

Supporting Information

Surfactant-free Synthesis of Reduced Graphene Oxide Supported Well-defined Polyhedral Pd-Pt Nanocrystals for Oxygen Reduction Reaction

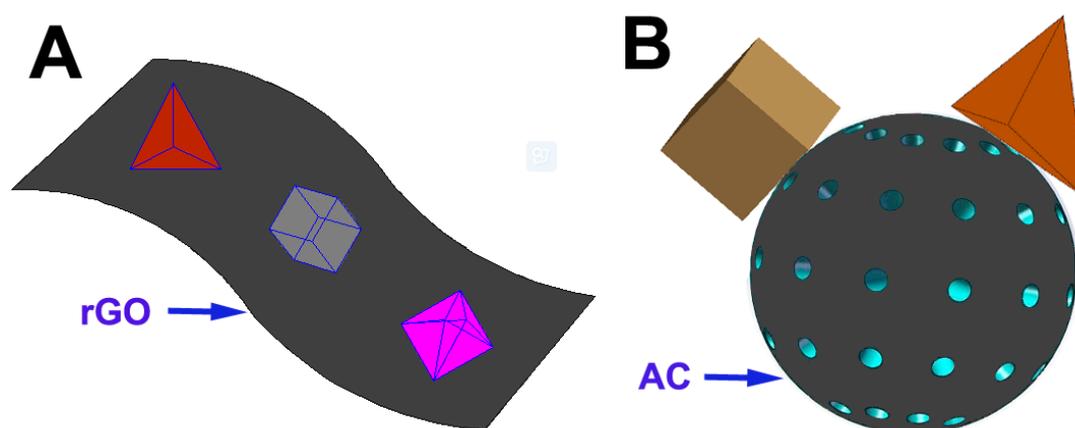
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1. Supplementary Figures



Scheme S1. The effect of the surface contact between the nanocrystals and the substrate on the elastic tension of the edges and corners.

Discussion for Scheme S1: As shown in **Scheme S1A**, when the surface was smooth for the rGO nanosheets, the polyhedral nanocrystals can be anchored on the substrate by fully contact. In contrast, when the surface was bend and rough for the spherical carbon materials (**Scheme S1B**), the nanocrystals would be not contact fully with the carbon substrate, which will make the edges and corners be hung and increase the elastic tension of Pd-Pt nanocrystals.

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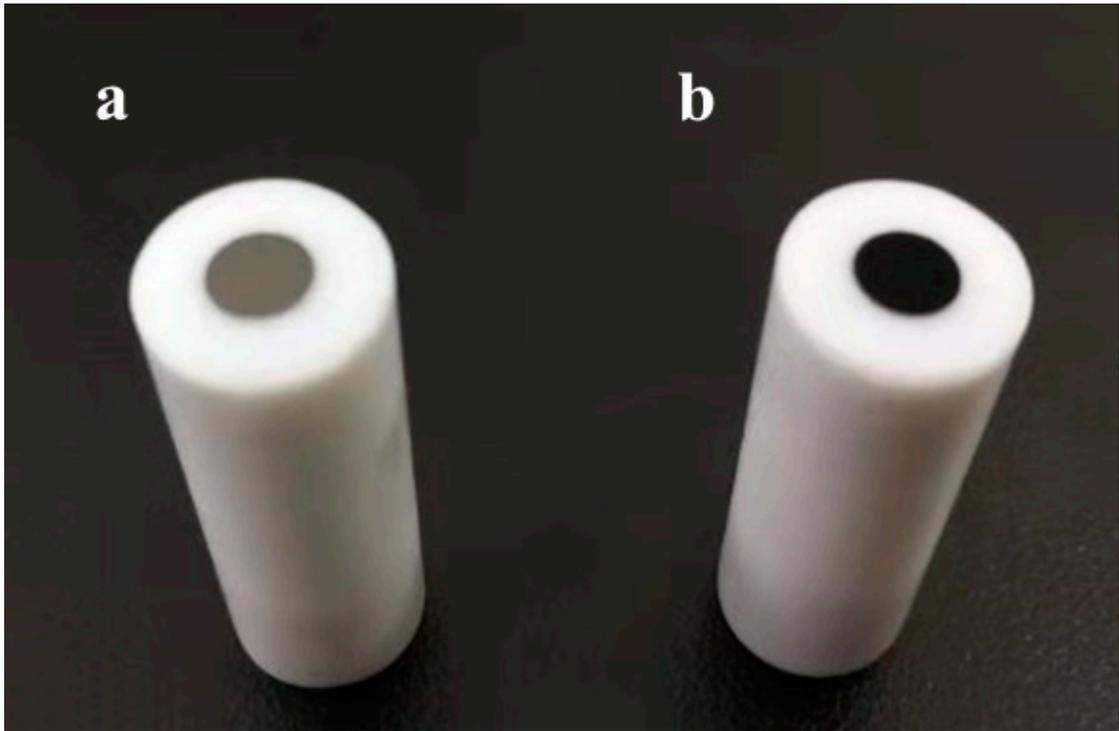


