemical and Biomolecular Engineering, Yonsei University, Seoul 03722, Republic of Korea

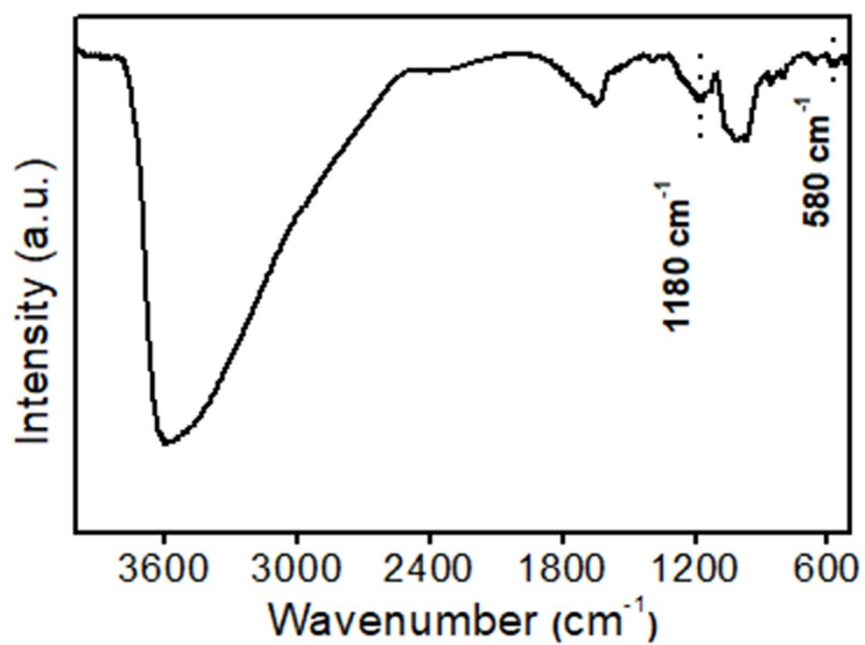
Corresponding authors: E-mail: pjyoo@skku.edu and blim@skku.edu.



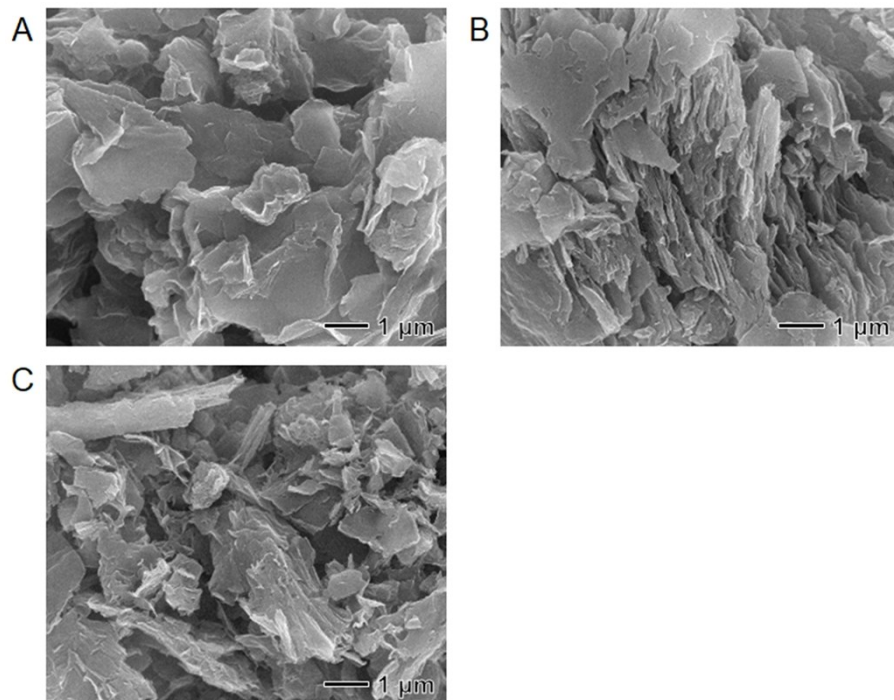
**Fig. S1** Digital images of GO and 2AT and the intermediates obtained after the addition of different precursors. It also shows the pyrolysed product.



