# Electronic Supplementary Material

## Fully Inkjet-Printed Multilayered Graphene-Based Flexible Electrodes for Electrochemical Performance

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**Figure S1**

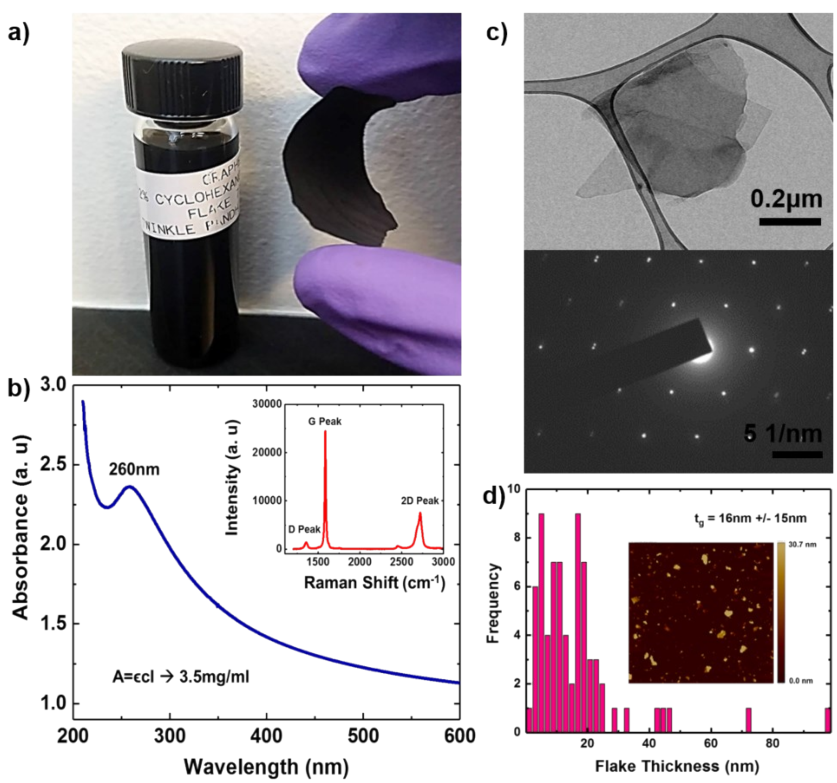

**Figure S1**: Graphene ink characterization **(a)** optical image graphene ink **(b)** UV-Visible absorption spectra and Raman spectra is seen (inset) for graphene/EC flakes on $SiO_2$ **(c)** TEM images and diffraction pattern of graphene flakes **(d)** AFM flake height distribution and (inset) AFM scanned image.

**Figure S2**

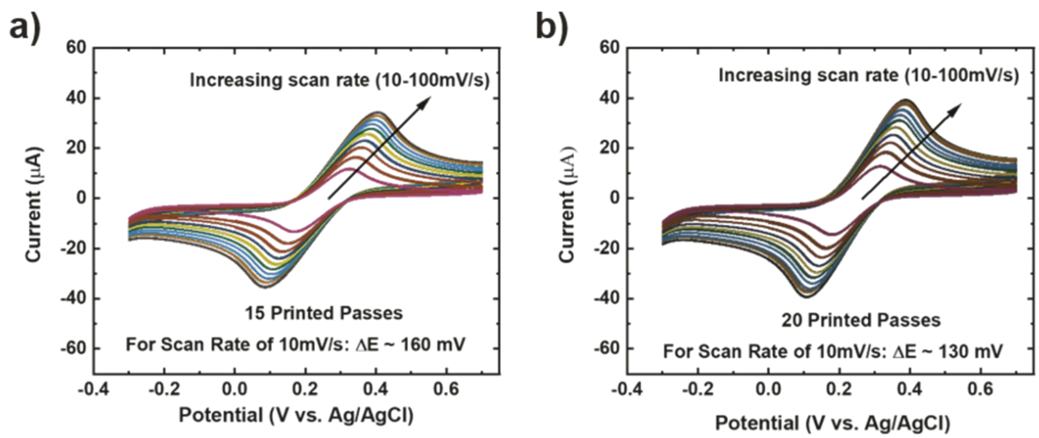


**Figure S2**: (**a**) 15 printed passes and (**b**) 20 printed passes cyclic voltammetry (CV) scans for 5 mM $K_3[Fe(CN)_6]$ in 1 M KCl as the supporting electrolyte with increasing scan rate from 10 to 100 mV/s

