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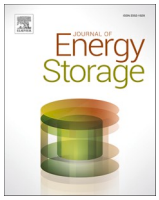

Research papers

Zinc gallium oxide/g-CN nanocomposites as emerging electrode materials for asymmetric supercapacitors: Insights into their enhanced charge storage behaviour

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ABSTRACT

The development of hybrid nanostructured electrodes has emerged as an effective approach to address the inherent limitations of individual electrode materials in supercapacitors. Hybrid nanocomposites integrating $ZnGa_2O_4$ with graphitic carbon nitride (g-CN) are developed to enhance the electrochemical performance of supercapacitor electrodes. Comprehensive structural and surface characterizations (XRD, FT-IR, FE-SEM, TEM, XPS and BET) elucidate the phase purity, crystallinity, functional groups, surface morphology, oxidation states and porosity of the synthesized materials, which thereby confirms the successful integration of g-CN with $ZnGa_2O_4$ in the prepared materials. Electrochemical studies infer that the fabricated g-CN@$ZnGa_2O_4$ (1.5:1) electrode delivers the highest specific capacitance of about 594.0 F g^{-1}, significantly outperforming $ZnGa_2O_4$ and g-CN@$ZnGa_2O_4$ (1:1) electrodes. This enhanced charge-storage behaviour is due to reduced charge-transfer resistance, improved electronic transport, and facilitated ion diffusion arising from effective interfacial interaction between $ZnGa_2O_4$ and g-CN. Furthermore, the asymmetric supercapacitor device assembled using the fabricated g-CN@$ZnGa_2O_4$ (1.5:1) electrode delivers specific capacitance of about 85.33 F g^{-1} with excellent cyclic stability, retaining 69.0% of its initial capacitance after 5000 charge–discharge cycles at 5 A g^{-1}. It also exhibits coulombic efficiency of about 96.0%, energy and power densities of about 26.66 Wh kg^{-1} and 750.0 W kg^{-1} respectively. These results demonstrate that g-CN incorporation represents an effective strategy for engineering $ZnGa_2O_4$-based composite electrodes with enhanced charge storage capability, positioning the developed materials as promising candidates for advanced supercapacitor systems.

1. Introduction

The transition toward sustainable and high-efficiency energy technologies have become imperative owing to the rapid scarcity of fossil energy resources, escalating environmental pollution, and the accelerating impacts of global warming. Developing new concepts and innovative routes for solving energy crisis is the most essential need to mitigate the challenges and secure a more sustainable future. [1–3] In this context, supercapacitors have attracted considerable attention owing to their superior power density compared to batteries, higher energy density than conventional dielectric capacitors, excellent cyclic stability and reliable performance. These attributes enable their widespread applications in power electronics, computer memory backup systems, large-scale transportation systems such as subway trains and buses, energy storage for intermittent power sources including wind turbines, and smart grid technologies. [4] The efficiency of a supercapacitor is primarily determined by the electrode material, electrolyte type, and device configuration. Among them, the electrode material

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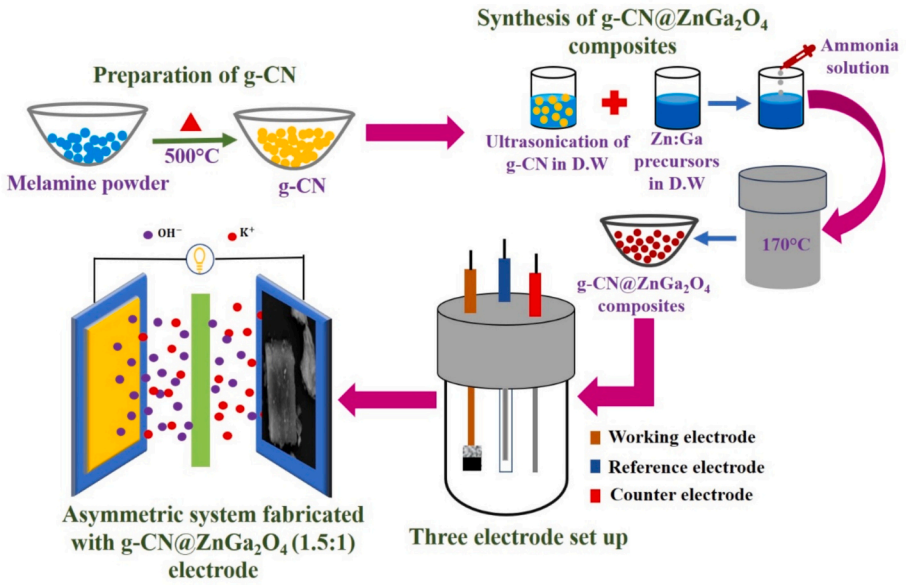

Fig. 1. Schematic illustration of g-CN@$ZnGa_2O_4$ nanocomposites as efficient electrodes for asymmetric supercapacitor application.

