

SYNTHESIS AND CHARACTERIZATION OF rGO AND NICKEL-COBALT OXALATE COMPOSITE MATERIALS FOR SUPERCAPACITOR ELECTRODE APPLICATIONS

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Abstract

In this work, a composite material comprising nickel-cobalt oxalate and graphene (rGO@NiCo-Ox) was synthesized via a hydrothermal method for application as a supercapacitor electrode. Comprehensive characterizations of the structural and compositional properties were conducted using X-ray diffraction (XRD), Raman spectroscopy, transmission electron microscopy (TEM), energy-dispersive X-ray spectroscopy (EDS), and thermogravimetric analysis (TGA). The electrochemical behavior was systematically evaluated by cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS). The experimental results reveal that the incorporation of graphene significantly reduces both the electrical resistance and particle size of the nickel-cobalt oxalate component, resulting in the rGO@NiCo-Ox composite demonstrating markedly improved electrochemical performance compared to pristine NiCo-Ox. Notably, the rGO@NiCo-Ox electrode achieves a maximum specific capacity of 652.65 C g⁻¹ at a current density of 1 A g⁻¹, representing an enhancement of 123% over the capacity of NiCo-Ox. In addition, the composite exhibits superior cycling stability relative to the pure NiCo-Ox electrode. These findings suggest that the integration of battery-type active materials with graphene offers a compelling strategy for augmenting the electrochemical properties of single-component systems, and highlight the rGO@NiCo-Ox composite as a highly promising electrode material for high-performance supercapacitors.

Keywords: Supercapacitor; nickel-cobalt oxalates; nickel-cobalt oxalates/graphene.

1. Introduction

Supercapacitors are widely recognized as promising next-generation energy storage devices due to their outstanding advantages, including high specific capacitance, long cycle life, abundant raw materials, and facile fabrication technology [1], [2]. Consequently, a numerous study on supercapacitors has been increasing in recent years. Similar to other energy storage systems, the electrochemical performance of a supercapacitor is primarily determined by its electrode materials. Therefore, research efforts have largely focused on developing advanced electrode materials. Currently,

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supercapacitor electrode materials could be generally classified into three main categories: (i) electric double-layer capacitor (EDLC) materials, (ii) pseudocapacitive materials, and (iii) battery-type materials [1], [3]. EDLC materials operate through the adsorption and desorption of electrolyte ions at the electrode-electrolyte interface. Among materials, carbon-based materials are the most typical representatives of this group [4], [5]. Pseudocapacitive materials, in contrast, store charge via reversible redox reactions and intercalation/deintercalation processes occurring within the electrode, i.e., RuO_2 and MnO_2 [6], [7]. Battery-type materials exhibit Faradaic charge storage behavior, characterized by distinct potential plateaus in the charge-discharge curves. This group typically includes transition metal oxides [8], [9], layered double hydroxides (LDHs) [10], [11], and transition metal sulfides [3], [12].

Each type of electrode material possesses its own unique advantages. EDLC materials provide high power density and excellent cycling stability but suffer from relatively low energy density. Therefore, they make them suitable mainly for applications requiring rapid power delivery over short durations. Pseudocapacitive materials exhibit significantly higher energy density than EDLCs, however, their power density and long-term stability are somewhat limited. Battery-type materials, benefiting from multiple oxidation states of transition metal ions, can achieve the highest energy density among supercapacitor electrodes, though their charge-discharge rate capability remains a major challenge. Nevertheless, battery-type materials are still regarded as a highly promising class of electrodes capable of simultaneously meeting the demands for both high energy and high-power densities, provided that their intrinsic limitations can be effectively mitigated. Therefore, numerous strategies have been proposed to enhance the charge-discharge kinetics of battery-type electrode materials for supercapacitor applications, such as metal-ion doping [13], [14], morphology control [15], [16], and hybridization with EDLC-type materials [17], [18].

Metal oxalates have recently garnered considerable interest as electrode materials due to their high specific capacitance, excellent electronic conductivity, cost-effectiveness, and environmental compatibility [19], [20]. Of these, nickel and cobalt oxalates are recognized as particularly promising candidates. For instance, cobalt oxalate nanostructures have demonstrated a remarkable specific capacitance of 1269 F g^{-1} at 6 A g^{-1} [21], while nickel oxalate has achieved capacitive values as high as 951 F g^{-1} at 2 A g^{-1} [22]. In recent work, mixed nickel-cobalt oxalates were successfully synthesized with various compositional ratios [23]. The results indicated that the electrochemical performance of all Ni-Co oxalate composites surpassed those of their pure nickel or cobalt counterparts, and the performances were highly dependent on the Ni:Co molar ratio. Notably, composites with Ni:Co ratios between 3:2 and 2:3 delivered

the most outstanding electrochemical characteristics, with specific capacitances ranging from 2018.5 to 2088.1 F g⁻¹ at a scan rate of 10 mV s⁻¹ (or 1219.6-1365.9 F g⁻¹ at 1 A g⁻¹), which represent improvements of approximately 200–600% over the single-cation oxalates. These examples demonstrate that introducing multiple transition-metal cations into a single material is a straightforward and effective strategy for enhancing electrochemical performance.

