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Supporting Information (SI)

Novel Graphene-Like Nanosheets Supported Highly Active Electrocatalysts with Ultralow Pt Loadings for Oxygen Reduction Reaction

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Additional results:

The average size of this $Pd_{10}Pt_1$ NPs was measured to be 4.36 nm by the TEM measurement from 60 randomly selected nanoparticles **(Figure S1)**. The result is well in accordance with that of XRD analysis.

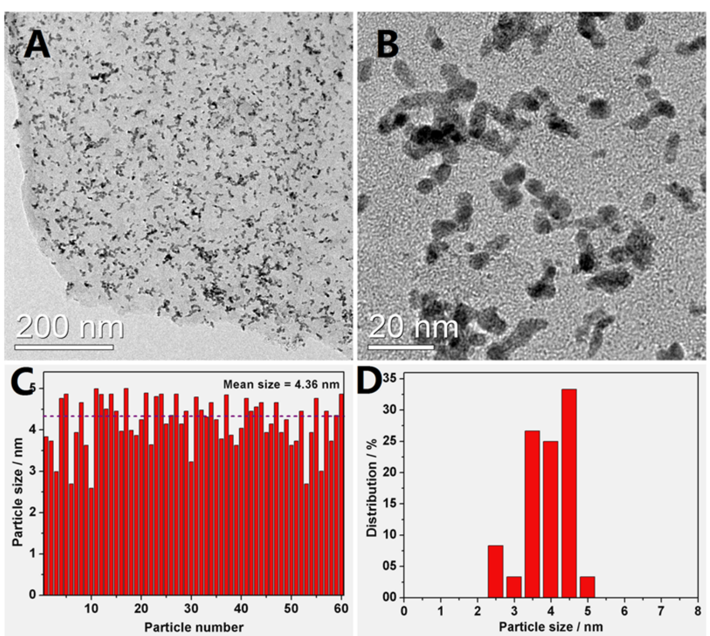

Figure S1. Particle size distribution of $Pd_{10}Pt_1$ NPs by TEM measurement.

The analysis of energy-dispersive X-ray spectroscopy (EDS) based on a selected area TEM image is presented in **Figure S2**. The EDS measurement confirmed that this selected area contains elements of C, O, Pd, Pt and Cu with mass ratio of 12.3 : 1.87 : 44.01 : 4.70 : 37.39 and atomic ratio of 46.71 : 5.44 : 19.28 : 1.13 : 27.44. The mass ratio of Pd and Pt from EDS is 9.36, which is a little lower than that of ICP-AES analysis (29.19 wt.% Pd + 2.85 wt.% Pt, the mass ratio of Pd and Pt = 10.24).

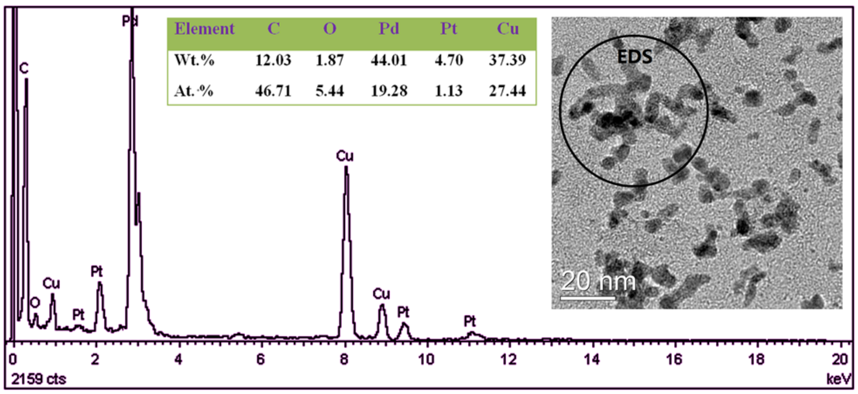

Figure S2. TEM selected-area EDS analysis of the $Pd_{10}Pt_1$/AGNs.

$Pd_{10}Pt_1$/RGO catalyst with the same metal loadings was also prepared by using reduced graphene oxide (RGO) as a support for comparison. **Figure S3**A is the XRD pattern of $Pd_{10}Pt_1$/RGO and **Figure S3**B is the TEM image of $Pd_{10}Pt_1$/RGO. It is shown in **Figure S3**A that the diffraction peak the RGO (2theta=20-30 degrees) is very broad due to their partially restored graphitic structures. **Figure S3**B shows the massive $Pd_{10}Pt_1$ nanoparticles and slightly agglomerated on the RGO. **Figure S3**C shows the ORR performance of $Pd_{10}Pt_1$/AGNs compared with $Pd_{10}Pt_1$/RGO and the **Figure S3**D shows the Nyquist plots obtained from the electrochemical impedance spectroscopy (EIS) of $Pd_{10}Pt_1$/RGO and $Pd_{10}Pt_1$/AGNs.

