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Supporting Information

Graphene Quantum Dots/$Co(OH)_2$ Electrode on Nanoporous Au-Ag Alloy for Superior Hybrid Micro-supercapacitor

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1. Calculation of mass loading

Power and energy densities are important parameters to evaluate the performance of as-fabricated supercapacitors, which can be appreciated according to mass loading, area and volume. Here, it is difficult for us to directly weigh the loading mass of active material. Instead, we can calculate the loading mass according to a previously reported approach (*Adv. Mater.* 2013, 25, 3302; *Adv. Mater.* 2014, 26, 4279).

(1) Loading mass of $Co(OH)_2$. Assuming 100% current efficiency via the reactions, $NO_3^-+7H_2O+8e^- \rightarrow NH^{4+}+10OH^-$ and $Co^{2+}+2OH^- \rightarrow Co(OH)_2$, the mass of electrodeposited $Co(OH)_2$ can be calculated using the equation,

$$m = \frac{ItM}{neN_A},$$

where I is the applied current, t is the deposition time, M is the molecular weight of $Co(OH)_2$, n is the number of transferred charge, e is the electron charge and N_A is the Avogadro's number. As a result, the mass of $Co(OH)_2$ was calculated to be 0.24 mg.

(2) Loading mass of GQDs. We calculate the loading mass of GQDs according to the ratio of Co: C from XPS results (Figure 1 in the manuscript) as,

Atom	Integrated area (S)	Sensitivity factor (I)	Estimated Mass
Co	116190	3.255	0.15mg
C	5616	0.296	0.016mg

Here, we assume that all carbon information comes from GQDs, so that the mass of GQDs is about 0.016 mg, and the whole loading mass of active electrode, GQD/$Co(OH)_2$, is 0.256 mg. Note that the calculated results are the largest mass of active electrode, and the actual mass should be less than this result.

2. Charge balance the between positive and negative electrodes

To assemble safe hybrid micro-SCs using GQDs/$Co(OH)_2$ electrode, it is important to balance the charge between positive and negative electrodes. Figure S5(a) shows the CV curves of GQDs/$Co(OH)_2$ and AC electrode at a scan rate of 50 mV s^{-1} in 3 M KOH electrolyte, which indicate a stable cell voltage of 1.4 V. Figure S5(b) shows the GCD curves of each electrode vs. SCE, from which we can see that total charge balance is achieved between the positive and negative electrodes, which can avoid the device damage. The specific capacitance of AC and the specific capacity of GQDs/$Co(OH)_2$ at a current density of 1 mA cm^{-2}, are 100 F g^{-1} and 308 C g^{-1}, respectively. Therefore, an optimal GQDs/$Co(OH)_2$ to AC mass ratio of 0.32 enables charge flow balance between the positive and negative electrodes ($Q^+ = Q^-$) according to the equation $\frac{m_+}{m_-} = \frac{C_{sp-} \times \triangle V_-}{C_+}$, where m_+ is the mass of GQDs/$Co(OH)_2$, m_- is the mass of the AC, C_{sp-} is the specific capacitance of AC, C_+ is the specific capacity of

GQDs/$Co(OH)_2$ and ΔV_- is the potential drop upon discharging (excluding the IR drop) of AC.

