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Comparative study of Supercapacitive Properties of Nanostructured Sulfides of Molybdenum, Tungusten and Chromium

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ABSTRACT: Nanostructured sulfides of molybdenum (MoS_2), tungsten (WS_2), and chromium (Cr_2S_3), have emerged as promising electrode materials for supercapacitor applications due to their unique layered structures, tunable electronic properties and excellent electrochemical performance. This comparative study systematically investigates the supercapacitive properties of these metal sulfides through analysis of their synthesis methods, structural characteristics, electrochemical behaviour and energy storage mechanisms. MoS_2 nanomaterials demonstrate exceptional specific capacitance, while WS_2 nanostructures exhibit very good capacitance values with superior cycling stability. Cr_2S_3-based electrodes, though less extensively studied, show remarkable potential when combined with conductive matrices. The study reveals that all three sulfides exhibit hybrid charge storage mechanisms involving both electrical double-layer capacitance (EDLC) and pseudocapacitance through reversible Faradaic redox reactions. Critical performance parameters including energy density, power density, rate capability, and cyclic stability are comparatively analysed. The findings indicate that while MoS_2 offers higher intrinsic capacitance, WS_2 demonstrates superior long-term stability, and Cr_2S_3 shows promise when appropriately engineered with conductive substrates.

KEY WORDS: Capacitance, electrochemical, electrode, metal sulfide

I. INTRODUCTION

The escalating global energy demand and the urgent need for sustainable energy storage solutions have driven intensive research into advanced electrochemical energy storage devices. Supercapacitors, which are also known as electrochemical capacitors, have garnered significant attention as promising energy storage systems that bridge the gap between conventional capacitors and batteries. These devices offer unique advantages such as high power density, rapid charge-discharge rates, excellent cycling stability exceeding 30,000 cycles, and long operational lifetimes compared to traditional batteries. However, the relatively low energy density of supercapacitors compared to batteries remains a critical limitation that restricts their widespread practical applications [1, 2].

II. RELATED WORK

The performance of supercapacitors is determined by the electrode materials employed. Transition metal sulfides (TMSs) have emerged as a promising class of pseudocapacitive materials for supercapacitor electrodes due to their high theoretical capacitance, excellent redox reversibility, superior electrical conductivity compared to metal oxides, and earth-abundant availability. Among various TMSs, molybdenum disulfide (MoS_2), tungsten disulfide (WS_2), and chromium sulfide (Cr_2S_3) represent particularly interesting candidates due to their layered crystal structures analogous to graphene, which facilitate efficient ion intercalation and charge storage [3].

MoS_2 and WS_2 belong to the family of transition metal dichalcogenides (TMDCs), characterized by a unique S-M-S trilayer atomic structure with weak van der Waals forces between layers. This layered morphology provides high surface areas, abundant active sites for electrochemical reactions, and favourable ion transport pathways. Recent studies have demonstrated that MoS_2-based electrodes can achieve specific capacitances ranging from 395 to 576 F g^{-1} depending on morphology and synthesis conditions. WS_2 electrodes exhibit excellent cycling stability with capacitance retention exceeding 93% after 5000 cycles, though their specific capacitance values are generally lower than MoS_2 [4].

Chromium sulfide, particularly Cr_2S_3, has received comparatively less attention in the supercapacitor literature despite its unique magnetic and electronic properties. Recent investigations have revealed that manganese-doped Cr_2S_3 combined with reduced graphene oxide (rGO) can achieve remarkable specific capacities of 886.7 F g^{-1} at 5 mV s^{-1}, with

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outstanding energy density of 121.91 Wh kg^{-1} and power density of 1040 W kg^{-1}. The ferrimagnetic semiconductor nature of rhombohedral Cr_2S_3 and its easily exfoliatable layered structure make it an intriguing candidate for electrochemical energy storage applications [5].

