Supporting Information

Tuning the Selectivity and Activity of Electrochemical Interfaces with Defective Graphene Oxide and Reduced Graphene Oxide

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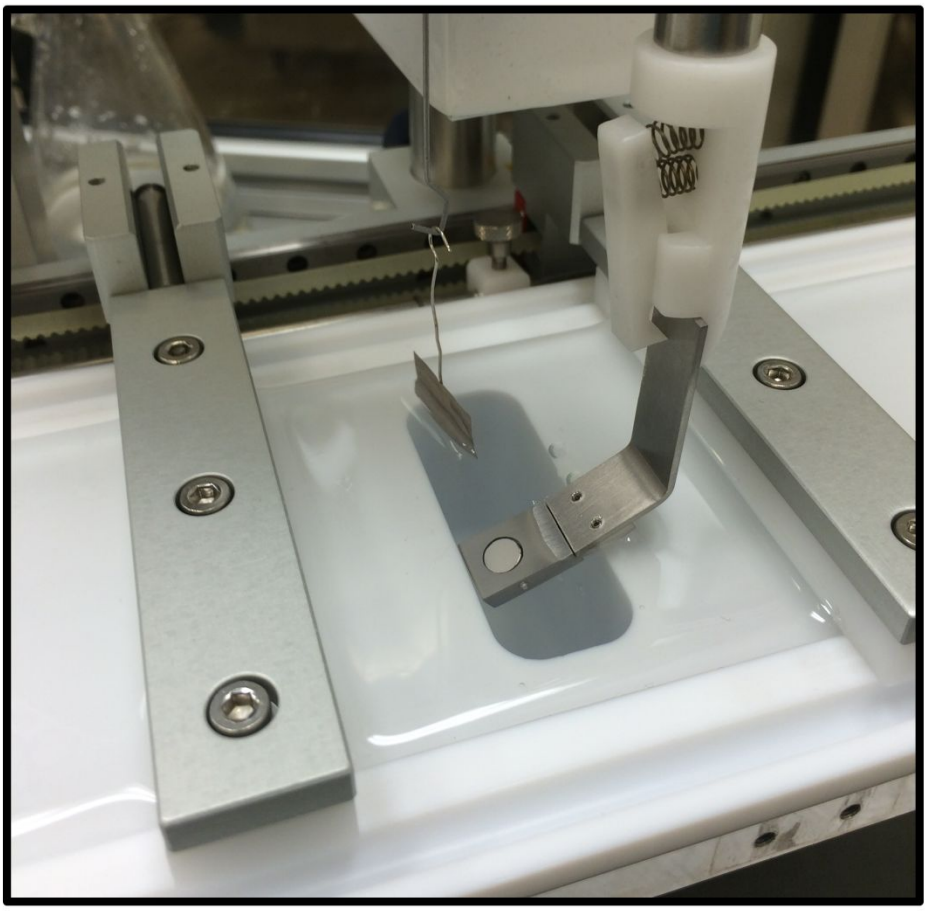

Figure S1. Photograph of Langmuir trough depicting compression barriers, Wilhelmy plate for surface pressure measurements, and the stainless-steel holder for dipping Pt disk electrodes.

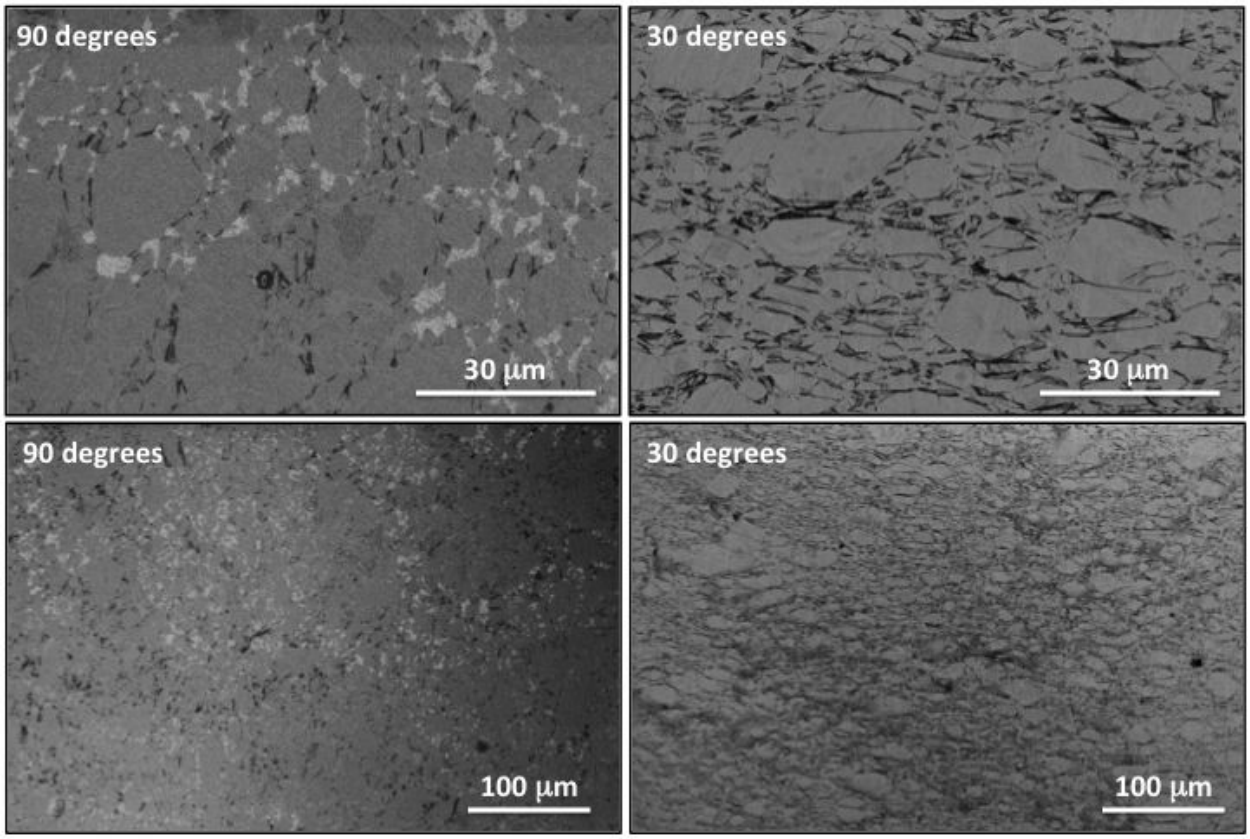

Figure S2. Comparison of representative SEM images for Pt disk dipped at 30° and 90° relative to the air-water interface. Clearly, the sample dipped at 30° shows higher and more uniform coverage. The bright areas obvious in the 90° angle dipped sample indicate holes in the GO monolayer. Because the Pt disk electrodes were secured to a holder with a gap between the holder and the crystal, the GO monolayer is strained more than in a typical LB dip on a flat, continuous substrate.

