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RESEARCH

Synergistic integration of MWCNT-decorated $MnFe_2O_4$/MoS_2 composite electrode for high-performance asymmetric supercapacitors

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Abstract
Asymmetric supercapacitors (ASCs) constructed from metal oxide–metal sulfide–carbon composites have been identified as high-performance energy storage devices due to their ability to combine fast redox kinetics with excellent electrical conductivity and structural stability. In the present study, a ternary $MnFe_2O_4$/MoS_2@MWCNT composite was synthesized via a hydrothermal-ultrasonication route and employed as the positive electrode material. The $MnFe_2O_4$ spinel oxide contributes rich redox activity, MoS_2 offers layered architecture for enhanced ion diffusion, and MWCNTs act as a conductive matrix that improves electron transport and mechanical integrity. Activated carbon (AC), with its enhanced surface area and porosity, was employed as the anode. The resulting ASC cell delivered a high energy density of 47.5 Wh kg^{-1} at a power density of 750 W kg^{-1} and demonstrated stable performance over extended cycles stability with maintaining 95.9% capacitance after 10,000 cycles. This study highlights the potential of designing hybrid electrode systems by integrating transition metal oxides, sulfides and carbonaceous materials for next-generation supercapacitor applications.

Keywords $MnFe_2O_4$ · Energy density · Electrochemical performance · Hydrothermal · Asymmetric supercapacitor · Energy storage device

Introduction

In recent years, the rapid pace of technological innovation aimed at enhancing human comfort has led to a significant increase in global energy consumption. This growing demand places considerable pressure on the world's limited fossil fuel reserves, highlighting the urgent need for eco-friendly, efficient, and reliable energy storage technologies [1, 2]. Consequently, there has been increasing interest in developing advanced power storage devices that are not only environmentally sustainable but also lightweight, portable, and capable of delivering energy quickly and consistently

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[3–5]. Among the various energy storage options, supercapacitors have emerged as a highly promising technology due to their exceptional power density, fast electrochemical response, and long operational life. These devices are generally classified into two main types based on their charge storage mechanisms: electrochemical double-layer capacitors (EDLCs) and pseudocapacitors (PCs). EDLCs store energy via physical adsorption and desorption of ions at the electrode–electrolyte interface, whereas pseudocapacitors rely on rapid Faradaic redox reactions of electrochemically active materials [6–8].

While EDLCs typically offer longer cycle life, they often fall short in terms of specific capacitance when compared to pseudocapacitors. To overcome this limitation, hybrid systems that combine features of both types have been developed, offering a balance between high energy output and sustained electrochemical durability [9, 10]. In this context, MFe_2O_4-type spinel ferrites, where M corresponds to divalent transition metal ions such as Mn^{2+}, Co^{2+}, Ni^{2+}, Zn^{2+}, or Cu^{2+} have gained considerable attention as pseudocapacitor electrode materials. Their ability to undergo multiple redox transitions and maintain electrochemical stability makes them well-suited for modern power storage device [11, 12]. $MnFe_2O_4$ (MFO), a bimetallic oxide, has been widely studied for its eco-friendly nature and its ability to undergo multiple oxidation states, which promote efficient electrochemical activity. Manganese ferrite ($MnFe_2O_4$) is widely considered a promising pseudocapacitive material attributable to its chemical and physical properties. Its spinel structure offers excellent structural stability, while the presence of Mn^{2+}/Mn^{3+} and Fe^{2+}/Fe^{3+} redox couples facilitates fast Faradaic reactions [13]. $MnFe_2O_4$ also exhibits good electrical conductivity, moderate band gap, high theoretical capacitance, and strong magnetic and thermal stability. Additionally, it is low-cost, environmentally benign, and abundant, making it ideal for scalable and sustainable advanced storage architectures [14, 15]. In earlier studies, Z. Ansari et al. [16] reported that $MnFe_2O_4$ nanofibers delivered a specific capacitance (Cs) of 365 F g^{-1} at 1 A g^{-1}, along with notable long-term stability. Wenjuan Yang et al. [17] demonstrated that $MnFe_2O_4$ can be employed in symmetric supercapacitors operating over an extended potential range of 1.6 V, achieving a capacitance of 0.92 F cm^{-2} at 3 mA cm^{-2}. Rushiraj P. Bhosale et al. [18] observed that the optimized $MnFe_2O_4$ showed Cs of 227 F g^{-1} at 2 A g^{-1}. Despite the redox-rich nature and high theoretical capacitance of $MnFe_2O_4$, its charge storage performance is still hindered by limited conductive behaviour and sluggish ion diffusion.

To overcome these challenges, introducing MoS_2 and MWCNTs into the composite structure offers a highly effective strategy. MoS_2 contributes additional pseudocapacitive behaviour and a large surface area due to its layered structure, which provides abundant active sites and facilitates fast ion intercalation [19, 20]. For instance, Samira Sharifi et al. [21] developed a $CoFe_2O_4/MoS_2$ composite exhibiting a remarkable enhancement in Cs from 500 to 1013 F g^{-1}. Similarly, Pratik V. Sinde et al. [22] reported that a $MoS_2/NiFe_2O_4$ hybrid delivered 246.68 F g^{-1} at 0.5 A g^{-1}. In another study, Zeyad M. Abdulhamid et al. [23] demonstrated that MoS_2–$ZnFe_2O_4$ nanocomposites achieved an impressive 2077 F g^{-1} at 25 A g^{-1}. These observations highlight the interactive contribution of MoS_2 and various ferrite materials, confirming the viability of integrating MoS_2 with $MnFe_2O_4$ for significantly enhanced supercapacitor performance. However, while MoS_2 enhances pseudocapacitance and ion transport, its intrinsic poor electronic conductivity can still limit overall efficiency. To overcome this limitation, scientific efforts have focused on developed composites by integrating $MnFe_2O_4$ nanoparticles with conductive carbon framework like graphene and carbon nanotubes (CNTs) [24, 25]. These carbon additives not only improve electron transport but also help in tailoring the morphology of the composite, such as forming porous networks, increasing active surface sites and enhancing the accessibility of electroactive sites.

