

Supporting online information

N-doped Graphene Nanosheets as Metal-Free Catalysts of  
Air-Electrode in Li-air Fuel cell under Acidic Electrolyte

Eunjoo Yoo<sup>a</sup>, Junji Nakamura<sup>b</sup>, Haoshen Zhou<sup>a\*</sup>

<sup>a</sup>Energy Technology Research Institute, National Institute of Advanced Industrial Science and Technology, Umezono 1-1-1, Central 2, Tsukuba, Ibaraki 305-8568, Japan

<sup>b</sup> Graduate School of Pure and Applied Science, University of Tsukuba, Tsukuba, Ibaraki 305-8573, Japan

Corresponding author. TEL/FAX: +81-29-861-5648/+81-29-861-3489  
E-mail address: hs.zhou@aist.go.jp

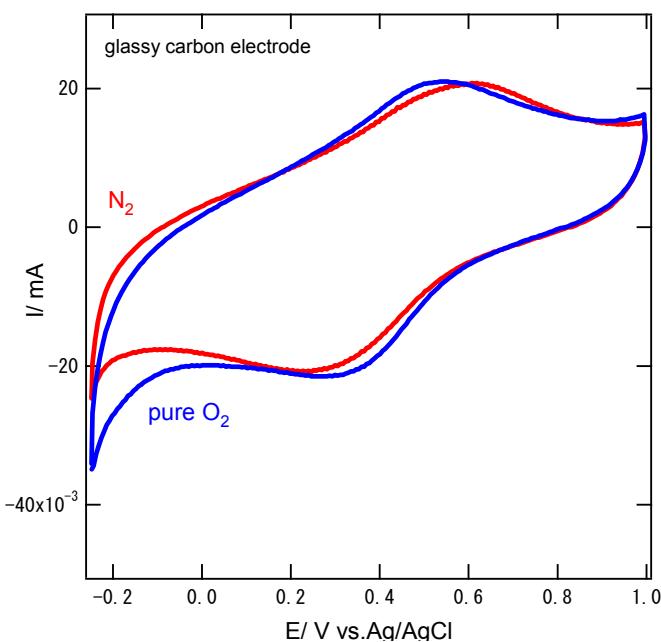


Fig. S1 the cyclic voltammetry curves for ORR at a glassy carbon electrode in  $1 \text{ moldm}^{-3} \text{ Li}_2\text{SO}_4 + 0.5 \text{ moldm}^{-3} \text{ H}_2\text{SO}_4$  electrolyte, rotating rate after  $\text{N}_2$  and pure  $\text{O}_2$  bubbling for 30 min: 500 rpm, scan rate:  $20 \text{ mVs}^{-1}$ .

Figure S1 shows the cyclic voltammetry curves for ORR at a glassy carbon electrode. There is no significantly difference of activity of ORR in the both gas at a glassy carbon electrode. Thus, it indicated that a glassy carbon electrode didn't have catalytic activity toward ORR under the acidic condition.

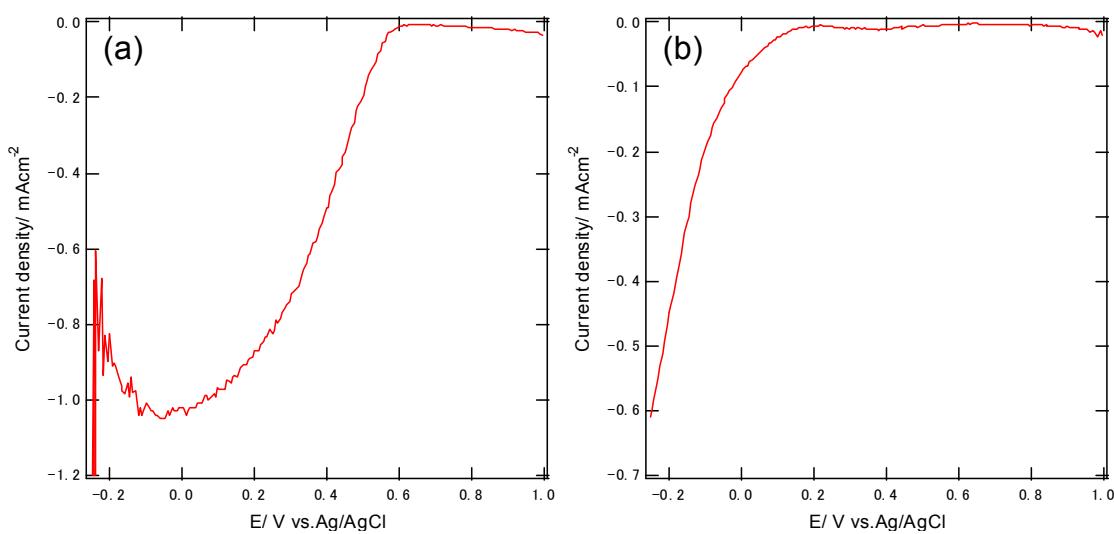


Fig. S2 the steady-state voltammograms of 20 wt%Pt/CB (a) and the GNSs (b) in 1  $\text{mol dm}^{-3}$   $\text{Li}_2\text{SO}_4 + 0.5 \text{ mol dm}^{-3}$   $\text{H}_2\text{SO}_4$  electrolyte, scan rate: 20  $\text{mV s}^{-1}$ , rotating rate: 500 rpm.

