Ultrathin Ni(OH)₂ layer coupling with graphene for fast electron/ion transport in supercapacitor

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ABSTRACT Integration of fast electrochemical double-layer capacitance and large pseudocapacitance is a practical way to improve the overall capability of supercapacitor, yet remains challenging. Herein, an effective cyanogel synthetic strategy was demonstrated to prepare ultrathin Ni(OH)₂ nanosheets coupling with conductive reduced graphene oxide (rGO) (rGO-Ni(OH)₂) at ambient condition. Ultrathin Ni(OH)₂ nanosheet with 3–4 layers of edge-sharing octahedral MO₆ maximally exposes the active surface of Faradic reaction and promotes the ion diffusion, while the conductive rGO sheet boosts the electron transport during the reaction. Even at 30 A g⁻¹, the optimal sample can deliver a specific capacitance of 1119.52 F g⁻¹, and maintain 82.3% after 2000 cycles, demonstrating much higher electrochemical capability than bare Ni(OH)₂ nanosheets. A maximum specific energy of 44.3 W h kg⁻¹ (148.5 W kg⁻¹) is obtained, when assembled in a two-electrode system rGO-Ni(OH)₂/rGO. This study provides an insight into efficient construction of two-dimensional hybrid electrodes with high performance for the new-generation energy storage system.

Keywords: two-dimensional nanomaterials, Ni(OH)₂ nanosheet, graphene, cyanogel synthetic strategy, supercapacitor

INTRODUCTION

Energy storage system has drawn unprecedented interests in academia and industry, owing to the ever-increasing demand for back-up power sources, large-scale storage of sustainable energy, electric vehicles, and portable electronic devices [1–6]. Among them, supercapacitors with high power density and exceptional cycle stability are

especially attractive for worldwide researchers. Based on the reaction mechanism, two types of supercapacitors are well defined, pseudocapacitors and electrochemical double-layer capacitors (EDLCs) [7,8]. EDLCs store energy by reversible ion adsorption at the electrolyte/electrode surface, which highly depends on the specific surface area. In contrast, pseudocapacitors work through Faradaic redox reactions both on and near the surface of the electroactive materials, which usually have larger capacitance than that of EDLCs [9–11]. However, the relatively low energy density is a common bottleneck for both types of supercapacitors, severely limiting their widespread applications.

To address this key issue, extensive studies have been done to develop high-performance electrode materials, including conducting polymers [12,13], heteroatom-doped carbon materials [14–19], transition metal phosphates [20], and transition metal oxides/hydroxides [21–27]. Lin *et al.* [28] reported a nitrogen-doped mesoporous carbon, of which the specific capacitance can reach to 855 F g⁻¹ at 1 A g⁻¹ in 0.5 mol L⁻¹ H₂SO₄ solution with the specific energy of 41 W h kg⁻¹ (19.5 W kg⁻¹) in 2 mol L⁻¹ Li₂SO₄ electrolyte. Recently, Wang's group [29] took advantages of hypercrosslinked polymers to endow N,S-doped porous carbon with the capacitance of 464 F g⁻¹ at 0.2 A g⁻¹. Self-assembly and cross-linking of conductive polymers (polyaniline (PANi), polypyrrole, etc.) into three-dimensional (3D) hydrogel electrodes were adopted for supercapacitors by Moussa *et al.* [30]. The best hydrogel electrode for the assembled flexible supercapacitors was the as-prepared PANi nanofibers with the

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capacitance of 492 F g^{-1} at 1 A g^{-1} .

Among the electrode materials for supercapacitors, transition metal hydroxides/oxides (e.g., Co(OH)_2 , Ni(OH)_2 , RuO_2 , MnO_2 , NiO , and Co_3O_4) [31–36], exhibit unique merits, such as high theoretical capacitance, reversible redox reaction, and multiple oxidation states. Thereinto, nickel hydroxide is an attractive material, thanks to its cost-effectiveness, well-defined redox behavior, high theoretical capacitance (2082 F g^{-1}) and eco-friendliness [37–39]. Wei *et al.* [40] prepared an assembly of flower-like Ni(OH)_2 under mild conditions. By assembling Ni(OH)_2 with carbon tube, the electrode yields a larger capacitance of 2225.1 F g^{-1} at 0.5 A g^{-1} . The specific capacitance of the hierarchical porous Ni(OH)_2 nanosheets, which was fabricated by an aqueous co-precipitation method followed by aging treatment, can reach to 2378.7 F g^{-1} at 1 A g^{-1} . Despite the important progress, there is still a large room to maximize the utilization of near-surface and to enhance the electrical conductivity of Ni(OH)_2 ($\sim 10^{-17} \text{ S cm}^{-1}$).