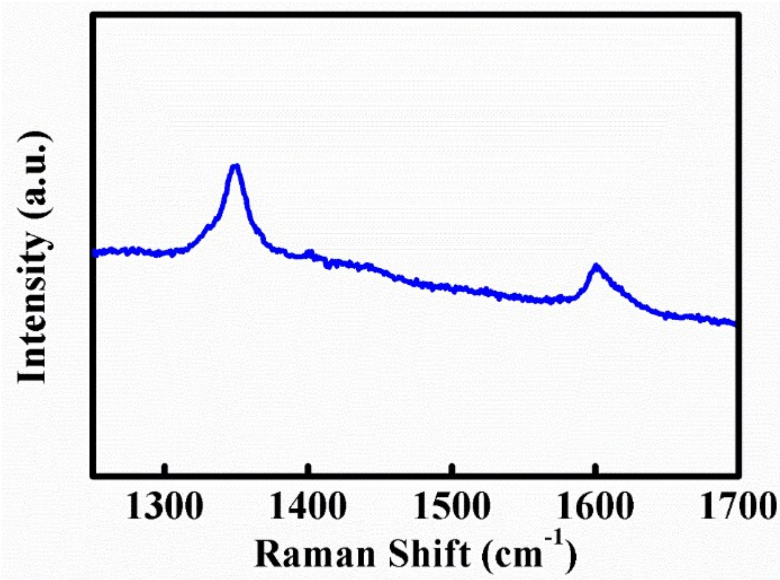

Figure S1 Raman spectra of pure GQDs in solution.

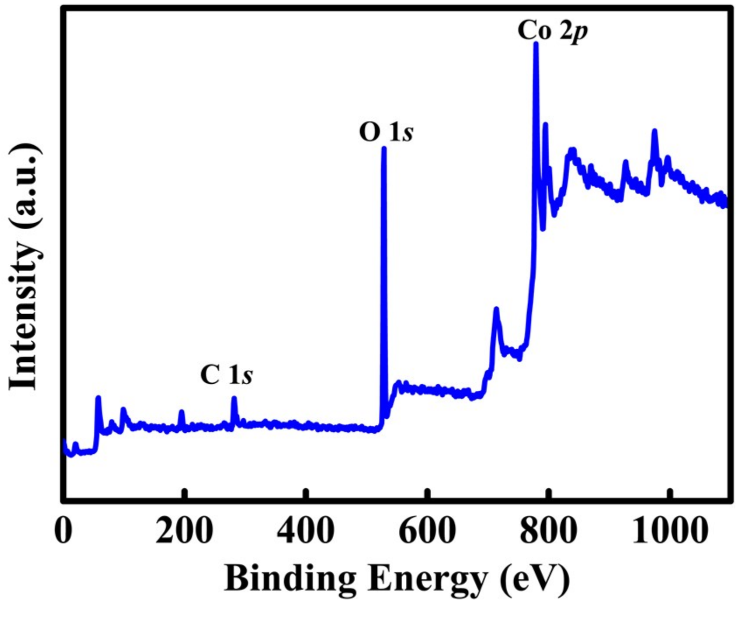

Figure S2. XPS spectra of GQDs/$Co(OH)_2$ nanosheets showing the presence of C 1*s*, O 1*s* and Co 2*p*.

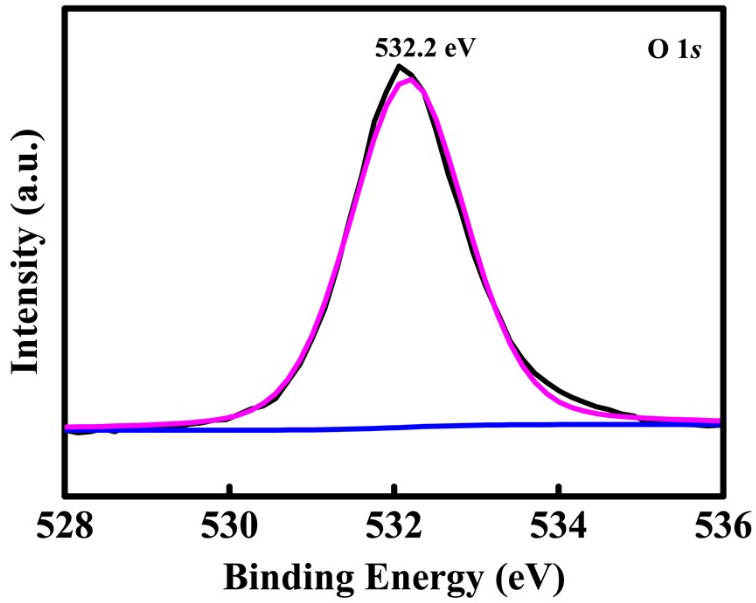

Figure S3. O 1*s* XPS spectra of GQDs/$Co(OH)_2$ highlighting the peak at 532.2 eV.

