

## Supporting information of

### Optimizing the Interfacial Electron Transfer Capability of Single Layer

### Graphene by Thermal Annealing

#### Experimental details

**SI-1 Reagents and materials** Potassium chloride (KCl), Potassium ferricyanide ( $\text{K}_3\text{Fe}(\text{CN})_6$ ), isopropanol ( $(\text{CH}_3)_2\text{CHOH}$ ) were all purchased from Sinopharm Co., China. Commercially available monolayer CVD SLGr on  $\text{SiO}_2/\text{Si}$  wafer were provided by Huicheng graphene Co., China. Before use, acetone or chloroform was used to remove the residual PMMA. All the aqueous solutions were all prepared with deionized water (18.2M $\Omega$  cm, Milli Q, Millipore Co.,).

**SI-2 Annealing of the SLGr on  $\text{SiO}_2/\text{Si}$  wafer** Before annealing, the wafer with the pristine SLGr was rinsed with isopropanol and blown dry in  $\text{N}_2$  promptly to avoid any solution residue, and then was cut into the square slides with a edge lenth of 5 mm. The slides were put in to a programmed furnace (SK-G06123K, Tianjin Zhonghuan, China) for thermal annealing. To eliminate residual impurities in the heating tube, an extra one-hour Ar flow was employed before heating. Then, the temperature elevated with a moderate rate about 10  $^\circ\text{C}/\text{min}$ . To exclude the effects of air and water, annealed SLGr electrodes were preserved in a vacuum desiccator.

**SI-3 Electrochemical measurements** An Autolab Electrochemical Work Station (PGSTAT302N, Metrohm Autolab, Switzerland) was used to measure the electrochemical properties of the graphene in a three-electrode system in electrolyte containing 1 mM  $\text{K}_3\text{Fe}(\text{CN})_6$  and 0.1 M KCl at room temperature. Before experiments, the SLGr slide was moved out of the vacuum desiccator, connected with a copper foil mechanically at one end, sealed with an insulative membrane with an exposure area of 2mm by 5mm, and was ready for the working electrode. A Pt plate (5 mm by 10 mm) and an Ag/AgCl electrode were adopted as the counter electrode and the reference electrode. A series of scan rates from 5  $\text{mV s}^{-1}$  to 100  $\text{mV s}^{-1}$  was performed in the cyclic voltammetry (CV) measurements. The data from 5  $\text{mV s}^{-1}$  was used to determine the kinetic rate by Nicholson's method and also simulated by KISSA-1D<sup>©</sup> software.

**SI-4 Characterizations** The optical images of graphene were obtained using an optical microscope (Olympus K-100, Olympus Co., Japan) and a confocal laser microscopy setup (VKX250, Keyence Co.). Single spot Raman spectra and Raman mapping were both performed on Xplora (Horiba Jobin Yvon, France) with 532 nm laser excitation. The laser spot size is about 2  $\mu\text{m}$  using a 50 $\times$  objective (NA = 0.55).