Graphene has also emerged as a leading EDLC-type material for supercapacitor applications, owing to its exceptional surface area, superior electrical conductivity, chemical stability, and mechanical strength. However, van der Waals interactions readily cause graphene layers to re-stack, which substantially decreases the accessible surface area and thus the attainable specific capacitance [24]. Recent research reveals that transition metal oxides, hydroxides, and oxalates can be efficiently grown onto graphene sheets, effectively preventing re-stacking and maintaining high surface accessibility. The integration of redox-active materials with graphene enables multiple charge storage mechanisms in a single composite structure, a strategy proven to enhance the overall electrochemical properties of electrode materials.

Despite these advances, the simultaneous use of multiple enhancement approaches for supercapacitor electrode materials has not been thoroughly explored. One particularly promising methodology is to combine both cation diversification and mechanism hybridization within a single material system. Therefore, synthesizing a composite composed of high-performance nickel-cobalt oxalate and highly conductive graphene is anticipated to yield rGO@NiCo-Ox, a material with superior electrochemical properties, higher energy density, and excellent cycling stability, positioning it as a highly attractive candidate for next-generation supercapacitor electrodes.

2. Experimental details

2.1. Chemicals

The chemicals used in this study include nickel (II) nitrate hexahydrate (Ni(NO₃)₂·6H₂O, Sigma-Aldrich); cobalt(II) nitrate hexahydrate (Co(NO₃)₂·6H₂O, Sigma-Aldrich); oxalic acid dihydrate (H₂C₂O₄·2H₂O, Sigma-Aldrich); hydrochloric acid (HCl, Sigma-Aldrich); potassium hydroxide (KOH, Sigma-Aldrich); nickel foam (Ni, TOB Machine); super conductive carbon black (Super P, Thermo Fisher Scientific); polyvinylidene fluoride (PVDF, Thermo Fisher Scientific); and N-methyl-2-pyrrolidone (NMP, Thermo Fisher Scientific). All chemicals were of analytical grade and used without further purification.

2.2. Synthesis of materials

Synthesis of rGO/nickel-cobalt oxalate composite:

The rGO/Ni-Co oxalate composite was synthesized via a co-precipitation process combined with a hydrothermal treatment. The synthesis procedure consists of the following steps:

Step 1: Graphene oxide (GO) was synthesized using the modified Hummers' method [25]. A total of 0.12 g of GO was dispersed in 20 mL of deionized water using ultrasonication for 30 minutes to obtain a homogeneous GO suspension;

Step 2: A mixture of 1.744 g of $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and 1.164 g of $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ was dissolved in the GO suspension under magnetic stirring at 300 rpm for 30 minutes to obtain solution A;

Step 3: Separately, 3.782 g of $\text{H}_2\text{C}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$ was dissolved in 20 mL of deionized water under magnetic stirring at 300 rpm for 30 minutes to obtain solution B;

Step 4: Solution B was slowly added dropwise into solution A under continuous stirring to initiate the co-precipitation process. The mixture was stirred for an additional 30 minutes to form a homogeneous suspension;

Step 5: The resulting mixture from Step 4 was transferred into a 50 mL Teflon-lined stainless-steel autoclave and subjected to hydrothermal treatment at 160°C for 10 hours, where GO is converted into reduced graphene oxide (rGO) and physical bonds is formed between rGO and NiCo-Ox [26], [27]. The autoclave was then naturally cooled to room temperature;

Step 6: The obtained product was filtered and washed several times with deionized water until the pH of the filtrate reached 7, followed by vacuum drying at 80°C for 3 hours. The final product was the rGO/Ni-Co oxalate composite (rGO/NiCo-Ox).

Synthesis of nickel-cobalt oxalate (NiCo-Ox):

For comparison, the Ni-Co oxalate (NiCo-Ox) sample was synthesized under identical experimental conditions as described above, except that GO was omitted.

2.3. Characterization details

Some characteristic properties:

The structural and physicochemical characteristics of the synthesized materials were analyzed using XRD (BRUKER D8 diffractometer with $\text{Cu K}\alpha$ radiation), TGA (PerkinElmer PYRIS 6 TGA analyzer), Raman spectroscopy (Horiba XPLORA+ spectrometer), TEM (JEOL JEM-2100 microscope), and EDS (HITACHI S-4800 high-resolution SEM).