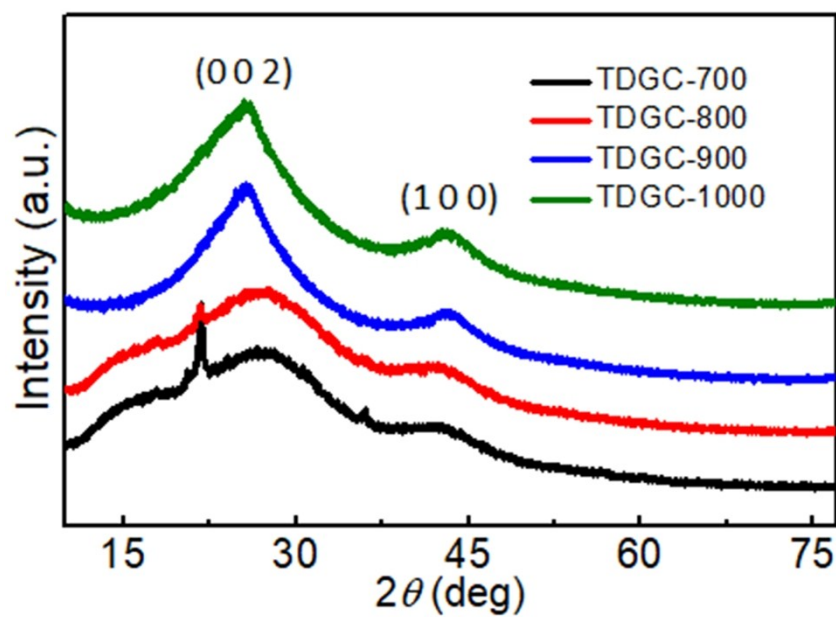
**Fig. S2** XRD patterns of 2AT, PEI, and PA



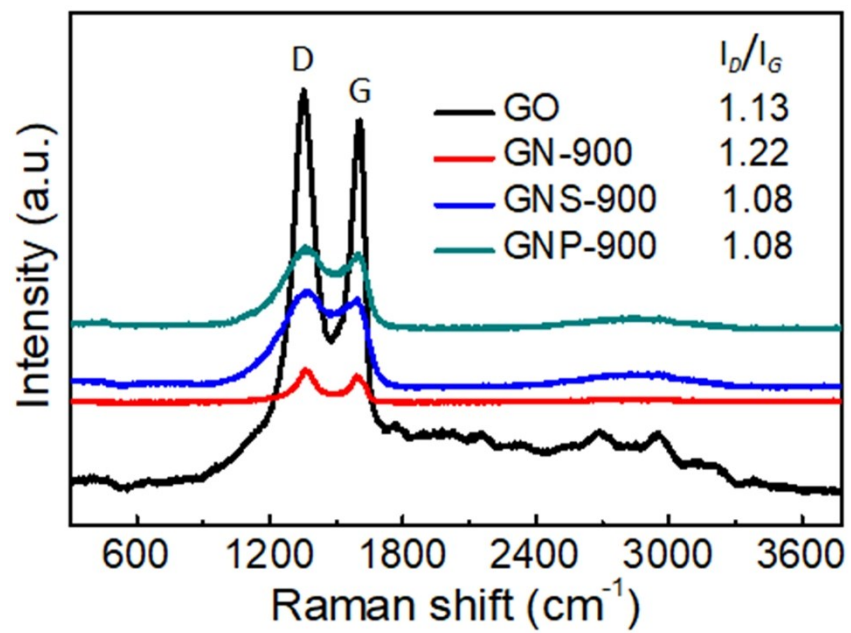
**Fig. S3** FT-IR of phytic acid (PA).



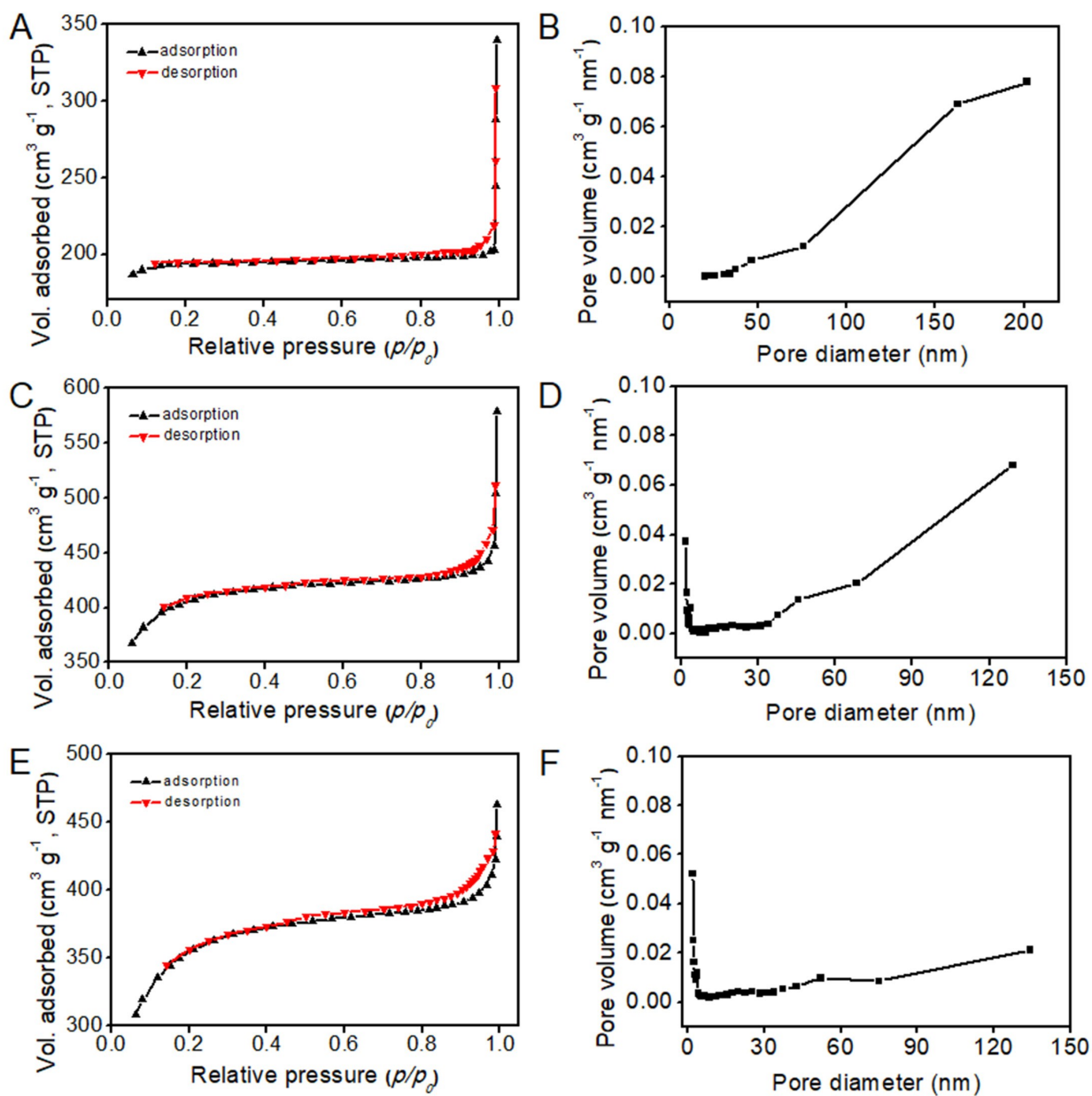
**Fig. S4** SEM images of (A) GO, (B) GO-2AT, and (C) GO-2AT-PEI.