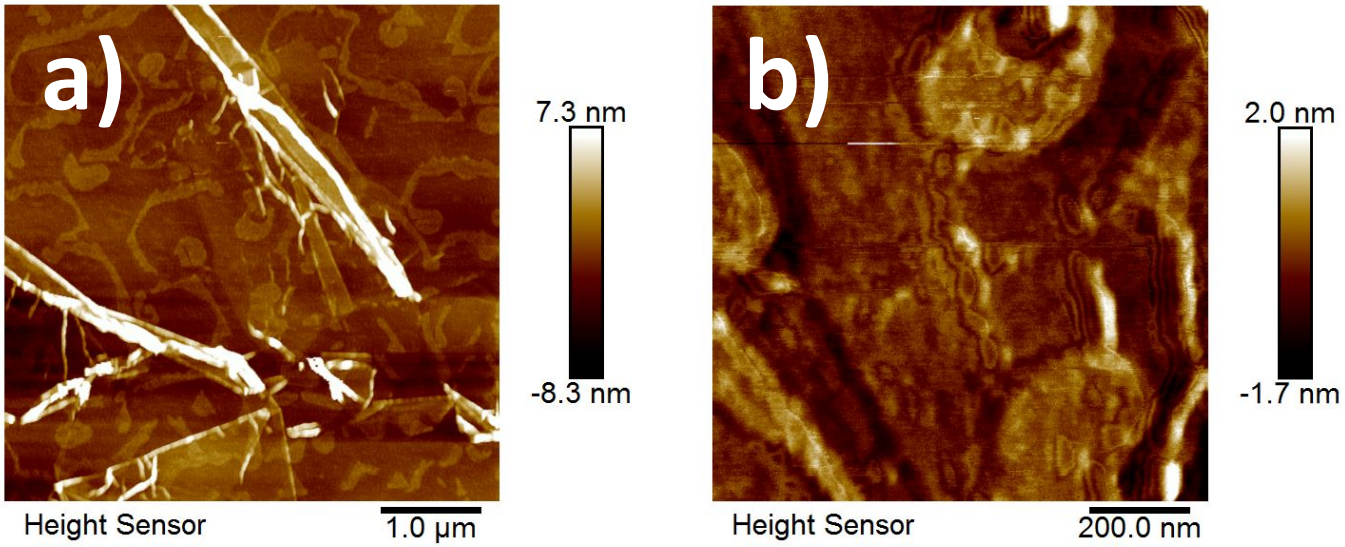

Figure S3. AFM images of GO on Pt disk. a) wrinkled area, where two GO flakes overlap, and b) basal plane on GO flake.

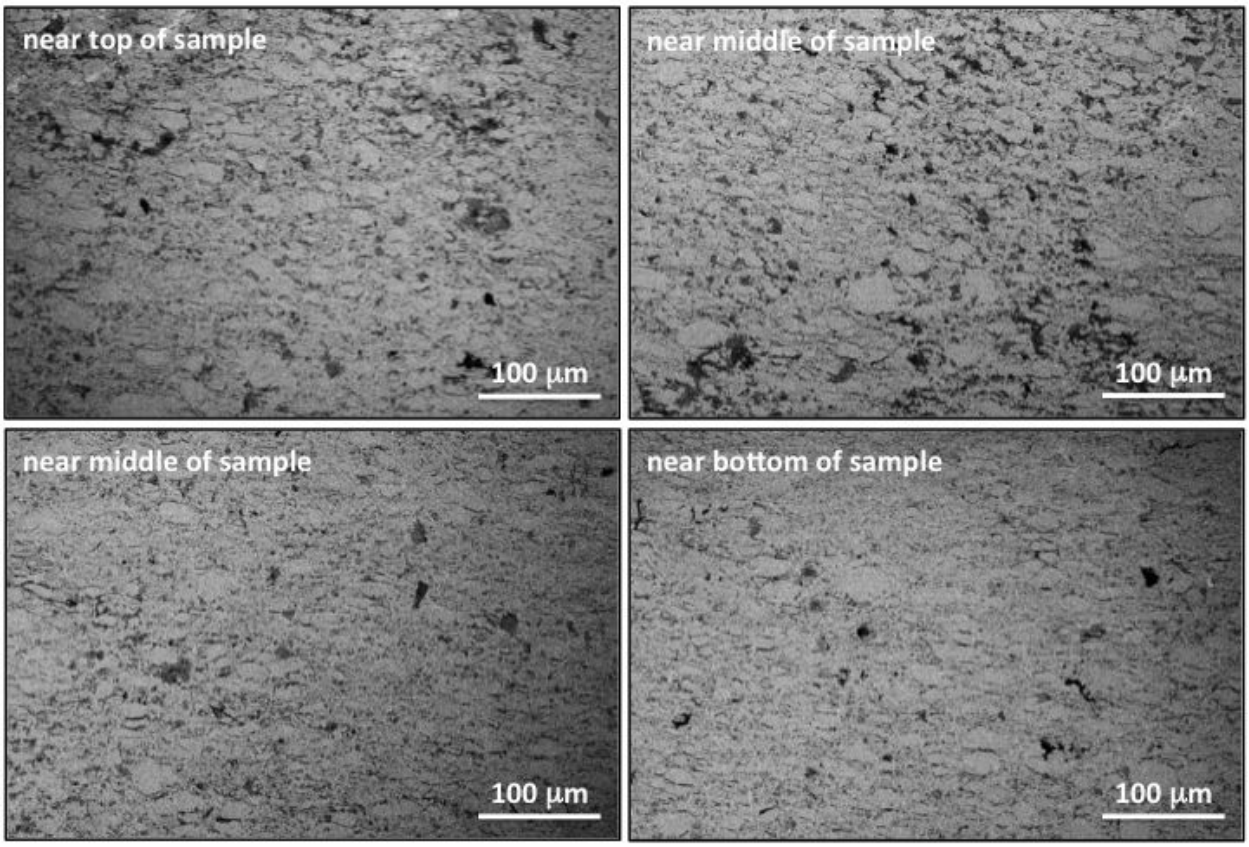

Figure S4. Representative SEM images of GO monolayers transferred to a Pt disk. Images were recorded at the top of the sample, near the middle of the sample, and near the bottom of the sample along the centerline of crystal. Coverage is very high and uniform with few pinholes in the monolayer. Holes in the monolayer are bright, as shown in Figure S2 for the sample dipped at 90°.