**Figure S3**

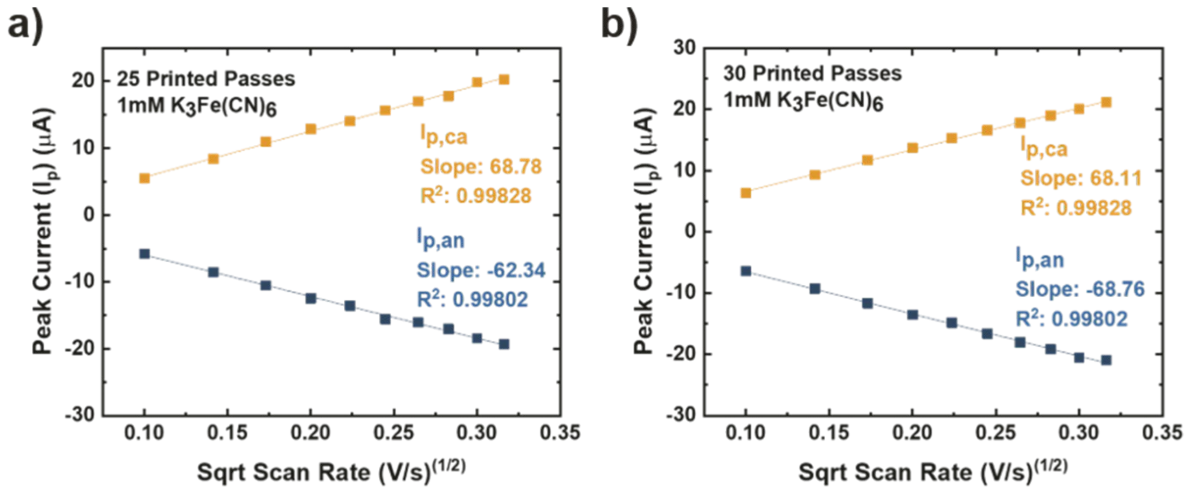


**Figure S3**: Extracted peak current ($I_p$) versus square root of scan rate $(V/s)^{1/2}$ data from the CV measurements of (**a**) 25 printed passes and (**b**) 30 printed passes.