Herein, we report a simple cyanogel synthetic strategy to directly fabricate ultrathin Ni(OH)_2 nanosheets coupling with reduced graphene oxides (rGO). Ni(OH)_2 nanosheets with several layers of edge-sharing octahedral MO_6 maximally expose the reaction surface, while the conductive rGO boosts the electron transport during the electrochemical reaction. The optimal sample, 5% rGO- Ni(OH)_2 , demonstrates the best rate performance and capacity retention, especially at 30 A g^{-1} still retaining 82.3% after 2000 cycles. This can be credited to the synergy effect from the rGO and ultrathin Ni(OH)_2 nanosheets, which simultaneously improves the electrical conductivity and superficial reaction kinetics of electrode. The scalable synthetic method and the resultant superior electrochemical performance provide a new pathway to prepare the cost-efficient electrode materials for the future.

EXPERIMENTAL SECTION

Reagents and chemicals

Sodium borohydride (NaBH_4) and nickel(II) chloride hexahydrate ($\text{NiCl}_2 \cdot 6\text{H}_2\text{O}$), were bought from Sinopharm Chemical Reagent Co., Ltd. (China). GO was obtained from Nanjing JCNANO Technology Co., Ltd. (China). Potassium hydroxide (KOH) was purchased from Alfa Aesar (China). Potassium cyanonickelate (II) ($\text{K}_2\text{Ni}(\text{CN})_4 \cdot x\text{H}_2\text{O}$) was bought from Macklin Biochemical Co.,

Ltd. (China).

Syntheses of the rGO- Ni(OH)_2 nanosheets

The rGO- Ni(OH)_2 nanosheets were prepared by modifying the procedure reported in the previous work [41–43]. Briefly, 1 mL of NiCl_2 (1 mol L^{-1}) was mixed with different amounts of GO at room temperature to form a uniform solution, following by the addition of 1 mL of $\text{K}_2[\text{Ni}(\text{CN})_4]$ (0.5 mol L^{-1}) to generate a grey GO- $\text{K}_2[\text{Ni}(\text{CN})_4]$ - NiCl_2 cyanogel. After that, NaBH_4 (0.13 g mL^{-1} , 40 mL) solution was poured into the grey GO- $\text{K}_2[\text{Ni}(\text{CN})_4]$ - NiCl_2 cyanogel under vigorous stirring for 8 h at 50°C . At last, the black solid powder was centrifuged out and washed with deionized water for six times and ethanol for two times, and dried at 40°C for 5 h to obtain different $X \text{ wt\%}$ rGO- Ni(OH)_2 ($X=3, 5$ and 7) nanocomposites.

Preparation of the working electrode

The electrode was constituted by mixing the electrode materials, acetylene black and polytetrafluoroethylene (8:1:1) in ethyl alcohol to form a slurry. After then, the mixture was dropped onto a 1-cm^2 Ni foam and dried at 80°C for 12 h. After drying, it was pressed at 10 MPa. The mass loading was controlled to be about 0.5 mg cm^{-2} .

Structural characterizations

The DX-2700 X-ray diffractometer with Cu K α radiation was used to test the X-ray diffraction (XRD). The AXIS ULTRA spectrometer was used to test the X-ray photoelectron spectroscopy (XPS) with Al K α radiation. The binding energy was amended at 284.8 eV using the C 1s spectrum. Scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDX, Quanta 200) images were taken by SU-8020. Brunauer-Emmett-Teller (BET) surface was examined by N_2 adsorption-desorption *via* ASAP 2460. Transmission electron microscopy (TEM) and high-resolution TEM (HR-TEM) images were characterized by TECNAI G 2 F20. The thickness of the electrode material was examined by atomic force microscopy (AFM, Dimension Icon.).