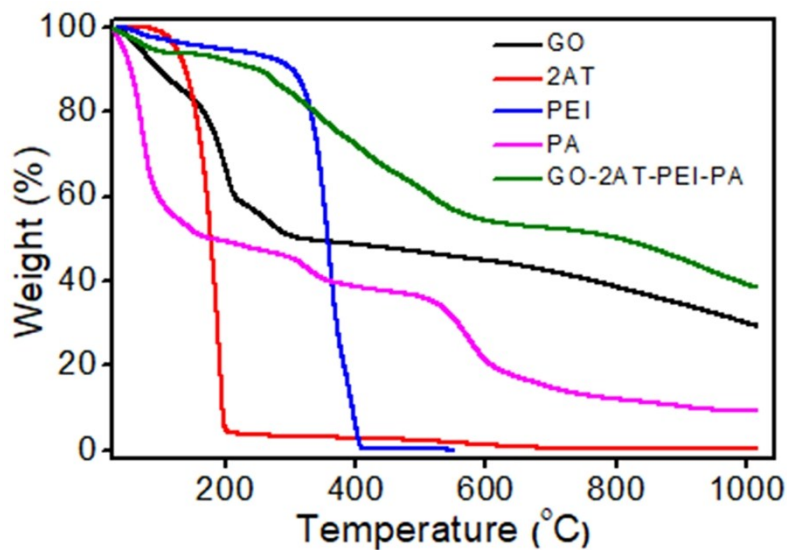
**Fig. S5** XRD patterns of TDGC-700, TDGC-800, TDGC-900, and TDGC-1000.



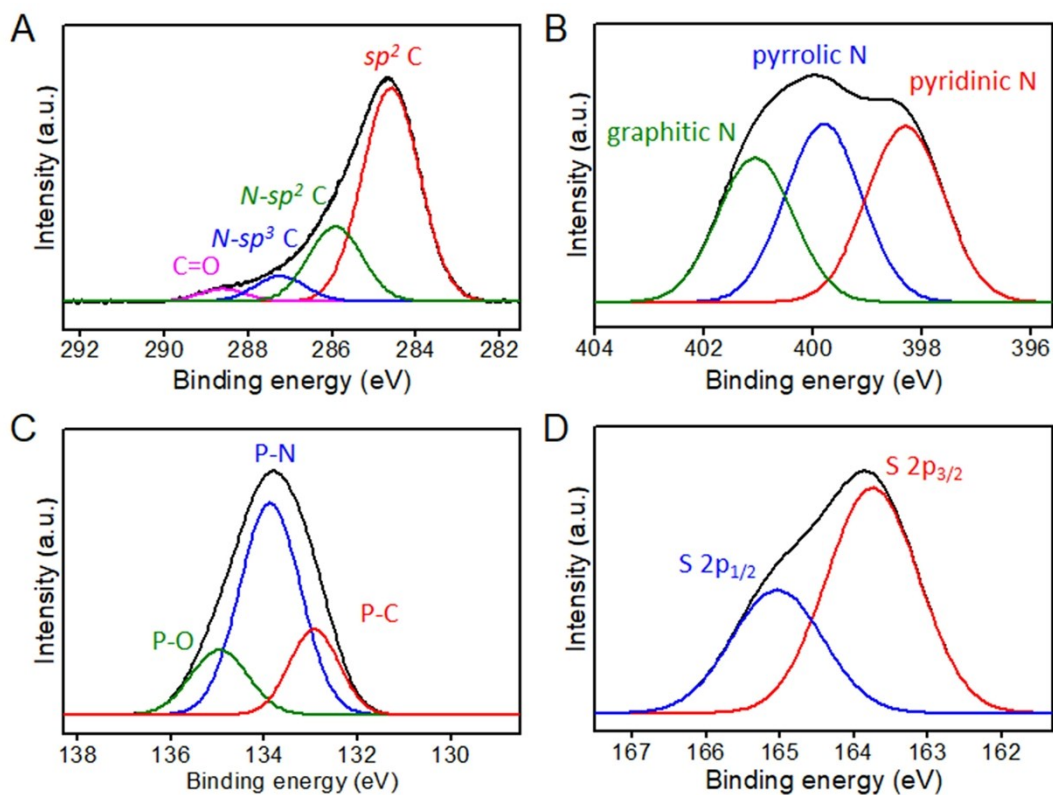
**Fig. S6** Raman spectra of GO, GN-900, GNS-900 and GNP-900.



