Pulse-Reversal Deposition of Nickel Sulfide Thin Film as an Efficient Cathode Material for Hybrid Supercapacitors

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In this study, nickel sulfide (Ni3S2) thin film was electrodeposited on Ni foam substrate by using a facile potentiodynamic (PD) and pulse-reversal (PR) method, respectively. After optimization of deposition parameters, the optimized Ni3S2 electrode was prepared by using PR mode under −1 V pulse-on potential, 0 V pulse-reversal potential, duty cycle of 0.5 and pulse frequency of 0.1 Hz. It delivered an impressive specific capacity up to 179.5 mAh g−1 at current density of 2 A g−1. The hybrid supercapacitor (SC) was assembled by using the optimized Ni3S2 electrode as cathode and activated carbon fiber cloth (ACFC) as anode, respectively. The Ni3S2/ACFC hybrid SC exhibited a high specific capacity of 26.7 mAh g−1 at charge-discharge current density of 2 A g−1 and displayed a maximum energy density of 26.4 Wh kg−1 at 1978 W kg−1 based on the total mass of active material of 9.2 mg. Furthermore, the Ni3S2/ACFC hybrid SC showed an impressive excellent cycling performance in 1 M KOH aqueous electrolyte at current density of 2 A g−1, with 97% specific capacitance retained after cycling of 400–5000 consecutive charge/discharge tests.

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Experimental

Preparation of Ni3S2 electrode.—The Ni3S2 films were deposited on to the nickel foam (10 mm × 10 mm × 1.7 mm, 94 PPI) substrate by PD and PR deposition modes in the deposition bath containing 50 mM NiCl2 · 6H2O (Arcos, 97%) and 1 M thiourea (TU) (Arcos, 

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Table I. Parameters for the electrodeposition of various Ni3S2 electrodes.

|       | Pulse on (V) | Pulse reverse (V) | Duty cycle | Pulse frequency (Hz) | Deposition cycle | Mass Loading (mg) |
|-------|--------------|------------------|------------|----------------------|-----------------|------------------|
| NSPD  | –            | –                | –          | –                    | 6               | ~1.4             |
| NSPR-1| −0.9         | 0                | 0.5        | 1/10                 | 900             | ~0.5             |
| NSPR-2| −1           | 0                | 0.5        | 1/10                 | 420             | ~1.4             |
| NSPR-3| −1           | −0.1             | 0.5        | 1/10                 | 360             | ~1.4             |
| NSPR-4| −1           | 0                | 0.3        | 1/10                 | 900             | –                |
| NSPR-5| −1           | 0                | 0.7        | 1/10                 | 360             | ~1.4             |

99%). All electrodeposition were performed in a conventional three-compartment cell consisting of a Ni foam substrate as working electrode, a saturated silver/silver chloride (Ag/AgCl) as reference electrode, and a Pt wire as counter electrode using a computer-controlled CHI 627D (CH Instrument) electrochemical analyzer. Prior to the preparation of the Ni3S2 electrodes, Ni foam substrates were ultrasonically washed with 6 M HCl, deionized water and ethanol for 15 min, respectively. After that, the pre-cleaned Ni foams were dried under vacuum at 60 °C for 12 h. The PD deposition of Ni3S2 thin film was conducted within the potential range between −1.2 V and 0.2 V vs. Ag/AgCl at a scan rate of 5 mV s−1 for 6 sweep cycles. The Ni3S2 electrodeposition by PR deposition mode was performed by adjusting a variety of deposition parameters including pulse-on potential, pulse-reversal potential and duty cycle. It should be noted that the pulse frequency of PR mode was set at 0.1 for all PR deposition in this work. The duty cycle and pulse frequency in this study are defined as following:

\[
duty\ cycle = \frac{t_{\text{pulse-on}}}{t_{\text{pulse-on}} + t_{\text{pulse-reversal}}} \quad [1]
\]

\[
pulse\ frequency = \frac{1}{t_{\text{pulse-on}} + t_{\text{pulse-reversal}}} \quad [2]
\]

where \(t_{\text{pulse-on}}\) and \(t_{\text{pulse-reversal}}\) represents the period at pulse-on potential and pulse-reversal potential, respectively. The corresponding parameters of PR deposition mode for preparing Ni3S2 electrodes were listed in Table I. It should be noticed that the Ni3S2 electrodes prepared by using PD and PR modes were designated as NSPD and NSPR-1~NSPR-5. After the electrodeposition, the as-deposited Ni3S2 electrodes were rinsed with deionized water and subsequently dried in a vacuum at ca. 9.2 mg. The sandwich-type hybrid SCs was further encapsulated using commercial activated carbon fiber cloth (ACFC) with a specific surface area of 2000 m2 g−1 was purchased from Taiwan carbon technology company and directly used as anode material for hybrid SCs.

