

Hybrids of porous NiMn₂O₄@reduced graphene **oxide composites for asymmetric supercapacitor applications**

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ABSTRACT

A distinctive hierarchical hybrid nanostructure which consists of ultrathin and vertical NiMn₂O₄ nanosheet grown on reduced graphene oxide (NiMn₂O₄ NSs[®]) rGO) is synthesised by a facile hydrothermal procedure. The surface morphologies, structure, and elemental composition of the materials are examined by XRD, Raman spectroscopy, FESEM, and HRTEM and XPS. The active materials ($NiMn₂O₄@rGO$) are coated on nickel foam electrode to test the capacitance activity from cyclic voltammetry (CV), galvanostatic charge–discharge (GCD), and electrochemical impedance spectra (EIS) techniques with 1.0 M KOH. The specific capacitance of NiMn₂O₄@rGO is 1891 Fg⁻¹ at 1 Ag⁻¹. After 5000 cycles, the specific capacitance retention rate of NiMn₂O₄@rGO is 80% at 20 Ag⁻¹. Besides, the assembled NiMn₂O₄@rGO//AC asymmetric supercapacitor (ASC) displays the maximum energy density of 49.8 Whkg⁻¹ at a power density of 880.5 Wkg⁻¹. Signifcantly, an ultralong cycling life of 98% capacitance retention is achieved for the ASC device after 5000 charge/discharge cycles at 2 Ag^{-1} . These results showed that rGO addition has increased the conductivity of NiMn_2O_4 , and thus it can be suggested as a new technique to improve the capacitance. Moreover, the prominent cycling stability and high energy density indicate that the composites possess potential application as excellent electrode material.

1 Introduction

Since the world's fuel reserves are dwindling and pollution levels are rising to an all-time high, scientists have been compelled to explore novel methods of harnessing alternative energy. However, their intermitent nature in terms of energy production still limits their dominance over conventional coal and oil and gas energy sources, despite the fact that this energy has the potential to mitigate the major energy problem

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in the world. In this prospect an efficient energy storage device can be savior. Currently, scientists all over the world are putting in place various efforts to create energy storage systems such as the Na-ion batery and fuel cells. Due to their rapid charging−discharging cycle, large power density, modest energy density, and long cycle life [\[1](#page-9-0), [2\]](#page-9-1), supercapacitors are garnering a lot of interest. Carbon-based electrodes are used in commercial supercapacitors because of their low contact resistance and high surface area, both of which contribute to the device's high performance [[3](#page-9-2)[–5\]](#page-9-3). Recently, it has been discovered that mixed transition metal oxides (MTMOs), an emerging category of announcing electrode materials for a variety of energy-related uses, are produced by the sophisticated synthesis of two inexpensive transition metal oxides, resulting in the creation of single-phase MTMOs with higher electric and electrochemical characteristics than the single part. For example, CoFe_2O_4 , MnFe_2O_4 , $NiCo₂O₄$, MnCo₂O₄, CoMn₂O₄, and NiMn₂O₄ have been utilised as the active electrode materials for supercapacitor applications [[6](#page-9-4)[–8](#page-9-5)].

Among these, $NiMn₂O₄$ stands out for having a special spinel crystal structure and having outstanding electrochemical properties due to its many benefts such as lower cost, sustainability, and ease of preparation. There have been some advancements made using $NiMn_2O_4$ materials. $NiMn_2O_4$ with a mesoporous structure has been described by Miao Zhang and colleagues [[9\]](#page-9-6), $NiMn₂O₄@CNT$ nanocomposite have been created by Nan's groups [[10](#page-9-7)], and K. Vijaya Sankar's groups have looked into the electrochemical intercalation/de-intercalation process of it for supercapacitors [[11\]](#page-9-8). Similar to other transition metal oxides, nevertheless, its uses are constrained by its low particular area of surface, poor dispersion, and generally poor conductivity [[12\]](#page-9-9). We focus on integrating promising electrode materials with carbon materials in order to address the aforementioned limitations. An essential form of carbon known as graphene has a hexagonal 2D layer structure made up of sp2 carbon atoms [[13\]](#page-9-10). Graphene is a promising candidate for hybrid hierarchical nanostructures composites with TMO because of its unusual structural characteristics, which include a large surface area, exceptional electricity conductivity, excellent