Figure S1. Photographs of the bare glass carbon electrode (GC) and the catalysts coated GC.

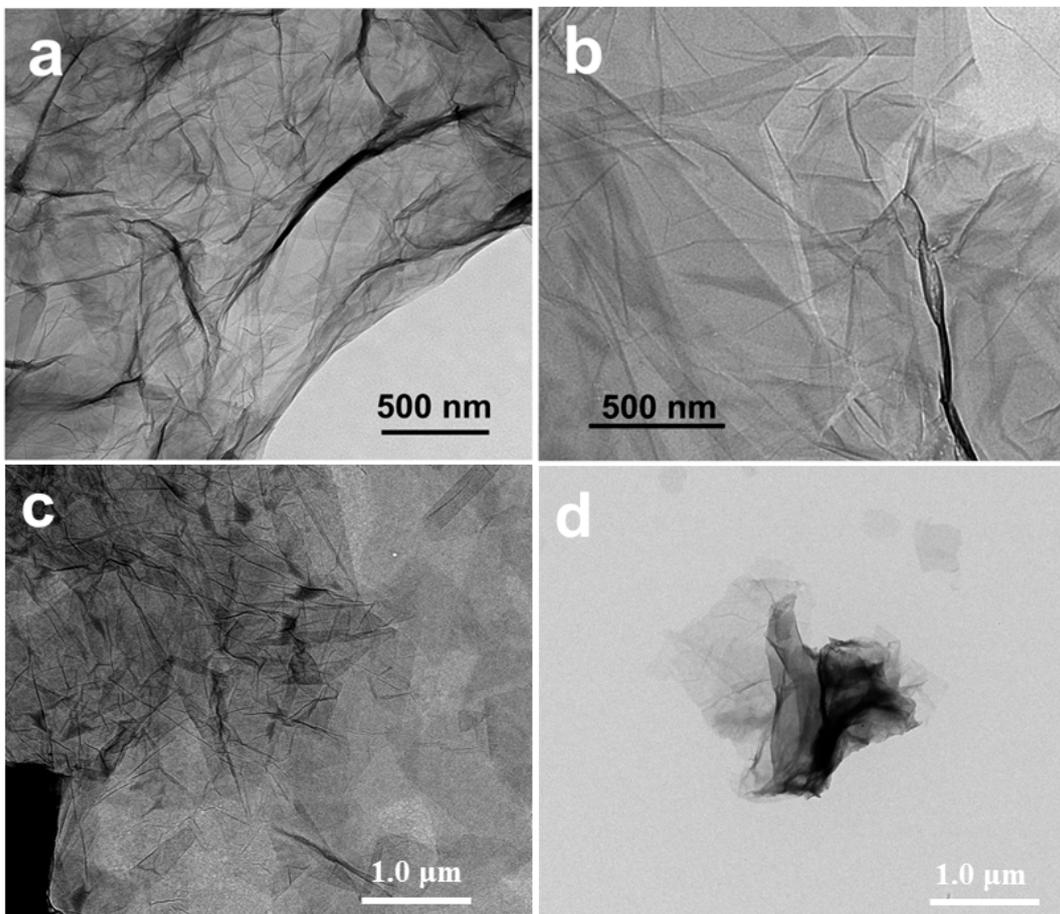


Figure S2. TEM images of the (a, c) GO500-1 and (b, d) GO3500-1 substrates with (a, b) high magnification and (c, d) low magnification.