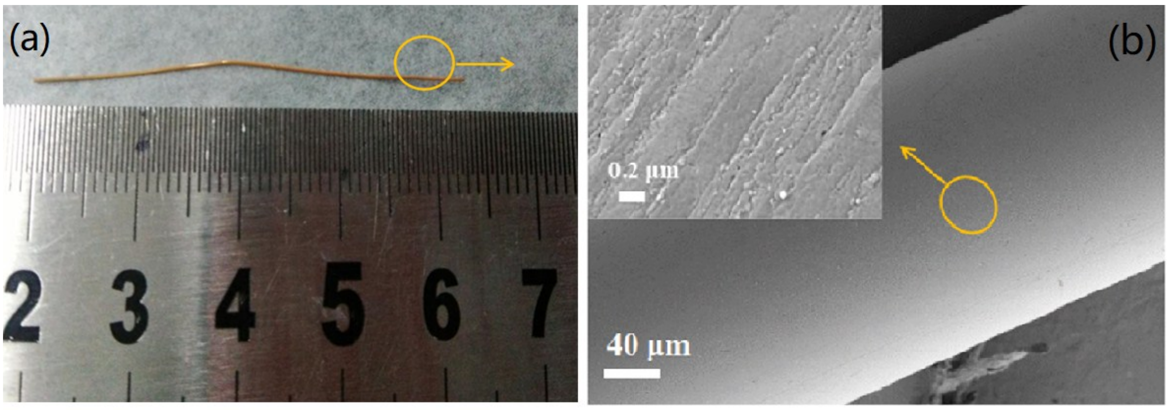

Figure S4. (a) Real picture of Au-Ag alloy wire before chemical etching. (b) SEM image of the part highlighted in (a) at low and high (Inset) magnification.

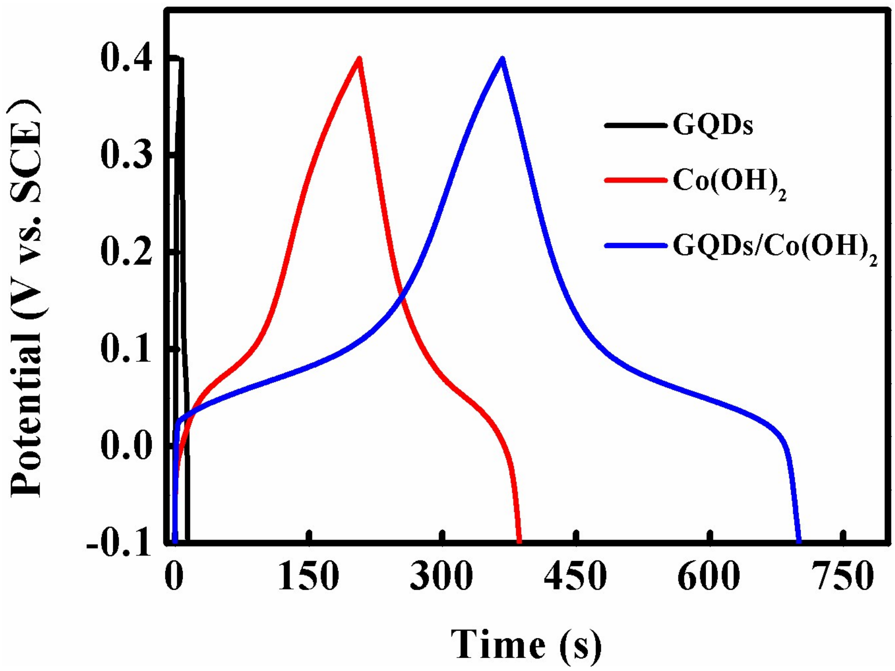

Figure S5. GCD curves of GQDs, $Co(OH)_2$ and GQDs/$Co(OH)_2$ electrodes on NPAAW.