Crystal Structure and Morphology

All three metal sulfides share a common layered crystal structure, though with distinct variations in their atomic arrangements and electronic properties. MoS_2 and WS_2 naturally adopt a 2H-phase hexagonal structure where the metal atoms (Mo or W) are sandwiched between two layers of sulphur atoms in a trigonal prismatic coordination. The layers are held together by weak van der Waals forces with interlayer spacing of approximately 0.65 nm, facilitating easy exfoliation into few-layer or monolayer nanosheets. The 2H phase exhibits semiconducting behaviour with indirect band gaps of 1.2 eV for multilayer structures and direct band gaps of approximately 1.8 eV for monolayers [6]. A metallic 1T phase can be obtained through chemical exfoliation or phase transformation, which exhibits octahedral coordination and significantly enhanced electrical conductivity (10-100 S cm^{-1}) compared to the 2H phase (0.2 S cm^{-1}). The 1T-phase MoS_2 and WS_2 demonstrate 100 times higher conductivity than their 2H counterparts, leading to superior supercapacitor performance. Studies have shown that 1T-WS_2 electrodes achieve specific capacitances significantly higher than 2H-WS_2 due to enhanced electron transport and increased active sites [7]. Chromium sulfide (Cr_2S_3) crystallizes in a rhombohedral structure and exhibits ferrimagnetic semiconductor properties. The compound possesses a layered structure like MoS_2 and WS_2, making it easily exfoliatable like graphite. Single-unit-cell thick Cr_2S_3 nanosheets demonstrate unique magnetic ordering with critical Néel temperatures, and the material can exist as a fully compensated ferrimagnet where opposing magnetic moments completely cancel out. This distinctive electronic structure contributes to its electrochemical properties, though the specific mechanisms require further investigation [8].

III. SIGNIFICANCE OF THE STUDY

This comprehensive review aims to systematically compare the supercapacitive properties of nanostructured MoS_2, WS_2, and Cr_2S_3 through analysis of their synthesis methodologies, structural characteristics, charge storage mechanisms, electrochemical performance metrics, and practical applications. Understanding the comparative advantages and limitations of these three metal sulfides is essential for rational design and optimization of next-generation supercapacitor electrode materials.

IV. METHODOLOGY

Synthesis Methodologies

Hydrothermal synthesis has emerged as the most widely employed method for preparing nanostructured metal sulfides due to its simplicity, cost-effectiveness, scalability, and ability to control morphology. For MoS_2 synthesis, ammonium molybdate [$(NH_4)_6Mo_7O_{24}\cdot 4H_2O$] or sodium molybdate serve as molybdenum sources, while thiourea (CH_4N_2S) or thioacetamide provides the sulphur source. The reaction typically occurs at temperatures between 180-240°C for 12-24 hours in Teflon-lined stainless-steel autoclaves. This process yields various morphologies including nanosheets, nanoflowers, microspheres, and nanodots depending on synthesis parameters such as temperature, precursor concentration, reaction time, and surfactants employed [9]. Wang et al. synthesized three-dimensional flower-like MoS_2 microspheres composed of nanosheets through hydrothermal treatment, achieving specific capacitances of 518.7 F g^{-1} at 1 A g^{-1}. The use of cetyltrimethylammonium bromide (CTAB) as a surfactant facilitates the formation of nanoflower morphologies with enhanced surface areas and more active sites compared to simple nanosheets. MoS_2 nanodots, produced through prolonged ultrasonication of nanosheets, demonstrate excellent specific capacitance of 395 F g^{-1} at 1.5 A g^{-1} due to their high surface-to-volume ratio [2].

Tungsten disulfide synthesis follows similar hydrothermal protocols using tungsten hexachloride (WCl_6) or sodium tungstate as tungsten sources combined with thioacetamide or thiourea as sulfur precursors. Synthesis at 240°C for 24 hours produces WS_2 nanorods with widths ranging from 7-9 nm and band gaps of 1.84 eV. The morphology of WS_2 can be tuned to nanoflakes, nanosheets, nanoflowers, or nanorods through adjustment of synthesis conditions. Chemical exfoliation methods can produce metallic 1T-phase WS_2 with superior conductivity for enhanced supercapacitor performance [10]. Chromium sulfide nanoparticles are synthesized through hydrothermal methods using chromium salts and sulphur sources under controlled conditions. Manganese-doped Cr_2S_3 with reduced graphene oxide (Mn-doped Cr_2S_3/rGO) nanocomposites are prepared by mixing chromium precursors, manganese dopants, sulphur sources, and graphene oxide in aqueous solution followed by hydrothermal treatment. The synthesis temperature, dopant concentration, and reaction duration critically influence the crystallinity, morphology, and electrochemical properties of the final product [11].