Figure S2(a) reveals the steady-state voltammograms of 20 wt%Pt/CB in the  $\text{O}_2$  saturated 1  $\text{mol dm}^{-3}$   $\text{Li}_2\text{SO}_4 + 0.5 \text{ mol dm}^{-3}$   $\text{H}_2\text{SO}_4$  electrolyte. The onset potential for oxygen reduction reaction (ORR) is at about 0.62 V *versus* Ag/AgCl for the 20 wt%Pt/CB. Fig. S2(b) also shows the steady-state voltammograms of GNSs. The onset potential for ORR of GNSs is at 0.2 V *versus* Ag/AgCl.

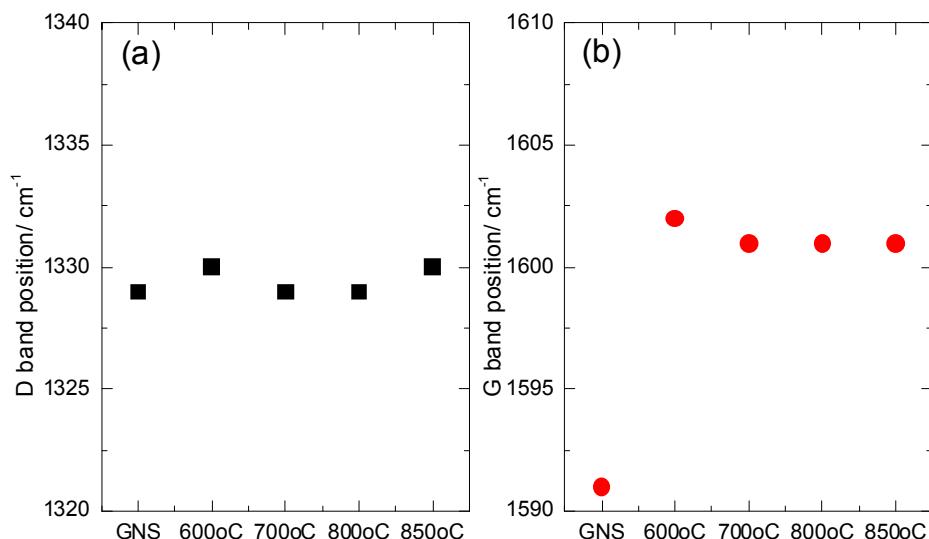


Fig. S3 the shift of D band (a) and G band (b) for GNSs and all N-doped GNSs as a function of nitrogen doping temperature.

Figure S3 shows the degree of shift for the D and G band as a function of nitrogen doping temperature by calculated for Raman spectra. As shown in Fig. S3(a), there is not observed the shift of the D-band for all measured sample. However, the G band of all N-doped GNSs was upshift by about 10  $\text{cm}^{-1}$  that of the GNSs.