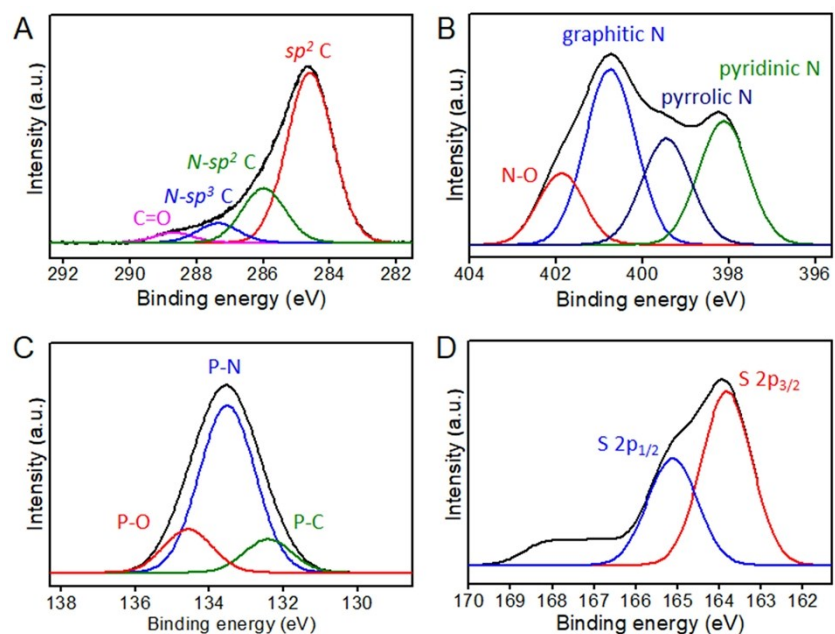
**Fig. S7** Nitrogen adsorption-desorption isotherms and pore size distribution of (A, B) TDGC-700, (C, D) TDGC-800, and (E, F) TDGC-1000.



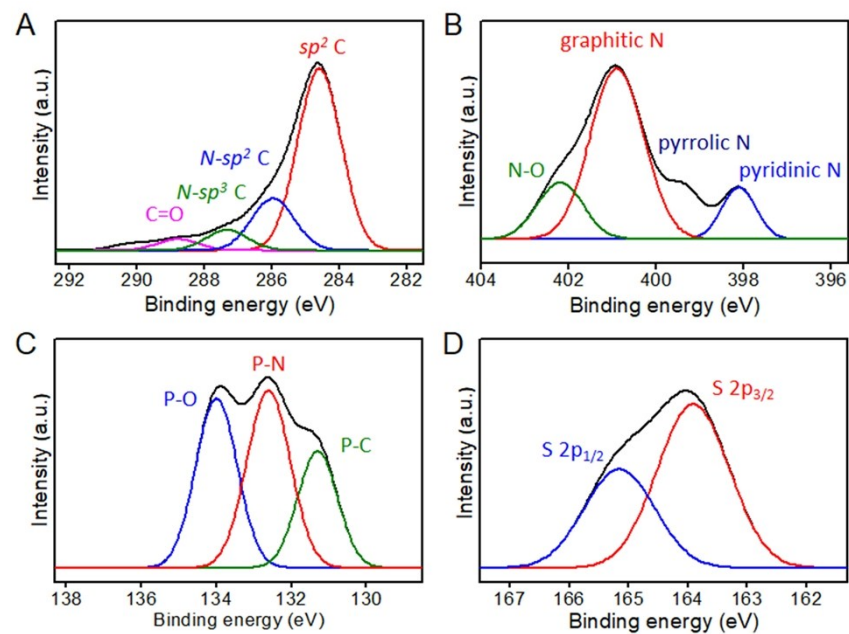
**Fig. S8** TGA traces of the GO, 2AT, PEI, PA, and GO-2AT-PEI-PA.