Electrochemical property:

The electrochemical performance of the materials was evaluated using cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS). All electrochemical measurements were performed in a three-electrode configuration, consisting of (i) a working electrode (prepared from the active material), (ii) a reference electrode (Ag/AgCl), and (iii) a counter electrode (platinum plate, $1 \times 1 \text{ cm}^2$). The electrolyte used was 3 M KOH, and all measurements were carried out using a Metrohm Autolab PGSTAT 302N electrochemical workstation. The specific capacitance (C_s , in C/g) of the electrode materials was calculated from the CV and GCD curves according to the following equations:

$$C_{s-CV} = \oint \frac{IdV}{m} \quad (1)$$

$$C_{s-GCD} = \frac{I \times t}{m} \quad (2)$$

where I is the discharge current (A), m is the mass of the active material loaded on the electrode (g), and t is the discharge time obtained from the GCD analysis (s). The working electrode was prepared as follows: (i) a homogeneous slurry was obtained by dispersing a mixture of the active material, Super P, and PVDF in a weight ratio of 80:10:10 in NMP using ultrasonication for 30 minutes; (ii) the resulting uniform slurry was then carefully coated onto a $0.5 \text{ cm} \times 0.5 \text{ cm}$ area of a nickel foam substrate (total size of $0.5 \text{ cm} \times 2 \text{ cm}$), followed by vacuum drying at 60°C for 5 hours. The mass loading of the active material on each electrode was approximately $0.8\text{--}1.0 \text{ mg/cm}^2$.

3. Results and discussion

3.1. Structural, microstructural and compositional properties

The phase structures of the synthesized materials were investigated via XRD, with the corresponding patterns depicted in Fig. 1(a). The XRD spectrum of the NiCo-Ox sample exhibits distinct diffraction peaks at 18.97° , 19.32° , 23.00° , 25.38° , 30.66° , 34.17° , 35.65° , 41.08° , 43.80° , 47.90° , 48.95° , and 51.36° . These peaks are indexed to the (-202), (200), (002), (-112), (-402), (-113), (021), (-314), (-223), (004), (-604), and (023) crystal planes of $\alpha\text{-NiC}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$ with a monoclinic structure assigned to space group C2/c (PDF-00-025-0581) [22], as well as $\alpha\text{-CoC}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$ (PDF-00-025-0251) [18]. In contrast, the XRD pattern of the rGO/NiCo-Ox composite obtains all the characteristic peaks attributed to NiCo-Ox, thus confirming the successful incorporation of the NiCo-Ox phase into the composite matrix. The inclusion of graphene oxide during the synthesis process does not disrupt the crystalline phase formation of NiCo-Ox. Notably, the characteristic diffraction peak for rGO is absent in the XRD pattern of the

rGO/NiCo-Ox composite, a phenomenon ascribed to the relatively low rGO content (approximately 3 wt%). Furthermore, the homogeneous coating of NiCo-Ox nanoparticles over the rGO nanosheets may suppress the rGO diffraction signals, a result consistent with prior literature reports [28].

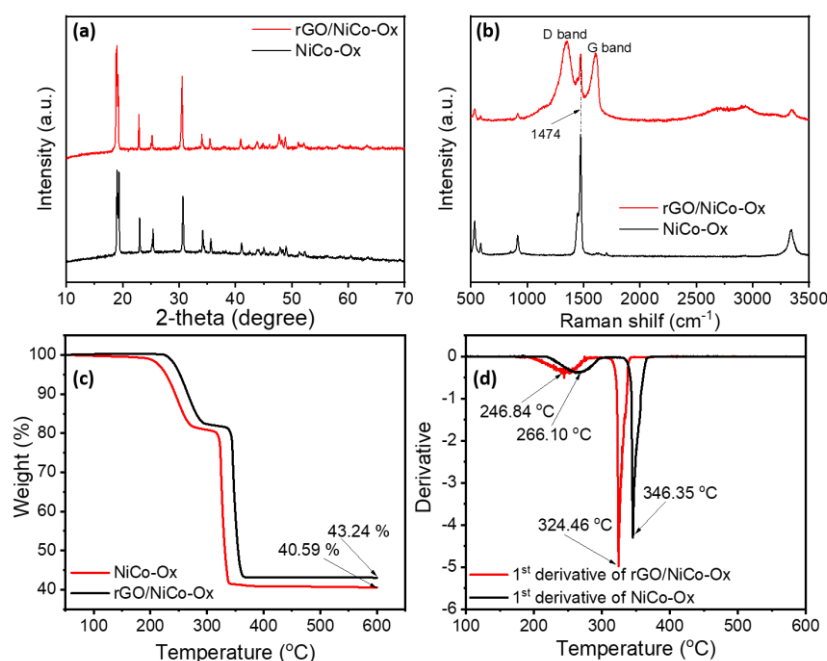


Fig. 1. Characteristic properties of synthesized materials: (a) XRD; (b) Raman; (c, d) TGA.

