In situ deposited multilayer integrated hydrogels for deformable and stretchable supercapacitors

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ABSTRACT Hydrogel systems promote the development of flexible energy storage devices because of their inherent mechanical elasticity and ionic conductivity. However, achieving stable energy storage capacity under violent mechanical deformation is still a challenge for hydrogel devices. In this work, an all-in-one integrated supercapacitor (AISC) was assembled using *in situ* deposited polyaniline/graphene oxide nanocomposites for both sides of the incorporated ionic hydrogel electrolyte. The assembly process of the AISC was greatly simplified, and the displacement and separation of the multilayer structured hydrogel complex were avoided during mechanical deformation. The hydrogel electrolyte with ionic additives exhibited strong adhesion and flexibility, and high ionic conductivity, thereby ensuring the excellent specific capacitance and rate performance of the AISC. The specific capacitances of the AISC were 222.8 mF cm^{-2} at the current density of 0.2 mA cm^{-2} and 151.7 mF cm^{-2} at 3.2 mA cm^{-2} . The capacitance retention rate was 68.1%. The energy density of a piece of the device reached $44.6 \mu\text{Wh cm}^{-2}$ at a power density of $120.0 \mu\text{W cm}^{-2}$. Moreover, reliable and reproducible energy storage was acquired under bending, compression, and stretching deformations. The AISC was also easily assembled in series to power a light-emitting diode (LED) light. This work provides a facile approach to the construction of flexible supercapacitors for the development of energy storage devices in flexible electronics.

Keywords: hydrogel electrolyte, flexibility, polyaniline/graphene oxide, supercapacitor

INTRODUCTION

Flexible electronic devices with prominent scalability and portability have been proposed for use in a wide array of wearable devices, electronic skins, smart textiles, and others [1,2]. The development of flexible electronic devices strictly relies on stretchable functional components and energy storage modules. In particular, the design of ductile deformation-tolerant energy storage devices has become a bottleneck in integrated flexible electronics. Flexible supercapacitors (SCs), which feature high

power density, fast charge-discharge capability, and long-term cycling stability, show particularly wide application prospects in flexible electronics [3].

Mechanically deformable electrode and electrolyte materials, and structures are the most critical factors in the assembly of flexible devices [4]. The most widely used electrode material in the field of flexible SCs is conductive polymer, which shows high electrical conductivity by the π - π stacking between polymer chains [5]. During the electrochemical reaction process, conductive polymers can store charges through reversible n-type or p-type ion doping and purification [6]. For example, Oh *et al.* [7] used an electrode fabricated with porous polyaniline (PANI) composites to assemble flexible SCs. However, the pseudocapacitance characteristics of conductive polymers are observed in limited ion exchange reactions during the charge transfer process, thereby resulting in poor rate performance. Moreover, the serious electrode cracking that occurs during ion doping and purification significantly reduces the stability and reliability of flexible SCs. With the critical requirements of flexible SCs in practical applications, recent studies mainly focused on enhancing the stretchability and rate performance of conductive polymers in flexible SCs [8].

One of the most feasible approaches is to incorporate conductive polymers into appropriate stretchable substrates [9]. Hydrogels, which are porous, stretchable, and hydrophilic polymer networks with high water content, can be utilized in electrode-electrolyte-electrode layered structures and show great promise in stretchable SCs [10]. Hydrogels are commonly applied as integrated separators and electrolytes because of their inherent electronic insulation and ion conductance features, and their enhanced ion conductivity relative to industrialized solid electrolytes [11]. With the appropriate incorporation of active materials, conductive hydrogels can also be utilized as stretchable electrodes in flexible SCs. For example, Yang *et al.* [12] proposed all-solid SCs with poly(3,4-ethylene dioxathiophene)/PANI hydrogel electrodes and polyvinyl alcohol/sulfuric acid (PVA/ H_2SO_4) hydrogel electrolytes. The all-solid SCs showed excellent energy storage performance and exhibited an energy density of 0.25 mWh cm^{-3} at a power density of $107.14 \text{ mW cm}^{-3}$. However, the sandwich-structured device was

