

Supporting Information

Experimental

Materials: K_2SiF_6 were obtained from Alfa Co., nylon filtration membrane (100 nm pore size, 25 mm membrane diameter) was purchased from Yibo Co. (Haining, China). $KAuCl_4$, graphite were purchased from Sigma. Multi-walled carbon nanotubes (MWNTs) were purchased from Shenzhen Nanotech Port Co., Ltd. (Shenzhen, China). Graphdiyne sample were prepared using a cross-coupling reaction using hexaethynylbenzene precursors on the surface of copper. All other reagents were of analytical grade and used without further purification. All aqueous solutions were prepared with Milli-Q water.

Synthesis of GDY: Graphdiyne (GDY) was synthesized on the surface of copper via a cross-coupling reaction using hexaethynylbenzene precursors, as demonstrated previously. In a typical synthesis, the monomer of hexaethynylbenzene was synthesized in good yield (62%) by adding tetrabutylammonium fluoride into a tetrahydrofuran solution of hexakis-[(trimethylsilyl)ethynyl]benzene for 10 min at 8°C. The GDY was successfully grown on the surface of copper foil in the presence of pyridine by a cross-coupling reaction of the monomer of hexaethynylbenzene for 72 h at 60°C under a nitrogen atmosphere. In the process of GDY formation, the copper foil serves as not only the catalyst for the cross-coupling reaction but also the substrate for the growth of GDY film. The copper foils covered by GDY were first washed with acetone followed by 80°C DMF under sonication for 1 hour to obtain black solid. The black solid was refluxed at 100°C for 2 h in 4 M sodium hydroxide, 6 M hydrochloric acid, 4 M sodium hydroxide solution, respectively, to remove the impurities and copper residue. The product was collected by centrifugation, washing with hot DMF (80°C), hot ethanol (70°C), and then dried to give pure GDY.

Liquid-phase exfoliation of GDY: In a typical experiment, 100 mg of Li_2SiF_6 was dissolved in 5 ml water, and then 5 mg bulk GDY was mixed into the solution. Liquid-phase exfoliation (LPE) was carried out by continuously stirring the as-prepared dispersion for 60 hours under ambient condition (25°C). The obtained dispersion was centrifuged at 500 rpm for 5 min to remove large precipitates. After that, the product was collected and filtered through a nylon membrane with 100-nm pore size and consecutively washed several times with deionized water and isopropanol by vacuum filtration. In order to measure the concentration of eGDY dispersions, the masse of dry filter membrane loaded with eGDY was carefully weighed and subtracted with the one of remaining nylon membrane after peeling off eGDY powder to give the net weight of the obtained eGDY.

Synthesis of Au loaded on substrates: In a typical synthesis of Au/eGDY, Au/GDY, Au/G, and Au/MWNT, a homogeneous aqueous dispersion of eGDY, GDY, G, MWNT (2 ml, 0.5 mg ml^{-1}) was, respectively, mixed with of KAuCl_4 (0.2 ml, 10 mM) aqueous solution under vigorous stirring for 10 min in an ice bath. Then, the reaction mixture was centrifuged and washed with water to remove unreacted reagents. The resulting products are denoted as Au/eGDY, Au/GDY, Au/G, and Au/MWNT, respectively.

Catalytic Reduction of 4-Nitrophenol (4-NP): The aqueous solution of NaBH_4 (0.65 ml, 0.2 M) and the corresponding nitroarenes (0.10 ml, 1 mM) were mixed with water (2.25 ml). The mixture was stirred for 5 min at room temperature. Then the homogeneous dispersion of Au/eGDY, Au/GDY, Au/G, and Au/MWNT was added, respectively; the adsorption spectra of the resulting solution were monitored by recording the UV-Vis spectra at short intervals in the range of 250-500 nm at room temperature.