The Raman spectra of the NiCo-Ox and rGO/NiCo-Ox samples, as shown in Fig. 1(b), exhibit two distinct peaks at 1329.5 cm^{-1} and 1597.1 cm^{-1} for the rGO/NiCo-Ox composite, corresponding to the D and G bands of carbon, respectively [29]. These bands are absent in the pure NiCo-Ox sample. Therefore, it provides a clear evidence for the successful incorporation of rGO within the composite structure. Both NiCo-Ox and rGO/NiCo-Ox materials also displays a peak at 1474 cm^{-1} , attributed to the stretching vibration of the C-O bond in oxalate ions [30]. The TGA and DTG curves for NiCo-Ox and rGO/NiCo-Ox are presented in Fig. 1(c) and (d). All samples demonstrate a minor weight loss of approximately 1% below 200°C , which is associated with the evaporation of adsorbed moisture. A substantial weight loss throughout $200\text{--}300^{\circ}\text{C}$ is observed in Fig. 1(d), which corresponds to the release of two crystallization water molecules from the $\text{AC}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$ structure ($\text{A} = \text{Ni, Co}$) as described by [31]: $\text{AC}_2\text{O}_4 \cdot 2\text{H}_2\text{O} \rightarrow \text{AC}_2\text{O}_4 + 2\text{H}_2\text{O}$. Following the dehydration process, the residual mass of both samples stabilizes around 81–82% of the initial value, consistent with stoichiometric expectations for $\text{AC}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$ (Ni:Co molar ratio = 3:2).

In the subsequent range of 320–370°C, decomposition of oxalate groups occurs accompanied by the thermal degradation of the rGO carbon framework within the rGO/NiCo-Ox composite. Notably, the rGO/NiCo-Ox sample exhibits an additional weight loss of approximately 2.65% compared to the pure NiCo-Ox specimen, attributable to the combustion of the rGO moiety. This mass loss permits an estimation of the rGO content at about 2.65%, which correlates well with initial feed amounts. Post-decomposition, the remaining mass stabilizes at 43.24% for NiCo-Ox and 40.59% for rGO/NiCo-Ox, indicating the formation of thermally resilient metal oxide phases such as NiO and Co₂O₃, with possible minor contributions from Co₃O₄ [31].

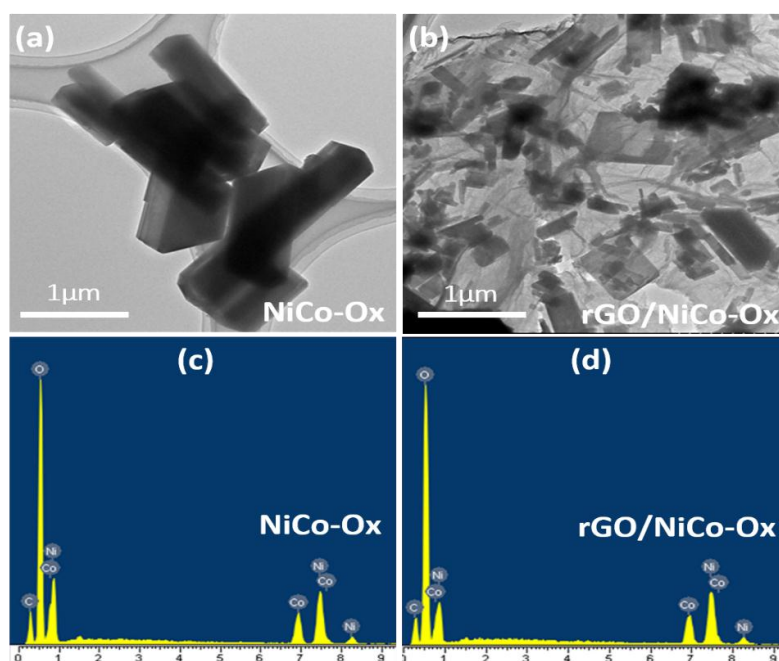


Fig. 2. TEM images and EDS spectra of the synthesized materials: (a, c) NiCo-Ox; (b, d) rGO/NiCo-Ox.