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found to remain problematic in long-term applications. First, a certain pressure on the device was required along with the charge-discharge process to achieve a close contact between the electrodes and the electrolytes [13]. Second, the delamination and displacement of the laminated structure took place under large deformations, resulting in an obvious decrease in energy storage performance [14]. Therefore, exploiting novel configurations is a top priority for achieving seamless connection and conformal deformation between the electrodes and electrolytes in stretchable SCs. Research has shown that the all-in-one assembly in flexible SCs is an effective way to reduce performance dissipation under mechanical deformation. Hu *et al.* [15] prepared all-in-one configured hydrogel SCs through a one-step deposition of nanocomposites on both sides of a polyacrylic acid (PAA) hydrogel. The device could work at scanning rates of up to 10 V s^{-1} with reserved characteristics of tension and compression. Guo *et al.* [16] prepared integrated SCs by depositing PANI and single-walled carbon nanotubes on two layers of PVA/ H_2SO_4 hydrogel film; the specific capacitance of the fully configured SCs reached 15.8 mF cm^{-2} at a current density of 0.044 mA cm^{-2} . In addition, a large number of hydrogen bonds provided by the physically crosslinked hydrogel promoted an enviable self-healing property in the device. To further optimize the electrochemical performance of the integrated devices, the authors introduced redox additives into the hydrogel electrolyte. Han *et al.* [17] designed a redox bromide ion-compensated hydrogel electrolyte (SA-Zn-Br) for flexible Zn ion hybrid SCs; SA-Zn-Br displayed a maximum energy density of 605 W h kg^{-1} at a power density of 1848 W kg^{-1} and superior voltage of 2.6 V . The additional redox reactions at the electrode-electrolyte interface from the incorporated redox additives significantly increased the specific capacitance and energy density of the flexible SCs [18].

Herein, we assembled an all-in-one integrated SC (AISC) *via* a laminated hydrogel system, in which a PANI/graphene oxide (GO) hydrogel electrode was *in situ* deposited on both sides of a piece of hydrogel electrolyte (GELE). Ionic additives were incorporated into the hydrogel electrolyte to form multiple crosslinking networks, which greatly improved the mechanical and electrochemical properties of the AISC. The specific capacitances of the fully configured SC were 222.8 and 151.7 mF cm^{-2} at current densities of 0.2 and 3.2 mA cm^{-2} , respectively. The area energy density reached $44.6\text{ }\mu\text{W h cm}^{-2}$ at the power density of $120.0\text{ }\mu\text{W cm}^{-2}$. In practice, the PANI/GO-GELE device displayed excellent mechanical resistance to bending, compression, and tension with optimized electrochemical performance. With the excellent adhesive properties of the hydrogel, the device can be easily assembled in a series connection to power light-emitting diode (LED) lights.

EXPERIMENTAL SECTION

Materials

Acroleic acid (AA, 99%), acrylamide (AM, 99%), and *N,N*-methylene bisacrylamide (MBA) were purchased from Sigma-Aldrich. Sodium alginate (SA), CuSO_4 , and CuBr_2 were obtained from Aladdin Chemical Co., Ltd. Aniline, sulfuric acid (H_2SO_4), and ammonium persulfate (APS) were purchased from Yonghua Fine Chemical Co., Ltd. The GO suspension was obtained from Zhongke Yueda (Shanghai) Material Technology Co., Ltd. All chemical reagents were of analytical grade and used without

further purification.