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Electrochemical Performance and Charge Storage Mechanisms

Charge Storage Mechanisms

Transition metal sulfides exhibit hybrid charge storage mechanisms involving both electrical double-layer capacitance (EDLC) and pseudocapacitance through Faradaic redox reactions. EDLC arises from the electrostatic accumulation of ions at the electrode-electrolyte interface without charge transfer, providing rectangular cyclic voltammetry (CV) curves and linear galvanostatic charge-discharge (GCD) profiles. Pseudocapacitance originates from fast and reversible Faradaic redox reactions at or near the electrode surface, involving electron charge transfer between the electrode and electrolyte [12]. For metal sulfides, pseudocapacitance can occur through three primary mechanisms: (1) surface redox reactions involving rapid oxidation-reduction of metal centers at different valence states, (2) ion intercalation/deintercalation within the layered structure, and (3) underpotential deposition of ions. The layered crystal structure of MoS_2, WS_2, and Cr_2S_3 facilitates intercalation of electrolyte ions (OH^-, H^+, K^+, Na^+) between the sulfur layers, enabling charge storage throughout the material volume rather than only at the surface [13]. In alkaline electrolytes (typically 1-6 M KOH), metal sulfides undergo reversible redox reactions according to the general equation: $MS + OH^- \leftrightarrow MSOH + e^-$, followed by $MSOH + OH^- \leftrightarrow MSO + H_2O + e^-$. These Faradaic processes contribute significantly to the total capacitance, often exceeding the EDLC contribution by 10-100 times. The multiple oxidation states available to molybdenum (0, +2, +3, +4, +5, +6), tungsten (0, +2, +3, +4, +5, +6), and chromium (0, +2, +3, +6) enable rich redox chemistry that enhances charge storage capacity [14]. Studies using cyclic voltammetry reveal that MoS_2 and WS_2 electrodes exhibit quasi-rectangular CV curves with redox peaks, indicating combined EDLC and pseudocapacitive behaviour. The redox peaks correspond to reversible Faradaic reactions of the metal centres with hydroxide ions in the electrolyte. At high scan rates, the diffusion-controlled processes become limited, and the capacitance decreases due to insufficient time for ion intercalation into inner layers [15].

V. EXPERIMENTAL RESULTS AND DISCUSSION

Comparative Analysis and Structure-Property Relationships

Electrochemical Impedance and Charge Transfer Kinetics

Electrochemical impedance spectroscopy (EIS) provides critical insights into charge transfer kinetics, ion diffusion processes, and interfacial properties of supercapacitor electrodes. Nyquist plots of metal sulfide electrodes typically show a semicircle in the high-frequency region corresponding to charge transfer resistance (RCT), followed by a Warburg impedance region representing ion diffusion, and a nearly vertical line at low frequencies indicating capacitive behaviour. The equivalent series resistance (ESR), determined from the x-intercept of Nyquist plots, reflects the combined resistance of electrolyte, electrode materials, and contact resistance. Lower ESR values correlate with better conductivity and superior electrochemical performance. MoS_2 electrodes show charge transfer resistance typically ranging from 3.4 to 4.9 Ω depending on morphology and synthesis conditions. WS_2 exhibits significantly lower charge transfer resistance (3.9 Ω) compared to MoS_2 (21 Ω), explaining its superior rate capability [16]. Chromium sulfide-based composites demonstrate low charge transfer resistance when combined with conductive substrates. The Mn-doped Cr_2S_3/rGO composite showed improved charge transfer kinetics attributed to the conductive graphene network facilitating electron transport. The unique structural properties of these composites provide extended ion diffusion pathways that enhance ionic conductivity and electrochemical performance [17].