Electrochemical measurements

The galvanostatic charge-discharge (GCD), cycling voltammetry (CV) and electrochemical impedance spectroscopy (EIS) were tested in a three-electrode system using 6 mol L^{-1} KOH as the electrolyte. Pt foil was used as the counter electrode, and Hg/HgO as the reference electrode. GCD curves were obtained *via* LAND CT-2001A instrument. CV and EIS were obtained by using a CHI

660D electrochemical workstation. CV curves were tested with the scanning rate of 2, 5, 8, 10 and 15 mV s^{-1} . GCD was tested at 1–30 A g^{-1} . The voltage range was from 0 to 0.5 V. At the open-circuit potential, the EIS was conducted by exerting alternating current amplitude of 5.0 mV with the frequency ranging from 100 kHz to 0.01 Hz. The capacitance (C , F g^{-1}) was calculated based on the following equation:

$$C = I \Delta t / m \Delta V, \quad (1)$$

where ΔV is the potential (V), m is the mass loading of electrode materials (g), Δt stands for the discharge time (s), and I stands for the discharge current (A).

In the two-electrode system, the asymmetric supercapacitor (ASC) was assembled using 5% rGO-Ni(OH)₂ and graphene as anode and cathode, respectively. The energy density (E , Wh kg^{-1}) and power density (P , W kg^{-1}) were computed as follows:

$$E = (C \times \Delta V^2) / 2, \quad (2)$$

$$P = E \times 3600 / \Delta t. \quad (3)$$

RESULTS AND DISCUSSION

Syntheses and characterizations of rGO-Ni(OH)₂ composites

The typical synthesis approach of the Ni(OH)₂ nanosheets coupling with rGO is illustrated in Fig. 1a. GO was firstly mixed and stirred with NiCl₂ to form a uniform solution, followed by the addition of K₂Ni(CN)₄. Then, the above solution gradually formed GO-NiCl₂-K₂Ni(CN)₄ cyanogel by the ligand-substitution reaction between NiCl₂ and K₂Ni(CN)₄, due to the high water affinity and particular 2D structural units of K₂Ni(CN)₄. After several hours, a certain amount of NaBH₄ aqueous solution was added into cyanogel under stirring. At last, black rGO-Ni(OH)₂ powder was harvested by centrifugation and vacuum drying. By changing the mole ratio, different weight percentages of rGO can be incorporated into the final products, which was designated as X wt% rGO-Ni(OH)₂ (X=3, 5 and 7). For comparison, bare Ni(OH)₂ nanosheets (Ni(OH)₂-NS) can be obtained

Figure 1 (a) Schematic illustration of the typical preparation of ultrathin rGO-Ni(OH)₂. (b) SEM image, (c) STEM-EDX mapping image, (d) TEM image, (e) HR-TEM image, and (f) AFM image of typical 5% rGO-Ni(OH)₂.

by the same procedure but without the addition of GO, while Ni(OH)_2 particles ($\text{Ni(OH)}_2\text{-NP}$) were gained without the addition of both GO and $\text{K}_2\text{Ni(CN)}_4$.

The SEM image reveals the panoramic morphology of the 5% rGO- Ni(OH)_2 , displaying wrinkled 2D nanosheets (Fig. 1b). Bare $\text{Ni(OH)}_2\text{-NS}$ also shows obvious lamellar structure arising from the planar units of $\text{K}_2\text{Ni(CN)}_4$, but a small number of nanoparticles are present because of the absence of rGO (Fig. S1a). 3% rGO- Ni(OH)_2 also presents a tiny number of nanoparticles, while 7% rGO- Ni(OH)_2 shows remarkable thicker layers (Fig. S2). It implies that the addition of rGO is beneficial for the uniform nucleation and growth of $\text{Ni(OH)}_2\text{-NS}$, because rGO sheets may supply more active sites to adsorb metal ions [44]. In contrast, without $\text{K}_2\text{Ni(CN)}_4$ and rGO, $\text{Ni(OH)}_2\text{-NP}$ exhibits aggregated nanoparticles (Fig. S1b). Fig. 1c presents the scanning TEM (STEM) image and the corresponding EDX mapping images, suggesting the homogeneous dispersion of C, O, and Ni and the uniform coupling of rGO with $\text{Ni(OH)}_2\text{-NS}$. TEM image also reveals the voile-like characteristic of 5% rGO- Ni(OH)_2 (Fig. 1d), proving the ultrathin nature of $\text{Ni(OH)}_2\text{-NS}$.