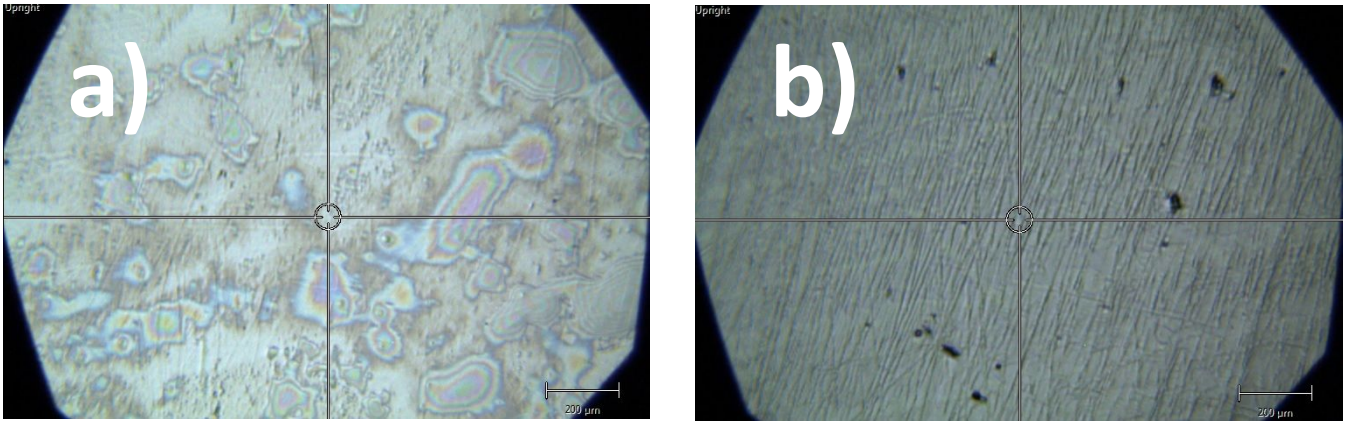

Figure S5. Optical Microscope images of GO on Pt disk after Langmuir-Blodgett deposition: **a)** as prepared, and **b)** after drying in vacuum at 40^{o}C.

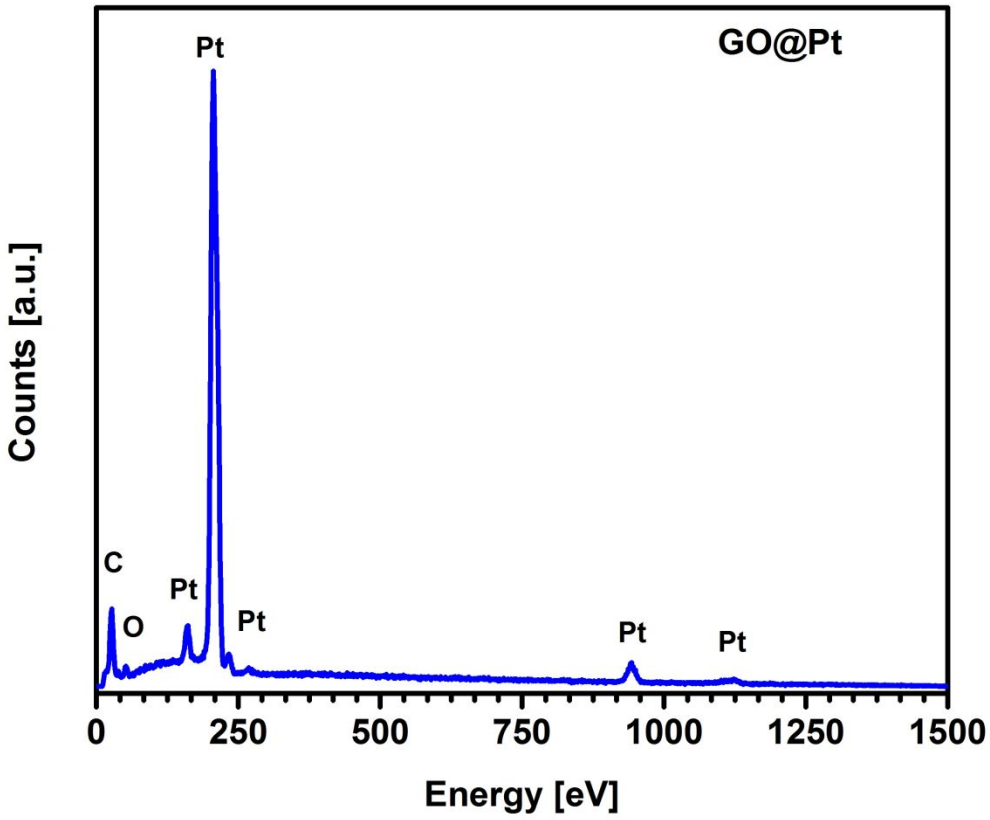

Figure S6. EDS spectrum taken on a GO monolayer transferred to a Pt disk. Only Pt, C, and O are detected.