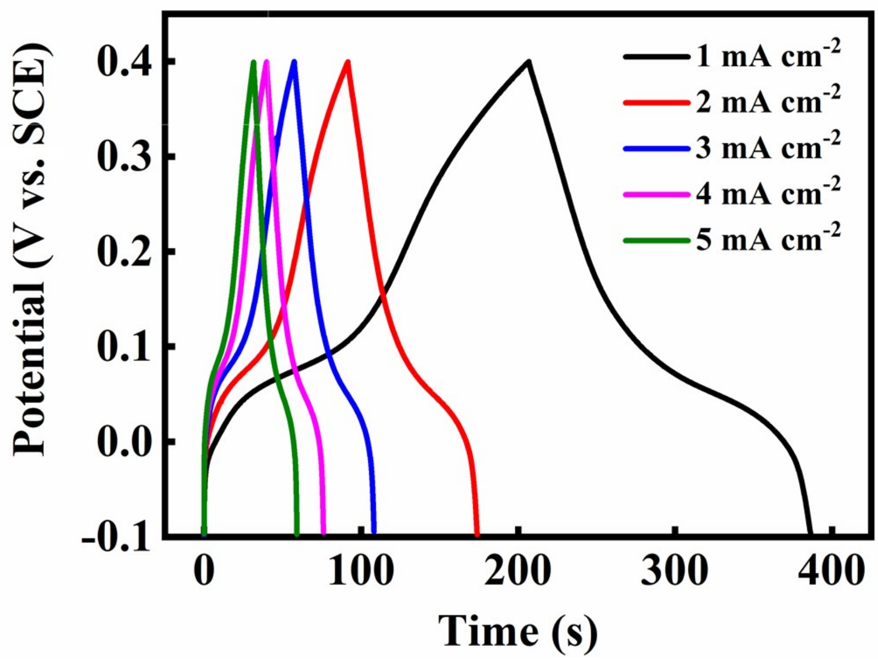

Figure S6. GCD curves of the $Co(OH)_2$ electrode at different current densities.

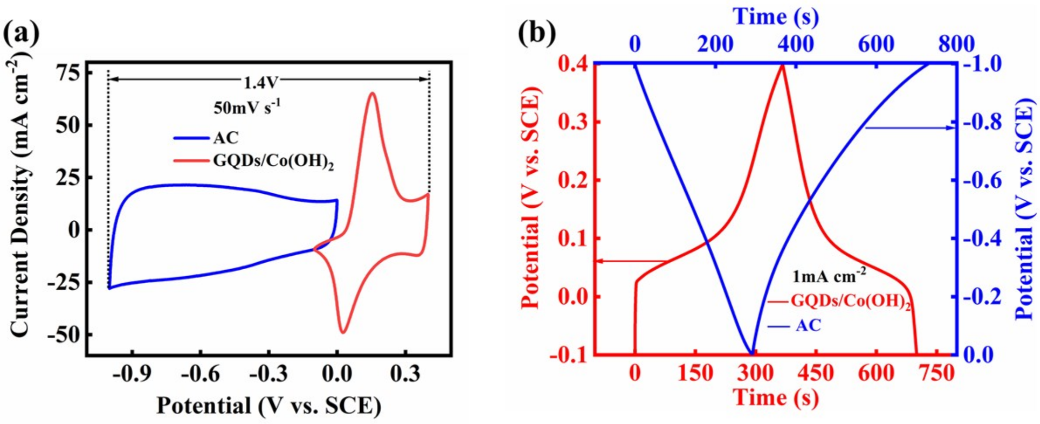

Figure S7. (a) CV curves of the $GQDs/Co(OH)_2$ electrode and the AC electrode at a scan rate of 50 mV s^{-1} in 3 M KOH electrolyte. (b) GCD curves of the $GQDs/Co(OH)_2$ electrode and the AC electrode at a current density of 1 mA cm^{-2} in 3 M KOH electrolyte.

