Electronic Supplementary Information (ESI)

Asymmetric electrochemical capacitors with high energy and power density based on graphene/CoAl-LDH and activated carbon electrodes

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1. Thermogravimetric (TG) analysis of RGO/LDH and LDH

Fig. S1 (a) TG curves of RGO/LDH and LDH from 30 °C to 800 °C. (b) XRD pattern of the decomposition product of RGO/LDH after TG measurement.

Figure S1 (a) shows the TG curves of RGO/LDH and LDH, which were tested in air atmosphere from 30 °C to 800 °C at the heat rate of 20 °C/min. the weight loss of LDH can be divided into two stages. The first stage of weight loss of about 12% below 190 °C is due to the removal of interlayer water molecules.¹ The second stage of weight loss is due to the dehydroxylation and the removal of interlayer anions¹. The TG curve of RGO/LDH also shows two stages which are similar to those of LDH. Moreover, the decomposition of RGO in RGO/LDH also occurs during the TG analysis. Figure S1 (b) shows the XRD pattern of the decomposition product of RGO/LDH after TG analysis. The diffraction peaks can be assigned to the spinel structure of Co₂AlO₄ (PDF card no. 38-0814). Then it can be confirmed after the TG analysis that the decomposition product of RGO/LDH is Co₂AlO₄ spinel, and the molar ratio of Co and Al is 2:1 in RGO/LDH. From the TG curve of LDH and

RGO/LDH, the weight loss of LDH is about 31% during the TG analysis, thus it can be calculated that the Co_2AlO_4 occupies about 69% in LDH. The weight loss of RGO/LDH is about39%, thus Co_2AlO_4 is 61% in RGO/LDH. So the weight percentage of LDH in RGO/LDH can be calculated as 88% whereas the mass ratio of RGO and LDH is 12%/88%=0.136.

(a) Reduced graphene oxide (b) Reduced graphene oxide (c) Go (c) Go

2. The SEM images and XRD pattern

Fig. S2 (a, b) SEM images of RGO, (c) SEM image of GO and (d) XRD pattern of GO.

Figure S2 (a, b) shows the SEM images of RGO, which is the product of RGO/LDH after 3 h reaction with 2 M HCl. During the reaction the RGO agglomerate into smaller size of about 10 um. Figure S2 (c) shows the SEM image of GO. The image is not so clear which is due to the poor electric conductivity of GO. From the image, we can see the size of GO is about 50 um, much larger than that of RGO. Figure S2 (d) is the XRD pattern of GO. The strong (001) peak is the characteristic peak of GO

3. The AFM image of GO



Fig. S3 AFM image of GO nanosheets. From the AFM image of GO, it is obvious that the thickness of GO is about 1nm.



4. Energy-dispersive X-ray spectroscopy (EDS) of RGO/LDH

Fig S4 (a) SEM image of RGO/LDH and (b) EDS spectrum of RGO/LDH.

The results of energy-dispersive X-ray spectroscopy (EDS) of RGO/LDH also confirmed that the molar ratio of Co and Al is about 2:1 in RGO/LDH, which is consistent with the TG analysis.



5. Electrochemical properties of RGO/LDH

Fig. S5 (a) CV curves of RGO/LDH at different scan rates ranging from 5 to 80 mV/s. (b) Specific capacitance of RGO/LDH ECs at different scan rates.

The RGO/LDH electrodes are measured within a potential window from -0.1 V to 0.55 V vs. SCE at a scan rates ranging from 5 mV/s to 80 mV/s. The CV curves of RGO/LDH in figure S5(a) exhibit a pair of cathodic and anodic peaks, indicating the existence of fast Faradic redox reactions during CV tests. Figure S5(b) shows the dependence of specific capacitance of RGO/LDH ECs at different scan rates. The specific capacitance of RGO/LDH reaches 510F/g at the scan rate of 5mV/s. The specific capacitance of RGO/LDH decreases with the increase of scan rate. When the scan rate reaches 80 mV/s, the specific capacitance remains 138F/g.

Notes and References

1. X. Dong, L. Wang, D. Wang, C. Li and J. Jin, *Langmuir*, 2012, **28**, 293-298.