Preparation of GELE film

First, 1 g of AM was dissolved in 4 mL of deionized (DI) water, and the resulting mixture was then added with 4.5 g of SA aqueous solution (5%) under vigorous stirring. Thereafter, 2 mL of AA was added to the aforementioned solution. The mixed solution was sonicated for 30 min to ensure a uniform dispersion. Then, 0.2 g of APS and 5 mg of MBA were dispersed in a mixture solution *via* ultrasonic treatment. The mixed solution was injected into a mold and polymerized at 55°C for 2 h . The obtained hydrogel precursor (GELEP) was removed from the mold and washed with DI water to remove the unreacted monomer. Finally, 0.48 g of CuSO_4 and 0.08 g of CuBr_2 were dispersed in 20 mL of DI water. The GELEP film was soaked in the solution for 24 h to fully achieve material balance. The product was then named GELE and used as the hydrogel electrolyte. The introduction of Cu^{2+} changed the color of the hydrogel from white to light blue. The size of the GELEP and the immersion time were strictly controlled to ensure the introduction of the same amount of ions.

Preparation and electrochemical characterization of PANI/GO-GELE AISC

In brief, the PANI/GO-GELE hydrogel complex was obtained by *in situ* depositing PANI/GO nanocomposites onto the two-sided faces of the GELE film. The electrode materials of aniline and GO were firmly *in situ* polymerized and deposited onto the two sides of the GELE separator. Solution A was prepared by mixing 1 mL of aniline and 2 mL of GO solution (3 mg mL^{-1}) into 2 mL of $1\text{ mol L}^{-1}\text{ H}_2\text{SO}_4$ solution with constant stirring in an ice water bath for 10 min . Solution B was prepared by dissolving 0.56 g of APS in 2 mL of $1\text{ mol L}^{-1}\text{ H}_2\text{SO}_4$ solution cooled at 0°C . Solutions A and B were merged at 0°C , and the GELE was immersed into the mixture of solutions A and B at -7°C for 30 min . After the polymerization, the PANI-decorated PANI/GO-GELE with bilateral PANI/GO electrode materials was obtained. Finally, the all-in-one flexible SC was assembled by connecting both ends of the PANI/GO electrode with carbon cloth as the current collector.

Material characterization

The crystalline structures of the nanocomposite were profiled by X-ray diffraction (XRD) (SmartLab 3KW). The microstructure and elemental analysis of the hydrogel were characterized by scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS) (Hitachi, S-4800, Japan). The mechanical properties of the hydrogel were measured using a test platform with a stepping motor (ESM302, Mark-10) and a force gage (M4-2).

Electrochemical characterization

The electrochemical performance of the AISC was measured by cyclic voltammetry (CV) and galvanostatic charge/discharge (GCD) in a two-electrode configuration by using an electrochemical workstation (Chenhua CHI660E, Shanghai) and a NEWARE battery testing system. The GCD curve measured on the specific capacitance was calculated as follows [19]:

$$C = \frac{I \times \Delta t}{S \times \Delta V}, \quad (1)$$

where I is the applied current, Δt is the discharge time, ΔV is the charge-discharge voltage range, and S is the area involved in the chemical reaction.

The energy density (E) and power density (P) of the device were further calculated through the following [20]:

$$E = \frac{1}{2} C \Delta V^2, \quad (2)$$

$$P = \frac{E}{\Delta t}. \quad (3)$$

RESULTS AND DISCUSSION

Preparation and characterization of PANI/GO-GELE AISC

The fabrication process of the PANI/GO-GELE hydrogel complex and AISC is presented in Fig. 1. First, the monomers of AA, AM, APS, and MBA were dissolved in the SA solution and copolymerized at 55°C for 2 h to form the hydrogel electrolyte precursor GELEP. Then, the GELEP film was immersed in the $\text{CuSO}_4/\text{CuBr}_2$ solution for 24 h to form GELE [21]. In this step, the oxygen-containing polar groups on the molecular chain of SA, including hydroxyl ($-\text{OH}$) and carboxyl ($-\text{COO}^-$), were easily coupled with the Cu^{2+} ions under mild conditions. The redundant Cu^{2+} further coordinated with the carboxyl groups from the PAA chain through ionic interactions [22]. Third, the GELE film was immersed in an acidic solution of aniline and GO nanosheets for *in situ* deposition. In the acidic environment, PANI was quickly polymerized, thereby forming sufficient hydrogen bonds with oxygen-containing functional groups on the surface of GO, and was then deposited onto the two sides of the GELE film, as reflected by the color changes and cross-sectional structure evolution in the images in Fig. S1 [23]. The PANI/GO network established an extensive interaction with the GELE film to reduce the interfacial impedance and ensure rapid