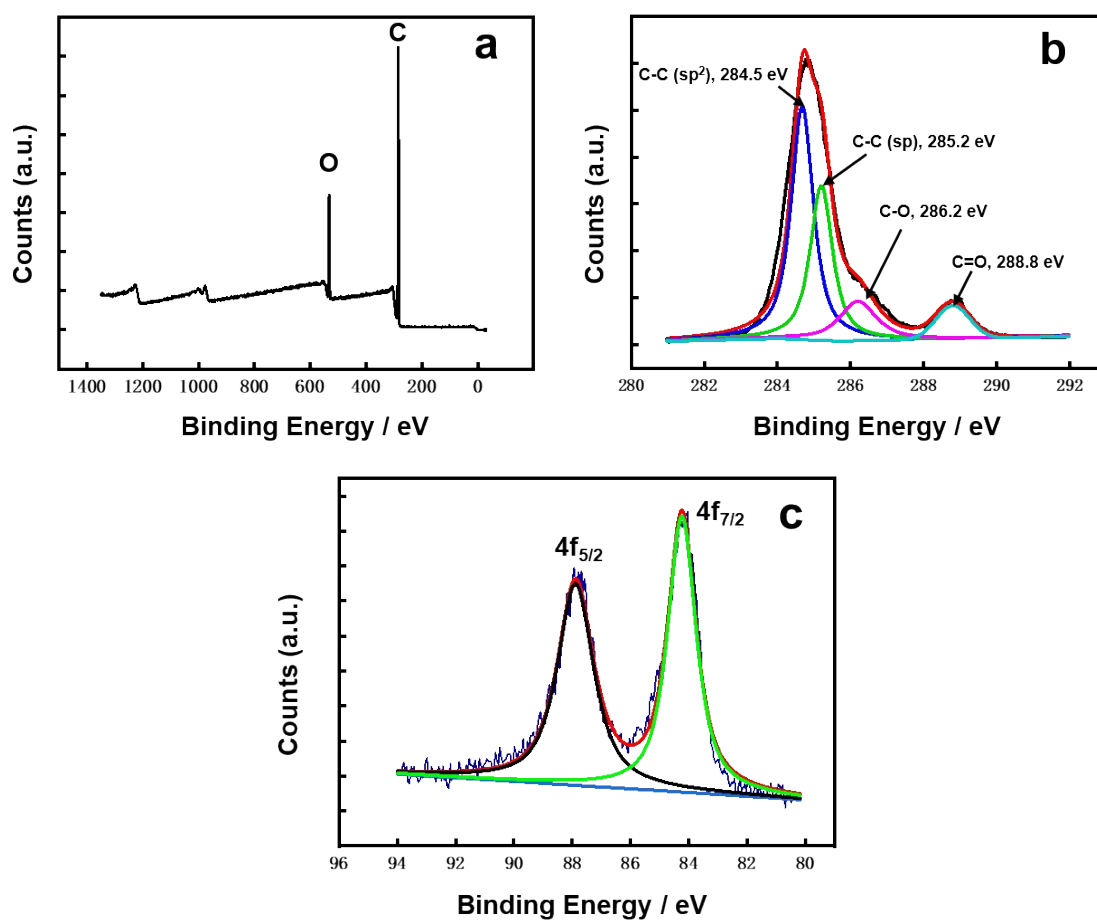


Fig. S1 XPS spectra of (a) eGDY, (b) C1s of eGDY and (c) Au 4f of Au/eGDY.