chemical substances and thermal stability, and wide potential window [[14\]](#page-9-11). The development of graphene/TMO hybrid materials has received a lot of atention recently, and as a result, such substances perform electrochemically beter than their component parts individually. By virtue of its oxygen-containing groups, graphene provides a 2D foundation for the construction of conductive networks, inhibits the volume fuctuation and aggregation of TMO, and assures reliable electrical connections between graphene and TMO [[15](#page-9-12), [16](#page-9-13)]. Although there have only been a few papers on $NiMn_2O_4$ graphene composite materials for super-capacitors, it is safe to assume that these materials will have improved electrochemical performance. Thi Ngo [[17](#page-9-14)] designed a 3D structure with excellent capacitance (396.85 Fg^{-1}) and very high glucose sensitivity (1310.8 µAmM⁻¹ cm⁻²), all with a quick reaction time of 3.5 s. Energy density of 60.69 Whkg⁻¹ and high capacitance of 151 Fg⁻¹ were achieved for CNT@NiMn₂O₄ nanocomposites [[10](#page-9-7)]. The specific capacitance of 30 wt% rGO/NiMn₂O₄ nanorods composites was measured at 693.00 Fg^{-1} [[18\]](#page-9-15). Here, we use a simple technique to synthesise $NiMn₂O₄@rGO composites.$ The mellow, simple, and inexpensive method used to prepare the product shows promising electrocatalytic activity and chemical inertness. Results from an electrochemical study showed that the composites have a specific capacitance of 693.00 Fg^{-1} , which is excellent for supercapacitors, and a notable cycling stability. Power density was 10 kWkg⁻¹, and energy density was 43 Whkg $^{-1}$, both of which are extremely impressive for a man-made product. Additionally, at 1 Ag^{-1} , the ASC device maintained 99% coulombic efficiency while retaining 90% of its capacitance after 10,000 cycles.

2 Materials and methods

2.1 Reactants and materials

Manganese nitrate ($Mn(NO₃)₂·6H₂O$, purity 99.99%), nickel nitrate (Ni $(NO₃)₂·6H₂O$, purity 99.99%), and sodium hydroxide (NaOH) were obtained from Adamas Reagent Co., Ltd. Graphene oxide slurry (LN-10R, purity 99.99%) was purchased from Levson Co., Ltd. AC (Activated Carbon, purity 99.99%) was purchased from Fuzhou Yihuan Carbon Co., Ltd. Nickel foam was obtained from Shanxi Lizhiyuan Batery Material Co., Ltd. All chemicals were used as received without any further purifcation.

2.2 Synthesis of $NIMn_2O_4$ and $NIMn_2O_4@rGO$ **composites**

Graphene oxide (GO) was synthesised from graphite powder using a reconfgured Hummers method [[19](#page-9-16)]. To reduce rGO from GO, 30 ml of GO aqueous suspension (2 mg/ml) were dissolved in 120 ml of ethanol with ultrasonic stirring for 20 min. The above solution was used to dissolve and stir 1 mmol of $Ni(NO₃)₂·6H₂O$ and 2 mmol of $Mn(NO₃)₂·4H₂O$. The solution was mixed for 20 min under strong magnetic stirring. The solution was then dripped with 2 mL of NaOH. The remedy was collected and placed in a 100 mL reactor before being heated to 160 °C in an oven for 12 h. The eventual results reactant was freeze-dried after being washed multiple times with deionised water and ethanol. The resulting $\text{NiMn}_2\text{O}_4/\text{rGO}$ powders underwent a two-hour calcination process at 500 °C (heating rate 0.5 °C min⁻¹). Pure NiMn₂O₄ was prepared by following the same procedure except the addition of GO. The ratio between GO and NiMn_2O_4 was 1:3.

