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Interfacial engineering of $NiCo_2S_4$/graphene quantum dots for enhanced charge transport and high-performance asymmetric supercapacitors

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ABSTRACT

Rational design of composite electrode materials based on bimetallic sulfides integrated with functionalized graphene quantum dots (GQDs) for advanced electrode materials with enhanced features for high-energy storage remains a significant challenge. In particular, engineering ternary nickel cobalt sulfides ($NiCo_2S_4$, NCS) combined with GQDs to boost interfacial electrochemistry for optimal capacitive electrode performance is indispensable. In this work, hierarchical NCS/GQDs nanocomposites were successfully synthesized via a one-step hydrothermal synthesis followed by a controlled high-temperature sulfurization process at different time intervals. The incorporation of GQDs significantly improves the conductivity, structural stability, and availability of charge storage sites in the NCS/GQDs electrodes. Interestingly, the synergistic interaction between NCS and GQDs during synthesis led to the formation of well-developed two-dimensional nanosheet architectures. As a result, the NCS/GQDs@1h electrode delivers a specific capacitance of 1618 F g^{-1} at 0.5 A g^{-1}, demonstrating excellent electrochemical performance. Owing to enhanced electron transport, improved electrical conductivity, and an abundance of active sites for charge storage, the asymmetric supercapacitor (ASC) device delivers a maximum energy density of 32.7 Wh/kg at a power density of 472.9 W/kg. Furthermore, due to strong structural integration between NCS and GQDs, the NCS/GQDs@1h demonstrates excellent structural robustness even after 10,000 charge-discharge cycles, with a high capacitance retention (~98.9%). The cooperative effect between NCS and GQDs not only enhances the structural robustness but also provides efficient electron transport pathways at the interface, highlighting the potential of the NCS/GQDs@1h electrode as a promising candidate for efficient charge storage in advanced supercapacitor applications.

1. Introduction

The continuous increase in population, along with industrialization and advancements in modern electronics technology, has led to a rise in energy consumption. This surge presents significant issues, including resource depletion and environmental pollution. To date, fossil fuels remain the dominant energy source for energy production, and an excess of their use results in significant emissions of greenhouse gases, which subsequently lead to climate and environmental threats. Accordingly, the development of sustainable and efficient energy storage systems, renewable energy technologies, as well as next-generation electronic devices, as an alternative to conventional technologies, has attracted significant research attention [1,2]. The existing energy storage devices, including batteries, fuel cells, supercapacitors (SCs), and thermal energy storage systems, have been extensively investigated [3,4]. In recent years, these energy storage devices have been widely used in numerous electronic devices, including digital cameras, mobile phones, laptops, aircraft, and electric vehicles [5–7]. Among them, SCs have emerged as promising energy storage devices because of their high-power density, fast charge–discharge rates, long cycle life, and environmentally friendly nature [8,9]. The electrochemical performance of SCs is strongly governed by the properties of the electrode materials, particularly those based on transition metal oxides (TMOs), transition metal hydroxides (TMOHs), and conducting polymers, which have been

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