Supplementary information

Enhanced Electrochemical Oxygen Reduction Reaction by Restacking of N-doped Single Graphene Layers

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Table S1. Abbreviated names of the prepared catalysts according to further treatment steps of NGr.

Catalysts[1]	1st heat-treatment			2nd heat-treatment		
	H.T.[2]	materials	A.L.[3]	H.T.[2]	materials	A.L.[3]
NGr	-	-	-	-	-	-
NGr-H	O	-	-	-	-	-
NGr-H_M	O	Co-Fe	-	-	-	-
NGr-H_ML	O	Co-Fe	O	-	-	-
NGr-H_{DM}	O	Co-Fe/DCDA	-	-	-	-
NGr-H_{DM}L	O	Co-Fe/DCDA	O	-	-	-
NGr-$H_{DM}L H_{DM}$	O	Co-Fe/DCDA	O	O	Co-Fe/DCDA	-
NGr-$H_{DM}L H_{DM}$L	O	Co-Fe/DCDA	O	O	Co-Fe/DCDA	O

[1]The name of catalysts were abbreviated as treatment steps of reduced GO (NGr); heat-treatment without any additives (H), heat-treatment with metals (H_M), DCDA (H_D), or metal-DCDA composite (H_{DM}), and acid-leaching (L).

[2]H.T.: heat-treatment.

[3]A.L.: acid-leaching.

Table S2 Compositions and BET surface areas of the prepared catalysts.

catalysts	GO	NGr	NGr-H	NGr-H_M	NGr-H_{MD}	NGr-$H_{MD}LH_{MD}$
C[1]	65.2	83.7	95.0	93.3	90.5	91.4
O[1]	34.8	7.7	1.7	4.5	3.1	2.5
N[1]	-	8.6	3.3	2.2	6.4	6.1
O/C[2]	53.4	9.2	1.8	4.8	3.4	2.7
N/C[2]	-	10.3	3.5	2.4	7.1	6.7
Surface area	73	232	189	194	157	158

[1]at. %, [2]Atomic ratio (%), [3]m^2/g

Table S3 Electrochemical properties of the graphene derived catalysts for the ORRs in 1M $HClO_4$ electrolyte.

	NGr	NGr-H	NGr-H_M	NGr-H_{DM}	NGr-$H_{DM}LH_{DM}$
O. P.[1]	0.58	0.77	0.86	0.89	0.89
M. A.[2]	0	0.02	0.22	0.99	1.28
n[3]	3.58	3.29	3.89	3.92	3.88

[1]Onset potential (V, *vs.* RHE)

[2]Mass activity (mA mg^{-1}) at 0.75 V (V, *vs.* RHE)

[3]Number of electrons transferred at 0.4 V (V, *vs.* RHE)

Table S4 Proportion of various N-doping types in the graphene-derived catalysts obtained from the XPS results: pyridinic-N (N1); amide, amine, or pyrrolic-N (N2); graphitic-N (N3); and pyridinic-oxide (N4).

%	GO	NGr	NGr-H	NGr-H_M	NGr-H_{DM}	NGr-$H_{DM}LH_{DM}$
N1	-	26.7	50.4	54.4	60.8	59.5
N2	-	46.5	-	-	-	-
N3	-	46.6	35.3	31.9	27.3	28.7
N4	-	10.2	14.3	13.7	11.9	11.9

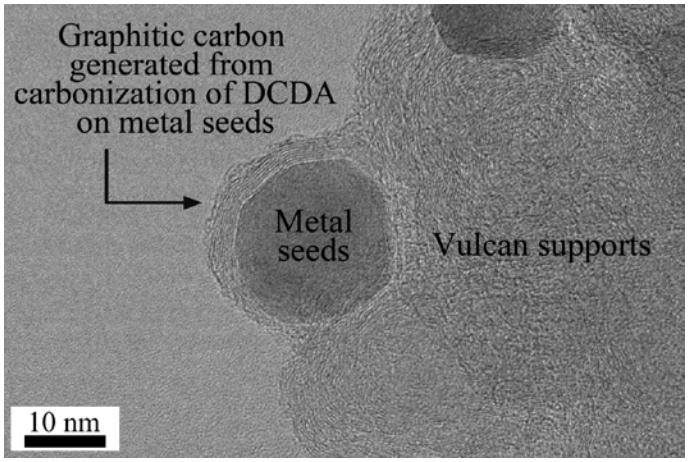

Fig. S1 TEM image of the graphitic carbon layers deposited on the Fe metal nanoparticles *via* carbonization of the DCDA on the metal seeds. This material was obtained *via* pyrolysis of the DCDA on Fe_2O_3/Vulcan XC-72R at 900 ^oC under an Ar flow, as reported previously (*Int J Hydrogen Energ* **37**, 4563-4570 (2012)).