Assembly of hybrid SCs.—The sandwich-type hybrid SCs were fabricated by employing the Ni3S2 prepared by PR deposition mode as cathode, ACFC as anode and filter paper as separator, respectively. The total mass loading of active materials in the hybrid SC was controlled at ca. 9.2 mg. The sandwich-type hybrid SC was further encapsulated by the film of aluminum plastic. Afterward, the 1 M KOH electrolyte was injected into the hybrid SCs.

Characterization of Ni3S2 electrodes.—The crystal structure and surface morphology of the deposits were characterized by using a grazing incident X-ray diffraction (GIXRD, Rigaku D/TTTRAX III, Japan) and field-emission scanning electron microscope (FESEM, JSM-7000F).

The capacity performance of various Ni3S2 electrodes were investigated by cyclic voltammetry (CV) and galvanostatic charge/discharge tests in 1 M KOH aqueous electrolyte using the aforementioned electrochemical analyzer. The CVs were recorded within a potential interval ranging from −0.2 V to 0.6 V vs. Ag/AgCl at different scan rates from 5 mV s−1 to 50 mV s−1. The galvanostatic charge/discharge tests were performed within a potential range of 0–0.5 V vs. Ag/AgCl at charge/discharge current densities ranging from 2 to 32 A g−1. The electrochemical impedance spectroscopy (EIS) measurements were performed at a constant dc bias potential of 0.3 V vs. Ag/AgCl in the frequency range from 100 kHz to 10 mHz with an amplitude of 5 mV by using a potentiostate (IM 6, Zahner) equipped with a frequency analyzer (Thales). The resultant EIS spectra were then simulated by means of Zview software.

Results and Discussion

Characterization of Ni3S2 electrodes.—Fig. 1 shows the XRD patterns of the Ni3S2 electrodes prepared by using PD and PR deposition modes. It can be apparently found that all diffraction peaks of the deposits can be assigned to the pure phase of Ni3S2 [JCPDS NO. 44-1418] except to the three strong signals (2θ = 44.4°, 51.7° and 76.4°) originating from the Ni foam substrate. It should be noted that no XRD pattern is presented for NSPR-4 since almost no mass change is observed after the deposition. This can be explained by the reason that the dissolution rate at the reversal period is almost equal to the deposition rate at the pulse-on period with such low duty cycle. Figs. 2a–2e displays the FESEM images of various Ni3S2 deposits. Obviously, all Ni foam substrates are fully and uniformly covered by the Ni3S2 thin films. The corresponding magnified FESEM images are depicted in the inset of Figs. 2a–2e to further display the morphology details of the resultant Ni3S2 thin films. As expected, the surface morphology of the NSPD electrode (Fig. 2a) is observed to be constructed with nanoflakes, which is well consistence with our previous study.12 On the basis of the preliminary assessment, only small amount deposits on Ni foam substrates were observed while using the PR deposition mode, in which the pulse-on potential was set at more positive potential than −1 V vs. Ag/AgCl while accompanying with a constant pulse-reverse potential of 0 V vs. Ag/AgCl. This result indicates their low mass loading of Ni3S2 deposits on the Ni foam substrates. For instance, only ca. 0.5 mg of Ni3S2 was deposited on the substrate while setting the pulse-on potential at −0.9 V vs. Ag/AgCl for 900 PR cycles. Although the resultant NSPR-1 electrode demonstrates the similar surface morphology to that of the NSPD electrode (see Fig. 2b), it was found that the mass loading of Ni3S2 thin film just increased a little bit while even doubling the PR cycles, which cannot be increased to that of the NSPD electrode.
Figure 2. Low-magnification FESEM images of (a) NSPD, (b) NSPR-1, (c) NSPR-2, (d) NSPR-3 and (e) NSPR-5 Ni$_3$S$_2$ films deposit. The corresponding high-magnification FESEM images are depicted in the insets of Figs. 7a–7e.