charge transfer at the electrode/electrode interface. In particular, the quick deposition of PANI created a strong adhesion to the GELE film through ionic interaction and formed a distinct sandwich structure (Fig. 1b), which also prevented the infiltration of hydrophilic active materials into the electrolyte film and avoided short circuits in the device [24].

The PANI/GO-GELE hydrogel complex incorporated by multiple physical and chemical interactions showed excellent mechanical and electrical properties because of the GELE substrate that interacted with ions. As shown in Fig. S2, the hydrogel substrate, GELEP, and GELE showed remarkable mechanical deformation, which could sustain sufficient flexibility for stretchable SCs.

Fig. 2a shows that the integrated hydrogel device could withstand a weight of 500 g with negligible damage and revert to its original state after the removal of the external force. More importantly, the PANI/GO-GELE hydrogel complex (Fig. 2b) is easy to stretch in the original, twisted, knotted, and crossover states, thereby exhibiting excellent compression and tension properties. Meanwhile, the hydrogel complex presented good mechanical resistance to compression and quickly returned to its original states after bearing weights of 100, 200, and 500 g (Fig. 2c). Fig. 2d shows that the hydrogel adhered tightly to the carbon cloth, thereby enabling close contact with the current collector to ensure reliable electrochemical performance in the PANI/GO-GELE AISC.

Tensile tests were conducted to quantitatively assess the stretchability of the hydrogel complex. As shown in Fig. 2e, the stretchability of the hydrogel complex was greatly improved by the hydrogel electrolyte substrate because of the rapid chelating performance of Cu^{2+} with carboxylates and amino/hydroxyl groups and the synergistic effects of the multiple interacted PANI/GO networks [25]. Fig. 2f shows the XRD spectrum of

Figure 1 (a) Schematic and synthesis route of PANI/GO-GELE hydrogel complex. (b) Images (upper) and sketches (lower) of the assembly of the laminated AISC.

Figure 2 (a) PANI/GO-GELE hydrogel complex in sustaining a weight of 500 g. (b1–b4) Bare, crossed, twisted, and knotted stretches for the hydrogel complex. (c) Compression properties of the hydrogel complex under weights of 100, 200, and 500 g and its shape recovery performance. (d) Adhesive behavior of PANI/GO-GELE hydrogel complex with carbon cloth. (e) Tensile stress-strain curves of different hydrogels. (f) XRD spectrum of PANI.

PANI, with the broad peak at 26° indicating the successful polymerization and deposition of PANI in the hydrogel complex [26].

The microstructure of hydrogel substrates has been reported to exert a great influence on the electrochemical performance of integrated devices [27]. As shown in Fig. 3a, b, the inner morphology of the PANI/GO-GELE hydrogel complex was characterized by SEM after freeze-drying. The sponge-like hydrogel was uniformly distributed and porous. PANI nanofibers were entangled to form a conductive network across the planar hydrogel electrode. Pieces or small clusters of GO nanosheets were distinguished from the PANI networks in the enlarged images. The uniform dispersion of Br, Cu, C, O, and S were further proved by the EDS elemental mapping of the selected region shown in Fig. 3c–h and confirmed the good incorporation of Cu^{2+} , SO_4^{2-} , and Br^- into GELE. In addition, the three-dimensional (3D) porous laminated networks provided a sufficient attachment area for PANI growth. The channel structure improved the efficiency of mass transport, which further

enhanced the electrochemical performance of the AISC device [28]. In general, the uniform distribution of the active material in the electrode layer, the highly conductive 3D polymer network, and the abundant active sites provided by the porous and thick PANI/GO matrix ensured excellent electrochemical performance in the integrated device [29].

