Research Progress of All-In-One Supercapacitor

Yue Wang

School of Materials Science and Engineering, Henan Polytechnic University, Jiaozuo 454003, China.

Abstract: With the development of flexible electronics, supercapacitors have received great attention due to their own advantages, of which the rapidly developing all-in-one supercapacitors have received more favour from researchers in recent years. Because they not only can reduce the interfacial resistance between electrolyte and electrode, but also ensure stability in external deformation. This paper provides a comprehensive review of all-in-one supercapacitors. Firstly, the similarities and differences between conventional and all in one supercapacitors are compared, followed by a discussion of the three main assembly methods for all-in-one supercapacitors.

Keywords: All-In-One Supercapacitor; Preparation Methods

1. Introduction

In the past decades, coal, oil and gas have been the main sources of energy driving socio-economic development. However, the environmental pollution caused by these non-renewable energy sources has led to a shift towards renewable energy sources such as wind, solar and tidal energy, which in turn has led to a push for high performance energy storage devices to store these renewable energy sources. Supercapacitors are receiving increasing attention for energy storage in wearable electronic devices due to their faster charge and discharge rates than batteries, high power density and long cycle life. Some special structural designs of supercapacitors are able to match various complex deformations of electronic devices, especially the gel electrolyte-based all-in-one supercapacitors, which are the focus of current research in energy storage devices due to their high flexibility, low interfacial resistance and self-healing properties. This paper reviews the basic information of supercapacitors, the comparison of conventional and all-in-one supercapacitors, and we also analyses the latest designs.

2. Comparison of conventional supercapacitors and all-in-one supercapacitors

2.1 Conventional supercapacitors

The four components-electrode material, electrolyte, diaphragm and collector - are the main components of a conventional supercapacitor. The electrode material is crucial for generating pseudocapacitance and double layer capacitance. The most popular ones currently in research are carbon-based materials, conducting polymers, metal oxides, heteroatom-doped carbon materials, sulphides, MXenes, polymetallic oxides, metal-organic skeletal materials, black phosphorus, etc., whose main role is to provide capacitance to the supercapacitor during the charging/discharging process. Electrolytes are able to provide supercapacitor with the ions necessary to conduct. In conventional supercapacitor, electrolytes usually include aqueous electrolytes, organic electrolytes, and ionic liquids. The collector is the electrically conductive connection structure between the electrode material and the external connection point of the supercapacitor. During the charging and discharging process, the charge carriers are effectively pooled or transported mainly through the collector. The diaphragm is located between the positive and negative electrodes of the supercapacitor to avoid a short circuit due to direct contact between the two electrodes, and it also can ensure that the free transport of ions between the positive and negative electrodes.

2.2 All-in-one supercapacitors

All in one supercapacitor means that the electrode, electrolyte, separator and even collector are integrated on a flexible substrate. This approach not only reduces the interlayer interface resistance, but also avoids the use of redundant inactive substances, effectively improving the performance of the supercapacitor. Compared with the traditional sandwich structure supercapacitor, the design of the all in one supercapacitor has three advantages: firstly, it can solve the problem of multi-interface delamination between the electrode and electrolyte, and the interfacial resistance between the layers can be reduced. Secondly, the all in one supercapacitor will not lead to electrode detachment or interlayer sliding. Finally, the integrated structure can also be readily cut into various shapes and sizes according to the requirements. Typically, all in one supercapacitors are prepared using conductive polymers such as polyaniline, polypyrrole and PEDOT-PSS combined with a hydrogel matrix. Conductive polymers are grown on the surface of the GPE mainly by in situ polymerisation, which allows simultaneous deformation of the electrode and electrolyte.

3. Preparation methods of all-in-one supercapacitors

3.1 In-situ polymerization

In recent years, most all-in-one supercapacitors have been prepared mainly by in situ deposition of conductive polymers on gel electrolyte. In detail, the conductive monomer is infiltrated into the network of the gel electrolyte by soaking, which not only reduces the interfacial resistance between the electrolyte and the electrode, but also ensures that the all-in-one supercapacitor can withstand complex external deformation and damage without displacement or delamination, making it more suitable for energy storage device. Zhang[1] prepared an all-in-one supercapacitor by in situ polymerisation of pyrrole on a PVA /KCl gel electrolyte. The device achieved a large area capacitance of 224 mF/cm^2 and a significant energy density of 20 µWh cm^{-2}, and could exhibit excellent electrochemical stability at various bending angles (0°, 90° and 180°). In addition, in situ polymerisation of the electrolyte on the prepared electrode is also an effective method, which also ensures a tight contact between the electrode and electrolyte. Peng[2] formed an all-in-one supercapacitor with low-temperature tolerance by applying a PVA/NaCl/glycerol hydrogel premise solution to an activated carbon electrode in situ, which can be easily and rapidly moulded at room temperature. The device exhibited excellent flexibility and electrochemical performance at 23°C with a capacitance retention of 90.5%.

