## **Supporting Information**

## Boosting oxygen reduction activity of nano-graphene catalyst by charge redistribution at graphene-metal interface

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**Fig. S1.** TEM images and particle size distribution of (a) commercial Pd/C, (b) NGS\_ASP, (c) NGS@300, (d) NGS@500, (e) NGS@700, and (f) NGS@900.



Fig. S2. TEM image and particle size distribution of commercial Pt/C catalyst.



**Fig. S3.** Nitrogen adsorption-desorption isotherms of (a) NGS@900, (b) Pt/C, and (c) carbon support.

**Table S1.** Specific surface areas and total pore volumes of NGS@900, Pt/C, and carbon support(Vulcan carbon) obtained by Brunauer-Emmett-Teller (BET) analysis.

Sample	NGS@900	Pt/C	Carbon support
Specific surface area (m <sup>2</sup> /g)	163	204	244
Total pore volume $(cm^3/g)$	0.32	0.39	0.46



Fig. S4. XRD patterns of commercial Pd/C and NGS catalysts.



Fig. S5. (a) CVs and (b) CO stripping curves of NGS catalysts.



Fig. S6. Pd 3d core-level XPS spectra of NGS catalysts.



**Fig. S7.** (a) CVs and (b) CO stripping curves of commercial Pd/C and allyl mercaptan-modified Pd/C catalysts.

## **Koutecky-Levich plots:**

The number of electrons transferred for ORR on NGS@900 and Pt/C catalysts was calculated according to the Koutecky-Levich equation:

$$1/J = 1/J_D + 1/J_K = 1/(B\omega^{1/2}) + 1/J_K$$

$$B = 0.62 n F C_0 D_0^{2/3} v^{-1/6}$$

where  $J_D$  is the diffusion-limiting current,  $J_K$  is the kinetic current, F is the Faraday constant (96485 C mol<sup>-1</sup>),  $D_0$  is the diffusion coefficient of oxygen in 0.1 M KOH ( $1.9 \times 10^{-5} \text{ cm}^2 \text{ s}^{-1}$ ), v is the kinematic viscosity of electrolyte ( $1.09 \times 10^{-2} \text{ cm}^2 \text{ s}^{-1}$ ),  $C_0$  is the bulk concentration of oxygen in 0.1 M KOH ( $1.2 \times 10^{-6} \text{ mol cm}^{-3}$ ),  $\omega$  is the angular velocity (rad s<sup>-1</sup>), and n is the electron transfer number for the ORR.



Fig. S8. (a) LSV curves of NGS@900 at different rotation speed (400 to 2500 rpm) in O<sub>2</sub>-saturated 0.1 M KOH at a scan rate of 5 mV s<sup>-1</sup>. (b) Koutecky-Levich plots for NGS@900 catalyst at different potentials.



Fig. S9. (a) LSV curves of Pt/C at different rotation speed (400 to 2500 rpm) in  $O_2$ -saturated 0.1 M KOH at a scan rate of 5 mV s<sup>-1</sup>. (b) Koutecky-Levich plots for Pt/C catalyst at different potentials.



**Fig. S10.** (a) CVs and (b) CO stripping curves and (c) change in the metal surface areas of commercial Pt/C and NSG@900 before and after ADTs.

![](_page_12_Figure_0.jpeg)

**Fig. S11** ORR polarization curves of NGS@900 and Pt/C in 0.1 M KOH without (solid line) and with (dotted line) 0.5 M MeOH.