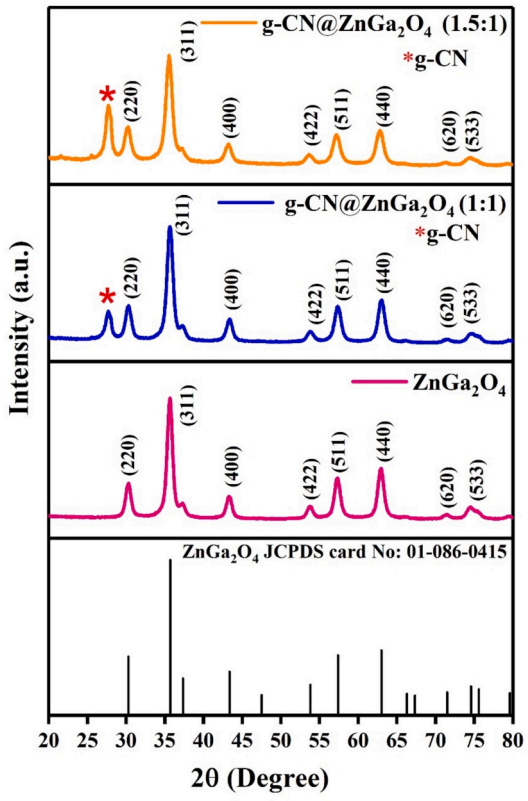

Fig. 2. Comparative XRD patterns of $ZnGa_2O_4$, g-CN@$ZnGa_2O_4$ (1:1) and g-CN@$ZnGa_2O_4$ (1.5:1) composites.

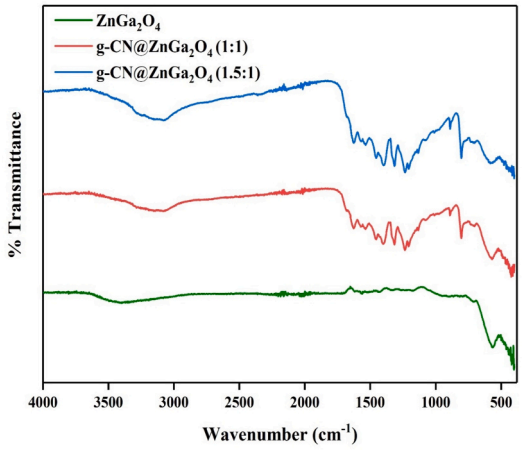

Fig. 3. Comparative FTIR spectra of $ZnGa_2O_4$, g-CN@$ZnGa_2O_4$ (1:1) and g-CN@$ZnGa_2O_4$ (1.5:1) composites.

plays a pivotal role in the performance of the supercapacitor. [5] Supercapacitor electrodes should ideally exhibit key characteristics such as high-power output, rapid charge-discharge kinetics, and extended cycle life, making them highly desirable for advanced energy storage applications. [6] Till date, a wide range of materials, including carbon nanomaterials, conductive polymers, and various transition-metal oxides, have been studied to assess their suitability for electrode material toward supercapacitor application. [7]

Transition-metal oxides (TMOs) have become increasingly significant in modern technology owing to their diverse physical, chemical, and optical properties. The presence of multiple oxidation states in transition metal oxides (TMOs) promotes efficient redox activity and tunable electronic properties, resulting in high specific capacitance and multifunctionality in energy storage applications. Recently, spinel-type binary metal oxides with the general formula $A^{2+}B_2^{3+}O_4$, [8] where A represents a divalent cation (such as Zn, Ni, Mn, or Cu) and B denotes trivalent cations (such as Co, Fe, Ga or Mn), have attracted considerable attention as promising electrode materials for supercapacitor application. [9,10] Their enhanced electrochemical performance originates from the mutual electronic interaction between the two metal cations, which optimizes charge transfer and redox activity. [11,12] The mixed oxide spinel structures feature highly adaptable octahedral and tetrahedral sites within their closely-packed oxygen frameworks, leading to the formation of tunable crystal structures. This flexibility allows for numerous possibilities in exploring new cation combinations with multiple oxidation states, which can be incorporated into these structures to enhance storage capacity. Various gallium-based ternary spinel oxides (MGa_2O_4), such as $ZnGa_2O_4$ and $CdGa_2O_4$, are used as wide-

F g^{-1}. Galvanostatic charge–discharge tests of the ASC device were conducted using a constant current calculated from the selected current density based on the total active mass of both electrodes ($m_+ + m_-$). Accordingly, all ASC electrochemical parameters, including specific capacitance, energy density, and power density, are normalized with respect to the total mass of active materials in both electrodes. Although the discharge voltage window includes the initial iR drop, its magnitude is small relative to the overall operating voltage range; therefore, its impact on the calculated capacitance, energy density, and power density is considered to be less.