Figures 2(a) and (b) show the TEM images of the NiCo-Ox and rGO/NiCo-Ox samples, respectively. The NiCo-Ox material exhibits irregular polyhedral morphologies with non-uniform particle dimensions ranging widely from approximately 200 nm to 2 μm (Fig. 2(a)). Moreover, the polyhedral particles tend to aggregate, which can be attributed to the absence of any surfactant during the synthesis process to control morphology. In contrast, the TEM image of the rGO/NiCo-Ox composite (Fig. 2(b)) reveals a uniform distribution of NiCo-Ox nanoparticles over the rGO nanosheets. The particle size of the NiCo-Ox component observed in the composite is primarily below 500 nm. Notably, the incorporation of rGO during synthesis leads to a significant reduction in particle size, improved uniformity, and suppressed aggregation of the

NiCo-Ox component. This observation can be explained as follows: The presence of GO layers in the hydrothermal process allows for the formation of numerous nucleation site; creating physical bonds to prevent agglomeration; and promoting the formation of smaller, uniform particles [27], [32]. These morphological improvements are expected to enhance the specific surface area and, consequently, the electrochemical performance of the composite material compared to the pristine NiCo-Ox sample.

The elemental composition of the synthesized materials was determined using the EDS, as shown in Fig. 2(c) and (d), and Tab. 1. The EDS spectra confirmed the presence of Ni, Co, C, and O elements in both the NiCo-Ox and rGO/NiCo-Ox samples, with no detectable impurities, indicating the high purity of the synthesized materials. An increased carbon content in the rGO/NiCo-Ox composite, relative to the pristine NiCo-Ox sample, further supports the successful incorporation of rGO. The Ni:Co atomic ratios were found to be 0.66:0.34 in both samples, which deviates slightly from the initial precursor stoichiometry. This minor difference likely arises from the surface-sensitive nature of EDS, which primarily analyzes the near-surface region rather than the bulk composition.

Tab. 1. Elemental composition ratios of the synthesized materials

Samples	Elemental composition percentage (%)				Ni:Co ratio
	Ni	Co	C	O	
NiCo-Ox	12.79	6.62	15.03	65.56	0.66:0.34
rGO/NiCo-Ox	9.90	5.19	20.75	64.35	0.66:0.34

To a sum-up, the results of XRD, TGA, Raman, TEM, and EDS analyses confirm the successful synthesis of NiCo-Ox and rGO/NiCo-Ox with high phase purity via a straightforward co-precipitation-hydrothermal approach. The good agreement of obtained data with previously reported findings demonstrates the reliability of these results. Incorporation of rGO into the composite introduces an additional electric double-layer capacitance (EDLC) mechanism and promotes particle size reduction, improved homogeneity, and suppressed aggregation of NiCo-Ox, attributes expected to enhance the composite's electrochemical performance over the pristine counterpart.

3.2. Electrochemical properties

The electrochemical performance of the synthesized materials was evaluated using CV, GCD, and EIS. All electrochemical measurements were performed in a three-electrode configuration with 3 M KOH as the electrolyte. Figure 3(a) presents the CV curves of both samples recorded at a scan rate of 20 mV/s. Well-defined redox peaks are observed for both materials, indicating that battery-type behavior dominates the charge storage mechanism.

The Faraday reaction between the active material and the electrolyte ions can be described by the following equations: $A(II)C_2O_4 \cdot 2H_2O + OH^- \leftrightarrow A(III)OHC_2O_4 \cdot 2H_2O + e^-$ [33], where A is defined as nickel and cobalt ions. The rGO/NiCo-Ox composite exhibits a broader CV loop and higher current density at the redox peaks compared to the pristine NiCo-Ox. This result suggests the enhanced electrochemical activity and superior energy storage capability. The specific capacitance calculated from the CV curves at 20 mV/s is 1277.5 C/g for rGO/NiCo-Ox and 982.9 C/g for NiCo-Ox. The superior electrochemical performance of the composite was further confirmed by GCD measurements under identical conditions (current density of 1 A/g, voltage of 0–0.45 V), as shown in Fig. 3(b). The rGO/NiCo-Ox electrode exhibits longer charge-discharge durations than the NiCo-Ox electrode, confirming its higher specific capacitance, consistent with the CV results. The specific capacitances derived from GCD curves were 652.65 C/g for rGO/NiCo-Ox and 528.75 C/g for NiCo-Ox at 1 A/g. The similar shapes of the CV and GCD profiles for both materials indicate that the contribution of the EDLC from the rGO component is relatively minor, which can be attributed to the low rGO content (< 3%) as previously confirmed by structural characterization.

