

Supporting information

Doping of graphene via adlayer formation of electrochemically reduced dibenzyl viologen

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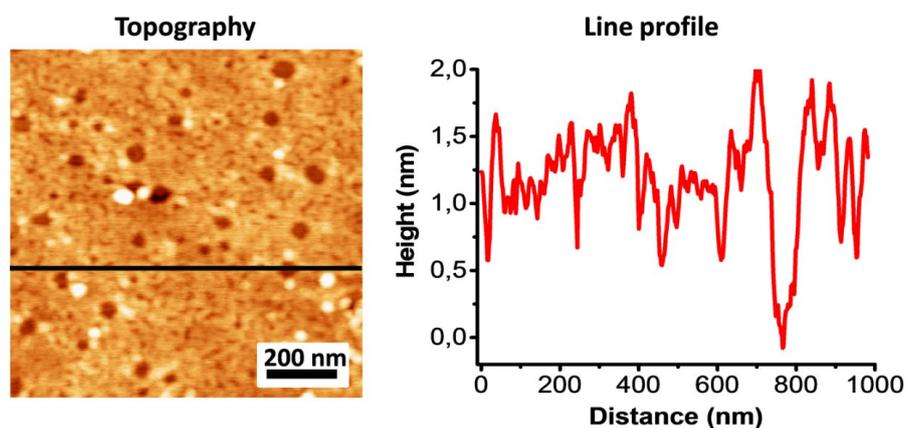


Figure S1: a-b) AFM images and line profile of DBV⁰ functionalized G-SiO₂ prepared at E = -0.85 V vs RHE. Due to the influence of the hydrogen evolution reaction the graphene surface is modified by disk-shaped features most likely generated by hydrogen nanobubbles.

Stacking phase forming on HOPG

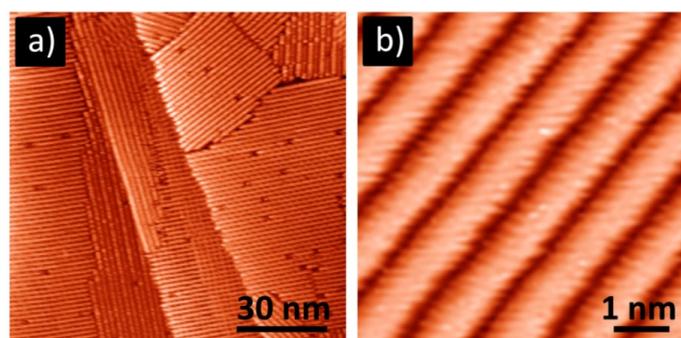


Figure S2: EC-STM images of the stacking phase of DBV⁰ on HOPG surface under electrochemical control: E = -0.6 V vs HRE, U_b = 100 mV, I_t = 0.1 nA. The results prove that the DBV⁰ species adsorb strongly on HOPG surface and align in rows within individual domains.

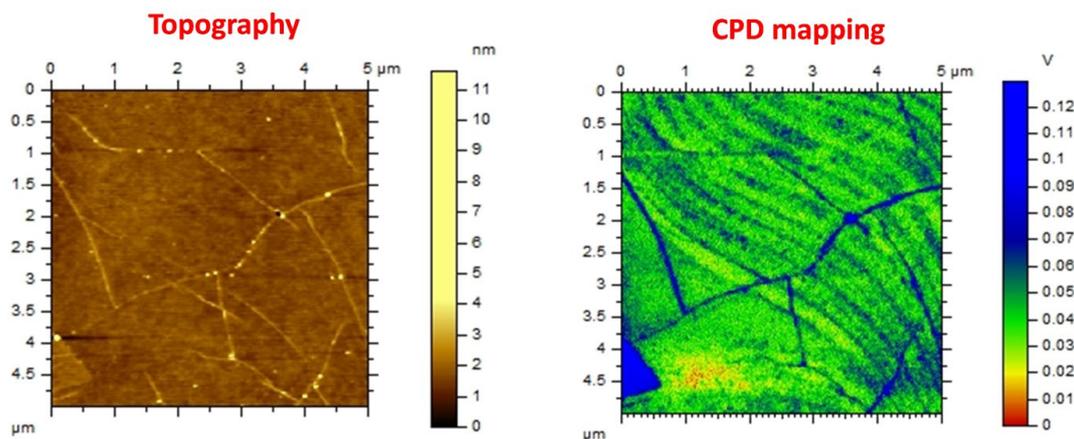


Figure S3: Topography and CPD mapping of pristine G-SiO₂ showing a significant change both of the contrast and the CDP at the wrinkles.