(c. 1.4 mg). As for employing pulse-on potential of $-1$ V vs. Ag/AgCl for the PR deposition mode, the mass loading of the NSPR-2 electrode can easily reach the level of the NSPD electrode by turning the PR cycles. As can be seen in Fig. 2c, more uniform Ni$_3$S$_2$ thin film with less formation of cracks is deposited on the Ni foam substrate and its microstructure is constructed with more ordered interlaced nanoflakes while compared with the NSPD and NSPR-1 electrodes. This nanostructure with ordered open pores would be beneficial to the electrolyte penetration within the electrodes. Consequently, the pulse-on potential is set as $-1$ V vs. Ag/AgCl for the optimized PR deposition mode. As adjusting the pulse-reverse potential from 0 V to $-0.1$ V vs. Ag/AgCl, it is observed that interlaced flake-like morphology disappears for the NSPR-3 electrode (Fig. 2d). While using the low duty cycle of 0.3 for the PR deposition (NSPR-4 electrode), the dissolution rate was found to be almost equal to the deposition rate and no mass loading before and after the deposition was presented. Nevertheless, the increase in duty cycle from 0.5 to 0.7, the resultant NSPR-5 electrode is composed of the obscure Ni$_3$S$_2$ flakes due to the increased deposition rate.

Electrochemical properties of the Ni$_3$S$_2$ electrode.— Fig. 3 depicts the CV curves of various Ni$_3$S$_2$ electrodes in 1 M KOH electrolyte.

Figure 3. CV curves of (a) NSPD, (b) NSPR-1, (c) NSPR-2, (d) NSPR-3 and (e) NSPR-5 electrodes measured at different scan rates in 1.0 M KOH aqueous electrolyte.
at various scan rates from 5 mV s\(^{-1}\) to 50 mV s\(^{-1}\). All curves exhibit similar shape, and the shape of the CV curves represents distinct battery-type characteristics, which is totally different from the closely ideal rectangular CV shape for EDLCs. In particular, only one pair of redox peaks is observed in the all CV curves within the potential range from −0.2 to 0.8 V vs. Ag/AgCl, indicating that the electrochemical capacity of the Ni\(_3\)S\(_2\) electrode mainly originates from the purely faradaic reaction. The electrochemical reaction corresponding to the redox peaks of Ni\(_3\)S\(_2\) electrodes in alkaline electrolyte can be expressed as follows:\(^{10,11}\)

\[
\text{Ni}_3\text{S}_2 + 3\text{OH}^- \leftrightarrow \text{Ni}_3\text{S}_2(\text{OH})_3 + 3\text{e}^- \quad [3]
\]

Typically, a higher current density and larger area surrounded by a CV curve indicate higher electrochemical capacity. It can be apparently found that NSPR-1 possesses the highest current density and the largest area under the current–potential curve among all electrodes. This signifies that NSPR-1 may have the highest specific capacity among all Ni\(_3\)S\(_2\) electrodes.\(^{26}\) Since the mass loading for the NSPR-1 electrode is much lower compared to other electrodes, it would be unfeasible for practical applications.\(^{27}\) Except the NSPR-1 electrode, the NSPR-2 electrode exhibits a much larger area surrounded by the current–potential curve than those of NSPD, NSPR-3 and NSPR-5 electrodes, suggesting its higher specific capacity and superior capacitive behavior.

The specific capacities of the different Ni\(_3\)S\(_2\) electrodes at different current densities can be calculated according to the following equation, and the results are summarized in Fig. 5.

\[
Q_m = \frac{I \times \Delta t}{m} \quad [4]
\]

Where is the specific capacity (mAh g\(^{-1}\)), \(I\) is the discharge current (mA), \(\Delta t\) is the discharge time (h), and \(m\) is the mass of the electroactive materials (g). The NSPD electrode shows specific capacity of 102.9 mAh g\(^{-1}\) at 2 A g\(^{-1}\), 92.2 mAh g\(^{-1}\) at 4 A g\(^{-1}\), 83.8 mAh g\(^{-1}\) at 8 A g\(^{-1}\), 74.4 mAh g\(^{-1}\) at 16 A g\(^{-1}\), and 60.4 mAh g\(^{-1}\) at 32 A g\(^{-1}\), respectively. The \(Q_m\) values of all Ni\(_3\)S\(_2\) electrodes are found to decrease with increasing charge/discharge current density. Among all Ni\(_3\)S\(_2\) electrodes, the NSPR-2 electrode exhibits the highest specific capacity of 179.5 mAh g\(^{-1}\) at 2 A g\(^{-1}\), while the capacity remains 105.9 mAh g\(^{-1}\) at 32 A g\(^{-1}\), presenting its excellent capacity performance. Therefore, the NSPR-2 electrode could be regarded as the optimized one of the Ni\(_3\)S\(_2\) electrodes prepared by the PR deposition mode.
Figure 5. The relationship between the specific capacity and charge/discharge current density for the NSPD, NSPR-2, NSPR-3 and NSPR-5 electrodes.