2.3 Electrode preparation

NiMn₂O₄@rGO active material flour, conducting additive (acetylene black), and binder (polyvinylidene fuoride) were mechanically mixed together in a mass ratio of 80:10:10 to create the working electrodes. Pt wire and Ag/AgCl electrode were used as counter and reference electrodes, respectively. The powder was then placed onto a piece of nicked foil current collector after being soaked in *N*-methyl pyrrolidone to create a homogenous slurry. An area of 1 cm^2 was loaded with 1.0–2.0 mg of the prepared sample on nicked foil. Cyclic voltammetry (CV), galvanostatic charge/discharge (GCD), and electrochemical impedance spectroscopy (EIS) measurements were performed on a CH Instrument Ins to compare electrochemical outcomes with a typical three system in electrolyte solution (2 M KOH). An electrochemical workstation, model CHI 660e.

3 Results and discussion

3.1 Powder XRD analysis

X-ray powder difraction (XRD) were used to analyse the composition and structure of the as-obtained samples on a PANalytical B.V. X'Pert PRO MPD using a CuKa radiation source (l = 1.54056 Å) worked at 40 kV and 40 mA. Figure [1](#page-2-0), shows the XRD patterns of GO, rGO, NiMn₂O₄, and NiMn₂O₄@rGO composites. An oxygen substituent is present seen between layers of GO, as evidenced by the characteristic peaks at $2\theta = 10.5^{\circ}$, which coincides with the (002). The disappearance of this peak in the rGO patern is indicative of a decrease in oxygen groups as a result of the hydrothermal treatment. The diffraction peaks at 18.4°, 30.3°, 35.4°, 42.9°, 57.1°, and 63.1° were found to be associated with the (111) , (220), (311), (400), (511), and (440) difraction planes, respectively. This may be an indication of $NiMn₂O₄$ formation (JCPDS no. 01-1110) [\[20\]](#page-9-17). Weak peaks for the rGO and $NiMn₂O₄$ aspects can be seen in the XRD pattern of NiMn₂O₄@rGO, suggesting the existence of both phases in the nanocomposite.

3.2 Morphological analysis

FESEM was used to analyse the nanostructures and morphologies of NiMn_2O_4 NSs@rGO nanocomposites and $NiMn₂O₄$ $NiMn₂O₄$ $NiMn₂O₄$ aggregates. Figure 2a shows that $NiMn₂O₄$ ultrathin nanosheets are uniformly distributed throughout the rGO supporting surface to create

Fig. 1 XRD pattern of the electrode samples **a** GO; **b** rGO; **c** NiMn₂O₄ $@rGO$

Fig. 2 a and **b** FESEM images of NiMn2O4@rGO with different magnifications; **c** and **d** FESEM image of NiMn₂O₄; **e** TEM image of $NiMn₂O₄@rGO$ and **f** $NiMn₂O₄$; Elemental mapping images of **g** Ni; **h** Mn; **i** O; and **j** C

 $NiMn₂O₄ NSs@rGO. Magnification is further increased$ to show that the $NiMn₂O₄$ nanosheets are formed perpendicular on the rGO substrate surface, resulting in the cross-linked network structure seen in Fig. [2](#page-3-0)b. As shown in Fig. [2c](#page-3-0), if there is no rGO supporting surface, $NiMn₂O₄$ nanosheets form spherical formations with a diameter of around 1.5 m when they come together. Figure [2](#page-3-0)d displays the microsphere's magnifed picture. TEM was used to do more research on the materials' structural traits and morphological specifcs. The NiMn₂O₄ nanosheets seen in Fig. [2e](#page-3-0) are growing randomly and vertically over rGO without any set shape, and they are around 3.2 nm in thickness. The $NiMn₂O₄$ nanosheets are made up of smaller nanoparticles and form a cross-linked network structure, as shown by Fig. [2](#page-3-0)f as well. The data shown above agree with those from the FESEM. The elemental mapping of $NiMn₂O₄@rGO$ was tested and the resultant images are shown in Fig. [2](#page-3-0)g–j. The elements are Ni, Mn, O, and C and are found uniformly dispersed on the composite sample.