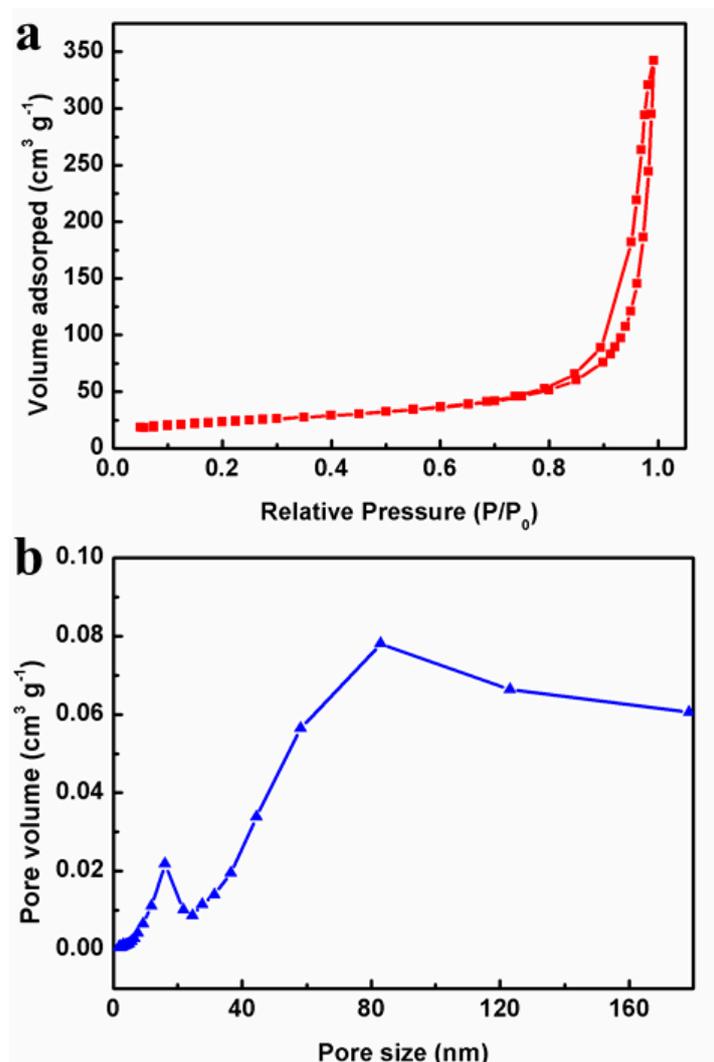


Figure S3. N₂ adsorption-desorption isotherms (a) and pore size distribution of the rGO500-3 substrate.