Some crumples are also observed due to the nanosheet stacking. The HR-TEM image confirms the overlapping of nanosheets and reveals clear lattice fringes (Fig. 1e). The enlarged area in the inset of Fig. 1e indicates that two cross crystal planes with the crystal face angle of 60.8° have a lattice distance of ~ 0.23 and ~ 0.26 nm, respectively, being assigned to (015) and (101) facets of $\alpha\text{-Ni(OH)}_2$ (JCPDS 38-0715). The AFM image shows the height profile of 5% rGO- Ni(OH)_2 (Fig. 1f) and reveals that the thickness is only 2.5 nm, about 3–4 layers of edge-sharing octahedral MO_6 (top-right scheme in Fig. 1a), further demonstrating the ultrathin nanosheets.

The crystal structure of all the as-prepared materials was tested by XRD. As shown in Fig. 2a, the typical diffraction peaks at 12.3° , 33.3° , 38.7° and 61.3° of 2θ are indexed to (003), (101), (015) and (113) diffraction peaks of $\alpha\text{-Ni(OH)}_2$ (JCPDS 38-0715) [45]. The characteristic peak located at 23.6° is assigned to rGO, demonstrating the successful reduction of GO. XPS was also employed to examine the chemical bonding states and composition in the samples. As shown in Fig. 2b, the Ni 2p, C 1s, and O 1s peaks are clearly observed. High-resolution Ni 2p

Figure 2 (a) XRD patterns of 5% rGO- Ni(OH)_2 , $\text{Ni(OH)}_2\text{-NS}$, and rGO. (b) Survey XPS spectra, (c) high-resolution Ni 2p spectra, and (d) N₂ isotherm profiles and (inset) pore size distribution of 5% rGO- Ni(OH)_2 and $\text{Ni(OH)}_2\text{-NS}$, respectively.

spectrum of 5% rGO-Ni(OH)₂ was fitted into a pair of Ni 2p_{1/2} (872.4 eV) and Ni 2p_{3/2} (855.7 eV) peaks (Fig. 2c). The distance between these two peaks ($\Delta E = 17.7$ eV) indicates that the dominant valence state is Ni²⁺ [46]. Compared with peaks in Ni(OH)₂-NS, there is a positive shift of 0.5 eV for 5% rGO-Ni(OH)₂, suggesting the electron transport from Ni(OH)₂ to rGO [47]. In Fig. 2d, the nitrogen sorption isotherm of 5% rGO-Ni(OH)₂ shows a hysteresis loop, which indicates the mesoporous structure and is beneficial for the electrolyte ion diffusion. As seen in the inset of Fig. 2d, the pore diameter distribution of 5% rGO-Ni(OH)₂ is concentrated at 2.5 nm. The specific surface area calculated from the desorption isotherm is 92.6 m² g⁻¹, larger than other rGO-Ni(OH)₂ composites as shown in Fig. S3b. But the surface area of the rGO-Ni(OH)₂ composites is smaller than that of Ni(OH)₂-NS (183 m² g⁻¹), which could be attributed to the large contribution of micropores in Ni(OH)₂-NS, as illustrated in the obvious adsorption at $P/P_0 = 0.01$.

Electrochemical performance of the rGO-Ni(OH)₂ composites in a three-electrode system