3.2 Physical adsorption

Considering the conductive polymer-based all-in-one supercapacitors always encounter swelling problems during the immersion process, the mechanical properties of the resulting all-in-one supercapacitors are degraded. Wang[3] have prepared all-in-one supercapacitors by a simple physical adsorption method. In detail, he first prepared a PVA/PAI film with adhesive properties and then hot pressed it onto the prepared electrode surface at 10 MPa and 80 °C to obtain an all-in-one supercapacitor with stretchable and self-healing properties. In addition, Chen[4] used PVA and poly(acrylamide-co-2-acrylamido-2-methylpropane sulfonic acid) (P(AM-AMPS)) and glycerol to prepare gel electrolytes with significant stretchability (over 894%), strong adhesion (37.00 kPa to carbon materials) and high ionic conductivity. All-in-one supercapacitors were obtained by uniformly coating carbon nanotube electrodes on both sides of the adhesive electrolyte. At a current density of 0.5 mA/cm^2, the device was able to display a high specific capacity of 85.25 mF/cm^2 and is able to maintain a stable specific capacitance when stretched (200% strain) or bent (0~180°)

3.3 Other preparation methods

In addition, some researchers have prepared all-in-one supercapacitors by pretreating and electrodepositing on the gel electrolyte. Inspired by skin structures, Fang[5] prepared an all-in-one supercapacitor by pretreating agar/polyacrylamide/lithium chloride (AG/PAAm/LiCl) gel electrolyte and affixing it to an activated carbon electrode. This design improved the electrochemical behaviour as well as specific capacitance and was able to maintain better performance under repeated use. Gao[6] designed a wearable all-in-one supercapacitor that integrated the collector fluid and electrodes into a flexible porous polyamide nanofibre film. The positive and negative electrodes were mainly fixed on both sides of the nanofibre film by electrodeposition. This segign simplifies the assembly process and reduces material costs without adding additional binders and metal collectors. In a neutral PVA/LiCl gel electrolyte, a volume specific capacitance of 3.1 F/cm^3 can be achieved at a current density of 0.5 A/g, and multiple devices in series can also achieve high output voltages.

4. Conclusion

This work makes a summary of all-in-one supercapacitors and reviews conventional supercapacitors. Meanwhile, they were compared specifically with conventional supercapacitors. The latest methods for the preparation of all-in-one supercapacitors are outlined, including chemical deposition, physical adsorption and electrodeposition.

References

[1] Zhang K, Zhang X, Zou B, et al. A leather-based electrolyte for all-in-one configured flexible supercapacitors[J]. Chemical Communications, 2022.

[2] Peng S, Jiang X, Xiang X, et al. High-performance and flexible solid-state supercapacitors based on high toughness and thermoplastic poly(vinyl alcohol)/NaCl/glycerol supramolecular gel polymer electrolyte[J]. Electrochimica Acta, 2019, 134874.

[3] Wang Y, Lv C, Ji G, et al. An all-in-one supercapacitor with high stretchability via a facile strategy[J]. Journal of Materials Chemistry A, 2020.

[4] Chen G, Hu O, Lu J, et al. Highly flexible and adhesive poly(vinyl alcohol)/poly(acrylic amide-co-2-acrylamido-2-methylpropane sulfonic acid)/glycerin hydrogel electrolyte for stretchable and resumable supercapacitor[J]. Chemical Engineering Journal, 2021, 131505.

[5] Fang L, Cai Z, Ding Z, et al. Skin-Inspired Surface-Microstructured Tough Hydrogel Electrolytes for Stretchable Supercapacitors[J]. ACS Applied Materials & Interfaces, 2019, 11(24):21895–21903.

[6] Gao T, Zhou Z, Yu J, et al. All-in-One Compact Architecture toward Wearable All-Solid-State, High-Volumetric-Energy-Density Supercapacitors[J]. ACS Applied Materials & Interfaces, 2018, 10(28):23834-23841.