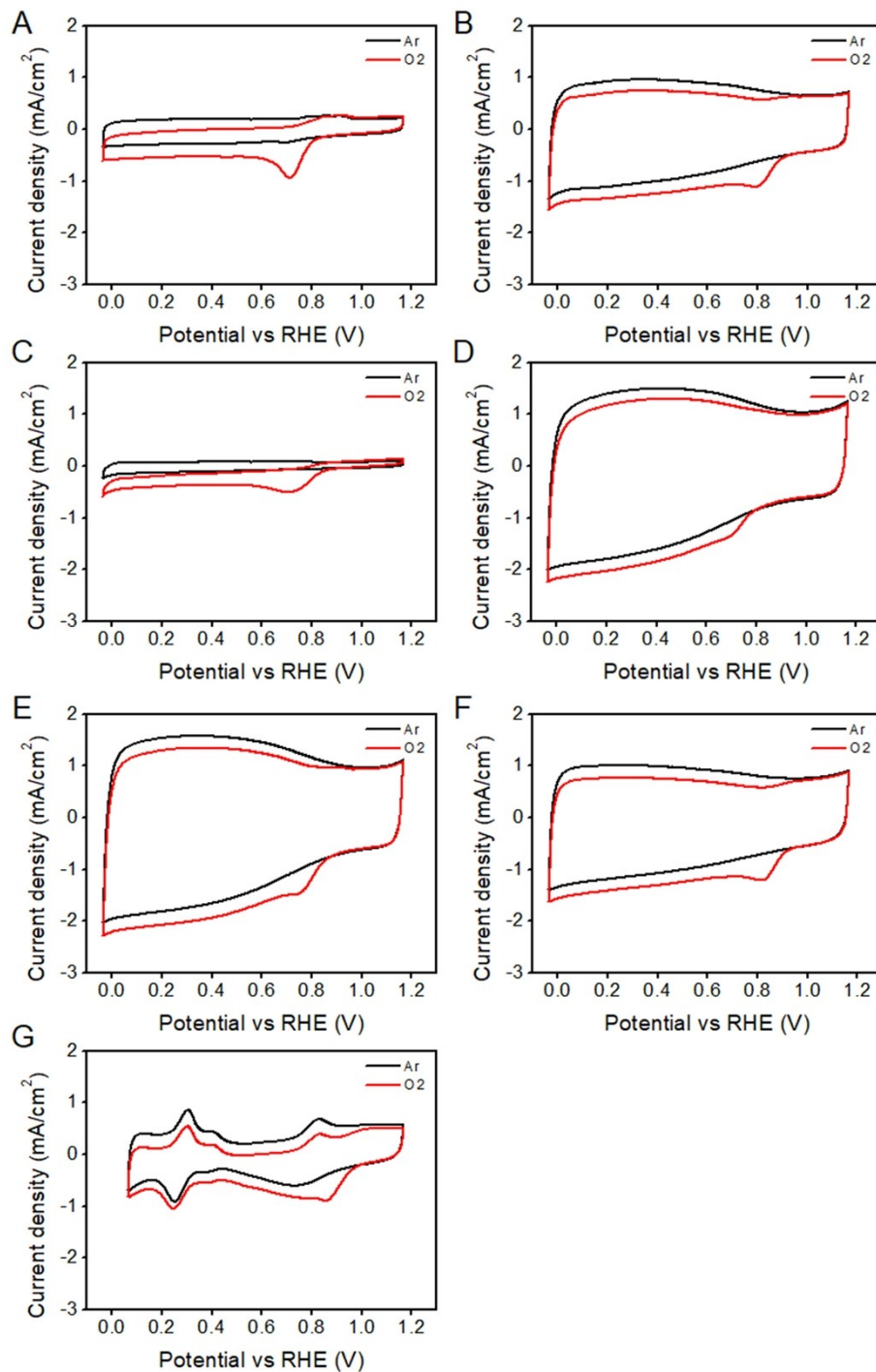
**Fig. S9** (A) C XPS 1s core level spectrum, (B) N XPS 1s core level spectrum, (C) P XPS 2p core level spectrum, and (D) S XPS 2p core level spectrum of TDGC-700.