Figure 6. Comparison of cycling performance for the NSPD and NSPR-2 electrodes at charge/discharge current density of 8 A g\(^{-1}\).

Figure 7. Nyquist plots of the NSPD and NSPR-2 electrodes at potential of 0.3 V (a) before and (b) after the 2500 cycles at a high current density of 4 A g\(^{-1}\). The inset in Fig. 7a is the equivalent circuit model used to simulate the resultant Nyquist plots.

To further compare the electrochemical stability of the optimized PR-deposited Ni\(_3\)S\(_2\) electrode (NSPR-2) with the NSPD electrode, the galvanostatic charge/discharge cycling tests were performed in 1 M KOH aqueous electrolyte at a constant current density of 8 A g\(^{-1}\) for 2500 consecutive cycles, as depicted in Fig. 6. Both electrodes display two-stage capacity decay, representing region I (1∼500 cycles) and region II (500∼2500 cycles), respectively. At the region I, the specific capacity of NSPD and NSPR-2 electrodes rapidly decreases, thus resulting in the decrease of nearly 32% and 45% of the corresponding initial discharge capacity. As for the region II, the relatively less decrease in capacity is observed with further cycling for another 2000 cycles, in which only approximately 17% and 28% of capacity loss for NSPD and NSPR-2 electrodes compared to that at the 500th cycle, respectively. After the consecutive 2500 cycles, the stable specific capacity of NSPD and NSPR-2 electrodes is found to be 51.2 and 54.5 mAh g\(^{-1}\), respectively.

To gain insight into the electrochemical kinetics and aged capacity performance of the NSPD and NSPR-2 electrodes before/after the cycling tests, EIS analyses were carried out in the frequency range of 10 kHz to 0.01 Hz. Fig. 7 shows the Nyquist plots the NSPD and NSPR-2 electrodes before and after the long-term cycling tests. The Nyquist plots are mainly composed of a semicircle at the high-frequency region and a straight line at low-frequency region. The inset in Fig. 7a is the equivalent-circuit model employed to simulate the resultant EIS spectra. The real axis intercept in high-frequency region represents the bulk solution resistance (\(R_s\)). The diameter of the semicircle in the high-frequency region is associated with the Faradaic interfacial charge-transfer resistance (\(R_{ct}\)). The constant phase element (CPE) is used to account for a double-layer capacitance, \(Z_W\) and \(C_F\) represent the Warburg impedance and the Faradaic pseudocapacitor, respectively. Before the cycling tests, the \(R_s\) values of NSPD (\(\sim1.43\) \(\Omega\)) and NSPR-2 (\(\sim1.32\) \(\Omega\)) electrodes are found to be similar. It is worthy noted that the NSPR-2 electrode possesses much lower \(R_{ct}\) value (4.18 \(\Omega\)) than that of the NSPD electrode (10.41 \(\Omega\)), indicating that the NSPR-2 electrode shows a higher rate of charge transport than the NSPD electrode. Therefore, the high initial discharge capacity of the NSPR-2 electrode can be ascribed to its excellent charge-transfer characteristics. After the cycling test, the NSPD and NSPR-2 electrodes still have similar \(R_s\) values, (\(\sim1.45\) \(\Omega\)) and (\(\sim1.34\) \(\Omega\)), respectively. However, the \(R_{ct}\) value of the NSPD and NSPR-2
electrodes are increased from 10.41 Ω to 12.38 Ω and 4.18 Ω to 11.44 Ω, respectively. It signifies that the NSPD and NSPR-2 electrodes have comparable $R_s$ and $R_{ct}$ values after the long-term cycling tests, therefore resulting in the similar discharge capacity after 2500 charge/discharge cycles.