The electrochemical property of the prepared electrode

materials was analyzed by GCD measurement at 1 A g⁻¹ (Fig. 3a). The optimal capacitance of 5% rGO-Ni(OH)₂ is 1562.4 F g⁻¹, which outperforms Ni(OH)₂-NS (1008.6 F g⁻¹), Ni(OH)₂-NP (969.1 F g⁻¹) and many other metal oxides (Table S1). This can be ascribed to the improved conductivity from the rGO and maximally exposed Ni(OH)₂ surface. The large capacitance of 3% rGO-Ni(OH)₂ (1370.9 F g⁻¹) and 7% rGO-Ni(OH)₂ (1347.28 F g⁻¹) can also confirm the synergy between the ultrathin Ni(OH)₂-NS and rGO, which is highly conducive to the charge transport and ion diffusion. Moreover, the high mechanical strength of the rGO also buffers the structural damage in the fast charge and discharge cycles, which is favorable to the stability [33,48,49]. Fig. 3b shows the GCD diagram of 5% rGO-Ni(OH)₂ under different current densities. The capacitance values are 1562.4, 1481.76, 1357.92, 1278.72, 1239.12, 1187.28, 1149.6 and 1119.52 F g⁻¹ at 1, 2, 5, 8, 10, 15, 20 and 30 A g⁻¹ in order, demonstrating its high rate performance, especially at 20 and 30 A g⁻¹. Fig. 3c exhibits the rate retention of the as-synthesized electrode materials. At different rates, 3% rGO-Ni(OH)₂ and 7% rGO-Ni(OH)₂ show a little inferior specific capacitance to 5%

Figure 3 (a) GCD curves of different electrode materials. (b) GCD curves of 5% rGO-Ni(OH)₂ at different current densities. (c) Specific capacitance of all the as-prepared samples at different current densities. (d) Cycling stability at the current density of 30 A g⁻¹. The inset shows the GCD curves around the 1000th cycle and 2000th cycle.

rGO-Ni(OH)₂, but better than bare Ni(OH)₂-NS. Fig. 3d illustrates the long-term durability of 5% rGO-Ni(OH)₂ and bare Ni(OH)₂-NS at 30 A g⁻¹ for 2000 cycles. While Ni(OH)₂-NS only maintains 51% of its initial capacitance, the capacitance value of 5% rGO-Ni(OH)₂ retains 82.3% after 2000 cycles, indicating optimal cycling stability. As illustrated in the inset of Fig. 3d, the charge-discharge curves at the 1000th cycle and 2000th cycle display no obvious changes.

To elucidate the electrochemical kinetics, CV was conducted and analyzed. The prominent redox peaks at

each scan rate, as shown in Fig. 4a, indicate the typical capacitive behavior, which originates from the invertible reaction [50]:

The redox peaks increase with the increase of scan rate, implying that there is an obvious reversibility in charge and discharge cycles. The sweep rates (v) and measured current (i) comply with the following power-law relationship [51,52]:

$$i = av^b, \quad (5)$$

Figure 4 (a) CV curves at different scan rates from 2 to 15 mV s⁻¹. (b) Corresponding log*i* versus log*v* plots at specific peak currents. (c) CV curve with the pseudocapacitive fraction shown by the shaded area at a scan rate of 8 mV s⁻¹. (d) The percentage of pseudocapacitive contributions at different scan rates for X% rGO-Ni(OH)₂. (e) Nyquist plots and (f) derived diffusion resistances of X% rGO-Ni(OH)₂ and Ni(OH)₂-NS.

$$\log i = \log a + b \log v, \quad (6)$$

where a and b are variable values. When b is equal to 0.5, it manifests the diffusion-controlled process corresponding to a sluggish reaction kinetics, while 1.0 stands for that the surface capacitive process is dominated with ultrafast reaction kinetics. The b values are 0.74 and 0.70, which were calculated by the slope of peak 1 and peak 2 as shown in Fig. 4b, respectively, demonstrating that the reaction kinetics for 5% rGO-Ni(OH)₂ is mainly a surface capacitive process and performs very fast.

The pseudocapacitive contribution was quantified as follow [53–55]:

$$i(V) = k_1 v + k_2 v^{1/2}, \quad (7)$$

where k_1 and k_2 are constants for a certain voltage. The $k_1 v$ represents the contribution of capacitive-controlled, whereas the $k_2 v^{1/2}$ is for the diffusion-controlled contribution. The capacitive-controlled subscription is 83.3% at 8 mV s⁻¹ for 5% rGO-Ni(OH)₂ as shown in the shaded area (Fig. 4c). Small crystalline and the enhanced electrical conductivity on the ultrathin layer are synergistically beneficial to the electron transport and reaction kinetics of Faradaic reaction. When the scanning rate changes from 2 to 15 mV s⁻¹, the pseudocapacitive contribution ratio improves from 72.2% to 84.6% (Fig. 4d), revealing that the multitude of charge storage in 5% rGO-Ni(OH)₂ is the pseudocapacitive process. A little larger contribution comes from pseudocapacitance for 3% rGO-Ni(OH)₂ (Fig. S4), because of the high content of Ni(OH)₂-NS, and vice versa for 7% rGO-Ni(OH)₂ (Fig. S5), confirming the charge storage mechanism too.

