Supporting Information

In-Situ Electrochemical Polymerization at Air-water Interface: Surface Pressure Induced, Graphene Oxide Assisted Preferential Orientation of Polyaniline

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Figure S1. (a-d) BAM images showing respective monolayers at air-water interface in different regions of pressure-area isotherm. Pressure-area isotherms for (i) only EGO at the interface (15 mL of 0.1 mg/mL EGO) and (ii) EGO/anilinium at the interface (5 mM anilinium hydrochloride in 0.1 M HCl and 7 mL of 0.1 mg/mL EGO).

Figure S2. (a) UV-Visible spectra of EGO monolayers on quartz substrate deposited using Langmuir trough (numbers given in the figure, 1-5, indicate the number of EGO monolayers on substrate) and (b) Plot of absorbance versus number of EGO monolayers (Absorption spectra were recorded using a Perkin Elmer lambda 35 UV/VIS double beam spectrometer (Spectrum GX, Switzerland. Measurements were carried out on $3 \times 1 \text{ cm}^2$ quartz plates).

Figure S3. Pressure-area isotherms for different concentrations of anilinium in 0.1 M HCl sub-phase and 7 mL of EGO (concentration - 0.1 mg/mL) at the interface (both lift off area and surface pressure increases with increase of anilinium concentration in the sub-phase).

Figure S4. Cyclic voltammograms for potentiodynamic polymerization of EGO/anilinium complex *in-situ* with applied surface pressure in the potential range (a) 0 to 1 and (b) 0 to 0.85 V. Concentration of anilinium chloride used in the sub-phase is 5 mM and 0.1 mg/mL of EGO colloid is used to spread on the water surface. Area of the trough used is 250 cm². Area of glassy carbon electrode used for *in-situ* polymerization is 1 cm².

The voltammograms scanned up to 1.0 V show decrease in currents as a function of number of cycles and is due to the degradation of the polymer. Similar observations have been reported in the literature.³¹ On the other hand, the voltammograms scanned up to 0.85 V shows an increase in currents as a function of number of cycles.

Figure S5. Raman spectra for EGO (red) and EGO/PANI (black) composite in the range 200-2000 cm^{-1} at excitation wavelength of 632 nm with accumulation time of 80 s. The peaks in the range 200-1300 cm^{-1} in EGO/PANI composite (that are absent on EGO) clearly indicates the formation of PANI by electrochemical polymerization of aniline. Blue and pink arrows in the spectra indicate peaks corresponding to PANI and EGO respectively in EGO/PANI composite.

Figure S6. Raman spectra of (a) *ex-situ* polymerized EGO/PANI and (b) *in-situ* polymerized EGO/PANI under surface pressure in the frequency range 1000-2000 cm^{-1} . The predominant bands are due to D and G bands corresponding to the carbon present in EGO. Note the high intensities of the bands.

Figure S7. Raman spectra of PANI for control experiments. (a) *Ex-situ* polymerization with anilinium hydrochloride in the electrolyte on bare GCE, (b) *ex-situ* polymerization of EGO/anilinium modified GCE without anilinium hydrochloride in the electrolyte and (c) *in-situ* polymerization under applied surface pressure with anilinium hydrochloride in the sub-phase and dodecylbenzenesulfonic acid at the air-water interface. Broad features around 450 and 850 cm^{-1} are obtained as in the case of bipolaronic structure.

Figure S8. Steady-state current vs. time response of (a) linear and (b) coiled forms of PANI/EGO modified GCE at -0.6 V vs. SCE in 0.1 M phosphate buffer solution (pH 6.9) upon successive addition of 10 mM H_2O_2 .