Electronic Supplementary Information (ESI)

High capacitive performance of flexible and binder-free graphene-polypyrrole composite membrane based on in-situ reduction of graphene oxide and self-assembly

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Fig. S1. Zeta potential profiles of graphene oxide dispersion (a) and polypyrrole nanowire dispersion (b). Fig. S1 shows the zeta-potential profiles of surface charges of GO dispersion. As a comparison, the zeta-potential profile of polypyrrole nanowire dispersion is shown in the same figure. The zeta potential of GO is about -52.5 mV, suggesting the negatively charged surface due to the ionization of functional groups. In contrast, the zeta potential of PPy is about +48 mV suggesting the positively charged nature. Thus, the electrostatic interaction may govern the initial self-assembly of GO and PPy nanowires.
Fig. S2. Core level C 1s XPS spectra of GO (a) and RGO-PPy membrane (b).

For the GO sample, the main peak centered at about 284.6 eV originates from the graphitic sp2 carbon atoms (Figure S2a). The binding energies located at 286.7 eV and 287.9 eV are due to carbon atoms connecting with oxygenate groups, such as C=O and O=C=O. The C 1s XPS spectrum of RGO sample exhibits the same peaks to those of the GO sample, but the intensity of the peaks related to oxygenate groups is much weaker than those of GO (Figure S2b), suggesting the considerable deoxygenation by the reduction process. The small peaks indicate the presence of residual oxygenate groups on the RGO sheets.
Fig. S3. SEM images (a-c) and digital picture (d) of RGO-PPy membranes with various thicknesses and shapes.

Fig. S4. Cycle voltammetry curves at various scan rates (a) and charge/discharge curves at various densities (b) of a two-electrode cell with RGO-PPy aerogel as both electrodes.
Fig. S5. Charge–discharge curves of two-electrode cells with symmetric RGO–PPy foam and RGO-PPy membrane, respectively. The IR drops for charge–discharge curves are shown in the Figure.

Fig. S6. Cycle voltammetric curves (a) and specific capacitance (b) of two-electrode cells with symmetric RGO–PPy foam (~1.7 mg cm\(^{-2}\)) and RGO-PPy membranes with different thicknesses.