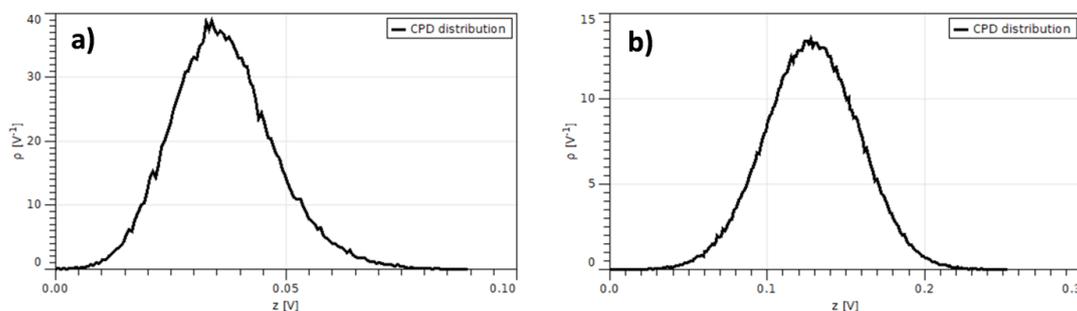


Figure S4: The CPD histograms of the KPFM images: a) pristine G-SiO₂ (corresponding to figure 3b) and b) G-SiO₂ exposed to DBV at E = -0.6 V (corresponding to figure 3h).

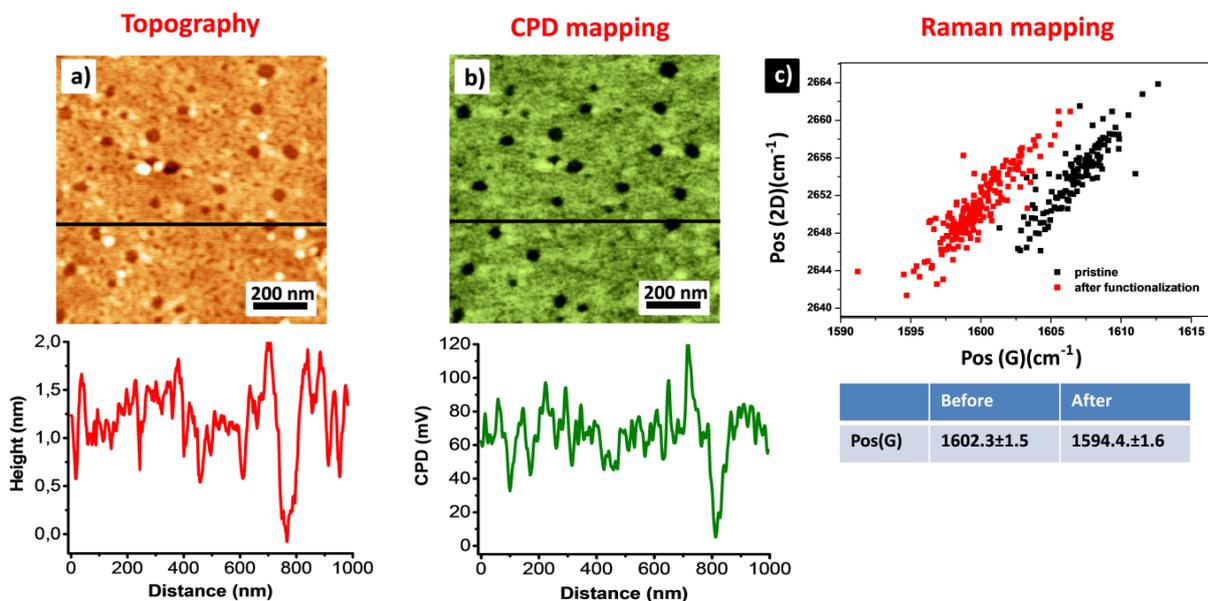


Figure S5: a-b) AFM images and CPD mapping of DBV⁰ functionalized G-SiO₂ prepared at E = -0.85 V vs HRE; c) Plots of Pos(2D) versus Pos(G) of the DBV⁰ modified G-SiO₂ and pristine G-SiO₂. Due to the influence of the hydrogen evolution reaction the graphene surface is not fully covered by DBV⁰ causing a decrease in the doping effect in comparison with the DBV⁰ functionalized G-SiO₂ prepared at E = -0.6 V vs HRE (mentioned in Figure 4e).