The energy and power density of the fabricated ASC device are determined from the eqs. (8) and (9) respectively.

$$Energy\ density\ (E) = \frac{C \times \Delta V^2}{7.2}\ \left(Wh\,kg^{-1}\right) \tag{8}$$

$$Power\ density\ (P) = \frac{3600\,E}{\Delta t}\ \left(W\,kg^{-1}\right) \tag{9}$$

where C (F g^{-1}) is the specific capacitance, ΔV denotes the potential range (V), and Δt represents the discharge time (t). [53] The assembled ASC device exhibits energy density of 26.66 Wh kg^{-1} along with maximum power density of about 750.00 W kg^{-1}. Cyclic stability is an important factor for the prolonged safety and durability of the supercapacitor device and can be evaluated through continuous charge-discharge cycles. Fig. 19 (d) illustrates the cyclic stability as well as the coulombic efficiency of the as-prepared g-CN@$ZnGa_2O_4$ (1.5:1) electrode. The electrode retains 69.0% of the original capacitance value even after 5000 cycles and a coulombic efficiency of about 96.0% at a current density of 5 A g^{-1}. The enhanced cyclic stability of the asymmetric supercapacitor (ASC) is attributed to the combination of the active material with a complementary electrode and the optimization of the electrolyte–electrode interface. This improves structural integrity, electronic conductivity, and ion transport, resulting in the cycling retention of 69.0% after 5000 cycles. Hence, the high retention capability highlights the superior electrochemical durability and conductivity of fabricated g-CN@$ZnGa_2O_4$ (1.5:1) as potential electrode material for long-term supercapacitor application. Table 4 represents electrochemical performance parameters of g-CN@$ZnGa_2O_4$ (1.5:1) asymmetric device performed at 1.5 V for 1 Ag^{-1} - 5 A g^{-1} current densities. The g-CN@$ZnGa_2O_4$ (1.5:1) composite exhibits superior specific capacitance compared to that of $ZnGa_2O_4$ and g-CN@$ZnGa_2O_4$ (1:1) electrodes, thereby highlighting the combined electrochemical contribution of g-CN in improving charge storage capabilities. Table 5 provides the comparison of the fabricated ASC system with the previous reports and the corresponding Ragone plot is shown in Fig. 20. The incorporation of g-CN enhances the electrical conductivity and facilitates efficient electron transport, leading to better electrochemical performance.

To further evaluate the structural stability, XPS analysis of g-CN@$ZnGa_2O_4$ (1.5:1) composite after electrochemical performance is carried out and is shown in Fig. 21. The survey spectrum confirms the presence of all constituent elements, including Zn, Ga, O, C, and N, indicating that the chemical composition of the material is well preserved after prolonged charge–discharge processes. A slight shift in Zn 2p spectra after cycling suggest changes in the local electronic environment of Zn, indicating its involvement in surface redox processes. No significant changes in the elemental composition were observed, suggesting good structural integrity and stability of the electrode during electrochemical operation. Hence, with its enhanced energy density, power density, extended cyclic stability, and superior charge storage properties, g-CN@$ZnGa_2O_4$ based composites emerge as a potent next generation electrode materials for supercapacitor application.

4. Conclusion

In this work, $ZnGa_2O_4$ and g-CN@$ZnGa_2O_4$ (1:1) and g-CN@$ZnGa_2O_4$ (1.5:1) nanocomposites were successfully synthesized through the hydrothermal route and evaluated as electrode materials for asymmetric supercapacitors. Structural characterization verified the formation of phase-pure cubic $ZnGa_2O_4$ and its effective integration with g-CN. Among the fabricated electrodes, the g-CN@$ZnGa_2O_4$ (1.5:1) electrode exhibited superior electrochemical performance, delivering the highest specific capacitance of about 594.0 F g^{-1}. The improved capacitance arises from reduced charge-transfer resistance (EIS), increased redox current (CV), and prolonged discharge time (GCD), indicating efficient electron transport and enhanced ion diffusion facilitated by the g-CN framework. The assembled asymmetric supercapacitor device demonstrated cyclic stability with 69.0% capacitance retention after 5000 cycles and a coulombic efficiency of 96.0%. Furthermore, the assembled asymmetric supercapacitor delivered an energy density of 26.66 Wh kg^{-1} and power density of about 750 W kg^{-1}. These results demonstrate that the incorporation of g-CN effectively enhances the electrochemical performance of $ZnGa_2O_4$-based electrodes through interfacial interaction and improved charge transport. Thus, the findings provide a foundation for the development of efficient hybrid electrode materials for next-generation energy storage technologies.

CRediT authorship contribution statement

Govarthini Seerangan Selvam: Writing – original draft, Methodology, Data curation. **Gunasekaran Subramani:** Writing – review & editing. **Muthukaruppan Alagar:** Writing – review & editing. **Mohammed Mujahid Alam:** Funding acquisition. **Mohamed Hussien:** Funding acquisition. **Thangaraju Dheivasigamani:** Writing – review & editing, Supervision, Methodology, Investigation, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.est.2026.122188.

Data availability

Data will be made available on request.

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