For example, Najmeh Foroutan et al. [26] showed that $MnFe_2O_4$/GO delivered 298 F g^{-1} at 1 A g^{-1} with stable cycling behaviour. K. Manimegala et al. [27] also revealed that $ZnCo_2O_4/MoS_2$@rGO attained a high Cs of 1764 F g^{-1} at 1 A g^{-1}. Kun-Yauh Shih et al. [28] prepared $MnFe_2O_4$/rGO hybrid material, which delivered a Cs of 196.6 F g^{-1} at 0.5 A g^{-1}. Jing Ran et al. [29] observed that $NiCo_2O_4$@MoS_2/RGO exhibited a Cs of 946 F g^{-1} at 1 A g^{-1}. Hamza Nawaz et al. [30] demonstrated that the $(MnFe_2O_4)_{0.60}$–$(CNTs)_{0.40}$ material achieved a high specific capacitance of 652 F g^{-1}. To address this, multi-walled carbon nanotubes (MWCNTs) are introduced as a conductive supportive network to facilitate rapid electron transport across the electrode. Their high aspect ratio, excellent electrical conductivity, and structural flexibility support effective charge percolation and mechanical stability of the composite [31, 32]. Furthermore, the integration of MWCNTs prevents agglomeration of both $MnFe_2O_4$ and MoS_2, enabling uniform dispersion and enhancing electrochemical utilization.

Therefore, the rational design of $MnFe_2O_4/MoS_2$@MWCNT composites integrates the redox activity of $MnFe_2O_4$, the layered structure and pseudocapacitance of MoS_2, and the conductive scaffold of MWCNTs to achieve enhanced electrochemical performance. In this work, we report the hydrothermal synthesis of morphology-controlled $MnFe_2O_4/MoS_2$@MWCNT composites, where $MnFe_2O_4$ forms nanospherical architectures anchored onto MWCNTs and coated with MoS_2 nanosheets, providing high surface area, efficient ion transport, and excellent structural

Table 2 Energy and power density comparison of the present device with previous reports for Ragone plot representation

S. No	Electrode	Energy density	Power density	Reference
1	$MnCo_2O_4/Ti_3C_2Tx$	40 Wh kg^{-1}	4828 W kg^{-1}	[57]
2	$ZnMn_2O_4$/RGO	40.2 Wh kg^{-1}	1125 W kg^{-1}	[58]
3	Gd-$ZnFe_2O_4$/CNT	40.025 Wh Kg^{-1}	279.78 W Kg^{-1}	[59]
4	$NiCo_2S_4/MoS_2$/MWCNT	23.73 Wh/kg	749.5 W kg^{-1}	[60]
5	$MnNi_2O_4$/PPY	35.9 W h kg^{-1}	802.9 W kg^{-1}	[61]
6	P-CNT@$MnCo_2O_4/Co_3O_4$	46.4 Wh kg^{-1}	800 W kg^{-1}	[62]
7	$MnFe_2O_4/MoS_2$@MWCNT	47.5 Wh kg^{-1}	750 W kg^{-1}	This Work

high energy density of 47.5 Wh kg^{-1} at a power density of 750 W kg^{-1}. Table 2 summarizes the energy and power density values of the present work in comparison with previously reported materials, as represented in the Ragone plot [57–62]. The $MnFe_2O_4/MoS_2$@MWCNT device demonstrates superior performance, delivering both higher energy and power densities than earlier studies. This enhanced performance arises from the complementary integration of redox-active $MnFe_2O_4$ and MoS_2 with highly conductive MWCNTs, which together create a hierarchical porous framework conducive to fast ion transport, efficient electron transfer, and stable electrochemical cycling. In summary, the assembled ASC device demonstrates outstanding capacitive performance, electrochemical characteristics and combining high energy and power densities. These electrochemical attributes make the $MnFe_2O_4/MoS_2$@MWCNT composite a promising candidate for high-performance next-generation hybrid capacitors.

Conclusion

In this study, a $MnFe_2O_4/MoS_2$@MWCNT composite electrode was successfully prepared via a simple hydrothermal technique followed by ultrasonication. The hybrid structure integrates the high redox activity of spinel-type $MnFe_2O_4$, the layered architecture and pseudocapacitive behaviour of MoS_2, and the excellent conductivity of MWCNTs. This well-designed composite offers abundant active sites, efficient ion diffusion, and rapid electron transport. Assembled into an asymmetric supercapacitor achieved a high energy density of 47.5 Wh kg^{-1} at a power density of 750 W kg^{-1}. It also exhibited excellent cycling stability, retaining 95.9 % of its capacitance over 10,000 cycles. Electrochemical impedance studies confirmed low internal and charge transfer resistance, both before and after long-term cycling. These findings highlight the synergistic interaction among the metal oxide, metal sulfide, and carbon components, demonstrating the $MnFe_2O_4/MoS_2$@MWCNT composite as a strong candidate for advanced energy storage systems.

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Data availability The data that support the findings of this study are available from the corresponding author, upon reasonable request.

Declarations

Ethical approval The authors declare that there are no human cells and tissues are used in this manuscript.

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