**Figure S4**

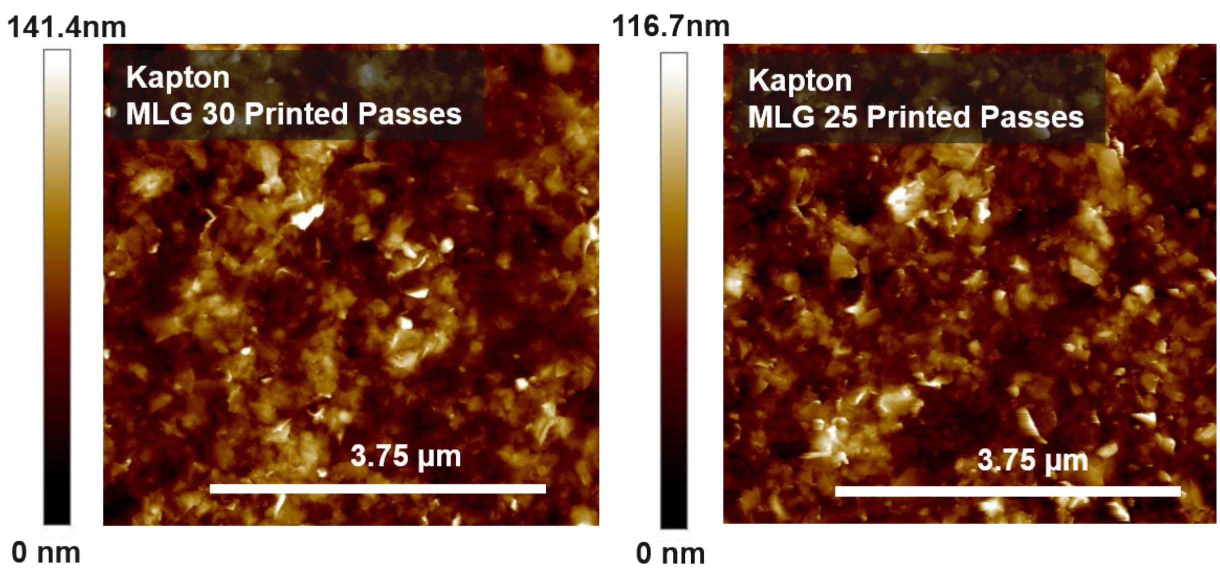


**Figure S4:** AFM scanned image of **a)** MLG 30 printed passes on Kapton and **b)** 25 printed passes on Kapton

**Figure S5**

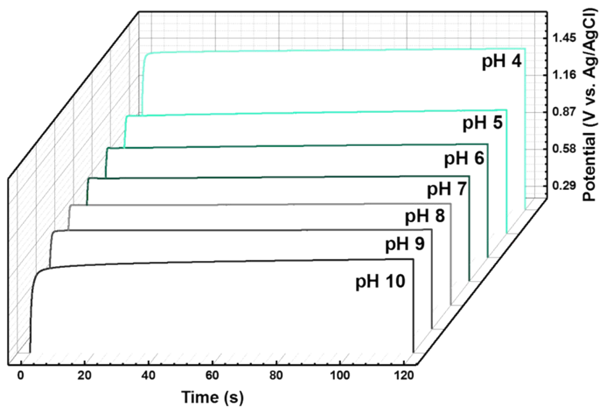


**Figure S5:** Raw static linear pH data vs. potential (potassium phosphate monobasic with sodium hydroxide commercial pH buffer solutions: 3-10 pH) using the 25 passes graphene printed electrode.

**Figure S6**

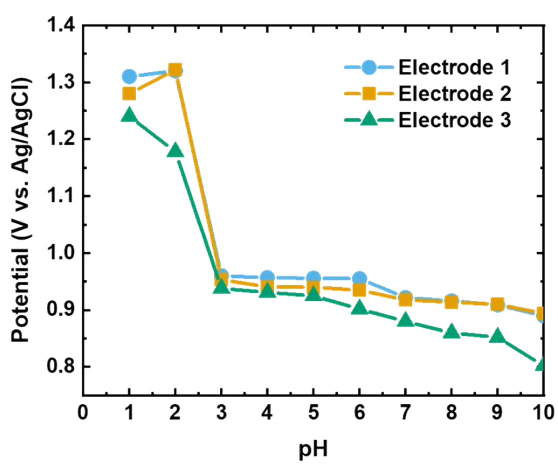


**Figure S6:** Static linear pH data vs. potential (potassium phosphate monobasic with sodium hydroxide commercial pH buffer solutions: 1-10 pH) using the 3 identical 25 passes graphene printed electrodes

**Supplementary Active Surface Area Calculations**

Using AFM analysis (below), we found the surface roughness of 25 printed passes and 30 printed passes.