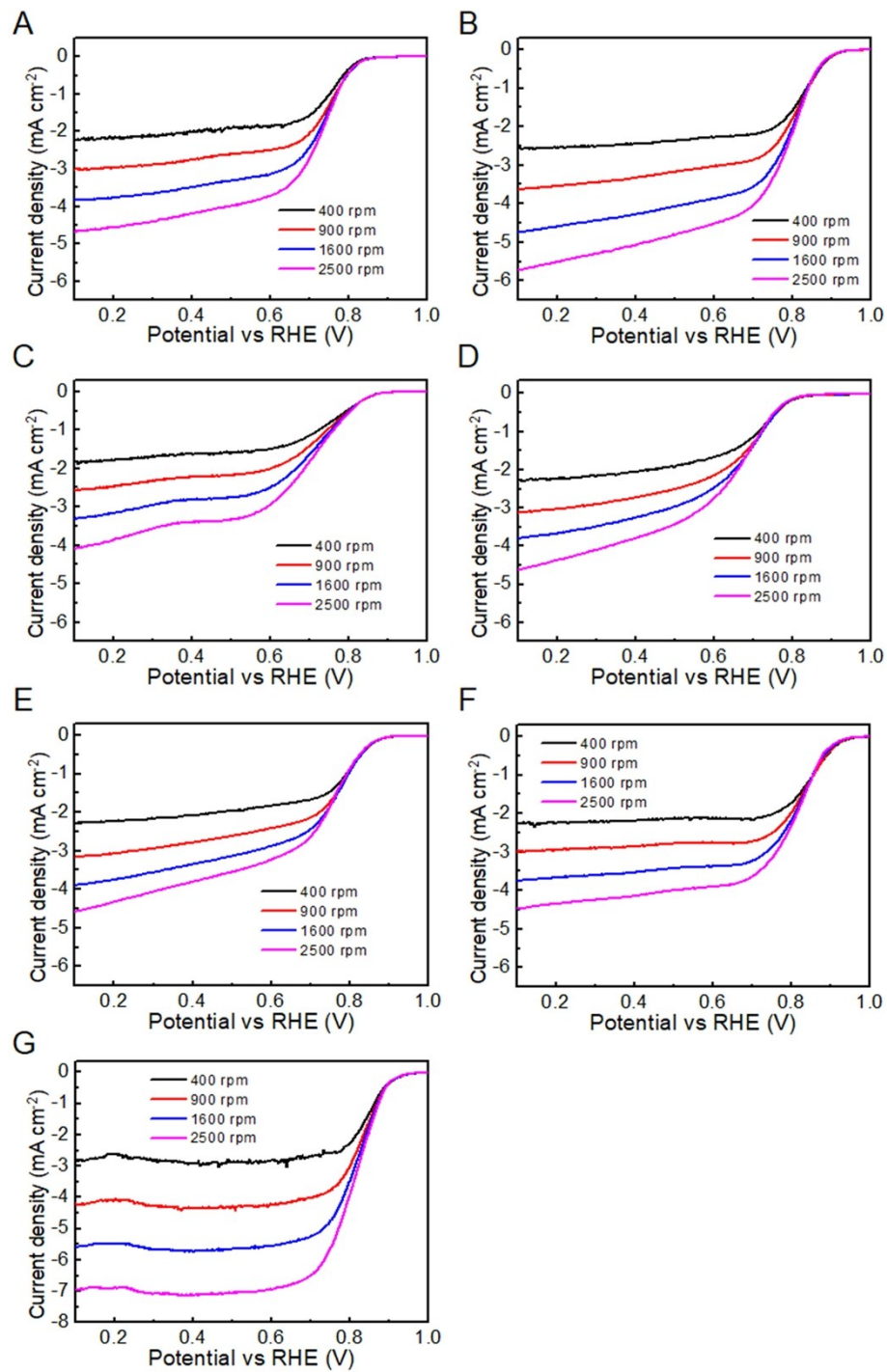
**Fig. S10** (A) C XPS 1s core level spectrum, (B) N XPS 1s core level spectrum, (C) P XPS 2p core level spectrum, and (D) S XPS 2p core level spectrum of TDGC-800.



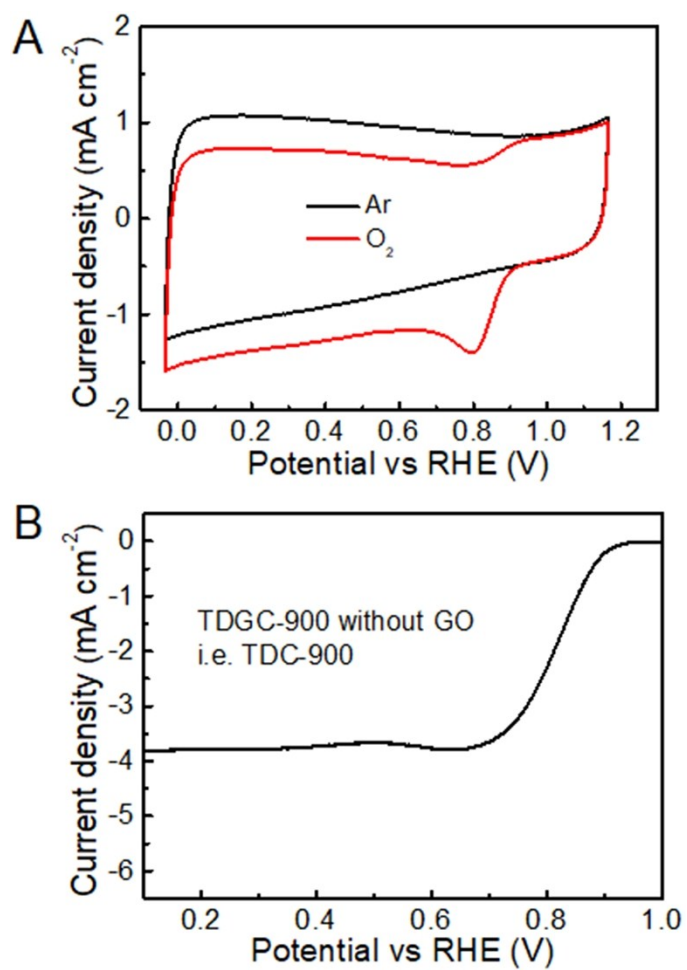
**Fig. S11** (A) C XPS 1s core level spectrum, (B) N XPS 1s core level spectrum, (C) P XPS 2p core level spectrum, and (D) S XPS 2p core level spectrum of TDGC-1000.