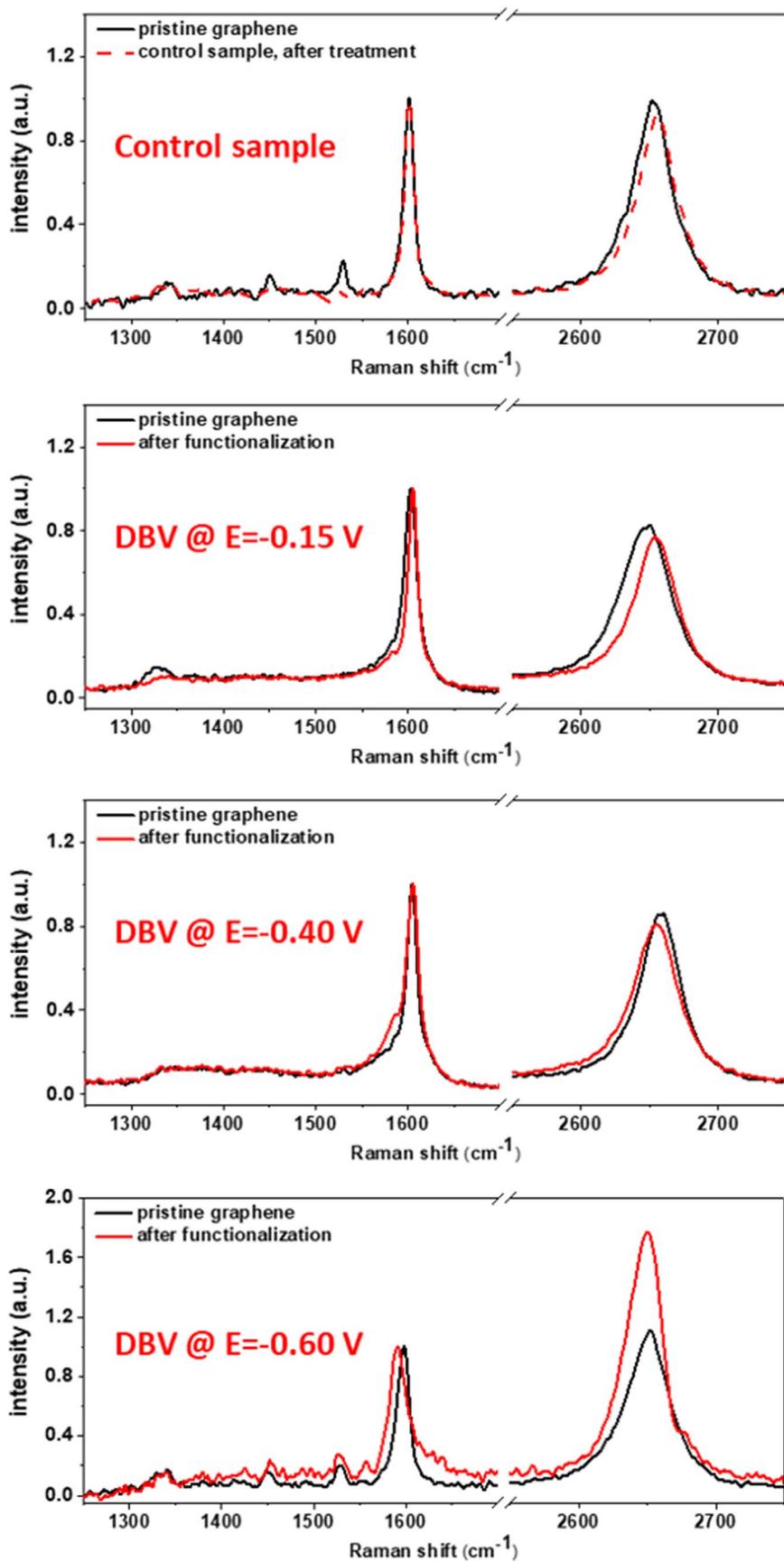


Figure S6: Raman spectra of graphene before and after treatment or functionalization with DBV.

Table S1: G and 2D peak positions, full width at half maximum (FWHM) of the G and 2D band and intensity ratios of the D and 2D band versus the G band, before and after treatment or functionalization with DBV.

		Pos(G)	Pos(2D)	FWHM(G)	FWHM(2D)	I(2D)/(G)	I(D)/I(G)
Control	Before	1600.6±1.5	2649.6±2.7	12.0±2.1	34.4±3.2	0.90±0.06	0.01±0.03
	After	1598.5±2.5	2648.4±2.9	12.0±2.9	34.8±4.3	0.90±0.08	0.01±0.03
DBV@-0.15V	Before	1605.3±2.4	2652.0±4.1	12.1±2.4	39.8±3.7	0.78±0.10	0.04±0.03
	After	1605.4±2.7	2654.4±4.3	13.0±3.7	39.5±3.3	0.87±0.08	0.05±0.03
DBV@-0.40V	Before	1604.3±1.8	2654.2±3.8	15.8±3.8	39.8±5.2	0.93±0.05	0.01±0.03
	After	1604.2±1.8	2653.5±2.9	14.6±3.3	37.5±2.9	0.84±0.08	0.04±0.04
DBV@-0.60V	Before	1599.7±1.4	2648.9±2.7	11.6±2.1	33.8±3.1	0.93±0.08	0.00±0.01
	After	1588.2±2.3	2642.7±3.6	17.4±3.3	38.9±2.7	1.61±0.27	0.03±0.06