To elaborate the reasons for the decayed capacity and increased $R_{ct}$ value after the long-term cycling tests, the composition and surface morphology of the NSPD and NSPR-2 electrodes were examined. Fig. 8a shows the GIXRD patterns of the cycled NSPD and NSPR-2 electrodes. It represents that the XRD patterns of both cycled electrodes still retain as their initial XRD patterns (Fig. 1), indicating the composition of both electrodes is without any change after 2500 charge/discharge cycling tests. Figs. 8b and 8c present the FESEM image of NSPD and NSPR-2 electrodes after 2500 cyclic charge-discharge tests. It can be observed that the pore size of interlaced nanoflakes of both NSPD and NSPR-2 electrodes becomes relatively larger after the long-term cycling test, which could be possibly ascribed to the volume expansion for the Ni-based electrodes after consecutive charge/discharge cycling tests. Consequently, both cycled NSPD and NSPR-2 electrodes have similar surface morphology. The evolution in surface morphology for both electrodes could be responsible to the significantly decreased capacity and the increased $R_s$ and $R_{ct}$ values after the cycling test. This also suggests that the capacity decay and the increased $R_s$ and $R_{ct}$ values for the electrodeposited Ni$_3$S$_2$ electrodes would significantly occur after long-term cycling tests regardless of deposition modes due to the surface evolution during charge/discharge cycling. The capacity of the electrodeposited Ni$_3$S$_2$ electrodes would reach stable while their surface morphology retains unchanged.

In order to evaluate its practical application, a hybrid SC device was assembled by using the optimized Ni$_3$S$_2$ prepared by the PR deposition mode (NSPR-2) as positive electrode and ACFC as anode, respectively. The electrochemical behavior of ACFC was firstly measured using CV method at different scan rates from 5 to 50 mV s$^{-1}$. It can be observed that the nearly rectangular-like shapes are observed in the operating potential range ($-1$ to $0$ V vs. Ag/AgCl), as shown in Fig. 9a. Furthermore, the galvanostatic charge/discharge curves of the ACFC electrode shows highly symmetric and linear curve at galvanostatic current densities ranging from 1 A g$^{-1}$ to 8 A g$^{-1}$, as shown in Fig. 9b. The ACFC electrode shows specific capacity of 34.7 mAh g$^{-1}$ at 1 Ag$^{-1}$, 32.6 mAh g$^{-1}$ at 2 Ag$^{-1}$, 30.4 mAh g$^{-1}$ at 4 Ag$^{-1}$ and 27.1 mAh g$^{-1}$ at 8 A g$^{-1}$, respectively. As for fabrication of the hybrid SC, the charge of the NSPR-2 cathode and the ACFC anode should balance according to the following equation:

$$m_+ + m_- = I_+ \times t_+ = I_- \times t_-$$

where $m$ (g) is the mass, $I$ (A) is the discharge current for positive (+) and negative (−), and $t$ is the discharge time (s) for positive (+) and negative (−) electrodes, respectively. The optimum mass loading ratio between NSPR-2 and ACFC should be $m_+/m_- = 0.18$ in the hybrid SCs device. Before evaluating hybrid SCs, the sum of the potential range of NSPR-2 cathode and ACFC anode were evaluated.
Figure 10. (a) CV curves of the ACFC electrode and NSPR-2 electrode measured in three-electrode cell in 1 M KOH aqueous electrolyte at scan rates of 50 mV s$^{-1}$. (b) CV curves of the NSPR-2//ACFC hybrid SC measured at various potential windows at a scan rate of 50 mV s$^{-1}$. (c) CV curves of the NSPR-2//ACFC hybrid SC measured with a potential window of 1.8 V at different scan rates.

by using CV measurements with the potential range from $-1$ V to 0.8 V vs. Ag/AgCl, as shown in Fig. 10a. Fig. 10b shows the CV curves of NSPR-2//ACFC hybrid SCs at various potential windows in 1 M KOH electrolyte at 50 mV s$^{-1}$. It clearly depicts that the NSPR-2//ACFC hybrid SC can deliver a stable working voltage of 1.8 V. Furthermore, Fig. 10c displays the CV curves of NSPR-2//ACFC hybrid SCs measured at different scan rate of 5, 10, 20 and 50 mV s$^{-1}$. Interestingly, the NSPR-2//ACFC hybrid SC exhibits both of the EDLC and Faradic electrode properties within the potential window of 0–1.8 V. Fig. 11a shows the galvanostatic charge/discharge