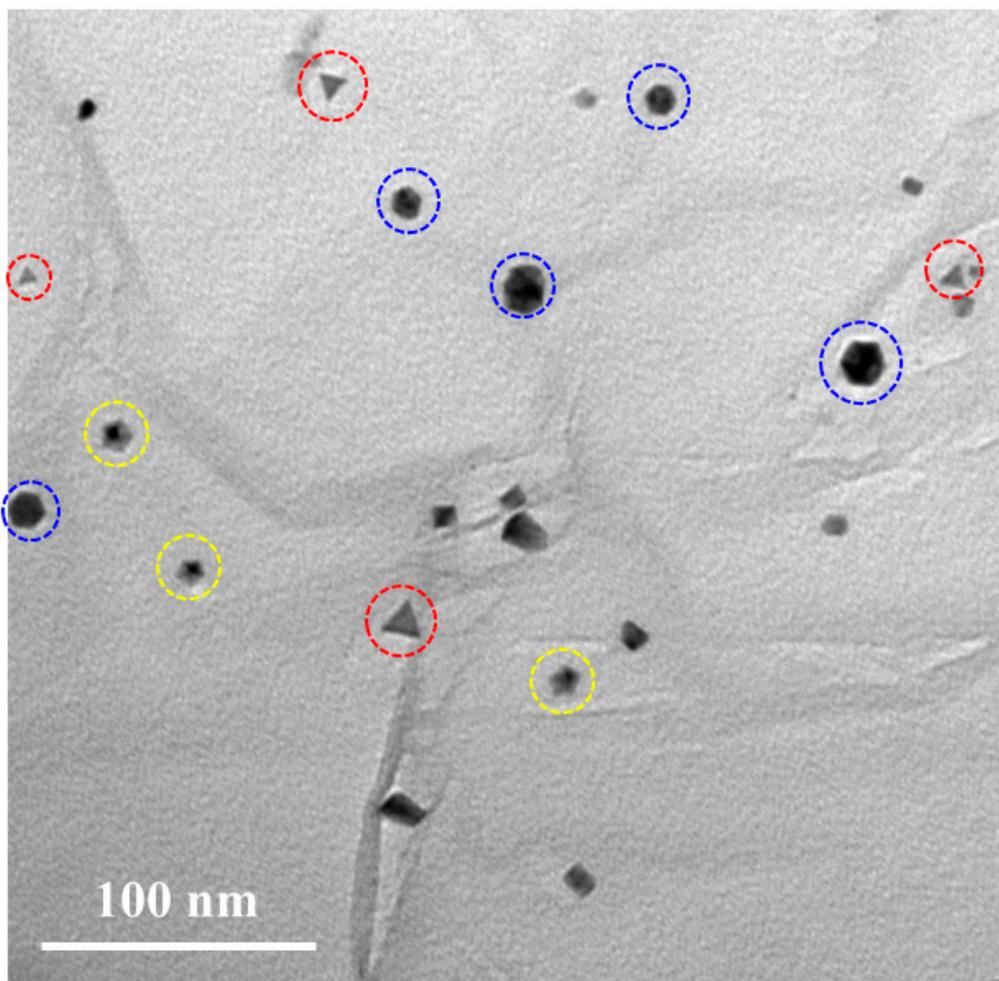


Figure S4. Magnification TEM image of Pd₆Pt₁/rGO500-1 electrocatalyst, corresponding to the **Figure 1a**. The red, blue and yellow dashed line circles represent for the tetrahedral, octahedral and five-fold twinned nanocrystals.