3.3 Raman spectra analysis

Raman spectra of rGO, NiMn₂O₄, and NiMn₂O₄@rGO composites are shown in Fig. [3](#page-4-0). Two distinct peaks, at 1339 cm⁻¹ and 1584 cm⁻¹, were found in rGO Raman spectra, which correlate to the D and G bands of rGO, respectively [[21,](#page-9-18) [22](#page-10-0)]. Assigning the broad peaks at 681 cm⁻¹ wavenumber to stretching vibrations of NiMn₂O₄ cubic spinel phases has been demonstrated [[23](#page-10-1)]. The Raman spectrum of the sample $NiMn₂O₄@$ rGO clearly shows that the distinctive bands of rGO and NiMn_2O_4 coexist, showing that they are well mixed, which is consistent with the above-mentioned XRD data. Additionally, the intensity ratio of D to G

Fig. 3 Raman spectra of the electrode samples

(ID/IG) for rGO and $NiMn₂O₄@rGO$ was around 1.08 and 1.13, respectively, indicating the presence of an interaction between rGO and $NiMn_2O_4$.

3.4 BET and XPS analysis

 $N₂$ sorption experiments were used to examine the $NiMn₂O₄$ and $NiMn₂O₄@rGO$ materials' tribological qualities (Fig. [4](#page-4-1)a). Type IV isotherms, typical of compounds [\[24](#page-10-2)[–28](#page-10-3)], can be applied to all of the aforementioned substances. It was determined that $NiMn₂O₄$ had an estimated SBET of 39.2 m 2 /g, while NiMn $_2\mathrm{O}_4$ @ rGO composites had an SBET of $101.7 \text{ m}^2/\text{g}$. The pore diameter distribution curves (Fig. [4](#page-4-1)a inset) show that mesopores, with sizes ranging from 2 to 50 nm, are present in all of the samples and result from the uneven layering of rGO. GO and rGO's respective C 1s XPS spectra are shown in Fig. [4](#page-4-1)b. During the hydrothermal treatment, GO was depleted as shown by a reduction in the strength of the C–O/C=O peaks in the rGO sample compared to that of GO. Figure [4](#page-4-1)c shows the survey spectra for NiMn2O4 and NiMn₂O₄@rGO materials. $Ni^{2+} 2p_{1/2}$ and $2p_{3/2}$ (centred at 874.1 and 856.3 eV, respectively) [\[29](#page-10-4)] and Mn^{3+} 2 $p_{1/2}$ and 2 $p_{3/2}$ (centred at 653.2 and 641.7 eV, to between) [[30](#page-10-5)] are credited to the peaks in the $NiMn₂O₄$ $NiMn₂O₄$ $NiMn₂O₄$ sample (Fig. 4d

Fig. 4 a BET surface and pore size curves (inset) of $NIMn_2O_4$ and $NIMn_2O_4$ and G_4 composite samples; **b** C 1s; **c** survey; **d** Ni 2p; **e** Mn 2p; and **f** O 1s spectra of $NiMn₂O₄$ and $NiMn₂O₄@rGO$ samples

and e, respectively). Sample $NiMn₂O₄'s Ni 2p$ and Mn 2p peaks, on the other hand, shifted towards that lower binding energy when compared to $NiMn_2O_4$; this diference suggests that graphene materials can boost the conduction electrons density of surface Ni and Mn atoms. $NiMn₂O₄'s$ O 1s high-resolution XPS spectrum (shown in Fig. [4f](#page-4-1)) [[31](#page-10-6)] exhibits three ftted peaks, which conform to adsorbed H_2O (OA, at 533.2 eV), hydroxyl oxygen (OH, at 532.0 eV), and lattice oxygen (OL, at 530.5 eV).