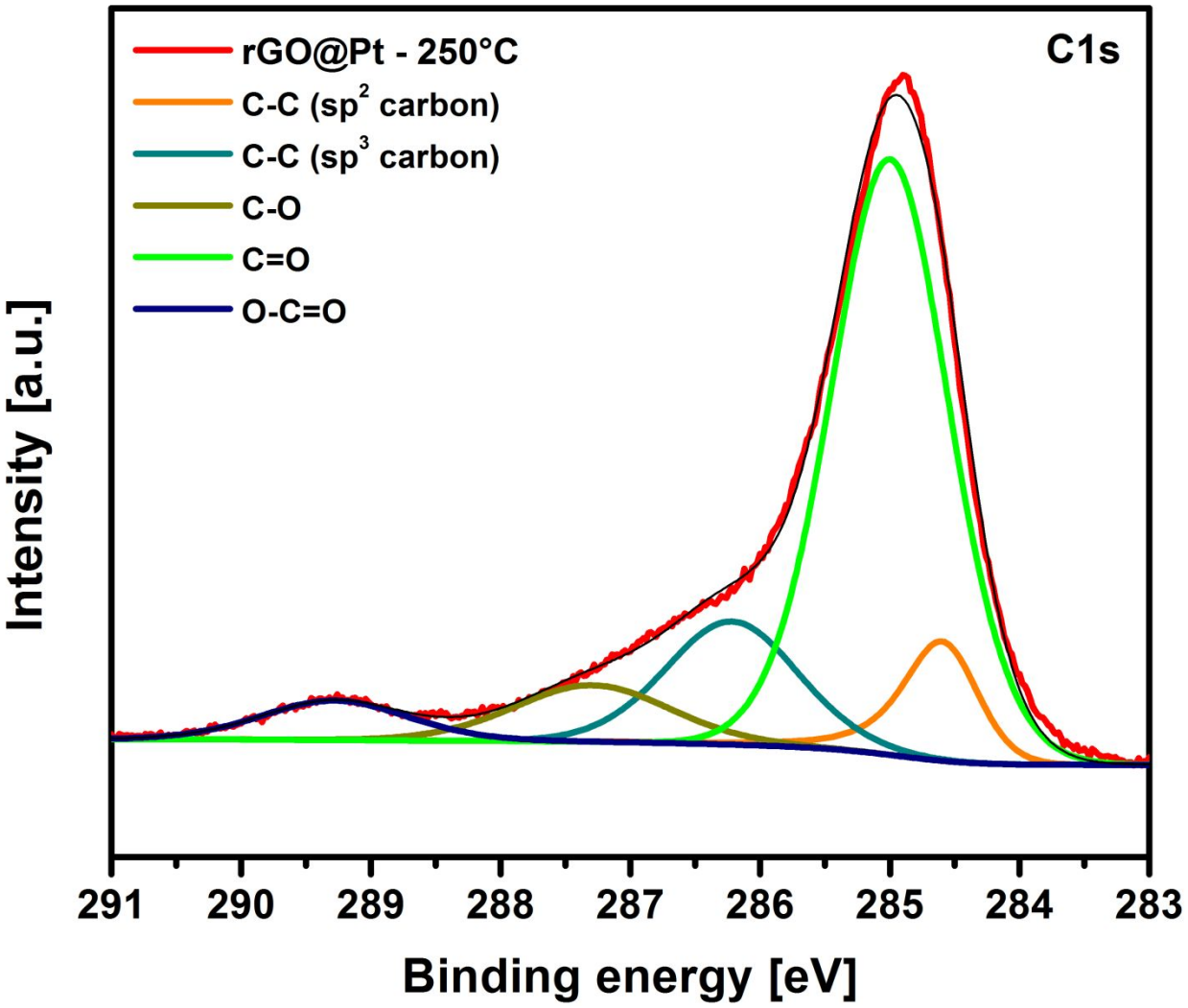

Figure S7. *C1s* XPS core level spectra with spectral deconvolution for GO@Pt annealed at 250°C. Below is C*1s* core level spectral deconvolution quantification.

Sample	Peak label	E_{bin} [eV]	Raw Area	%At. Conc.
rGO@Pt annealed at 250°C	C-C (sp^2)	284.60	4134.34	**7.68**
	C-C (sp^3)	285.00	34486.5	**64.07**
	C-O	286.22	8028.81	**14.92**
	C=O	287.29	4342.92	**8.07**
	O-C=O	289.28	2835.83	**5.27**

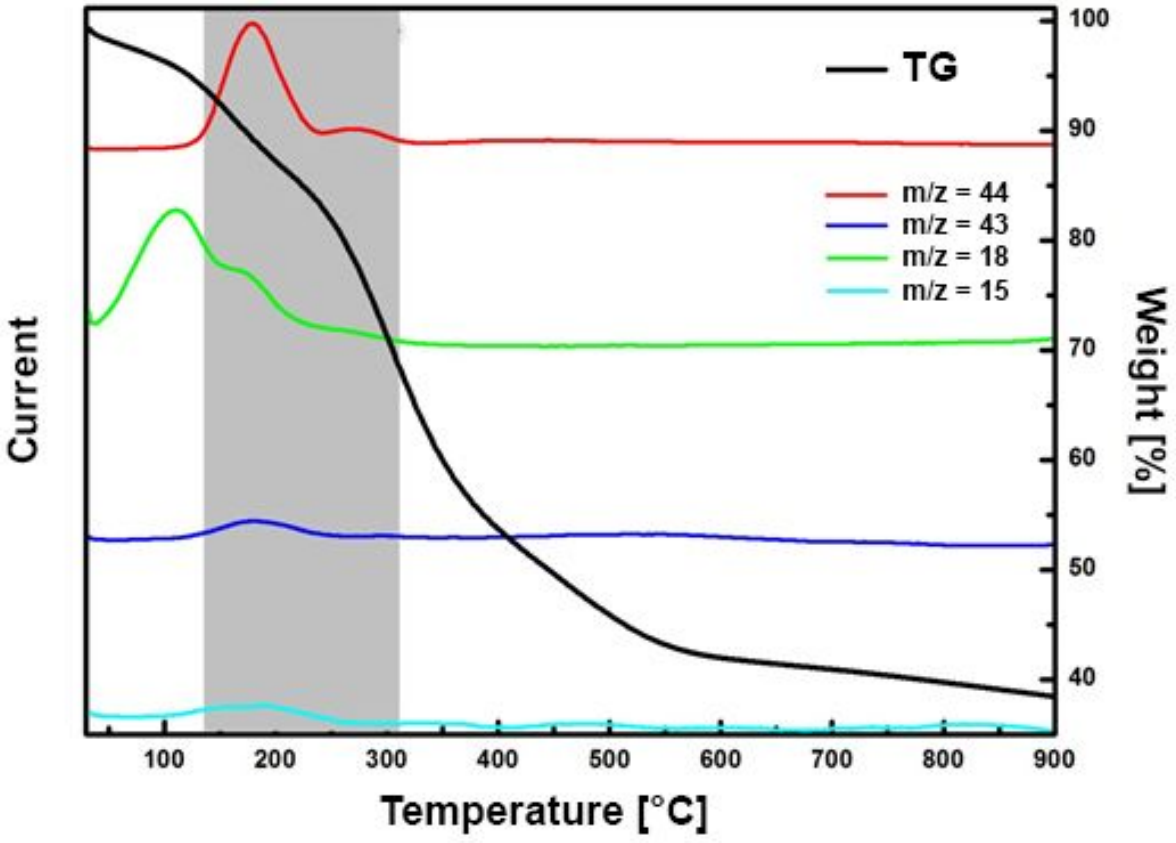

Figure S8. Thermogravimetric analysis coupled with mass spectrometry for bulk GO that was used Langmuir-Blodgett deposition. Analysis was done in Ar atmosphere with heating rate, 10 K/min and gas flow 50 mL/min. m/z 44 is representative for CO_2 evolution, m/z 18 for H_2O and m/z 15 form -CH_3.

Evolved gas analysis (EGA) experimental part.
Thermogravimetric measurements were performed on a Netzsch 449 F3 Jupiter instrument under a dynamic Ar (5.0) flow with a flow rate of 50 mL/min in a temperature range from 25°C to 900°C. A heating rate of 10 K/min was used. About 15 mg of sample was placed in an alumina (Al_2O_3) crucible. Mass spectrometry was performed simultaneously on avMS 403C Aëolos with a SEM channeltron detector and system pressure of $2x10^{-5}$ mbar. Gasses evolved under TG heat treatment were transferred to mass spectrometer through via quartz capillary (ID 75 μm) which was heated to 220 °C. The upper limit of the mass spectrometer detector was 100 AMU.

