Supporting Information

Tuning the oxygen functional groups in reduced graphene oxide papers to enhance the electromechanical actuation

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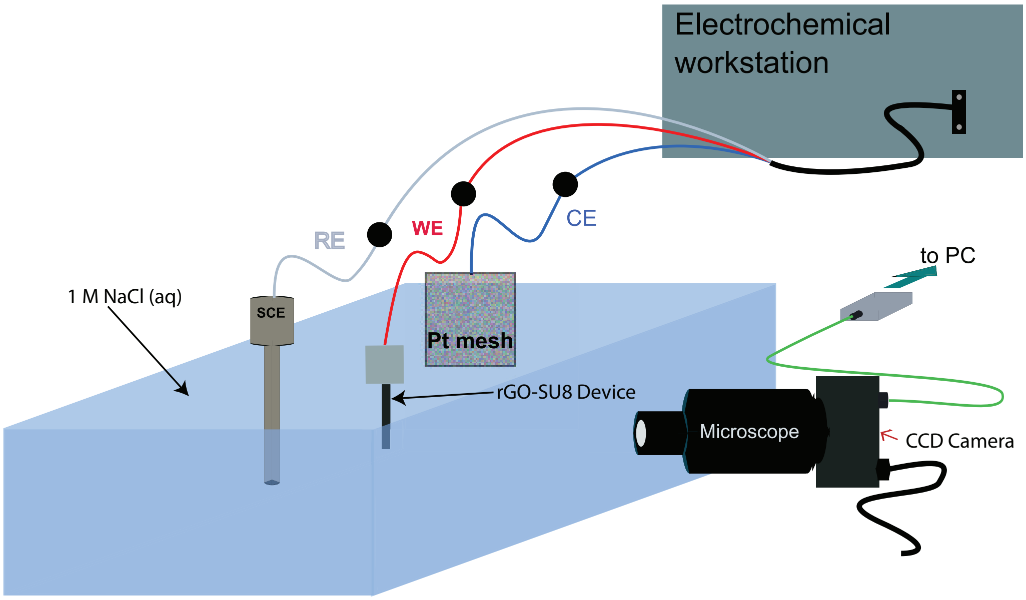

Figure 1 A sketch to demonstrate the experimental set up for measuring the electromechanical actuation of rGO papers. The bimporph device (Working Electrode), a platinum mesh (Counter Electrode), and a saturated calomel electrode (Reference Electrode) were immersed in 1 M NaCl aqueous electrolyte and connected to the electrochemical station. A CCD camera that was mounted on an optical microscope and connected to PC was used to record the tip displacement of the bimorph devices.

The experimental setup for electromechanical actuation of rGO papers is illustrated in Fig. 1. The electrochemical station and the CCD camera mounted on an optical microscope communicate with a computer for conducting square-wave voltage application via chronoamperometry and recording the actuation behaviour, respectively.

We adopted a similar way of applying an electric voltage to the bimorph actuators, as was employed by Liang et al.[1]. Figure 2(a) shows the square-wave voltage applied to our bimorph rGO paper devices. At one given voltage value, three cycles were applied at a frequency of 0.01Hz. After applying a positive voltage value, a negative voltage was applied. The magnitude of voltage gradually increased from 0.1V to 1V. Figure 2(b) shows the detected current results for the R_w0.52 hydrogel rGO paper bimorph device. Integrating the current results leads to the non-functional residue charges accumulated in the system as shown in Figure 2(c). Note that even though a zero voltage was applied during the discharging cycle for about 100s (a same time period as the charge cycle), it doesn't completely remove the residue charge in the system. As clearly seen in Fig. 2(c), this error continues to augment every time when the system was being charged. This is a common phenomenon during voltage application on graphene papers in aqueous electrolyte. A similar behaviour is seen in the study conducted by Liang et al., which clearly shows a higher area under the current vs time graph for the charge cycle compared that of the discharge cycle.[1] Hence, we calculated the amount of charge that was being discharged in the last discharging cycle to approximate the effective charges stored in the system, i.e., the difference of charge values between at the end of the 3rd charging cycle and at the end of the 3rd discharging cycle. This effective charge value was used to calculate the charge per-atom for each type of rGO papers in Figure 4 (b) in main text.