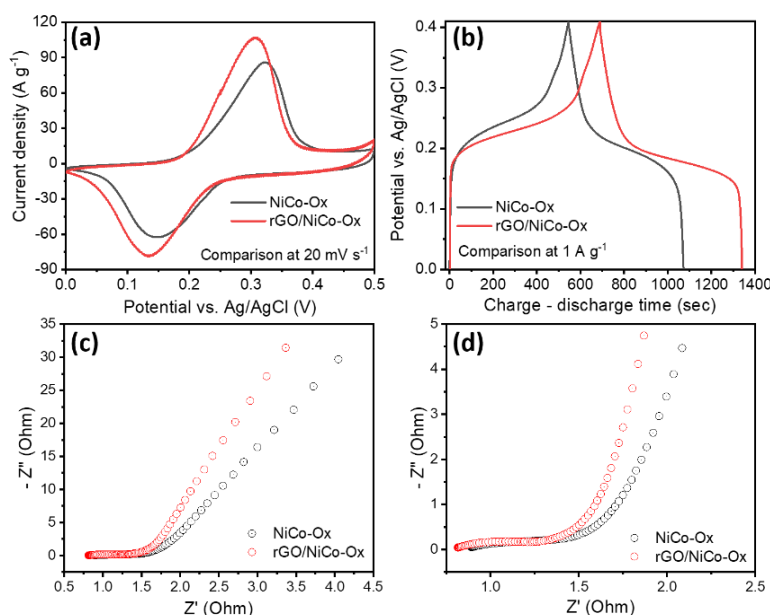


Fig. 3. Comparison of electrochemical properties of synthesized materials: (a) CV analysis; (b) GCD analysis; (c, d) EIS analysis.

The EIS analysis was conducted to further elucidate the role of rGO in enhancing the electrochemical performance of the composite material. As shown in Fig. 3(c), the Nyquist plots of both samples exhibit similar overall profiles, consistent with the comparable energy storage mechanisms discussed earlier. Each Nyquist plot consists of

a depressed semicircle in the high-to-medium frequency region and a straight line in the low-frequency region. Literature suggests that the intercept of the semicircle with the real axis at the high-frequency end represents the Ohmic resistance (R_s) of the electrochemical system, which is determined by the intrinsic resistance of the material and accounts for differences in R_s values. The intercept at the low-frequency end of the semicircle corresponds to the sum of R_s and the charge-transfer resistance (R_{ct}). As shown in Fig. 3(d), both R_s and R_{ct} values of the rGO/NiCo-Ox composite are smaller than those of the pristine NiCo-Ox, which explains the superior electrochemical performance of the composite electrode. Since both materials share the same Ni:Co ratio, this improvement can be attributed to the presence of the rGO component. The role of rGO in enhancing the electrochemical behavior can be interpreted as follows: (i) Morphological modification. The incorporation of rGO results in smaller and more uniformly distributed NiCo-Ox particles, which increases the electrode-electrolyte interfacial area and enhances electrochemical activity; (ii) Facilitated ion diffusion. The reduced particle size in the composite shortens the diffusion paths of electrolyte ions within the electrode, thus improving ion transport kinetics; (iii) Enhanced electrical conductivity. The conductive rGO layers provide efficient electron transport channels, leading to reduced overall resistance and charge-transfer resistance in the composite compared to pristine NiCo-Ox. Furthermore, the slope of the straight line in the low-frequency region of the Nyquist plot reflects the Warburg impedance, which is associated with the ion diffusion kinetics of the electrolyte within the electrode material. A steeper slope indicates more favorable ion diffusion and superior rate capability. As illustrated in Fig. 3(c) and (d), the rGO/NiCo-Ox composite exhibits a higher slope in this region than NiCo-Ox, suggesting improved ion diffusion behavior and faster charge-discharge kinetics.

The electrochemical performance of the rGO/NiCo-Ox composite was further evaluated by CV and GCD measurements under various conditions; the results are presented in Fig. 4. As shown in Fig. 4(a), the current density at the redox peak increases with increasing scan rate, indicating that the rGO/NiCo-Ox composite exhibits rapid reaction kinetics [23], [33]. However, the finite ion-diffusion kinetics into the electrode structure limit the time available for electrolyte ions to penetrate the electrode and react with the redox centers (Ni, Co) at high scan rates. Consequently, the oxidation and reduction peak potentials shift cathodically and anodically, respectively, as the scan rate increases. This is a behavior commonly reported for battery-type materials used in supercapacitors.

Figure 4(b) displays the GCD curves of the rGO/NiCo-Ox composite at different current densities. Distinct potential plateaus (stable voltage regions) are clearly visible during charging and discharging, which is consistent with the CV profiles and confirms the dominant contribution of Faradaic processes to charge storage. The specific capacitances calculated from CV and GCD at different conditions are summarized in Fig. 4(c) and (d) and Tab. 2. In all cases, the specific capacitance decreases as the current density or scan rate increases. Specifically, the specific capacitance of rGO/NiCo-Ox decreases from 652.65 C/g to 586.00 C/g when the current density increases from 1 A/g to 5 A/g, corresponding to a capacity retention of 89.8% under a fivefold increase in current density. From CV analysis, the specific capacitance decreases from 1362.47 C/g to 1078.20 C/g as the scan rate increases from 10 mV/s to 100 mV/s, corresponding to a capacity retention of 79.1% for a tenfold increase in scan rate. These results indicate that the rGO/NiCo-Ox composite possesses good rate capability, a critical property for supercapacitor applications because it enables the material to deliver high energy at high power.