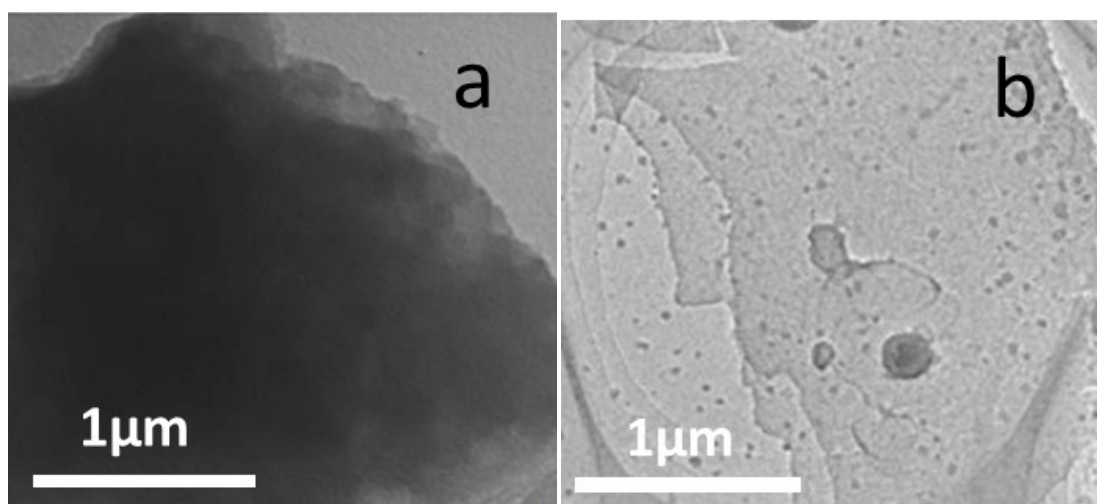


Fig. S2 TEM images of (a) bulk GDY powder and (b) eGDY flakes

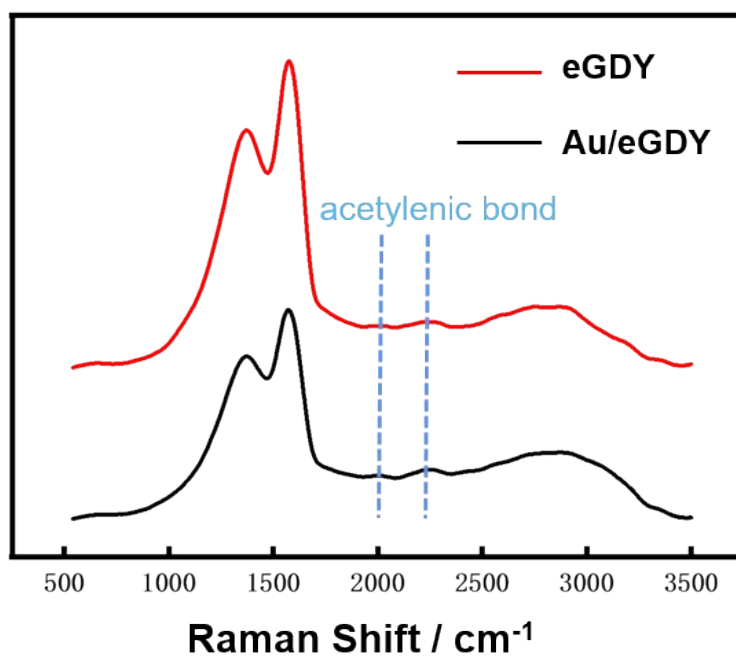


Fig. S3 Raman spectra of eGDY (red line) and Au/eGDY (black line).

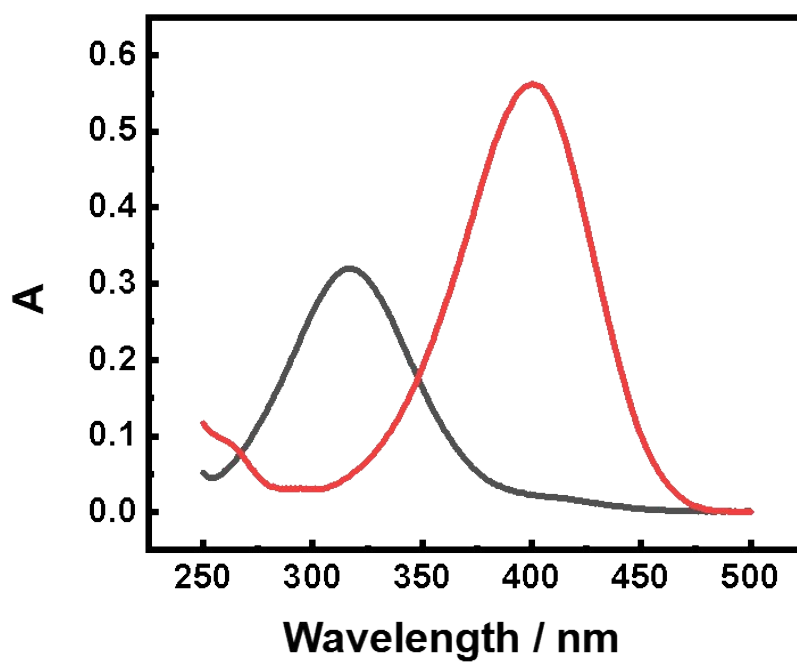


Fig. S4 UV-vis spectra of 4-NP (black line) and 4-nitrophenolate ion (red line).