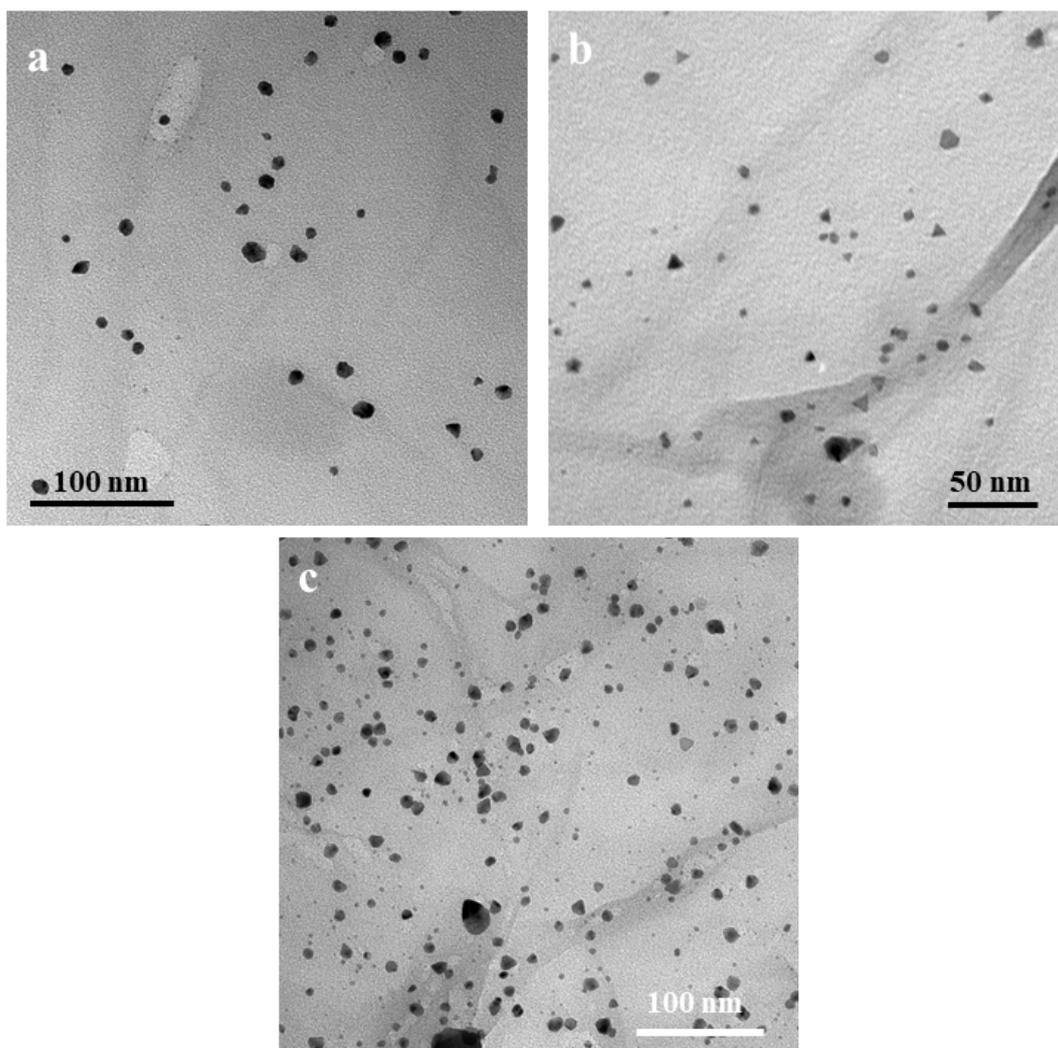


Figure S5. Magnification TEM images of the (a) Pd₆Pt₁/rGO500-3, (b) Pd₆Pt₁/rGO3500-1 and (c) Pd₆Pt₁/rGO3500-3 electrocatalysts, corresponding to the **Figure 2a-2c**.