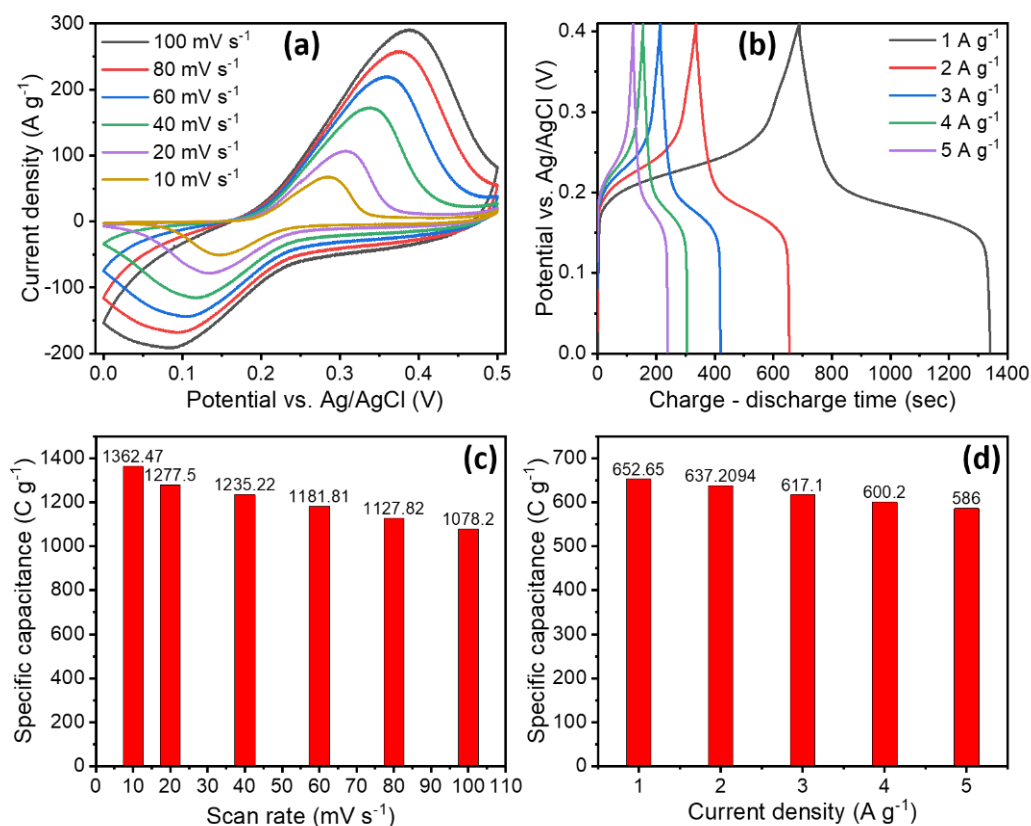


Fig. 4. Electrochemical performance of rGO/NiCo-Ox: CV curves (a) and corresponding specific capacitance (c) at different scan rate; GCD profiles (b) and corresponding specific capacitance (d) at different current density.

Tab. 2. Specific capacitances of rGO@NiCo-Ox calculated from CV and GCD analyses

Methods	Test conditions	Specific capacitances (C/g)
CV	10 mV/s	1362,47
	20 mV/s	1277,50
	40 mV/s	1235,22
	60 mV/s	1181,81
	80 mV/s	1127,82
	100 mV/s	1078,20
GCD	1 A/g	652,65
	2 A/g	637,21
	3 A/g	617,10
	4 A/g	600,20
	5 A/g	586,00