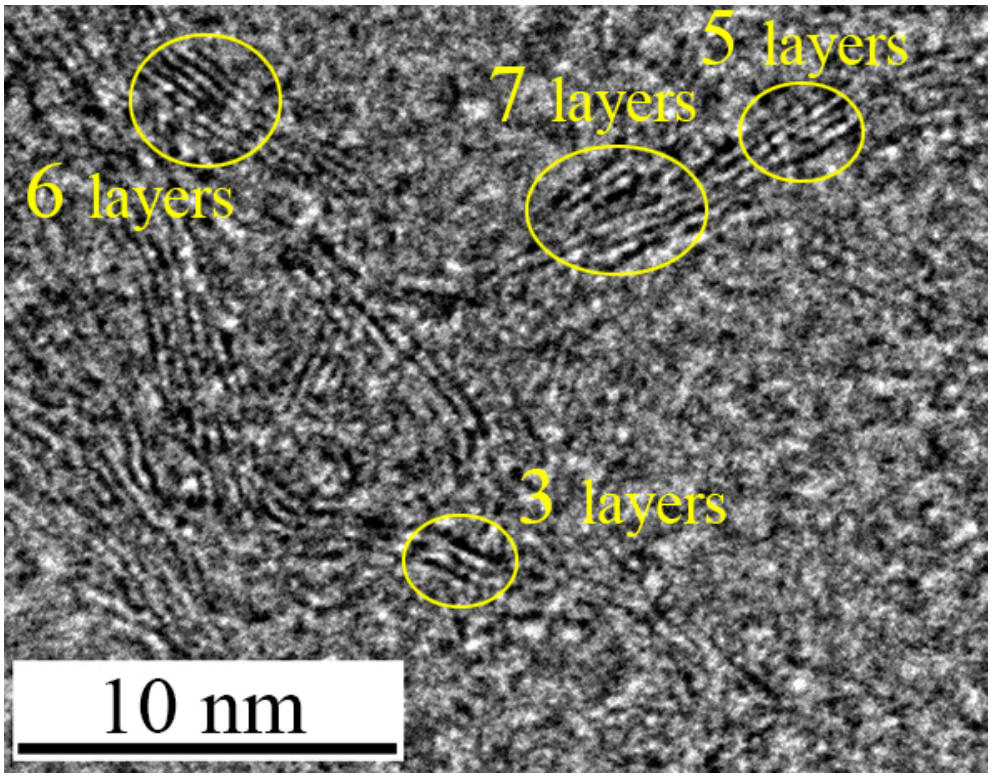

Fig. S2 TEM images of the NGr-$H_{DM}LH_{DM}$. Numbers in the figure indicate the number of graphene single layers in the catalysts indicated by red circles.