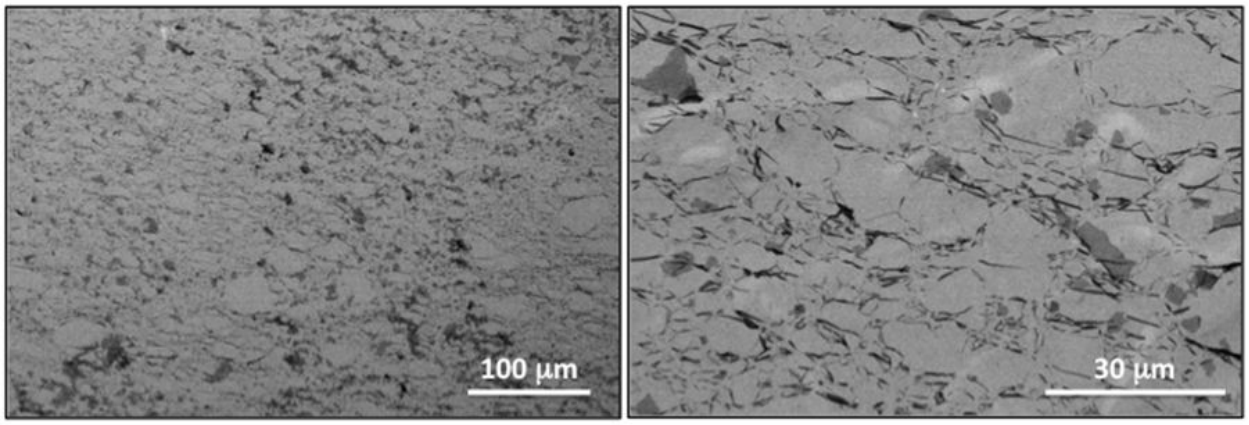

Figure S9. SEM images of an rGO monolayer. The morphology is very similar to the GO monolayers (Figure 1d).

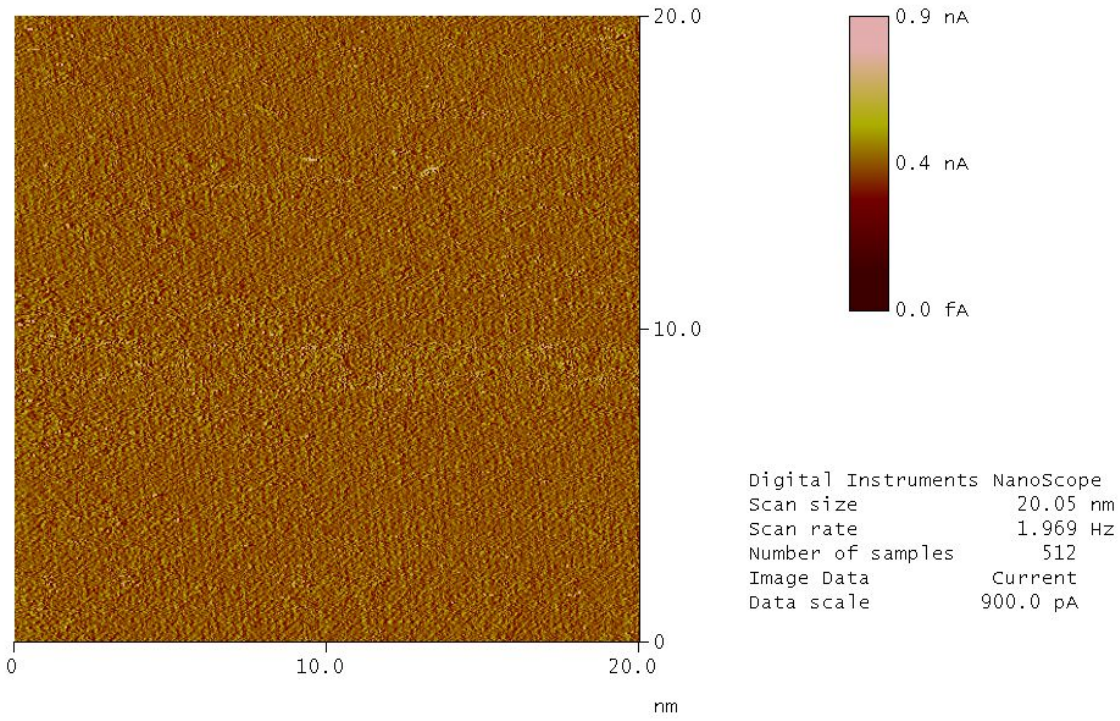

Figure S10. STM image of rGO on Pt disk.

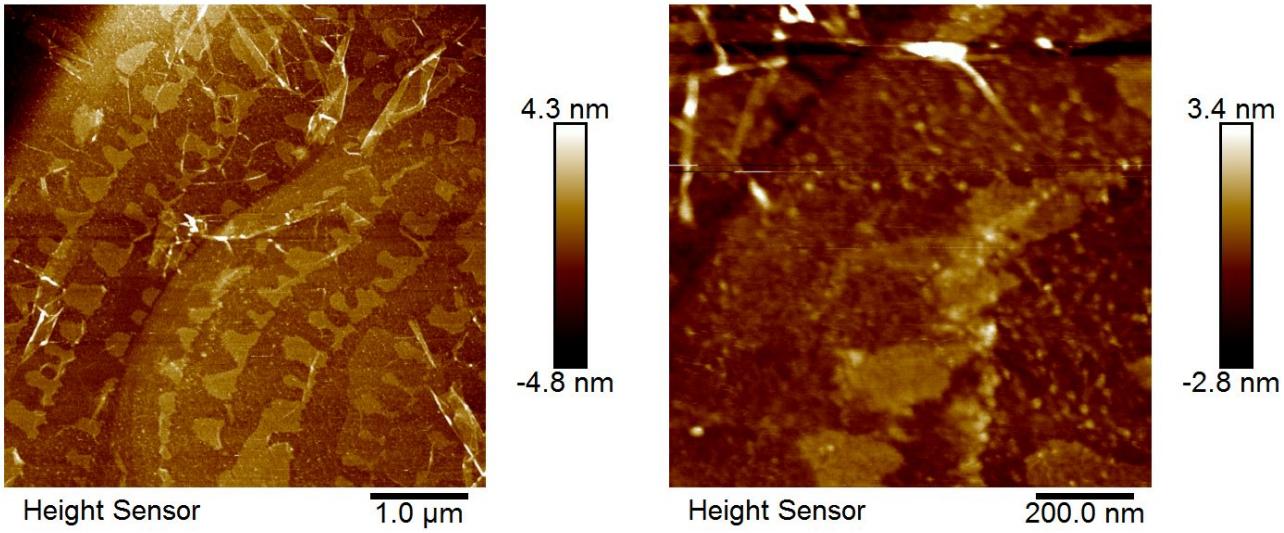

Figure S11. AFM images of rGO on Pt disk.