Energy Density and Power Density Performance

The energy density and power density of supercapacitor devices are typically represented through Ragone plots, allowing direct comparison with other energy storage technologies. Energy density (E) and power density (P) are calculated from galvanostatic charge-discharge curves using equations: $E = \frac{1}{2}CV^2$ and $P = E/\Delta t$, where C is specific capacitance, V is operating voltage, and Δt is discharge time [18]. MoS_2-based symmetric supercapacitors achieved energy densities ranging from 6.42 to 22 Wh kg^{-1} with power densities between 70 and 10,000 W kg^{-1}. These values significantly exceed conventional carbon-based supercapacitors (energy density 5-10 Wh kg^{-1}) while maintaining high power capability. The energy density of MoS_2 devices approaches that of nickel-metal hydride batteries (30-80 Wh kg^{-1}) while providing much higher power density [2]. WS_2-based supercapacitors demonstrate good energy-power balance with energy densities around 10-15 Wh kg^{-1} and power densities exceeding 1000 W kg^{-1}. While WS_2 electrodes typically show lower energy density than MoS_2, their superior cycling stability makes them attractive for applications requiring long operational lifetimes [15].

Cr_2S_3-based composites exhibit the highest energy density among the three metal sulfides, with Mn-doped Cr_2S_3/rGO achieving 121.91 Wh kg^{-1} at power density of 1040 W kg^{-1}. This exceptional energy density approaches lithium-ion battery performance while maintaining supercapacitor-level power density. However, achieving such performance requires careful compositional design and incorporation of conductive substrates [19].

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Asymmetric supercapacitor configurations, combining metal sulfide positive electrodes with carbon-based negative electrodes, can significantly enhance device voltage window and energy density. MoS_2/activated carbon asymmetric devices operating at 1.6-1.8 V demonstrated energy densities exceeding 30 Wh kg^{-1}. WS_2/graphene asymmetric supercapacitors achieved stable operation at elevated voltages with improved energy storage capability [20].

V. CONCLUSION AND FUTURE WORK

This comparative study has systematically analyzed the supercapacitive properties of nanostructured molybdenum, tungsten, and chromium sulfides, revealing distinct advantages and limitations of each material. MoS_2 demonstrates the highest intrinsic specific capacitance (395-576 F g^{-1}) among pristine metal sulfides, attributed to its rich redox chemistry, layered structure facilitating ion intercalation, and high density of active edge sites. WS_2 exhibits superior cycling stability (>93% retention after 5000 cycles) and lower charge transfer resistance, making it attractive for applications requiring long operational lifetimes despite somewhat lower capacitance. Cr_2S_3-based composites achieve exceptional performance (886.7 F g^{-1}, 121.91 Wh kg^{-1}) when appropriately engineered with conductive substrates and dopants, though environmental considerations require careful attention. All three metal sulfides exhibit hybrid charge storage mechanisms combining electrical double-layer capacitance and pseudocapacitance through reversible Faradaic redox reactions. Their layered crystal structures facilitate efficient ion intercalation and provide high surface areas for electrochemical reactions. Morphology critically influences performance, with three-dimensional hierarchical nanostructures (nanoflowers, nanosheets arrays) consistently outperforming bulk materials.

Composite formation with conductive matrices (graphene, carbon nanotubes, conducting polymers) significantly enhances performance by improving electrical conductivity, preventing restacking, and providing mechanical stability. Asymmetric device configurations enable wider voltage windows and higher energy densities, approaching battery-level performance while maintaining supercapacitor power characteristics. The findings of this study provide fundamental insights into structure-property relationships and offer guidance for rational design of next-generation supercapacitor electrodes. Continued research addressing challenges of conductivity, cycling stability, and scalable synthesis will facilitate practical implementation of these promising materials in commercial energy storage systems.

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