Electrochemical characterization of PANI/GO-GELE AISC

The integrated SC was assembled for the electrochemical tests to determine the electrochemical properties of the PANI/GO-GELE hydrogel complex. In the PANI/GO-GELE AISC system, the PANI/GO hydrogel was utilized as the electrode layer, the ion-interacted GELE film served as the solid electrolyte and separator, and the carbon cloth was adhered to the outer surface of the PANI/GO hydrogel and used as the current collector to form a solid-state energy storage device. Fig. 4a demonstrates the energy storage mechanism of the integrated device. The PANI stored energy through a redox process, which is detailed in Fig. S3. Ions were transferred to the conductive polymer elec-

Figure 3 (a, b) SEM images of the PANI/GO-GELE hydrogel complex after freeze-drying and its partial enlargement. (c–h) EDS elemental mapping of a selected region.

trodes during the oxidation process and released from the entire backbone into the electrolytes during the reduction process. Moreover, the Faraday reaction of the bromide ions in the electrode/electrolyte interface could be directly combined with the electric double-layer electrode to promote additional energy storage [17]. The energy storage mechanism of bromide ions could be proved as follows:

The serious electrode cracking during ion doping and purification is a major challenge affecting the electrochemical performance of the PANI electrodes [30]. Herein, the GO suspension was composited into the PANI network to improve the electrical conductivity and prevent the PANI network from cracking in the deprotonated state [31]. Fig. S4 compares the electrochemical performances of the AISC after the addition of GO on the basis of CV, GCD, and electrochemical impedance spectroscopy measurements. The PANI/GO electrode exhibited better specific capacitance and rate than the pure PANI electrode. For GELE, the multi-ion additives effectively increased the ion concentration in the electrolytes, thereby enabling rapid ion diffusion rates as a solid electrolyte to advance the rate performance of the AISC.

Fig. 4b demonstrates the CV curves of the PANI/GO-GELE AISC at scan rates ranging from 10 to 100 mV s⁻¹. The redox reaction process was observed through the CV curves, and the peak current and curve area gradually increased with the increase of the scan rates. This result is consistent with the

pseudocapacitance characteristic of PANI. The GCD curves at different area current densities are shown in Fig. 4c. The specific capacitances were calculated to be 222.8, 187.2, 163.2, 160.0, and 151.7 mF cm⁻² at 0.2, 0.4, 0.8, 1.6, and 3.2 mA cm⁻², respectively, with the capacitance retentions being 84.0%, 73.2%, 71.8%, and 68.1% (Fig. 4d). As shown in Fig. 4e, the capacitance retention for the PANI/GO-GELE AISC was 79.5 % after 300 cycles. The Ragone plot calculated from the GCD curve is shown in Fig. 4f. The area energy densities of a piece of the device could reach 44.6, 37.4, 32.4, 32.0, and 30.34 μW h cm⁻² at power densities of 120.0, 240.0, 476.4, 960, and 1919.6 μW cm⁻², respectively, thereby satisfying the energy density and power density required in full hydrogel-based flexible energy storage devices.

Electrochemical performance of the PANI/GO-GELE AISC under mechanical deformation

In the actual application of flexible electronics, the electrochemical performance of energy storage devices should be highly adaptable to complicated mechanical deformations under external stimuli [32].