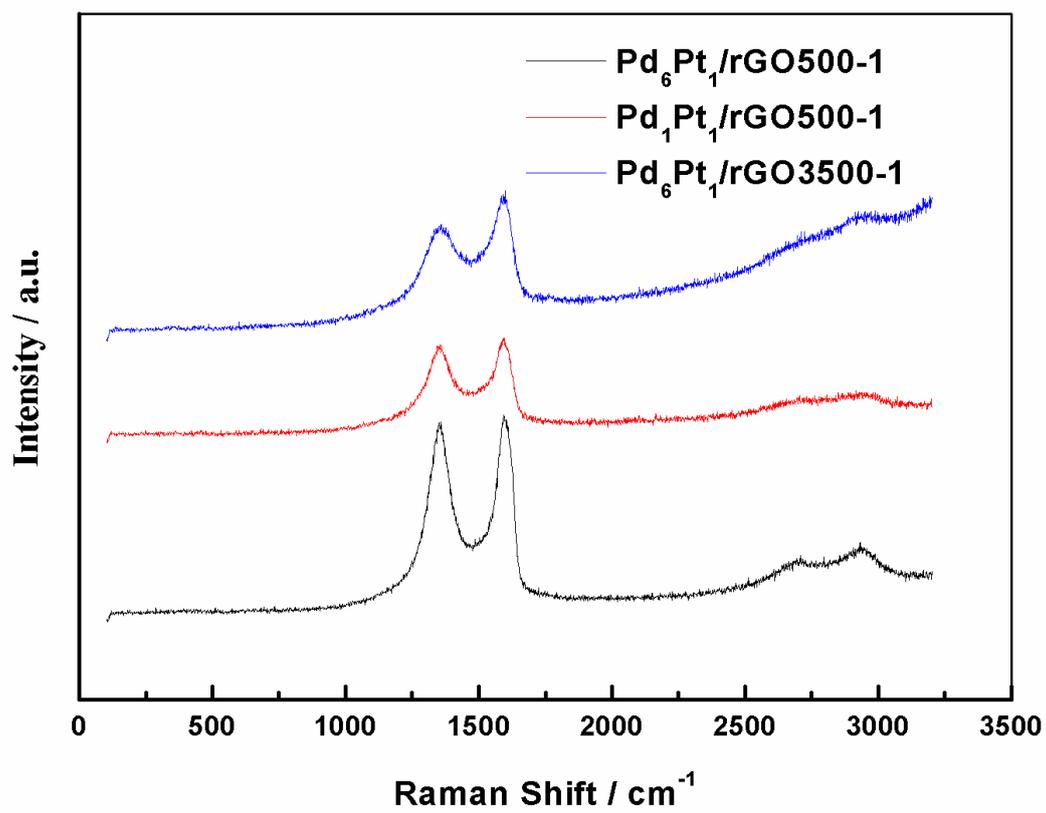


Figure S6. Raman spectra of the Pd₆Pt₁/rGO500-1, Pd₁Pt₁/rGO500-1 and Pd₆Pt₁/rGO3500-1 electrocatalysts.

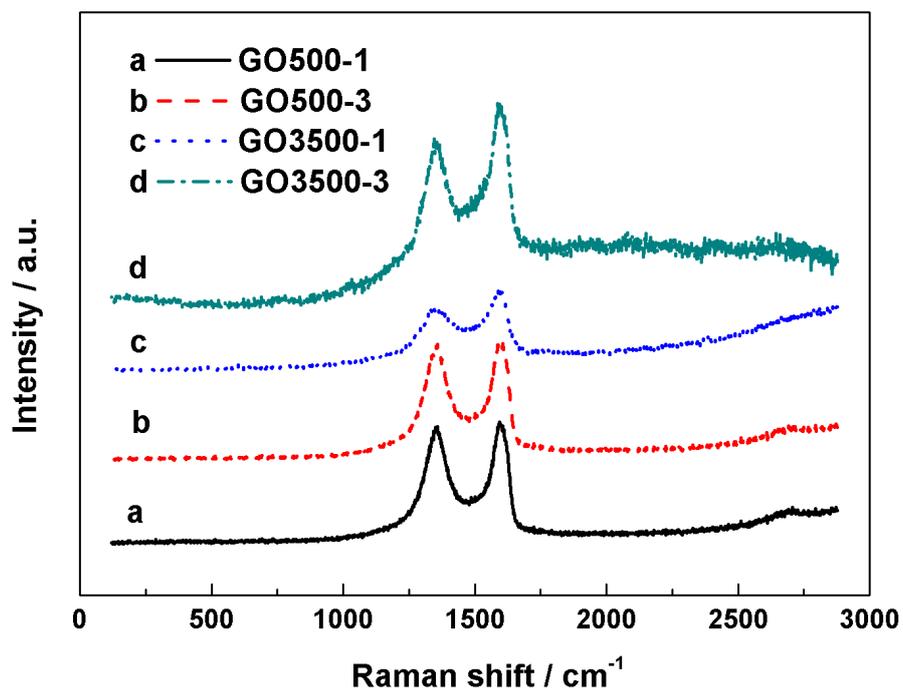


Figure S7. Raman spectrum of different GO samples derived from different graphite samples (with sizes of 500 mesh and 3500 mesh, respectively) with different treatment process (one time and three times acidified treatment, respectively) .