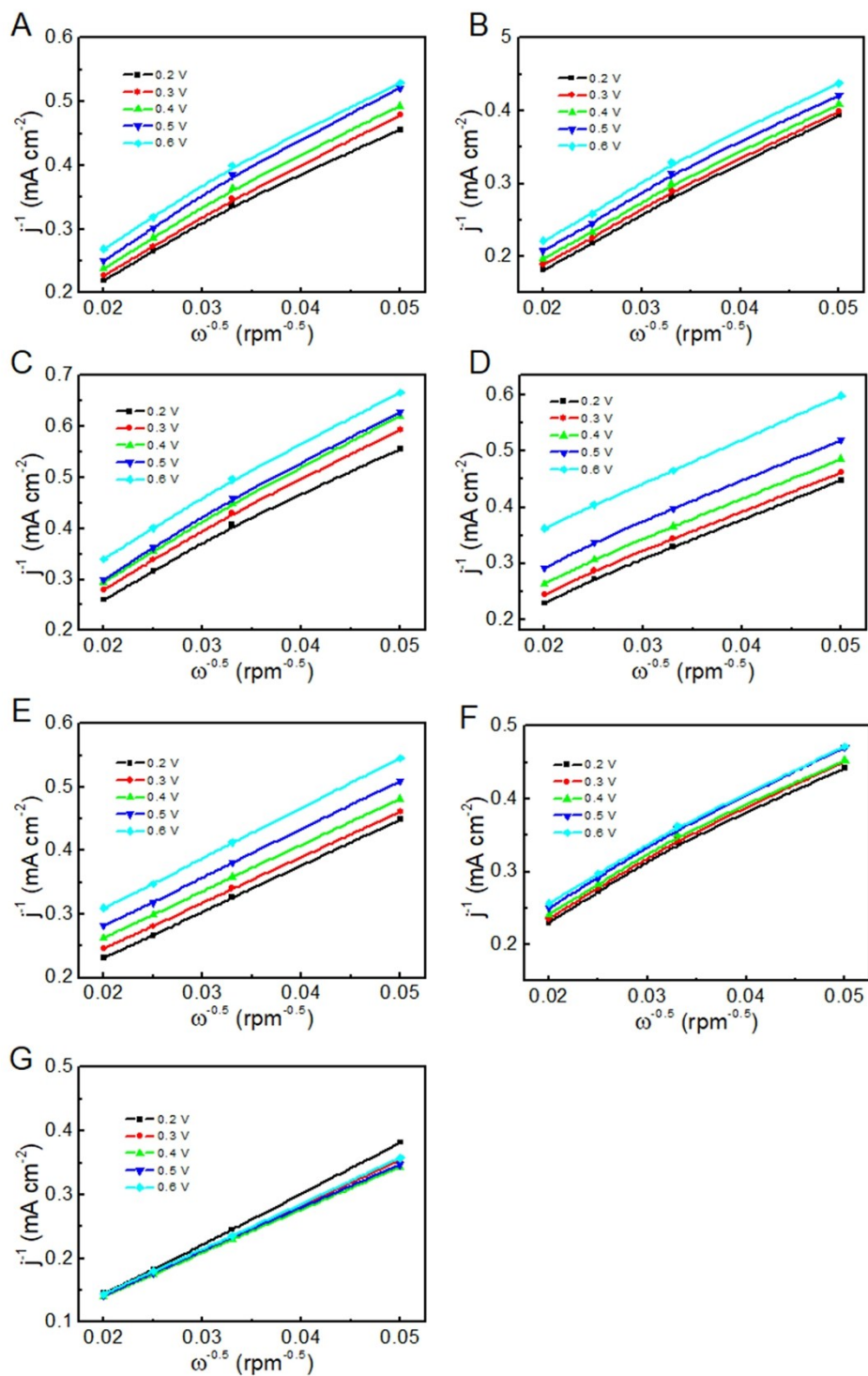
**Fig. S12** CV curves of (A) GN-900, (B) GNP-900, (C) GNS-900, (D) TDGC-700, (E) TDGC-800, (F) TDGC-1000 and (G) commercial Pt/C in Ar and O<sub>2</sub> saturated 0.1 M KOH solution.