Fig. 5a shows the testing assembly, in which one terminal of the SC was fixed by a small glass slide while the other terminal was rotated to control the bending angle. As demonstrated in Fig. 5b, the AISC showed excellent flexibility at bending angles of 30°, 60°, 90°, 120°, 150°, and 180°. No misalignment or delamination of the electrode and electrolyte could be distinguished even under 180° folding because of the *in situ* polymerization and quick deposition process of the active materials on the GELE film and the firm interfacial interaction in the sandwich

Figure 4 (a) Electrochemical energy storage mechanism of a PANI/GO-GELE AISC. (b) CV curves at different scan rates, (c) GCD curves at different current densities, (d) rate performance, (e) cycling stability at 3.2 mA cm^{-2} , and (f) Ragone plots of the assembled AISC.

Figure 5 (a) Schematic of a PANI/GO-GELE AISC for bending tests. (b) PANI/GO-GELE AISC bending at different angles. (c) CV curves at 100 mV s^{-1} , (d) GCD curves, and (e) calculated specific capacitances at 1.6 mA cm^{-2} for the PANI/GO-GELE AISC at different bending angles. (f) Consecutive CV cycles of the PANI/GO-GELE AISC bent at 90° at 100 mV s^{-1} .

structure. In the case of extreme bending at 180° , an almost negligible depression of the electrochemical performance was observed in the CV and GCD curves (Fig. 5c, d) and in the calculated specific capacitance (Fig. 5e). The slight increase of the specific capacitance might be attributed to the external force exerted on the device under bending. In addition, the electrochemical performance of the device showed high stability under

bending cycles. As shown in Fig. 5f, the CV curves remained steady in 10 consecutive cycles under bending at 90° , thus indicating the AISC's excellent electrochemical performance in the bending tests.

Fig. 6a shows a diagram of the electrochemical tests of the device under pressures of 0, 0.25, 0.5, 1, 2.5, and 5 kPa. The area enclosed by the CV curves in Fig. 6b gradually increased with

Figure 6 (a) PANI/GO-GELE AISC under pressures of 0, 0.25, 0.5, 1, 2.5, and 5 kPa. (b) CV curves at 100 mV s^{-1} , (c) GCD curves, and (d) calculated specific capacitances at 1.6 mA cm^{-2} of the PANI/GO-GELE AISC under different pressures.

the increase of pressure; such a result was consistent with the GCD curves in Fig. 6c and the calculated specific capacitance in Fig. 6d. Under compression at 5 kPa, the specific capacitance exceeded nearly 50% of that of the original device. The literature [33] shows that the adhesion of hydrogels is weakened under compression and that the adhesion property of polymers is inversely proportional to conductivity. Hence, ionic conductivity would apparently increase under compression. With an acceleration of the ion transmission rate at the electrode/electrolyte interface, the electrochemical performance of the integrated devices could be greatly enhanced. In addition, because of the high elasticity of hydrogels, the increase of external compression could shorten the ion diffusion distance to accelerate the redox reaction [34]. Furthermore, increasing compression could strengthen the interfacial contact between electrodes and electrolytes and thereby increase the accessible electrochemical active sites and accelerate charge transfer [35]. Such conjecture could be verified by the decrease of the internal resistance of the device under compression in Fig. S5. Therefore, the integrated AISC could display comparable or even enhanced energy storage performance through the introduction of compression.

The rapid development of flexible energy storage devices for practical applications is required to ensure excellent energy output under mechanical stretching. Fig. 7a shows a schematic of the AISC device at tensile strains of 25% and 50%, which are adequate for most types of human motion detection mechanisms in wearable devices. As shown by the CV curves in Fig. 7b, the enclosed area decreased with increasing tensile strains. Calculated from the GCD curves in Fig. 7c, the specific capacitances decreased to 114.9 and 94.5 mF cm^{-2} with strains of 25% and 50%, respectively (Fig. 7d). Hence, the GELE film with

multiple polymer crosslinking networks ensured the excellent toughness and mechanical strength of the hydrogel complex, inhibited the generation of microcracks on the PANI/GO hydrogel electrode during stretching, maintained the stability of the conductive network, and sustained the reliable electrochemical performance of the PANI/GO-GELE AISC device. These results indicate that the integrated SCs in the anti-deformation tests could meet practical requirements [36].

