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Layered construction of integrated sulfur-bridged CoNi-S/rGO architecture for enhanced electrochemical energy storage

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ABSTRACT

Transition metals chalcogenides (TMCs) have been recognised so far with their high Faradic activity, making them promising candidates for efficient charge transfer electrodes. However, their tendency to aggregate hindered their potential applications in supercapacitors. This study introduces a novel cathode material composed of CoNi-sulfides (CoNi-S) nanostructured flakes and reduced graphene oxide (rGO) sheets designed to be connected through additional sulfur atoms to enhance their conductivity and electroactive surface area for hybrid supercapacitors. Remarkable results were realised by forming a layered structure of CoNi-S/rGO in which rGO sheets wrap CoNi-S flakes. Interestingly, the CoNi-S/rGO composite exhibited a specific capacitance of 3308F g^{-1} (1654C g^{-1}) at 1 A g^{-1}, outperforming the performance of a single CoNi-S component, which recorded 2155F g^{-1} (1077.5C g^{-1}) at identical conditions. Both materials demonstrated exceptional high-rate capabilities, retaining about 70 % of their capacitance even at an elevated current density of 10 A g^{-1}. In a two-electrode coin cell system, the device showcased a high energy density of 50.2 Wh kg^{-1} at a power density of 750 W kg^{-1}. It maintained an impressive 84 % capacitance retention after enduring 35,000 cycles. These remarkable findings hold significant promise for advanced energy storage applications, marking substantial progress forward in hybrid supercapacitor technology.

1. Introduction

Significant electrochemical energy storage technology advancements have been made in the past decade. This progress has led to the emergence of various energy storage devices that have garnered attention and achieved significant milestones in fulfilling energy demands recently [1]. Aqueous hybrid supercapacitors offer promise in advancing renewable energy storage, employing capacitive materials as negative electrodes and battery-type materials as positive electrodes [2,3]. TMCs excel as superior pseudocapacitive materials due to enhanced redox chemistry, extended cycle life, added catalytic capabilities, and structural advantages like flexibility and high surface area, resulting in significantly higher specific capacitance than oxides and nitrides [4–6].

Transition metal sulfides (TMSs), like $NiCo_2S_4$, exhibit remarkable electrochemical activity and reversibility. However, single-metal sulfides need to be improved in catalytic activity due to poor conductivity and slow electron transfer. To overcome this, binary transition metal sulfides, such as $NiCo_2S_4$, integrate nickel's capacity with cobalt's stability, resulting in enhanced electrochemical performance. The incorporation of a second metal cation reduces the band gap, increases surface-active exposure, improves electron transport, and enhances structural stability [7–10].

Despite their promise, TMSs face challenges like aggregation, which can block active sites and hinder electron transfer. Additionally, their

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