Thesis Abstract

The rapid expansion of portable electronics, electric mobility, and renewable energy technologies has created a growing demand for high-performance energy storage systems like supercapacitors. However, achieving high energy density and power density while maintaining excellent rate capability remains a key challenge. Transition metal sulfides have attracted considerable attention as electrode materials due to their superior electrical conductivity and rich redox chemistry. In this context, the present thesis focuses on the systematic design and optimization of transition metal sulfide heterostructures to enhance electrochemical performance in supercapacitors.

The study begins with the investigation of metal sulfide heterostructure i.e. $Cu_2S/NiS/Ni_3S_4$ to evaluate whether multi-metal compositions can outperform single metal sulfides. It exhibits enhanced pseudocapacitive behaviour due to synergistic redox interactions among the constituent metals. To further improve electron transport, conductive carbon nanotubes were incorporated into a Cu_2S/Co_3S_4 composite, forming an interconnected conductive network that significantly reduced charge transfer resistance. Thereby it achieved a capacitance of 960 F g^{-1} with a capacitance retention of 91% after 20,000 cycles. Further improvement was achieved through optimization of synthesis conditions and phase engineering, enabling better control over crystal structure and electrochemical activity. Phase engineering was explored through a comparative investigation of the semiconducting 2H and metallic 1T phases of MoS_2. The superior conductivity of the 1T phase aided the design of a 1T-MoS_2@Cu_2S heterostructure, which demonstrated enhanced electrochemical activity and delivered an excellent energy density 144 Wh kg^{-1} and a high power density of 19 kW kg^{-1}. Furthermore, composition optimization was carried out in CdS/CuS heterostructures, where the formation of a p–n heterojunction promoted interfacial charge redistribution and improved electrochemical kinetics. Finally, electrolyte engineering was investigated using sodium-based water-in-salt electrolytes where CuS/CdS was used as an active electrode material to expand the electrochemical stability window. The assembled supercapacitor device using 17 m $NaClO_4$ operated at a voltage window of 2.75 V, achieving an energy density of 50 Wh kg^{-1} and a huge a power density of 27,500 W kg^{-1}. Overall, this thesis demonstrates that a multi-level optimization strategy involving material composition, conductive network formation, phase engineering, interfacial heterojunction design, and electrolyte selection can significantly enhance the electrochemical performance of sulfide-based supercapacitors, offering their applicability in real-world energy storage technologies such as hybrid electric vehicles, grid energy storage.

Keywords: Transition metal sulfide heterostructure, power density, synergistic effect, optimization