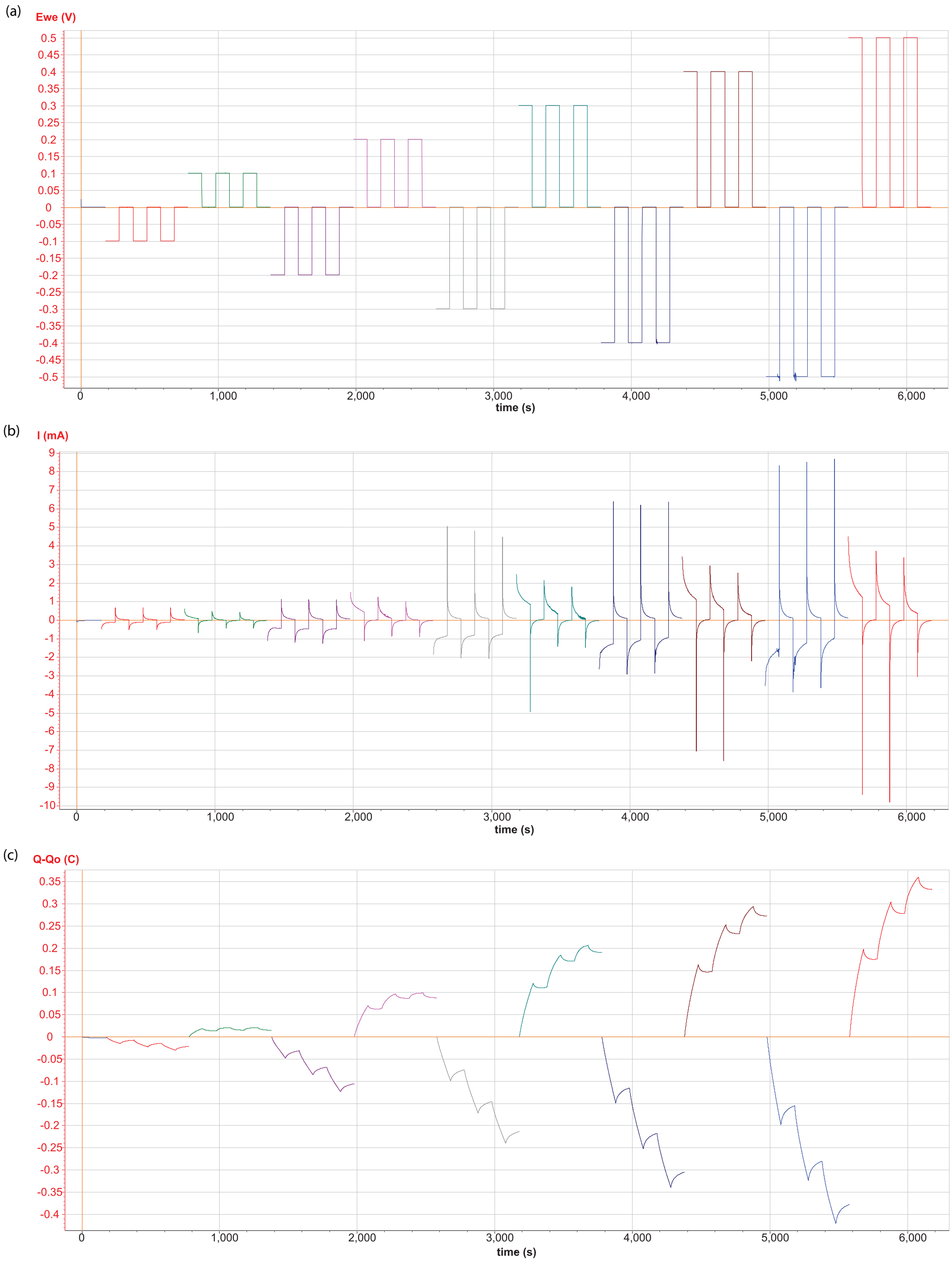

Figure 2 Raw data obtained during an actuation experiment for R_w0.52 hydrogel rGO paper. (a) Square-wave voltage application for three cycles at each voltage for 100 seconds of charge/discharge period. (b) The current flow of the system during the voltage application and (c)the amount of charge accumulated in the device during the voltage application, achieved by integrating the amount of current flow with respect to time.

References

[1] J. Liang, Y. Huang, J. Oh, M. Kozlov, D. Sui, S. Fang, R. H. Baughman, Y. Ma and Y. Chen, *Advanced Functional Materials*, 2011, **21**, 3778.