3.5 Electrochemical performance three‑electrode system

CV tests are made on a three-electrode setup with a 2 M KOH aqueous electrolyte to assess the electrode materials' electrochemical behaviour. The redox peaks in the CV curves of $NiMn₂O₄$ and $NiMn₂O₄$ @rGO can be seen in Fig. [5a](#page-5-0), b, which display the materials at a range of scan rates (10–100 mVs⁻¹) and potential windows (0.0–0.6 V). These maximums show that the Faradic reaction plays a signifcant role in regu-lating the capacitive behaviour [\[32\]](#page-10-7). NiMn₂O₄@rGO NFs electrode shows an increased incorporated area underneath the CV curve due to the addition of rGO. This is a result of the synergistic response of Ni–Mn oxides and rGO, and it indicates an increase in capacity. NiMn₂O₄@rGO electrode has a higher specific capacity than bare NiMn₂O₄ (901 Fg⁻¹ at 10 mVs⁻¹) at 1221 Fg^{-1} (Fig. [5](#page-5-0)c). Linear reliance of peak cathodic and anodic current densities on scan rates is shown in Fig. [5](#page-5-0)d. Also, it proves that the synthesised electrode materials are hybrid supercapacitors. The surface coverage (Γ*) of redox species over the electrode samples are calculated based on the previous reported work [[33](#page-10-8)]: we determine the concentrations of various species over electrode samples. The NiMn₂O₄@ rGO electrode has a higher calculated value of linear reliance of peak cathodic and anodic current densities on scan rates is shown in 5d. Also, it proves that the synthesised electrode materials are hybrid supercapacitors. The surface coverage (Γ^*) of redox species over the electrode samples are calculated based on the previous reported work $[33]$ $[33]$: we determine the concentrations of various species over electrode samples. The $NiMn₂O₄@rGO$ electrode has a higher calculated value of (Γ^*) (8.1105 molcm⁻²) than the bare NiMn₂O₄ electrode. NiMn₂O₄@rGO electrode may have a greater value of (Γ^*) because of the interconnected and combined surface of numerous hairy

Fig. 5 Electrochemical performance of $NiMn₂O₄$ and $NiMn₂O₄@rGO composite$ electrodes CV curves of **a** NiMn₂O₄; **b** NiMn₂O₄@ rGO with various scan rates; **c** specifc capacitance values as a function of various scan rates; **d** cathodic and anodic peak current density of NiMn₂O₄ and NiMn₂O₄[@] rGO composite electrodes

needle-like porous structures of $NiMn₂O₄$ and large surface area of rGO sheets. GCD measurements are used to learn more about the electrochemical behaviour of the synthesised materials. As can be seen in Fig., GCD measurements performed in the range of 0.0 to +0.06 V vs. Ag/ AgCl yielded the expected results. At a current density of 2 A/g , as shown in Fig. [6a](#page-6-0), the charge/discharge time for the $NiMn₂O₄@rGO$ electrode is significantly longer than that of pure $NiMn_2O_4$. GCD curves of $NiMn_2O_4@rGO$ electrodes at varying current densities are displayed in Fig. [6b](#page-6-0). All GCD curves are found to be nearly symmetrical, which indicates a high coulombic efficiency of the electrode materials across a wide range of current densities. NiMn₂O₄@ rGO electrode Cs = 1848 Fg−1, 1801 Fg−1, 1767 Fg−1, 1701 Fg⁻¹, and 1687 Fg⁻¹ at 1, 2, 6, 10, and 20 Ag⁻¹ current densities, respectively (Fig. [6](#page-6-0)c). Even at 20 Ag⁻¹ current density, the electrode maintained up to 80% of its Cs (353 Fg^{-1}). It is an indication of increased capacitive retention of electrode materials. Cycle life estimates for the NiMn₂O₄ and NiMn₂O₄@rGO composite electrode are shown in Fig. [6d](#page-6-0), e, which depicts the results of a continuous charge–discharge test at 1 Ag^{-1} . After one thousand charge–discharge cycles, the hierarchical