EIS test was further performed to investigate the kinetic processes. All samples present tiny incomplete semicircles with linear tails, which demonstrates a typical pseudocapacitive process (Fig. 4e). For all the samples, a small equivalent series resistance (ESR, intersection with the real axis) of ~1.6 Ω demonstrates low interface resistance on the surface between the electrode and electrolyte, and a fast ionic conductivity of the electrolyte [49]. Moreover, in the low-frequency region, 5% rGO-Ni(OH)₂ has a large slope, showing the low Warburg impedance (Z_w) of the diffusion of ions near the surface of the electrode. The diffusion resistance (σ), an important indicator for reaction kinetics, was quantified according to Equation (8) [29],

$$Z_w = \sigma \omega^{-0.5} - j \sigma \omega^{-0.5}, \quad (8)$$

where $\sigma \omega^{-0.5}$ belongs to the real part (Z'), and $-j \sigma \omega^{-0.5}$ represents the imaginary part (Z'') of impedance. As we can see from Fig. 4f, the σ of 5% rGO-Ni(OH)₂ is

1.23 Ω s^{-1/2}, much lower than the values of Ni(OH)₂-NS (3.40 Ω s^{-1/2}), 3% rGO-Ni(OH)₂ (3.61 Ω s^{-1/2}), and 7% rGO-Ni(OH)₂ (4.51 Ω s^{-1/2}), indicating the fastest reaction kinetics and optimal mutual effect between rGO and Ni(OH)₂-NS [56]. These results definitely prove that the ultrathin Ni(OH)₂ layers coupling with rGO supply more surface active sites to promote the electrode performance by speeding up charge transport and ion diffusion.

The improved electrochemical performance of rGO-Ni(OH)₂ can be attributed to the ultrathin 2D feature and the synergy effect between Ni(OH)₂-NS and rGO. Firstly, the 2D nanosheets enlarge the surface area and provide sufficient contact area between electrode and electrolyte for fast ion migration and efficient Faradaic reaction. Secondly, the addition of rGO is beneficial for the uniform nucleation and growth of Ni(OH)₂-NS, because rGO sheets supply more active sites to adsorb cations. Moreover, the rGO sheets, with the merits of high electrical conductivity and flexible nature, serve as a fast transportation highway for the released electrons and avoid the structure change during cycling.

Electrochemical performance of the rGO-Ni(OH)₂ composites in ASCs

To authenticate the practical application of the synthesized materials, an ASC (inset of Fig. 5a), named as rGO-Ni(OH)₂//rGO, was assembled with 6 mol L⁻¹ KOH as the electrolyte. The potential window (1.5 V) of the ASC was decided by the CV patterns of the cathode (-1.0–0 V) and anode (0–0.5 V) at 5 mV s⁻¹ (Fig. 5a). Besides, different CV curves at diverse voltage windows for the ASCs were also investigated to determine the potential window (Fig. S6). It is found that the oxygen evolution takes place when the potential is continuously increased to 1.6 V. Thus, 1.5 V is chosen as the voltage for the ASC. From Fig. 5b, the CV curves for ASCs remain well and no obvious peak shift occurs as the scanning rate gradually changes to 50 mV s⁻¹, suggesting the superior reversibility of the reaction.