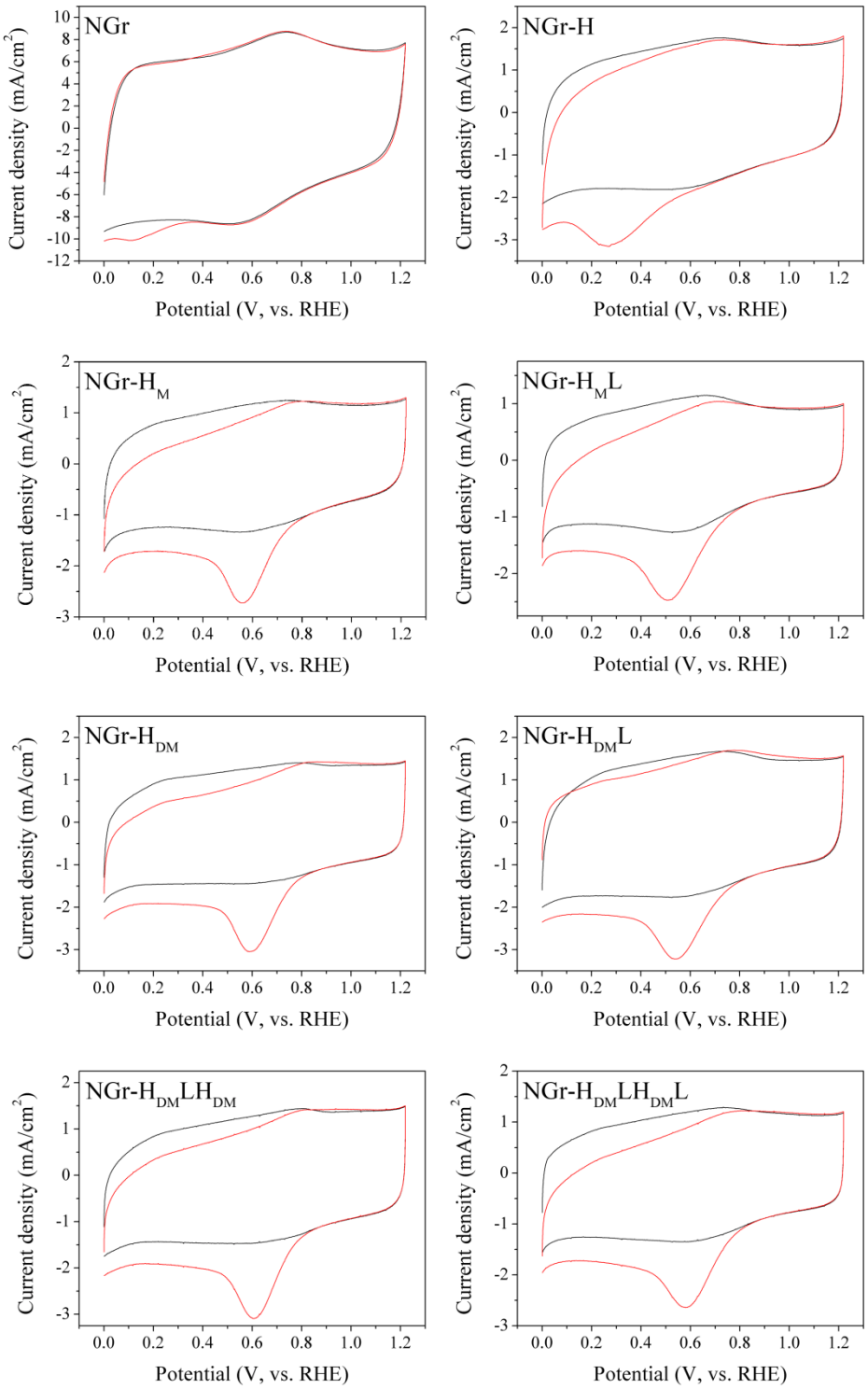

Fig. S3 Cyclic voltammetry results of the graphene-derived catalysts. The data were obtained in 1M $HClO_4$ purged with nitrogen (black line) or oxygen (red line) during 1h.

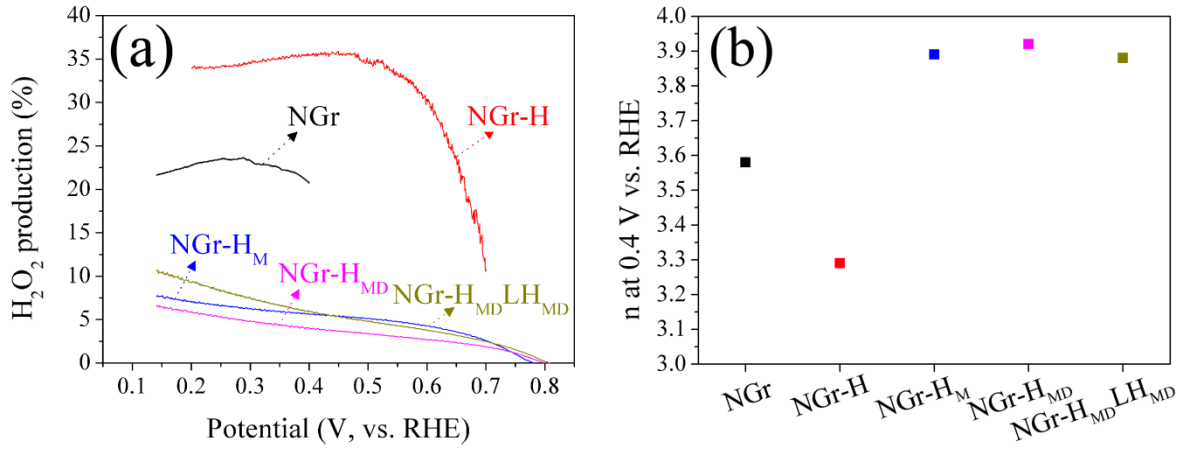

Fig. S4 The ORR pathway results of the prepared graphene-derived catalysts. (a) The H_2O_2 production yield during the ORRs obtained from the Pt-ring disk electrode, and (b) the number of electrons transferred in the ORRs at 0.4 V (*vs.* RHE).