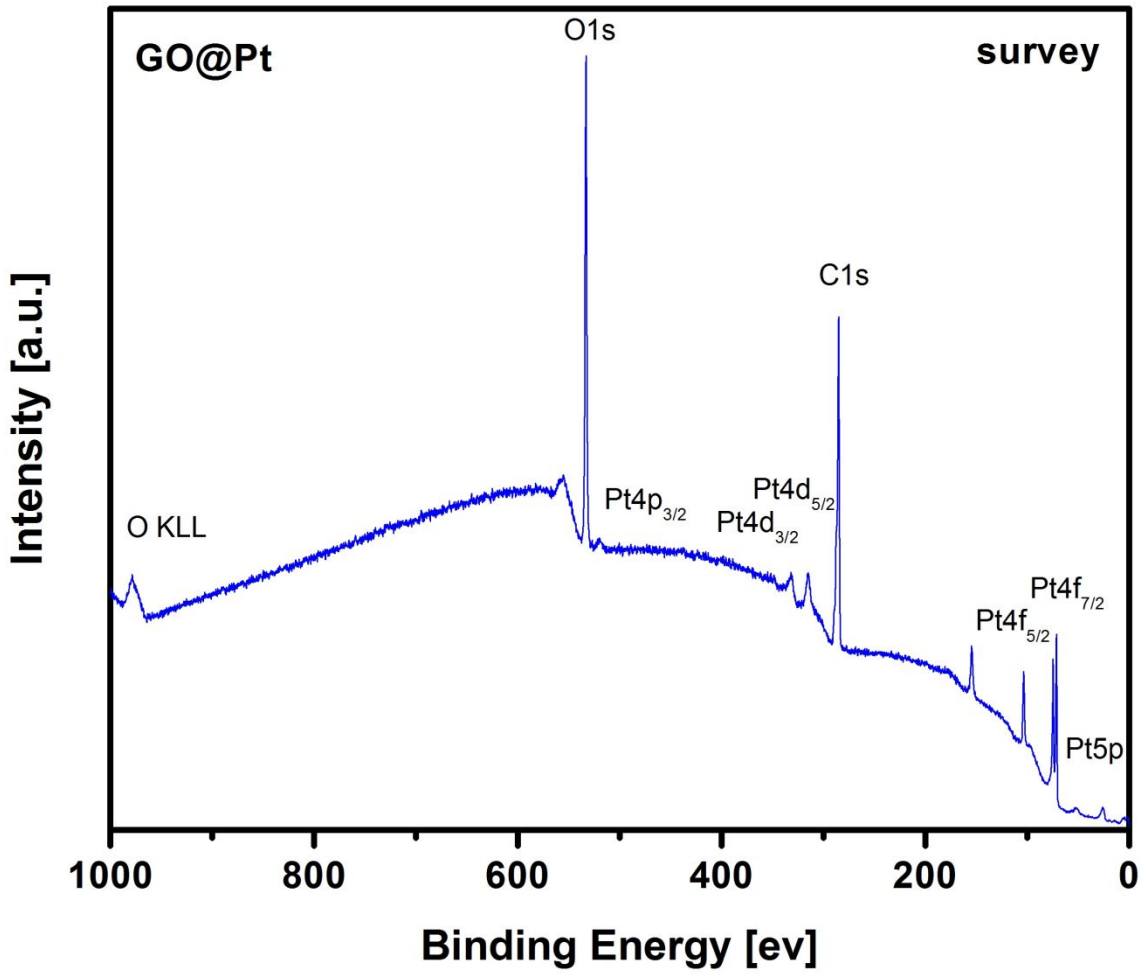

Figure S12. XPS survey spectrum of GO on Pt disk. Pt, C and O are detected.