Cycling performance is one of the most important parameters for evaluating materials used in electrical energy storage. The cycling stability tests were conducted for both materials to elucidate the crucial role of rGO in comprehensively enhancing electrochemical performance, and the results are presented in Fig. 5. It can be observed that the rGO/NiCo-Ox composite retains 89.41% of its initial capacitance after 3,000 cycles at a current density of 4 A/g, outperforming the 83.42% retention observed for pristine NiCo-Ox. These electrochemical characteristics clearly demonstrate that the incorporation of rGO not only improves the energy storage capability but also enhances the cycling stability of the rGO/NiCo-Ox sample compared with the bare NiCo-Ox sample. The superior electrochemical properties of rGO/NiCo-Ox compared to NiCo-Ox can be explained as follows: (i) The simultaneous combination of the battery-type material mechanism of the NiCo-Ox component and the EDLC mechanism of the rGO component allows for enhanced energy storage efficiency across the entire voltage range under investigation; (ii) the presence of rGO in the rGO/NiCo-Ox significantly reduced the size of the NiCo-Ox particles, which allowed for an increase in the contact surface area between the active material and the electrolyte; (iii) the formation of a physical link between NiCo-Ox particles and rGO layer, leading to synergistic effects like improved conductivity, structural stability, and facilitating charge transfer. The obtained results provide solid evidence that combining battery-type materials with carbon-based substrates represents an effective strategy for improving electrochemical performance. Furthermore, the rGO/NiCo-Ox composite is considered a promising electrode material for supercapacitors, capable of simultaneously delivering high energy and high power.

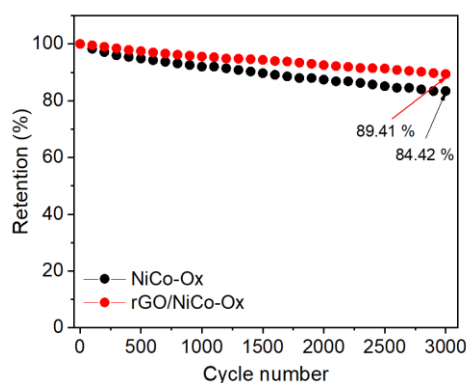


Fig. 5. Cycle performance of synthesized materials at a current density of 4 A/g after 3000 cycles.

4. Conclusion

In this study, a high-purity rGO/NiCo-Ox composite was successfully synthesized based on NiCo-Ox and rGO for application as a supercapacitor electrode. The characterization results revealed that the presence of GO during synthesis significantly reduced the particle size and aggregation of the NiCo-Ox phase in the composite compared with pristine NiCo-Ox. Consequently, the rGO/NiCo-Ox composite exhibited superior and comprehensive electrochemical performance. Specifically, the rGO/NiCo-Ox electrode delivered a specific capacitance of 652.65 C/g at 1 A/g and retained 89.41% of its initial capacity after 3,000 charge-discharge cycles at 4 A/g, which are markedly higher than those of pristine NiCo-Ox (528.75 C/g and 83.42%, respectively). These results illustrate the potential of the rGO/NiCo-Ox composite as a high-performance supercapacitor electrode material, offering both high energy density and high-power capability.

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TỔNG HỢP VÀ ĐẶC TÍNH HÓA VẬT LIỆU COMPOSITE rGO VÀ NIKEN-COBALT OXALAT CHO ỨNG DỤNG ĐIỆN CỰC SIÊU TỤ ĐIỆN

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Tóm tắt: Trong nghiên cứu này, vật liệu composite trên cơ sở niken-cobalt oxalat và graphene (rGO@NiCo-Ox) được tổng hợp bằng phương pháp thủy nhiệt để ứng dụng làm điện cực siêu tụ điện. Các đặc tính cấu trúc và thành phần được phân tích toàn diện bằng phương pháp nhiễu xạ tia X (XRD), quang phổ Raman, kính hiển vi điện tử truyền qua (TEM), quang phổ tán xạ tia X năng lượng (EDS) và phân tích nhiệt trọng lượng (TGA). Hành vi điện hóa được đánh giá một cách có hệ thống bằng phương pháp quét thế vòng tuần hoàn (CV), nạp-xả dòng không đổi (GCD) và phổ tổng trở điện hóa (EIS). Kết quả thực nghiệm cho thấy việc kết hợp graphene làm giảm đáng kể cả điện trở và kích thước hạt của thành phần niken-cobalt oxalat, dẫn đến vật liệu composite rGO@NiCo-Ox thể hiện hiệu suất điện hóa vượt trội hơn so với NiCo-Ox nguyên chất. Đáng chú ý, điện cực rGO@NiCo-Ox đạt dung lượng riêng tối đa là 652,65 C/g ở mật độ dòng điện 1 A/g, cao hơn 123% so với dung lượng riêng của NiCo-Ox nguyên chất. Ngoài ra, vật liệu composite cũng thể hiện độ ổn định chu kỳ vượt trội so với điện cực NiCo-Ox nguyên chất. Những phát hiện này cho thấy việc kết hợp vật liệu loại pin với graphene là một chiến lược đầy hứa hẹn để tăng cường các đặc tính điện hóa của hệ thống đơn thành phần, và nhấn mạnh vật liệu composite rGO@NiCo-Ox là một vật liệu điện cực rất triển vọng cho siêu tụ điện hiệu suất cao.

Từ khóa: Siêu tụ điện; niken-cobalt oxalat; niken-cobalt oxalat/graphene.

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