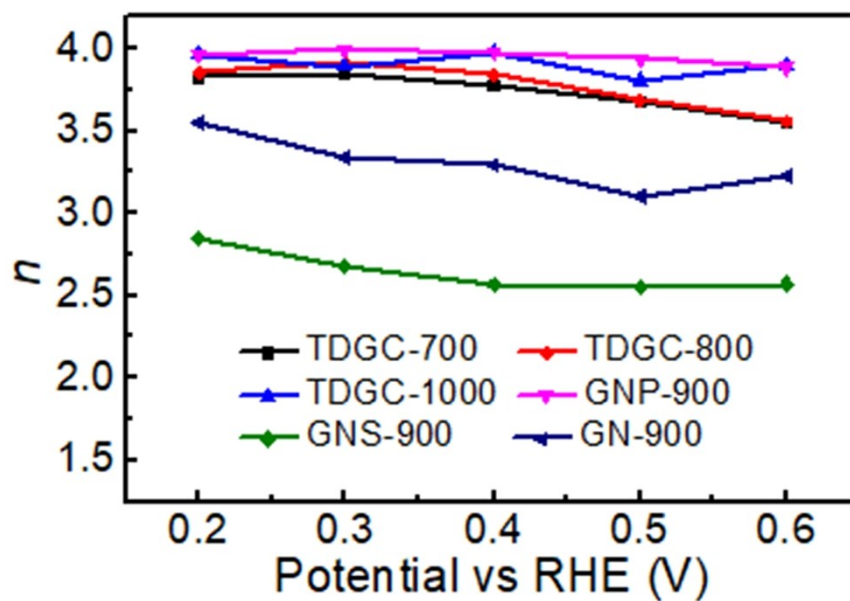
**Fig. S13** LSV curves of (A) GN-900, (B) GNP-900, (C) GNS-900, (D) TDGC-700, (E) TDGC-800, (F) TDGC-1000 and (G) commercial Pt/C in O<sub>2</sub> saturated 0.1 M KOH at various rotating speeds.



**Fig. S14** (A) CV curves in Ar and  $\text{O}_2$  saturated 0.1 M KOH solution and (B) LSV curve at an RDE (1,600 r.p.m.) in  $\text{O}_2$ -saturated 0.1 M KOH solution of TDGC-900 without GO.



**Fig. S15** K–L plots of (A) GN-900, (B) GNP-900, (C) GNS-900, (D) TDGC-700, (E) TDGC-800, (F) TDGC-1000 and (G) commercial Pt/C at various potentials.



**Fig. S16** Electron transfer number of TDGC-700, TDGC-800, TDGC-1000, GNP-900, GNS-900, and GN-900 at different potentials.

**Table S1.** BET specific surface area, pore diameter and pore volume of TDGC-700, TDGC-800, TDGC-900 and TDGC-1000.

Sample	BET specific surface area (m <sup>2</sup> g <sup>-1</sup> )	Pore diameter (nm)	Pore volume (cm <sup>3</sup> g <sup>-1</sup> )
TDGC-700	636	18, 50	0.31
TDGC-800	925	20, 51	0.71
TDGC-900	1107	5, 18, 118	0.87
TDGC-1000	1219	20, 55	0.65

**Table S2.** Oxygen reduction potential of GN-900, GNP-900, GNS-900, TDGC-700, TDGC-800, TDGC-900, TDGC-1000 and commercial Pt/C.

Sample	ORR potential (V vs. RHE)
GN-900	0.71
GNP-900	0.79
GNS-900	0.72
TDGC-700	0.71
TDGC-800	0.75
TDGC-900	0.82
TDGC-1000	0.80
Pt/C	0.85

**Table S3.** Quantification of different types of nitrogen in TDGC-700, TDGC-800, TDGC-900, and TDGC-1000.

Sample	Pyridinic N	Pyrrolic N	Graphitic N	N-oxide
TDGC-700	35.59	35.69	28.72	-
TDGC-800	23.30	21.71	40.37	14.62
TDGC-900	20.32	13.38	50.23	16.07
TDGC-1000	11.51	11.93	59.50	17.06

**Table S4.** Elemental composition of TDGC-700, TDGC-800, TDGC-900, and TDGC-1000.

Sample	C	O	N	P	S
TDGC-700	59.1	24.2	5.9	7.6	3.2
TDGC-800	75.2	16.0	3.1	3.6	2.1
TDGC-900	88.9	6.4	2.2	1.0	1.5
TDGC-1000	93.1	4.0	1.4	0.3	1.2

**Table S5.** Onset and half-wave potential of GN-900, GNP-900, GNS-900, TDGC-700, TDGC-800, TDGC-900, TDGC-1000 and commercial Pt/C.

Sample	Potential (V vs. RHE)	
	onset	half-wave
GN-900	0.87	0.75
GNP-900	0.94	0.81
GNS-900	0.89	0.72
TDGC-700	0.85	0.70
TDGC-800	0.90	0.77
TDGC-900	0.97	0.83
TDGC-1000	0.97	0.82
Pt/C	0.97	0.82