nanosheet array NiMn₂O₄@rGO composite still maintained about 90% of its maximum capacitance, proving its long-term electrochemical stability even at high current densities. The improved electrochemical performance of the composite samples is due to the following reasons: (1) The contact of the rGO nanosheets and the $NiMn₂O₄$ nanosheets provides the good structural stability and mechanical resilience, and prevents the pulverisation of $NiMn_2O_4$, which is the cause of the better electrochemical performance of the NiMn₂O₄@ rGO. (2) The rGO surface's capacitance for the Faradaic redox response and the rates of ion difusion and transport of electrons are signifcantly increased by the composition of the composite NiMn₂O₄@rGO. (3) The synergetic effect between $NiMn₂O₄$ and rGO, which siginifcantly improve the electrochemical performance due to the high electrical conductivity by the formation of ultra thin sheet like morphology. In order to beter understand the interfacial properties of $NiMn₂O₄@rGO$, an EIS analysis was conducted on the mixture. As shown in Fig. $6(f)$ $6(f)$, the Nyquist plots of the NiMn₂O₄@rGO composite electrode are a semicircle at high frequency region due to the Rct at the electrode interface, and a low-frequency inclined line

Fig. 6 GCD profile of **a** NiMn₂O₄; **b** NiMn₂O₄@rGO with various current densities; **c** specific capacitance values as a function of various current density; retention curves of **d** NiMn₂O₄; **e** NiMn₂O₄@rGO; **f** EIS curve of NiMn₂O₄ and NiMn₂O₄@rGO composite samples

Fig. 7 a schematic representation of the fabricated $NiMnO₄@$ rGO//AC ASC device; **b** CV curves of NiMn₂O₄ and AC; **c** CV curves of ASC with diferent sweep rates; **d** GCD curve of ASC

(W: Warburg impedance) that ascribes to the difusion of ions in the electrolyte to the electrode interface. The EIS spectra can be fitted to obtain an estimate of the semicircle's diameter, Rct. The NiMn₂O₄@ rGO electrode's low Rct value of 6.5 Ω demonstrates the improved electrical conductivity and low electrochemical resistance.

3.6 Electrochemical performance of NiMn₂O₄@ rGO//AC in a two‑electrode system

An asymmetrical supercapacitor (ASC) was constructed in 2 M KOH aqueous electrolyte with $NiMn₂O₄/rGO$ and activated carbon (AC) as the positive and negative electrodes, including both, to investigate the practical viability of the $NiMn₂O₄/$ rGO composite electrode. Figure [7a](#page-7-0) is a schematic of the fnal ASC device before it is used. The scan rate of 10 mVs−1 in Fig. [7b](#page-7-0) displays the CV curves of

with different current densities; **e** capacitance values as a function current density; **f** cycling performance of ASC (inset last 500 cycles)

Fig. 8 A Ragone plot (energy density vs. power density) for the $NiMn₂O₄@RGO//AC$ asymmetric supercapacitor device with various supercapacitor data from the literature and fabricated LED device (inset)

Electrode material	Electrolyte	3-Electrode specific capacitance	Refs.
$NiMn2O4$ microsphere	1 M Na ₂ SO ₄	768.9 Fg^{-1} @ 1 Ag ⁻¹	$\lceil 34 \rceil$
$NiMn2O4/rGO/PANI$	3 M KOH	757 Fg^{-1} @ 1 Ag^{-1}	$\left[35\right]$
$NiMn2O4$ on 3D N ₂ -doped graphene	3 M KOH	1308.2 Fg^{-1} @ 1 Ag^{-1}	$\lceil 36 \rceil$
$NiMn2O4 @ Cos Core–Shell microspheres$	2 M KOH	1751 Fg^{-1} @ 1 Ag^{-1}	$\left\lceil 37 \right\rceil$
MWCNT blended $NiMnO_3/NiMn_2O_4$	2 M KOH	1347 Fg^{-1} @ 5 Ag^{-1}	$\lceil 38 \rceil$
NiMn ₂ O ₄ @rGO	2 M KOH	1848 Fg^{-1} @ 1 Ag ⁻¹	This work