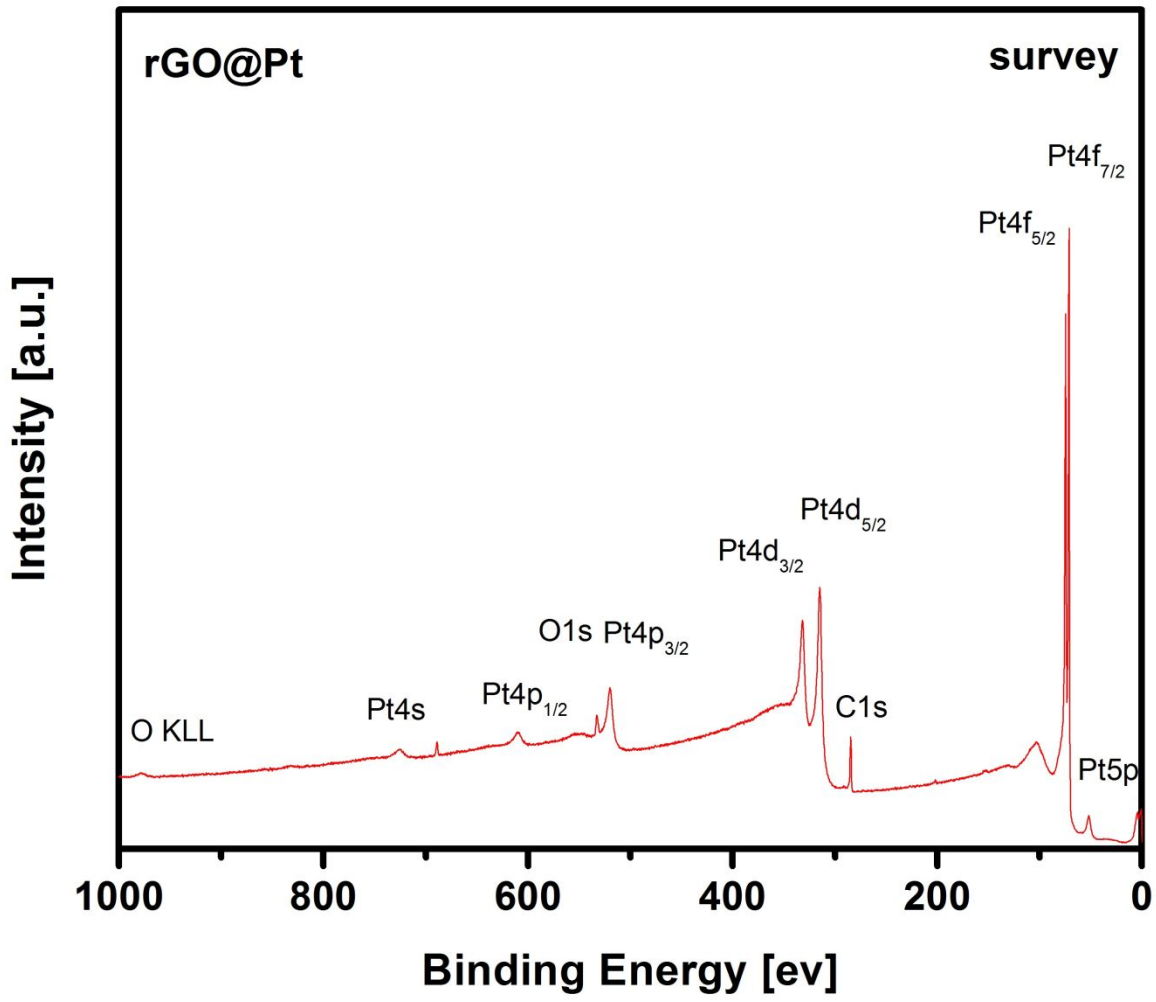

Figure S13. XPS survey spectrum of rGO on Pt disk. Pt, C and O are detected.

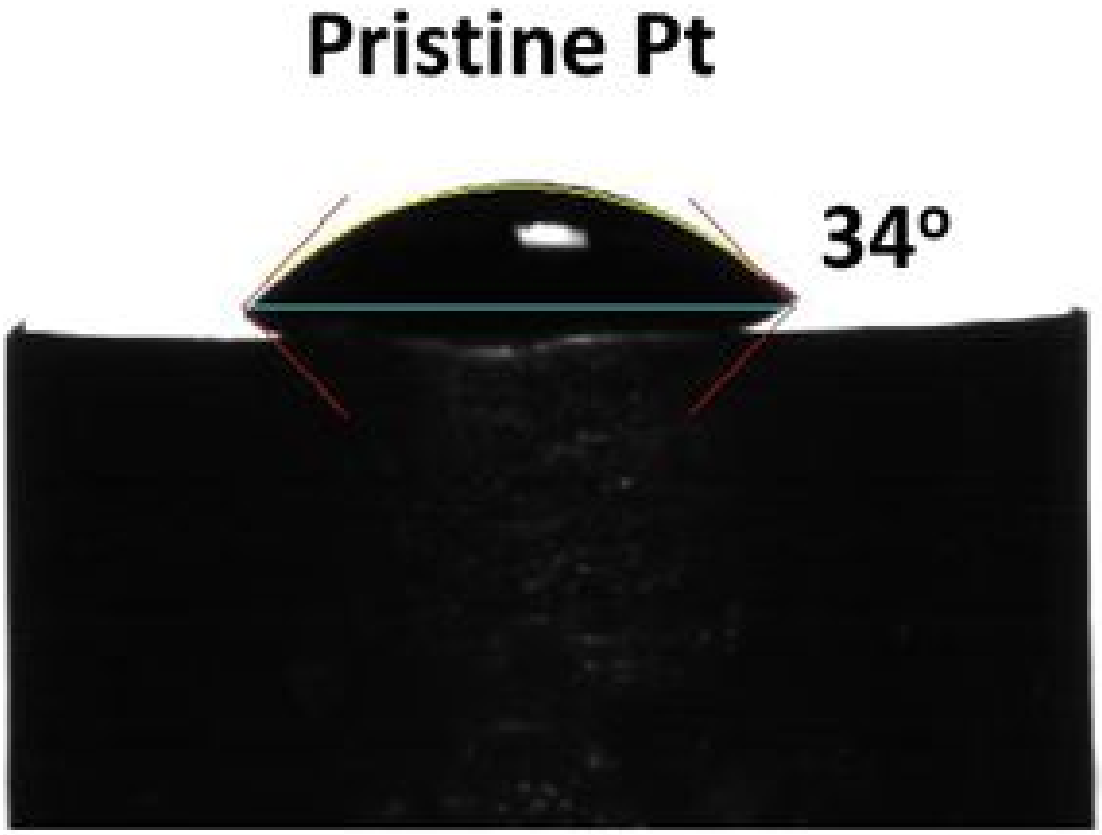

Figure S14. Contact angle measurements taken on a pristine Pt disk using a sessile drop technique before stabilized at 48°.

