Supporting information

Facile one-step synthesis of Co(OH)₂ microspheres/graphene composites for efficient supercapacitor electrode material

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 $Co(OH)_2/GN-1$ composite exhibit the better capacitive behavior. To further confirm the role of GN played in the composite, another experiment is designed in view of changing the amount of GN. $Co(OH)_2/GN-4$ and $Co(OH)_2/GN-5$ have been synthesized using 10 mg and 40 mg GO, respectively, in which the amount of other raw material remains the same to $Co(OH)_2/GN-1$ composite.



Fig. S1 SEM images of Co(OH)₂/GN-4 (a,b) and Co(OH)₂/GN-5 (c,d).

The results show that the $Co(OH)_2$ particles in $Co(OH)_2/GN-4$ and $Co(OH)_2/GN-5$ were relatively poorly-dispersed. When the amount of GO decreases to 10 mg, a large number of $Co(OH)_2$ particles agglomerates (Fig. S1a,b), however, when the amount of GO increases to 40 mg, more GN sheets restack (Fig. S1c,d), which possibly result in the inferior electrochemical performance.



Fig. S2 (a) Discharge curves of $Co(OH)_2/GN-1,4,5$ composites at the current density of 0.1 A/g , (b) The C_{spec} values of $Co(OH)_2/GN-1,4,5$ composites as a function of current density, (c) The cycling performance of $Co(OH)_2/GN-1,4,5$ composites at a constant current density of 1 A/g. (d) CV curves of $Co(OH)_2/GN-1,4,5$ composites at the scan rate of 50 mV/s.

The electrochemical performance of the composite electrode materials is measured by GCD, CV and EIS. The C_{spec} value is 216.4 F/g for Co(OH)₂/GN-4 and 230.3 F/g for Co(OH)₂/GN-5 at 0.1 A/g, which are much lower than that of Co(OH)₂/GN-1 (433 F/g) (Fig. S2a). In addition, for Co(OH)₂/GN-4 and Co(OH)₂/GN-5, 73.9% and 93.3% retention at 10 A/g are obtained, respectively, and 90.5% and 93 % retention of C_{spec} over 1000 cycles, which is lower than that of Co(OH)₂/GN-1 composite (Fig. S2b,c). It suggests that more uniform particle size distribution of Co(OH)₂ particles on the GN sheets and most optimized amount of GO is more important for the electrochemical performance of the composites. The intimate interaction between GN sheets and

well-dispersed $Co(OH)_2$ particles can minimize the interfacial resistance of the charge transfer process in $Co(OH)_2/GN-1$, which results in the best electrochemical performance.

Fig. S2d represents the CV plots of $Co(OH)_2/GN-1,4,5$ at scan rates of 50 mV/s. In the $Co(OH)_2/GN-1,4$ composite, a pair of obvious redox peak can be clearly observed. For $Co(OH)_2/GN-5$ composite, the redox peak is not very obvious, due to the amount of $Co(OH)_2$ is much less. The current density response for the $Co(OH)_2/GN-1$ composite electrode is much larger than that of $Co(OH)_2/GN-4$ and $Co(OH)_2/GN-5$ at the same scan rate, indicating that $Co(OH)_2/GN-1$ has a higher C_{spec} , which is consistent with the results of GCD.



Fig. S3 EIS of Co(OH)₂/GN-1,4,5 composites electrodes.

Fig. S3 shows Nyquist plots of the Co(OH)₂/GN-1,4,5 composites. It can be calculated that the R_s values for Co(OH)₂/GN-1,4,5 are 0.48, 0.67 and 0.49 Ω , respectively, and the Co(OH)₂/GN-1 has the smallest R_{ct} value (0.49 Ω) (0.70 Ω for Co(OH)₂/GN-4 and 0.51 Ω for Co(OH)₂/GN-5) and the low-frequency line is closer to the vertical, which suggests that the superior electronic conductivity of GN sheets is preserved after loading of Co(OH)₂/GN-1 composite electrode material. It is consistent with the GCD results, in which the unique structure of the Co(OH)₂/GN-1 composite leads to efficient utilization of such pseudo- and double-layer capacitors to result in the excellent supercapacitor performance.

All the results demonstrate that $Co(OH)_2/GN-1$ have the best electrochemical performance due to the optimized amount of GN. The synergistic effect of GN sheets and $Co(OH)_2$ particles can not only improve the electron conductivity but also prevent agglomeration of particles or irreversible restacking of GN sheets, in which GN can

intimately interact with the well-dispersed $Co(OH)_2$ particles to form an open pore system and minimizing the interfacial resistance of the charge transfer process to result in excellent capacitive behavior.