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 Pd₁₀Pt₁ 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 Pd₁₀Pt₁/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 Pd₁₀Pt₁/RGO catalyst at 0.9 V was only 870 mA mg⁻¹Pt, which is much lower than that of Pd₁₀Pt₁/AGNs catalysts (1934 mA mg⁻¹Pt) (see **Figure S3**C). **Figure S3**D gives the evidence that the ohmic resistance of the Pd₁₀Pt₁/AGNs is much lower than that of Pd₁₀Pt₁/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).