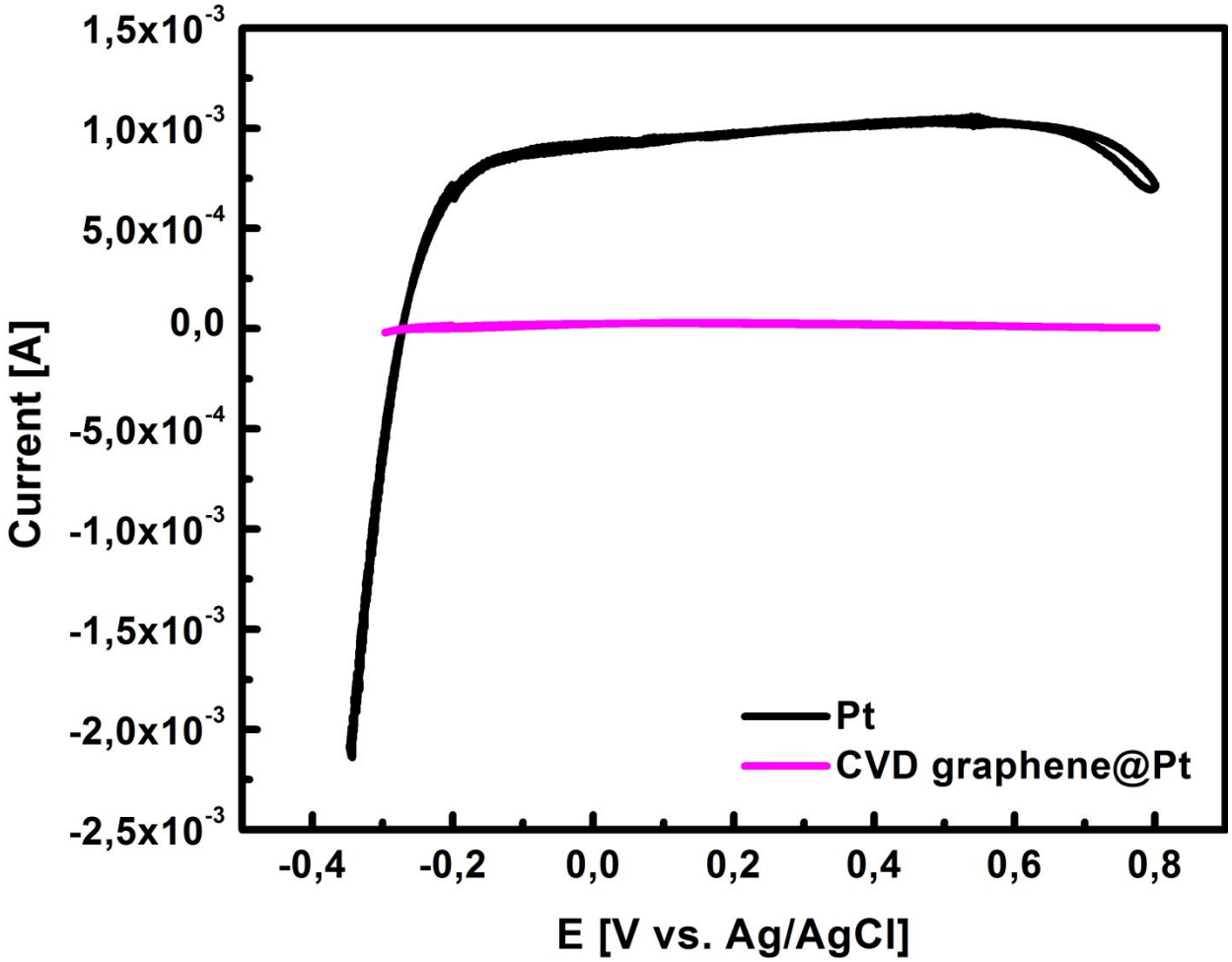

Figure S15: Polarization curves for the HOR for bare Pt (black line) and CVD grown graphene on Pt (pink line). CVD grown graphene on Pt exhibit no activity for HOR.

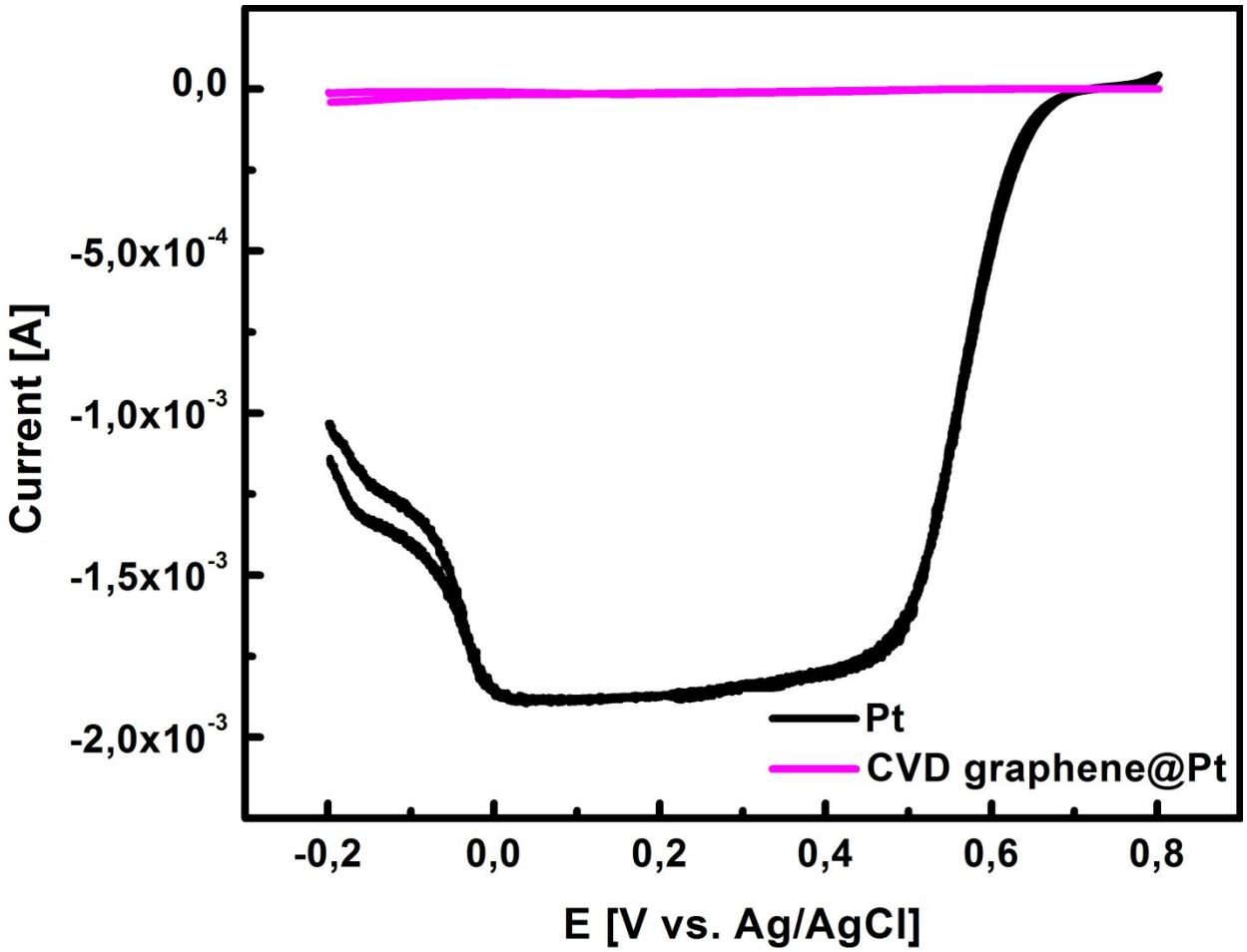

Figure S16: Polarization curves for the ORR for bare Pt (black line) and CVD grown graphene on Pt (pink line). CVD grown graphene on Pt exhibit no activity for the ORR.

a)

Sample	Peak label	E_{bin} [eV]	FWHM	Raw Area	%At. Conc.
GO@Pt disk	C-C (sp^2)	284.60	0.48	953.319	1.37
	C-C (sp^3)	285.20	1.29	39353.1	56.69
	C-O	286.29	1.50	10445.5	15.05
	C=O	287.45	1.50	13707.7	19.75
	O-C=O	289.38	1.50	4957.71	7.14

b)

Sample	Peak label	E_{bin} [eV]	FWHM	Raw Area	%At. Conc.
rGO@Pt disk	C-C (sp^2)	284.60	0.84	23233.6	96.40
	C-C (sp^3)	285.20	0	0	0
	C-O	286.29	0.89	811.691	3.37
	C=O	287.29	0.58	56.9117	0.24
	O-C=O	289.38	0	0	0

Table S1. C*1s* core level XPS spectral deconvolution quantification for a) GO@Pt and b) rGO@Pt surfaces.