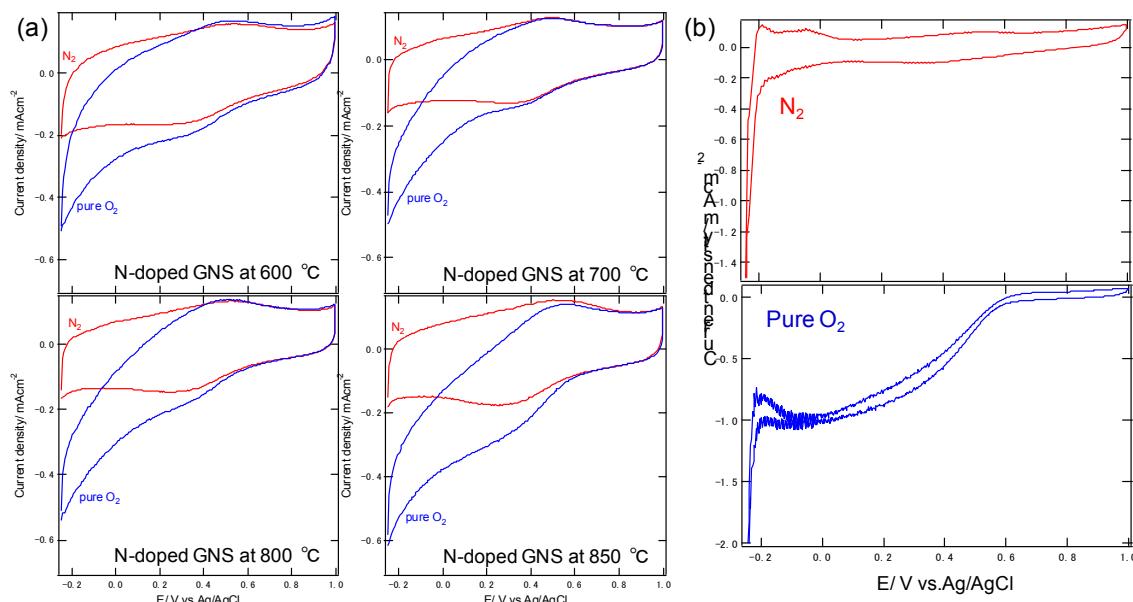


Fig. S4 Cyclic Voltammograms of the N-doped GNSs with different doping temperature (a) and 20wt%Pt/CB (b) in N<sub>2</sub> saturated (red line) or O<sub>2</sub> saturated (blue line) 1 mol dm<sup>-3</sup> Li<sub>2</sub>SO<sub>4</sub> + 0.5 mol dm<sup>-3</sup> H<sub>2</sub>SO<sub>4</sub>, scan rate: 20 mV s<sup>-1</sup>, rotating rate: 500 rpm.

Figure S4 shows the Voltammograms of the N-doped GNSs with different doping temperature (a) and 20wt%Pt/CB (b) in N<sub>2</sub> saturated (red line) or O<sub>2</sub> saturated (blue line) 1 mol dm<sup>-3</sup> Li<sub>2</sub>SO<sub>4</sub> + 0.5 mol dm<sup>-3</sup> H<sub>2</sub>SO<sub>4</sub>. The ORR current of all N-doped GNSs enhanced in O<sub>2</sub> saturated 1 mol dm<sup>-3</sup> Li<sub>2</sub>SO<sub>4</sub> + 0.5 mol dm<sup>-3</sup> H<sub>2</sub>SO<sub>4</sub>. For the 20wt%Pt/CB, as shown in Fig S4(b), the ORR current increased 5 times in O<sub>2</sub> saturated condition for comparison in N<sub>2</sub> saturated condition.

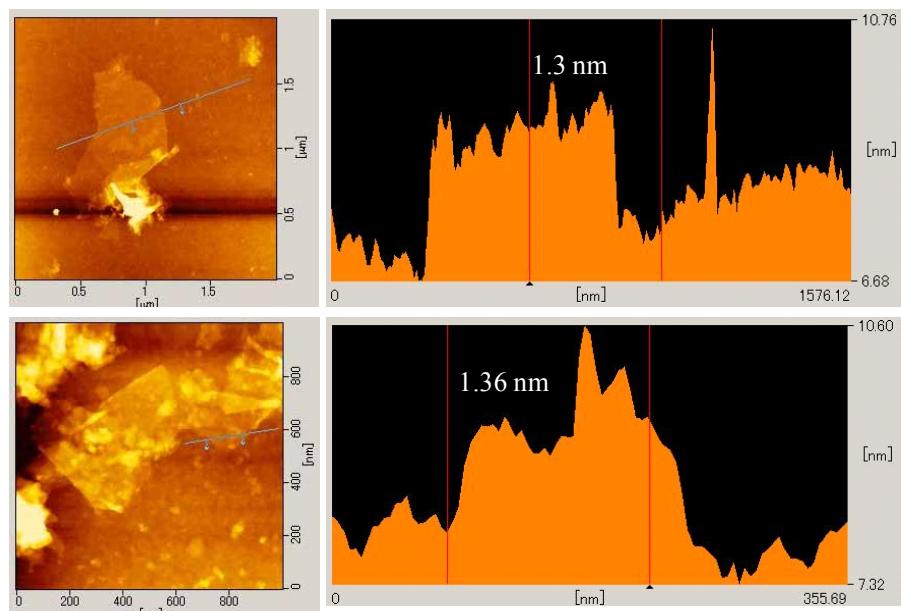


Fig. S5 AFM images of graphene nanosheets.

Figure S5 shows the AFM images of graphene nanosheets. The thickness of graphene nanosheets can be observed in the AFM images. It was found that the graphene nanosheets thickness ranged from 1.3 to 1.4 nm, which corresponds to an approximately about 4-5 layers, according to an interlayer spacing of 0.34 nm for graphite.