In Fig. 5c, GCD curves tested at different current densities indicate the capacitance values are about 152.8, 129, 85.2, 65.28, 56.64, 48 and 45.08 F g⁻¹ respectively, at 0.2, 0.5, 1, 2, 5, 8 and 10 A g⁻¹ in a wide voltage window of 0–1.5 V. Furthermore, the performance outperforms some previous results (Table S2). The cycling durability is presented in Fig. 5d. The capacitance increases gradually during the first 1000 cycles, and retains a stable level in the subsequent cycles. The capacitance almost remains 100% after cycling 2000 cycles (inset of Fig. 5d). This suggests that the as-assembled ASC with good cycling

Figure 5 (a) CV curves of individual positive and negative electrodes measured in a three-electrode system at 5 mV s^{-1} . (b) CV curves of the ASC at diverse sweep rates. (c) GCD curves of the ASC at diverse current densities. (d) Cycling performance of the ASC. The inset shows the GCD curves of the 1000th and 2000th cycle.

performance and high specific capacitance can serve as efficient and inexpensive energy storage equipment. The power density and energy density are depicted in the Ragone plot (Fig. S7). The specific energy of the assembled ASC achieves 44.3 W h kg^{-1} (148.5 W kg^{-1}), outperforming the previous results as shown in Table S2. Even when the power density is 7.25 W kg^{-1} , the energy density still maintains 14.1 W h kg^{-1} . Due to the synergistic effect of the modified conductivity and rapid ion migration in the 5% rGO-Ni(OH)₂ composite, the assembled ASC has remarkable electrochemical properties.

CONCLUSIONS

In summary, we successfully coupled ultrathin Ni(OH)₂ with conductive rGO using a NaBH₄ assisted cyanogel hydrolysis method. Benefiting from these advantageous features, The prepared sample possesses a high specific capacitance of 1119.52 F g^{-1} especially at 30 A g^{-1} , and maintains 82.3% after 2000 cycles, demonstrating much better retention and cycling durability than bare Ni(OH)₂-NS. The synergy effect between ultrathin Ni(OH)₂-NS and rGO is the main reason for its perfor-

mance improvement. By analyzing the reaction kinetics, it is found that the pseudocapacitive contribution accounts for 84.6% of the overall capacitance and the equivalent series resistance and σ are only $\sim 1.6 \Omega$ and $1.23 \Omega \text{ S}^{-1/2}$, respectively, demonstrating the improved electronic conductivity and the maximal exposure of active surface. The capacitance and energy density of the assembled ASC are 129 F g^{-1} at 0.5 A g^{-1} and 44.3 W h kg^{-1} (148.5 W kg^{-1}), indicating that it has a high application potential in the new-generation energy storage system.

Received 11 April 2020; accepted 11 June 2020;
published online 14 September 2020

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Acknowledgements This research was supported by the National Natural Science Foundation of China (21875133), Xijiang R&D Team (Wang X), the Science and Technology Program of Guangzhou (2019050001), and Science and Technology Commission of Shanghai Municipality (19ZR1479500).

Author contributions Zhang X, Wang H and Wang J contributed to the experimental planning, experimental measurements, data analyses and manuscript preparation; Shui L and Zhou G contributed to the data analyses and manuscript preparation; Wang X and Ma R contributed to

the experimental planning, data analyses and manuscript preparation. All authors contributed to the general discussion.

Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Supporting data are available in the online version of the paper.

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超薄 $\text{Ni}(\text{OH})_2$ 与石墨烯耦合用于超级电容器的快速电子/离子传输

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摘要 巧妙地将具有快速电化学过程的双电层电容与具有较大容量的赝电容相结合是提高超级电容器整体性能的实用途径, 但是, 实现这一目标仍具有挑战性。本文提出了一种氰胶-还原法, 并在常温下制备了与导电rGO偶联的超薄 $\text{Ni}(\text{OH})_2$ 纳米薄片二维复合材料($\text{rGO-Ni}(\text{OH})_2$)。超薄 $\text{Ni}(\text{OH})_2$ 纳米片具有3–4层共边八面体 MO_6 , 最大限度地暴露了法拉第反应的活性表面, 促进了离子扩散, 同时导电的rGO在反应过程中促进了电子的传递。在 30 A g^{-1} 下, 合成的 $\text{rGO-Ni}(\text{OH})_2$ 样品的电容值仍可达 1119.52 F g^{-1} , 经2000次循环后仍能保持82.3%的比容量, 表现出比 $\text{Ni}(\text{OH})_2$ 纳米片更好的电化学性能。组装的 $\text{rGO-Ni}(\text{OH})_2//\text{rGO}$ 双电极系统, 最大的能量密度为 44.3 W h kg^{-1} (148.5 W kg^{-1})。本工作为制备高性能二维混合电极提供了新的思路。