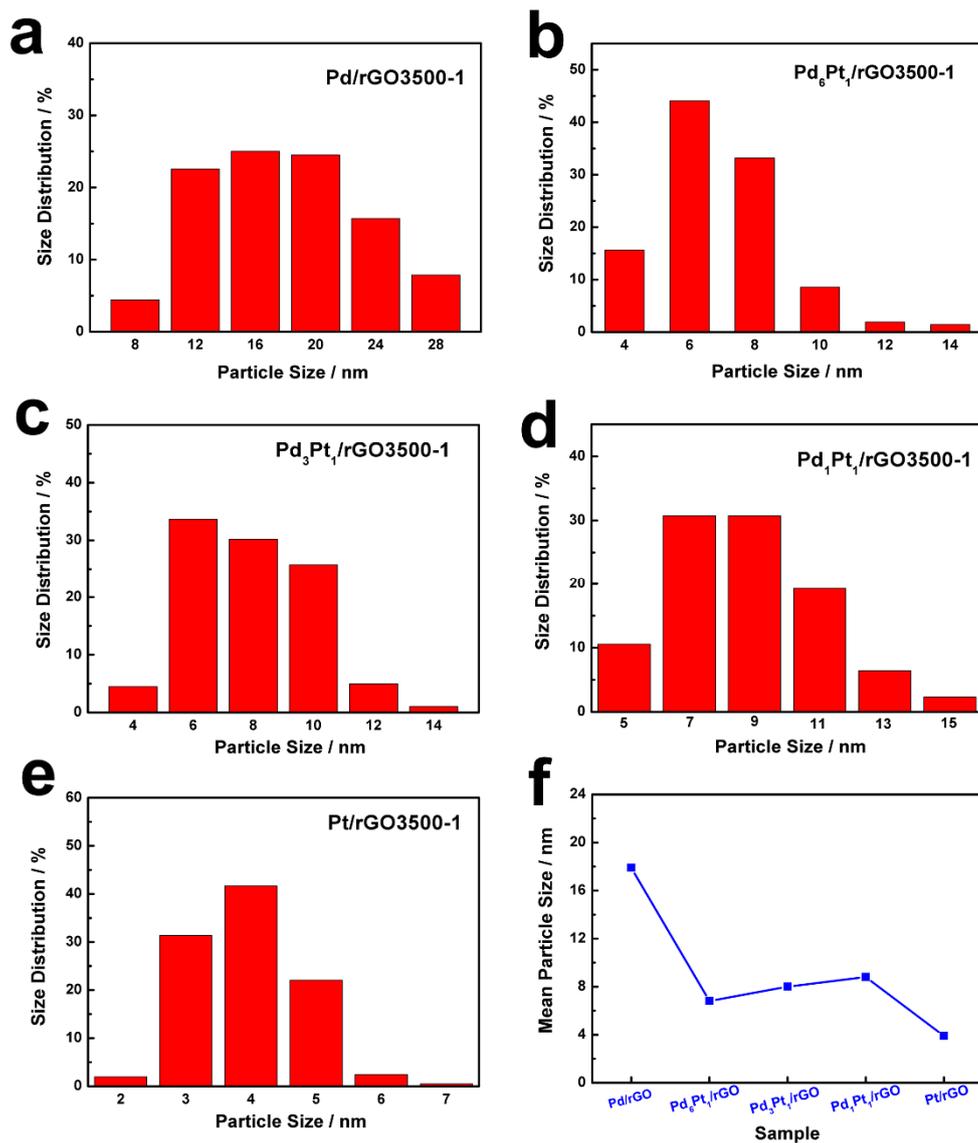


Figure S8. Particle size distributions of (a) Pd/rGO3500-1, (b) Pd₆Pt₁/rGO3500-1, (c) Pd₃Pt₁/rGO3500-1, (d) Pd₁Pt₁/rGO3500-1 and (e) Pt/rGO3500-1; (f) Mean particle sizes of Pd/rGO3500-1, Pd-Pt rGO3500-1 with different Pd/Pt ratios and Pt/rGO3500-1.