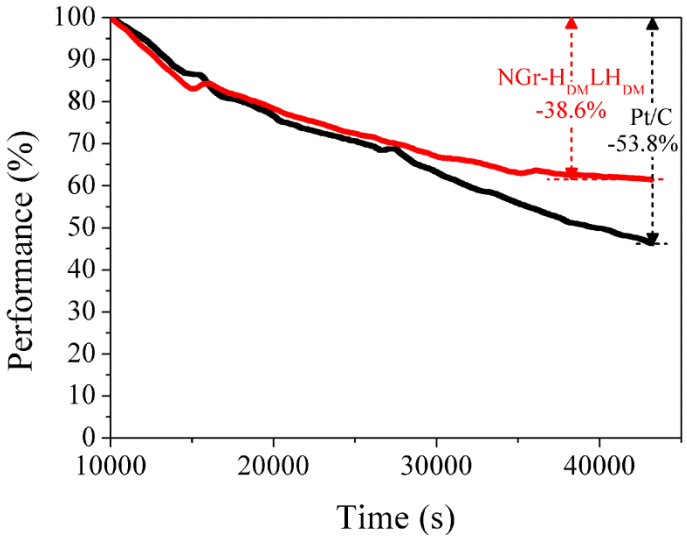

Fig. S5 Current-time chronoamperometric response (0.6 V, vs RHE) of NGr-$H_{DM}LH_{DM}$ and Pt/C for 10 h in 1M $HClO_4$ with continuous oxygen bubbling.

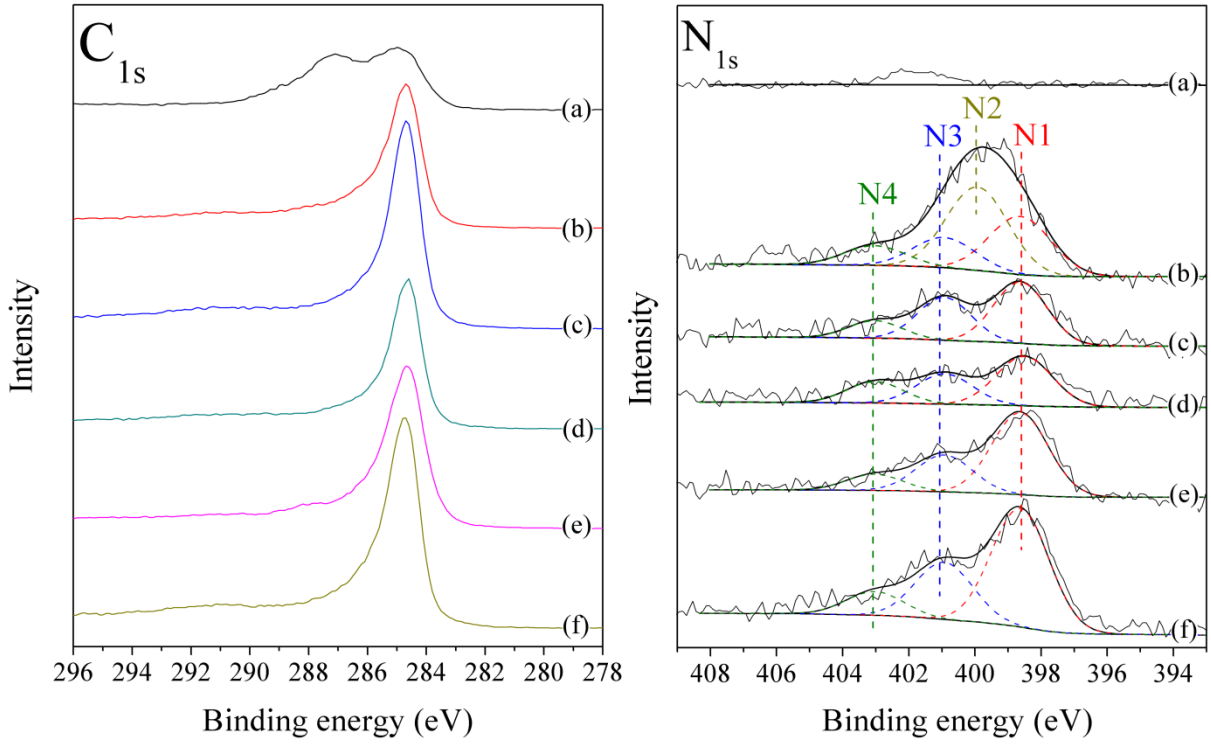

Fig. S6 XPS results of the prepared catalysts; (a) GO, (b) NGr, (c) NGr-H, (d) NGr-H_M, (e) NGr-H_{DM}, and (f) NGr-$H_{DM}LH_{DM}$. The XPS-N_{1s} results were deconvoluted with pyridinic-N (N1); amide, amine, or pyrrolic-N (N2); graphitic-N (N3); and pyridinic-oxide (N4).