1. **Surface Roughness Kapton**: Ra = 0.67± 0.05 nm Rq = 0.78 ± 0.09 nm

2. **Surface Roughness Inkjet-printed MLG 25 printed passes on Kapton**: Ra = 29.5± 0.2 nm Rq = 37.6 ± 0.5nm and surface area difference 22.2%

3. **Surface Roughness Inkjet-printed MLG 30 printed passes on Kapton**: Ra = 27.4± 0.2 nm Rq = 35.6 ± 0.5nm and surface area difference 20.8%

This suggests the overall area is 22.2% greater than the geometrical area. Similarly, for 30 print passes the overall area is 20.8% greater than the geometric area.

4. The electrochemical surface area was calculated by using Randles - Sevcik equation.

   $I_p = 2.69\times10^5 \times n^{3/2} A D^{1/2} v^{1/2} C$

   1 mM $K_3[Fe(CN)_6]$ with 1 M KCl as supporting electrolyte, at different sweep rates.

   At T = 298 K, area of the electrode surface is signified as A, the diffusion

   coefficient as D, i.e. $7.6 \times 10^{-6}$ $cm^2$ $s^{-1}$, sweep rate as v, and C is the concentration of

   $K_3[Fe(CN)_6]$.

5. From the slope of the cathodic current ($68.78\times10^{-6}$ and $68.11\times10^{-6}$ respectively) from the plots of $I_p$ vs. $v^{1/2}$ (25 and 30 printed passes (Supplementary Figure S2 a-b)).

6. The surface area of the O-ring was found to be $0.07 cm^2$ and the calculated electrochemical surface area was 0.093 $cm^2$ (25 printed passes) and 0.091 $cm^2$ (30 printed passes)

7. This simulation provided identical CV curves compared to the data for 25 and 30 printed passes respectively, as seen in Figures 3c-d.

8. Using the scan rate (10 mV/s) and electrochemical surface area of ~0.093 $cm^2$, the cathodic diffusion coefficient extracted for
   25 printed passes are **D** = $4.17\times10^{-6}$ $cm^2/s$
   30 printed passes **D** = $6.38\times10^{-6}$ $cm^2/s$ for

9. The average electron transfer rate constants,

   (25 printed passes) **k** =$1.125\times10^{-2}$ cm/s with average double layer capacitance of 43.4 μF and
   (30 printed passes) **k** =$7.34\times10^{-3}$ cm/s with average double layer capacitance of 45.5 μF, where active surface area of ~0.093 $cm^2$ and $\alpha$ = 0.5 (shows symmetric electron transfer).

**Figure S7**

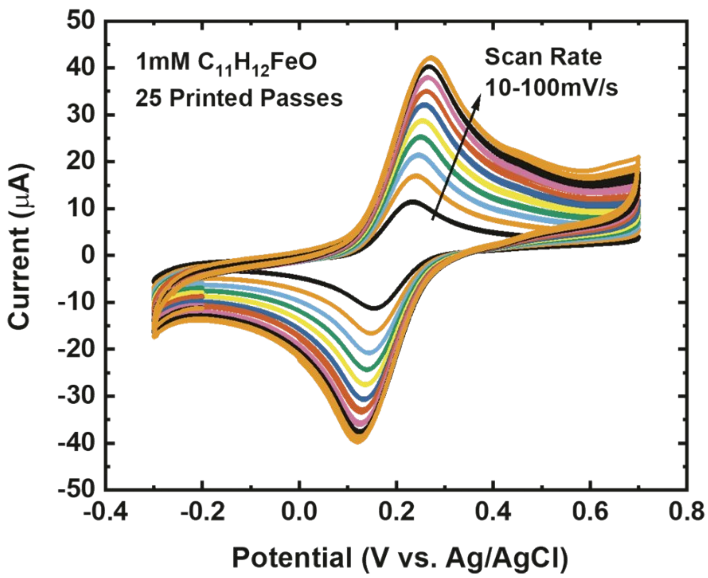


**Figure S7**: Graphene electrode cyclic voltammetry (CV) scan rate data for 1 mM $C_{11}H_{12}FeO$ in 1M KCl as the supporting electrolyte with increasing scan rate 10mV/s-100mV/s for 25 printed passes of graphene.