Table 1 Comparison of electrochemical performance between NiMn₂O₄@rGO and other Ni–Mn oxide/carbonaceous-based electrode materials

both the $NiMn₂O₄/rGO$ and the AC electrode. Given that the positive electrode has a potential window of 0 to 0.6 V and the negative electrode has a potential window of − 1.0 to 0 V, the estimated operating voltage of the $NiMn₂O₄/rGO/AC ASC$ is 1.8 V. The ASC's CV curves are shown in Fig. [7c](#page-7-0) in a variety of scan rates (10–100 mVs⁻¹). The electrochemical features of double-layer capacitance and pseudocapacitance are represented by a quasi-rectangular frame with prominent redox peaks. CV curves almost maintain their original shape without polarisation as the scan rate rises, indicating the device's high rate functionality. The GCD profle of an ASC device is depicted in Fig. [7d](#page-7-0). Using the GCD curves as depicted in Fig. At 1, 2, 6, 10, and 20 Ag^{-1} , the calculated specific capacitance of the ASC is 182.3, 166.8, 161.3, 145.1, and 121.6 Fg^{-1} , respectively (see Fig. [7e](#page-7-0)). A good rate achievement is indicated by a capacitance retention of around 72% as the current density is increased 20-fold from 1 to 20 A g^{-1} (Fig. [7](#page-7-0)f). Recent findings for the supercapacitor electrode NiMn_2O_4 @rGO are shown in Table [1](#page-8-0) [[34](#page-10-9)–[38](#page-10-10)]. The nanocomposite's enhanced conductivity and electron transfer path can be credited to the potent combination of rGO and NiMn_2O_4 , as demonstrated by the composite's remarkably high capacitance and capacitance retention. It also shows that the electrochemical doublelayer capacitance and the Faradaic redox reaction are facilitated by the $NiMn₂O₄'s$ structure being modified through rGO. The efficiency of an ASC device can be measured in part by its energy density and power density. Ragone plot (Fig. [8\)](#page-7-1) reveals that the $NiMn₂O₄@rGO/AC ASC$ device provides a high energy density of 49.8 Whkg⁻¹ at a power density of 880.5 Wkg−1. At a power density of 2100 Wkg−1, the energy density of 28.2 Whkg $^{-1}$ is still maintained. As an added bonus, the energy density of the results obtained in this work is signifcantly higher than that

of other $NiMn₂O₄$ -based ASC devices [[23,](#page-10-1) [39](#page-10-11)[–41](#page-10-12)]. Additional proof of its potential, two ASC devices linked in series can power a blue LED strip for 8 min or light a violet LED for roughly 20 min (inset Fig. [8](#page-7-1)).

4 Conclusions

A simple one-step hydrothermal method was used to satisfactorily concoct hierarchical NiMn₂O₄@rGO electrodes. The $NiMn₂O₄/rGO$ composite electrode achieves a high specific capacitance of 1848 Fg^{-1} and good rate abilities with 80% capacitance retention thanks to the novel efect of using electroactive $NiMn₂O₄$ as spacers to separate rGO nanosheets and the tight adhesion of the active materials on the current collector. Synergistic efects between the enhanced electric conductivity of rGO nanosheets and the redox characteristic of binary metal oxide are responsible for the good electrochemical conscious actions for $NiMn₂O₄@rGO$ electrode. These effects include an increase in the rate of ion difusions and electron transfers, an enhancement of the electrochemical double-layer capacitance for the surface of rGO, and a boost in the Faradaic redox reaction of near the surface of $NiMn_2O_4$. Based on these findings, $NiMn₂O₄@rGO material may be a promising$ option for high-performance materials for electrodes in energy storage systems.

Author contributions

PK and AJA: study conceptualisation and writing (original draft) the manuscript. PR and TK: data curation, formal analysis, and writing (review and editing).

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Data availability

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

Declarations

Conflict of interest The authors declare that there are no confict of interest regarding the research work reported in this manuscript.

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