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Communication

Simplified fabrication of high areal capacitance all-solid-state micro-supercapacitors based on graphene and MnO₂ nanosheets

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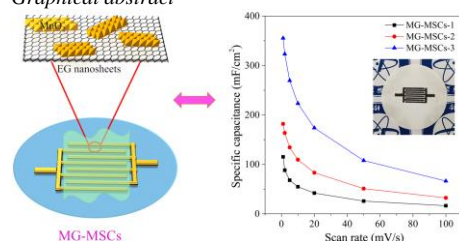
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Graphical abstract



A universal simplified strategy was developed to fabricate all-solid-state planar micro-supercapacitors with high areal capacitance (~ 355 mF/cm²), based on interdigital patterned films of 2D pseudocapacitive MnO₂ nanosheets and electrochemically exfoliated graphene.

ABSTRACT

All-solid-state micro-supercapacitors are acknowledged as a very promising class of microscale energy storage devices for directly integrating portable and wearable electronics. However, the improvement of electrochemical performance from materials to devices still remains tremendous challenges. Here, we demonstrate a novel and universal mask-assisted filtration technology for the simplified fabrication of all-solid-state planar micro-supercapacitors (MSCs) based on interdigital patterns of 2D pseudocapacitive MnO₂ nanosheets and electrochemically exfoliated graphene film as both electrode and current collector, and polyvinyl alcohol/LiCl gel as electrolyte. Remarkably, the resulting MSCs exhibit outstanding areal capacitance of ~ 355 mF/cm², which is among the highest values reported in the state-of-the-art MSCs. Meanwhile, MSCs possess exceptionally mechanical flexibility as high as $\sim 92\%$ of initial capacitance even at a highly bending angle of 180°, excellent cyclability with a capacitance retention of 95% after 3000 cycles, and impressive serial or parallel integration for modulating the voltage or capacitance. Therefore, our proposed strategy of simplified construction of MSCs will pave the ways for utilizing graphene and analogous pseudocapacitive nanosheets in high-performance MSCs.

Keywords: MnO₂ nanosheets Graphene All-solid-state Planar Micro-supercapacitors Energy storage

With the rapid development of wearable and portable electronics, microscale energy storage devices with multiple compatible features of lightweight, tailored size, outstanding flexibility, and high energy density have gained tremendous attentions. Recently, micro-supercapacitors (MSCs) are considered as a very promising class of on-chip energy storage devices for integrated electronics due to ultrahigh power delivery, excellent rate capability, robust mechanical flexibility, unique shape diversity and safety [1-3]. Despite remarkable advancements in the fabrication of nanostructured electrode materials and the manufacturing of novel devices, there are still huge challenges in the development of MSCs with high areal capacitance, which is a crucial performance metrics of MSCs [4,5]. To boost the areal capacitance, the key is to develop high-performance thick electrode materials with developed ionic and electronic conducting network, which however is still underdeveloped in this important research field.

Two-dimensional (2D) materials, characterized by nanoscale dimension in thickness and infinite length in the plane, are currently regarded as groundbreaking electrode candidates for MSCs because they present unique properties of large surface area, ultra-thinness, high mechanical flexibility, and unprecedented capacitance characteristics, and also demonstrate potential applications in electronics, transparent conducting electrode, composites, energy storage and conversion [6-10]. Conceptually, 2D materials mainly include graphene and graphene-like analogues (such as, transition metal oxides/hydroxides, metal sulfides, boron nitride, MXenes, phosphorene, thiophene) [11-13]. Among them, MnO₂ nanosheets have received extensive concerns for high-performance supercapacitors because of

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