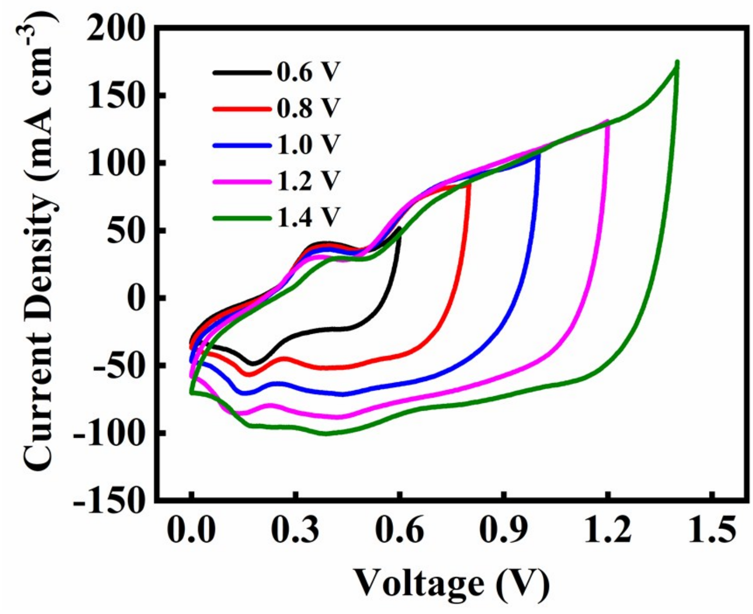

Figure S8**.** CV curves of $GQDs/Co(OH)_2$//AC hybrid supercapacitor within different voltage windows.

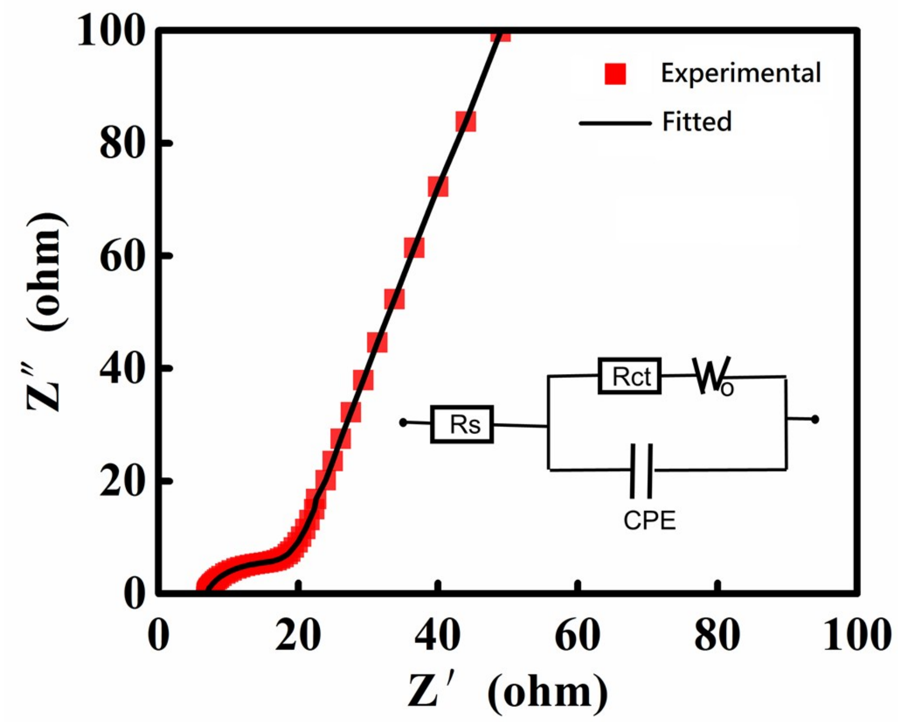

Figure S9. Nyquist plots for the GQDS/$Co(OH)_2$//AC hybrid supercapacitor device. Inset shows the equivalent circuit model.

Figure S10**.** Real picture of an all-solid-state flexible supercapacitor device and the photo of a LED powered by two in-series GQDs/$Co(OH)_2$//AC hybrid supercapacitors.