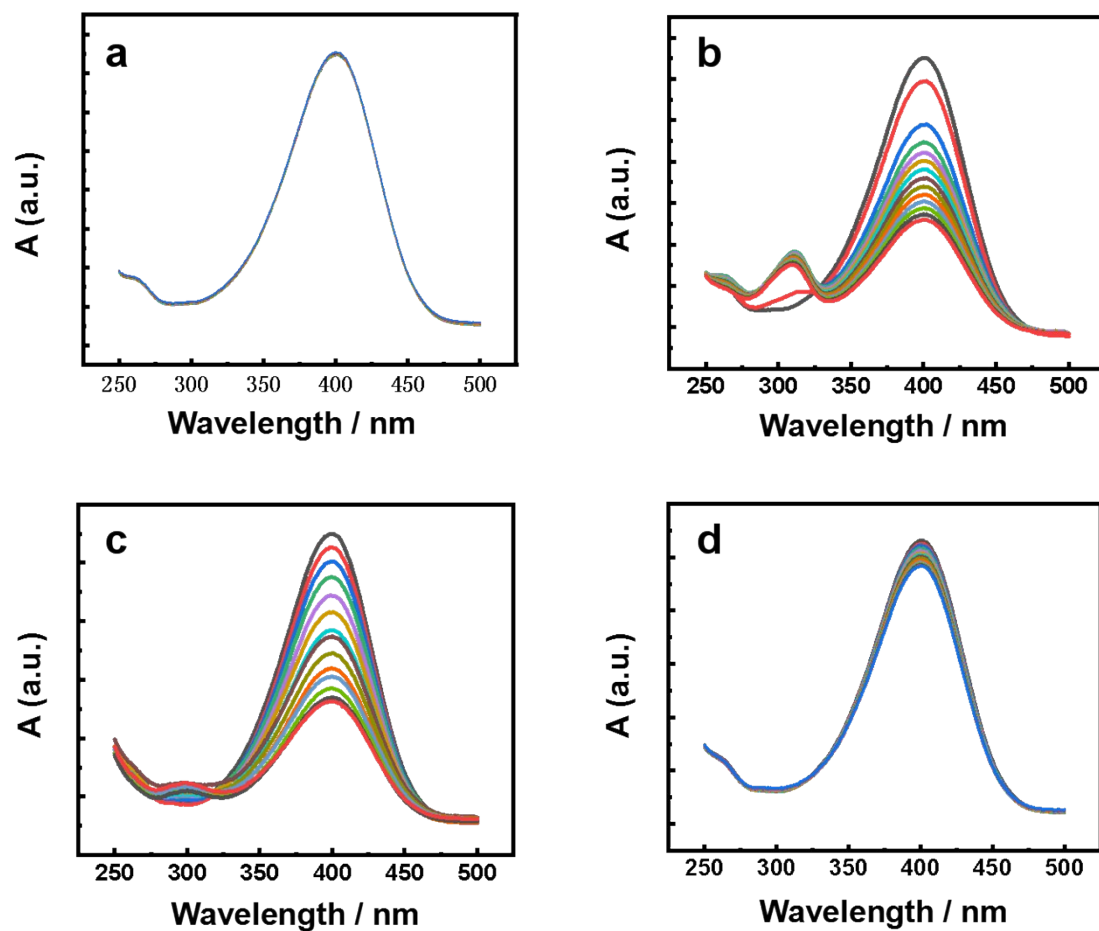


Fig. S5 Time-dependent UV-vis absorption spectra recorded during the catalytic reduction of 4-nitrophenol in the presence of (a) eGDY, (b) Au/GDY, (c) Au/G, (d) Au/MWNT