Figure 11. (a) Galvanostatic charge/discharge curves of the NSPR-2//ACFC hybrid SC at different charge/discharge current densities. (b) Ragone plot of the NSPR-2//ACFC hybrid SC compared with other hybrid SCs devices. (c) Cycling performance of the NSPR-2//ACFC hybrid SC at 4 A g$^{-1}$. (d) A photograph of a green LED lighted up by two NSPR-2//ACFC hybrid SCs in series.
curves of the NSPR-2/ACFC hybrid SCs measured at different current densities. As can be seen in Fig. 11a, the charge/discharge curves of NSPR-2//ACFC hybrid SCs show the behavior of the EDLC capacitors and battery-type electrodes within a stable potential window of 0–1.8 V. Moreover, the Qm values of NSPR-2/ACFC hybrid SCs at 2, 4, 8 and 16 A g⁻¹ are estimated to be 26.7, 23.8, 18.7 and 10.6 mAh g⁻¹, respectively. In Fig. 11b, the Ragone plot related to power densities (P) and energy densities (E) is further used to evaluate the performance of the NSPR-2/ACFC hybrid SCs. The energy density and power density of the hybrid SCs were calculated as follows:35

\[ E = \int_{t_1}^{t_2} (I \times V) dt \]  
\[ P = \frac{E}{\Delta t} \]

where \( Q_m \) is calculated based on the loading mass of active materials in anode and cathode, \( V \) is the voltage, \( t_1, t_2 \) is the start time and end time during discharging and \( \Delta t \) equal to \( (t_1-t_1) \) is the discharge duration, respectively. The corresponding energy density and power density at 2 A g⁻¹ are 26.4 Wh kg⁻¹ and 1978 W kg⁻¹, respectively. Even at a current density of 16 A g⁻¹, the energy density and power density still remain at 7.4 Wh kg⁻¹ and 11.1 kW kg⁻¹. It displays superior energy density and power density compared to other nickel oxide or sulfide-based hybrid SCs reported in literatures, such as NiO/carbon (11 Wh kg⁻¹ at 500 W kg⁻¹),35 Ni₃S₂/CNFs/CNFs (25.8 Wh kg⁻¹ at 425 W kg⁻¹)35 and Ni₃S₂/MWCNT-NC//AC (19.8 Wh kg⁻¹ at 798 W kg⁻¹).35 To further evaluate the electrochemical stability of the NSPR-2//ACFC hybrid SCs, the galvanostatic charge/discharge cycling tests were performed at a constant current density of 2 A g⁻¹ for 5000 consecutive cycles, as depicted in Fig. 11c. The 40% loss in \( Q_m \) value of NSPR-2//ACFC was observed after cycling of 1–400 charge/discharge cycles, which can be ascribed to the surface evolution of NSPR-2 electrode. Nevertheless, the relatively less decrease in capacity is found with further cycling for another 4600 cycles, in which only approximately 3% loss of specific capacity compared to that at the 400th cycle. To display the practical application of the NSPR-2//ACFC hybrid SCs, two hybrid SCs were assembled in series for lightening up a green light-emitting diode (LED) of driving voltage 3.0 V. Fig. 11d depicts that two NSPR-2//ACFC hybrid SCs in series can successfully light up the green LED.

Conclusions

In summary, the electrodeposited Ni₃S₂ thin films with excellent capacitive properties have been successfully prepared using a PR deposition mode. After the optimization of the deposition parameters, the optimized NSPR-2 electrode delivers remarkable specific capacity up to 179.5 mAh g⁻¹ at 2 A g⁻¹ and its discharge capacity even retains 105.9 mAh g⁻¹ at high charge/discharge current density of 32 A g⁻¹ in 1.0 M KOH aqueous electrolyte. Moreover, the specific capacity retention of the NSPR-2 electrode can still achieve ac.78% after cycling of 300–2500 cycles at a high current density of 8 A g⁻¹. Furthermore, the NSPR-2/ACFC hybrid SCs can be reversibly charged and discharged at a stable cell voltage of 1.8 V and generates an impressive specific capacity of 26.7 mAh g⁻¹ at charge/discharge current density of 2 A g⁻¹, delivering a maximum energy density of 26.4 Wh kg⁻¹ at a power density of 1978 Wkg⁻¹. More importantly, the NSPR-2/ACFC hybrid SC still retains 97% of 400th capacity after 5000 consecutive charge/discharge test at a charge/discharge current density of 2 A g⁻¹, indicating its great electrochemical stability. In views of the impressive capacitive performance for the NSPR-2/ACFC hybrid SC, it can be considered as a promising potential low-cost device for energy storage.

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