The mass activity (i_m) of the $Pd_{10}Pt_1$/RGO catalyst at 0.9 V was only 870 mA mg^{-1}_{Pt}, which is much lower than that of $Pd_{10}Pt_1$/AGNs catalysts (1934 mA mg^{-1}_{Pt}) (see **Figure S3**C). **Figure S3**D gives the evidence that the ohmic resistance of the $Pd_{10}Pt_1$/AGNs is much lower than that of $Pd_{10}Pt_1$/RGO measured by electrochemical impedance spectroscopy (EIS). The larger semicircles (larger charge transfer resistance (CTR)) and the larger x-intercept (larger equivalent series resistance (ESR)) were observed on $Pd_{10}Pt_1$/RGO, whereas, only small semicircle and x-intercept were shown for $Pd_{10}Pt_1$/AGNs. Therefore, it is reasonable that the $Pd_{10}Pt_1$/AGNs catalyst shows much higher activity (over twofold) than the $Pd_{10}Pt_1$/RGO catalyst, owing to the outstanding supporting effect and the better electronic conductivity of the AGNs.

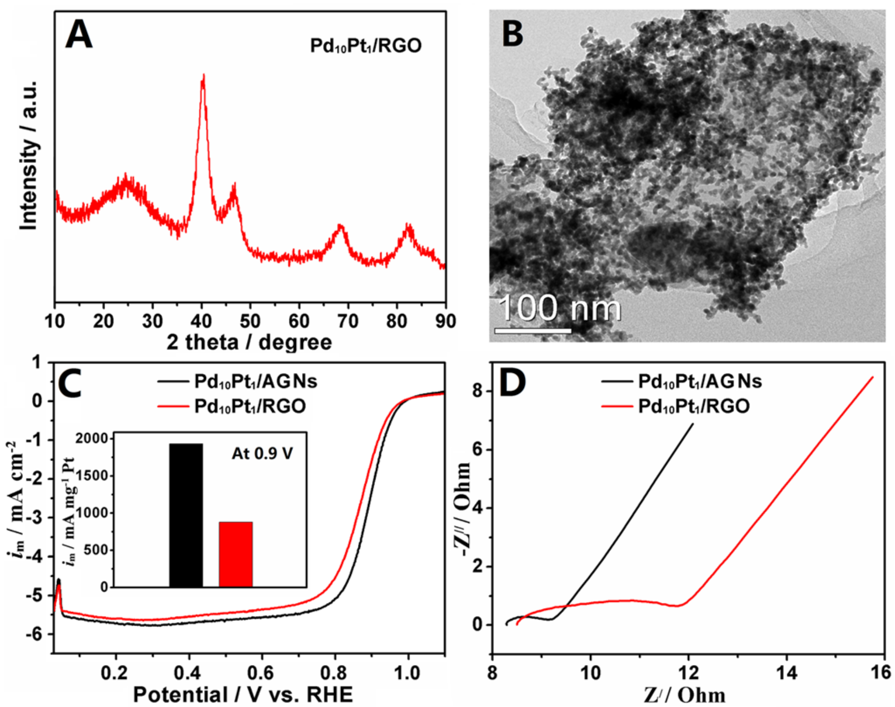

Figure S3. (A) XRD pattern of $Pd_{10}Pt_1$/RGO, (B) TEM image of $Pd_{10}Pt_1$/RGO, (C) ORR curves of $Pd_{10}Pt_1$/RGO and $Pd_{10}Pt_1$/AGNs, and (D) Nyquist plots obtained from the EIS of $Pd_{10}Pt_1$/RGO and $Pd_{10}Pt_1$/AGNs.

The stability of the commercial Pt/C catalyst much lower as shown in **Figure S4**. It lost about 30 % activity after 1,000 cycles which larger than that of the $Pd_{10}Pt_1$/AGNs after 10,000 cycles.

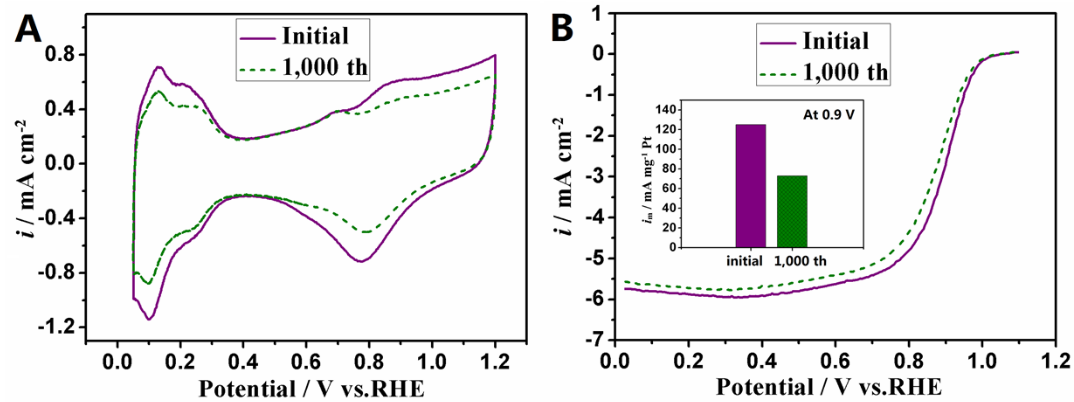

Figure S4. CV curves (A) and ORR curves (B) of commercial Pt/C catalyst (46.7 wt.%Pt, TKK, Japan).