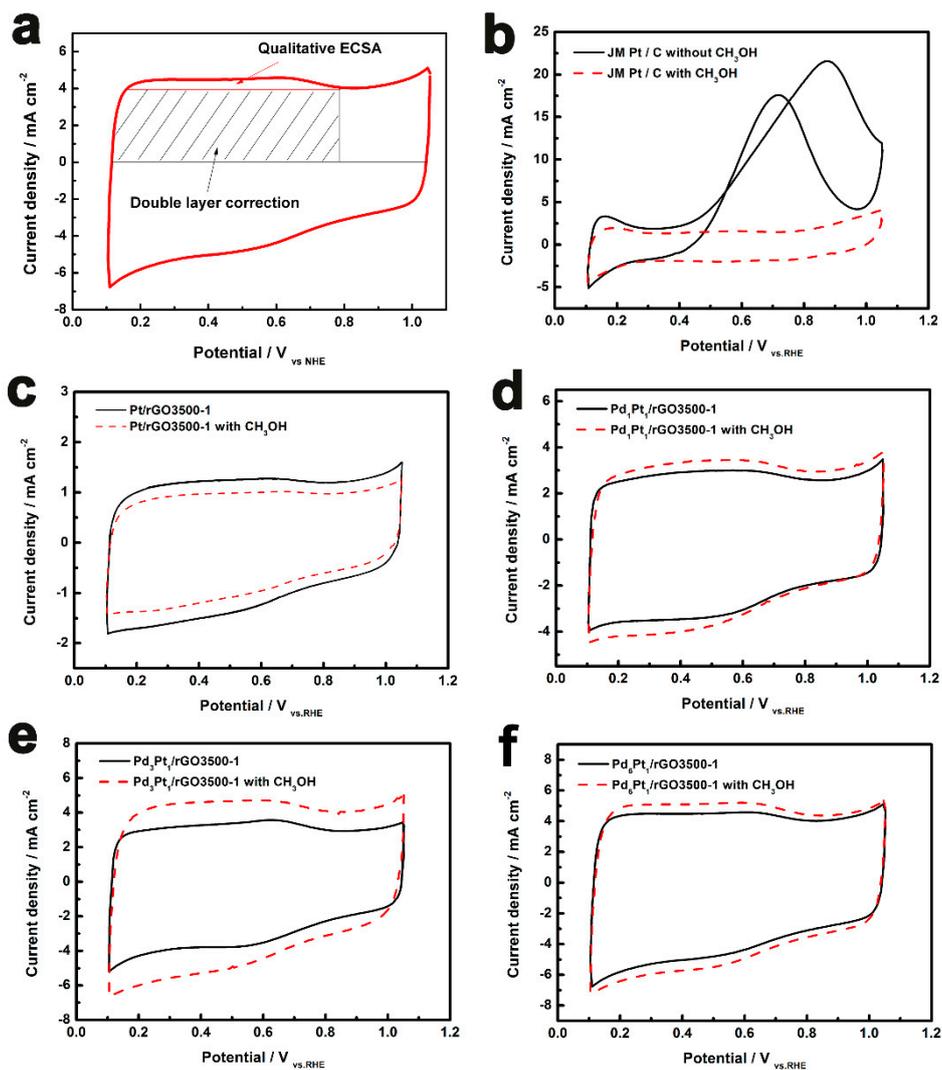


Figure S9. (a) Double layer corrected calculation for qualitative ECSA, CV curves of the (b) JM Pt/C, (c) Pt/rGO3500-1, (d) Pd₁Pt₁/rGO3500-1, (e) Pd₃Pt₁/rGO3500-1 and (f) Pd₆Pt₁/rGO3500-1 electrocatalysts in the 0.5 M H₂SO₄ solution without (black line) and with 0.5 M CH₃OH (red dashed line).

4. Supplementary Tables

Table S1. Lattice constant of rGO anchored Pd, Pt and Pd-Pt nanocrystals obtained with various rGO substrates and different Pd/Pt ratios.

Samples	Pd₆Pt₁/rGO500-1	Pd₆Pt₁/rGO500-3	Pd₆Pt₁/rGO3500-1	Pd₆Pt₁/rGO3500-3
(1 1 1) distance	0.2275 nm	0.2248 nm	0.2286 nm	0.2250 nm
Lattice constant	0.3940 nm	0.3893 nm	0.3958 nm	0.3897 nm
Crystallite size	6.8 nm	8.5 nm	2.1 nm	8.6 nm
Grain size	11.9 nm	8.9 nm	6.8 nm	9.2 nm
Samples	Pd/rGO3500-1	Pd₃Pt₁/rGO3500-1	Pd₁Pt₁/rGO3500-1	Pt/rGO3500-1
(1 1 1) distance	0.2255 nm	0.2264 nm	0.2262 nm	0.2253 nm
Lattice constant	0.3905 nm	0.3920 nm	0.3917 nm	0.3902 nm
Crystalline size	10.3 nm	7.4 nm	7.0 nm	5.1 nm
Grain size	17.9 nm	8.0 nm	8.8 nm	3.9 nm