The working voltage window was tested to further increase the energy density of stretchable SCs and expand the practical applications of the PANI/GO-GELE AISC. As shown in Fig. 8a, the operating voltage window could reach 1.5 V without severe polarization; the result is higher than that obtained with most integrated, flexible SCs [16,28,37]. Fig. 8b also shows convincing electrochemical performance in the CV curves under cycling at different scan rates. The expanded voltage window was attributed to a typical amphiphilic structure of the hydrogel electrolyte rich in multiple ions. The strong interaction between the abundant charged groups and the water molecules (76.0%) resulted in the expansion of hydrogen and oxygen evolution voltage [38]. As demonstrated in Fig. 8c, three pieces of AISCs could be easily connected in series owing to their inherently strong adhesive property, and the fully charged prototype could power a red LED light with steady energy output.

CONCLUSIONS

The integrated PANI/GO-GELE hydrogel complex was prepared into a sandwiched structure through a facile method. The assembled PANI/GO-GELE AISC showed excellent electrochemical performance with specific capacitances of 222.8, 187.2, 163.2, 160.0, and 151.7 mF cm^{-2} at 0.2, 0.4, 0.8, 1.6, and

Figure 7 (a) Schematic of PANI/GO-GELE AISC under different tensile strains. (b) CV curves at 100 mV s^{-1} , (c) GCD curves, and (d) calculated specific capacitances at 1.6 mA cm^{-2} for PANI/GO-GELE AISC under different tensile strains.

Figure 8 (a) CV curves of the PANI/GO-GELE AISC in different voltage windows at 50 mV s^{-1} . (b) CV curves of the PANI/GO-GELE AISC at different scan rates in the voltage range of 0–1.6 V. (c) Diagram of three pieces of AISCs in series connection to power LED light.

3.2 mA cm^{-2} , respectively. It also displayed a superior energy density of $44.6 \mu\text{Wh cm}^{-2}$ at a power density of $120.0 \mu\text{W cm}^{-2}$. Moreover, the AISCs revealed excellent mechanical deformation resistance to bending, compression, and stretching while maintaining reliable and reproducible electrochemical performance. Owing to the strong adhesion property of the hydrogel complex, the AISCs could be connected in series to power LED lights and thus demonstrate practical application prospects. The proposed PANI/GO-GELE AISC with remarkable electrochemical performances under mechanical deformation provides a novel approach to the development of flexible energy storage devices.

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

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原位沉积多层集成水凝胶用于可变形拉伸超级电容器

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摘要 因其固有的机械弹性和离子电导率, 水凝胶体系极大地促进了柔性储能设备的发展. 然而, 对于水凝胶装置而言, 在剧烈的机械变形下实现稳定的能量存储仍然是一个挑战. 本文通过在离子结合水凝胶电解质的两侧原位沉积聚苯胺/氧化石墨烯(PANI/GO)纳米复合材料, 组装一体化集成超级电容器(AISC), 简化了器件组装过程, 并避免了水凝胶储能器件在机械变形过程中多层结构的置换和分离; 同时, 离子添加剂水凝胶电解质表现出较强的粘附性、柔韧性以及高离子电导率, 确保了AISC优异的比电容和倍率性能. 其在 0.2 mA cm^{-2} 电流密度下比电容为 222.8 mF cm^{-2} , 在 3.2 mA cm^{-2} 电流密度下比电容为 151.7 mF cm^{-2} , 电容保持率为68.1%. 器件在 $120.0 \mu\text{W cm}^{-2}$ 的功率密度下, 能量密度达到 $44.6 \mu\text{Wh cm}^{-2}$. 此外, 在弯曲、压缩和拉伸的变形下可以获得稳定的能量存储, 串联组装的AISC可以为LED灯供电. 该工作在改善超级电容器机械性能的同时进一步提高了其电学性能, 为构建新型柔性超级电容器提供了一种新策略.