

Enhanced Electrochemical Energy Storage via FeCoS/RGO Composites[†]

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Abstract

Supercapacitors are considered a bridge between batteries and capacitors due to their significant energy density, as well as power density. Herein, we prepared two novel electrodes of Fe_{0.8}Co_{0.2}S and Fe_{0.8}Co_{0.2}S/rGO composites and analyzed their supercapacitor performance. The results indicated that Fe_{0.8}Co_{0.2}S/rGO, prepared through co-precipitation and annealing, exhibited a higher specific capacitance value and improved electrochemical properties in comparison to Fe_{0.8}Co_{0.2}S due to the synergistic effect of rGO with Fe_{0.8}Co_{0.2}S. X-ray diffraction (XRD) confirmed the desired phases of Fe_{0.8}Co_{0.2}S, while scanning electron microscopy (SEM) and energy-dispersive X-ray spectroscopy (EDS) verified the microstructures and desired elements. Cyclic voltammetry (CV) confirmed an enhanced oxidation current from +25 mA to +49 mA at 10 mV/s, while galvanometric charge–discharge (GCD) showed an enhanced discharge time from 78 s to 300 s. As a result, the specific capacitance and energy density were enhanced from 74.3 F/g to 285.7 F/g and 2.84 Wh/kg to 10.9 Wh/kg, respectively. This contributed to a more than 283% increase in specific capacitance, as well as energy density. Overall, Fe_{0.8}Co_{0.2}S/rGO shows great potential for small-scale energy storage devices.

Keywords: reduce graphene oxide; cyclic voltammetry; transition metal sulfides



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1. Introduction

The growing worldwide need for efficient and sustainable energy storage mechanisms underlines the imperative demand for innovative materials. Transition metal sulfides, well known for their remarkable electrical conductivity, excellent energy density, economic viability, mechanical strength, and ease of synthetic pathways, have become hopeful contenders [1,2]. Iron–cobalt sulfide (FeCoS) has become one of the promising contenders among them. The synergistic effect of iron and cobalt cations in the FeCoS structure is expected to provide excellent electrochemical performance, and thus it is an interesting material for future energy storage devices [3].

Many researchers favor transition metal sulfides (TMSs) from Bi, Co, Cd, Cu, Ni, and Fe due to their high specific capacitance, abundance, and feasibility. They outperform oxides and hydroxides [4]. This study focuses on Fe- and Co-based sulfides for their stable and conductive structures [5], high theoretical capacitance, fast Faradaic redox reactions, and ternary synergistic effects [6]. Further, optimizing TMS electrodes requires composite engineering and morphological control [7]. However, bimetallic Fe-Co sulfides remain understudied in energy storage, necessitating further exploration of Fe-Co sulfide composites.

A synergistic multicomponent approach enhances ionic/electronic conductivity, boosting energy storage potential [8]. Multicomponent TMSs surpass single-component sulfides in conductivity, stability, capacity, cycling performance, and efficiency, making them more viable for energy storage [8].

The optimization of FeCoS/rGO composites for energy storage involves the controlled morphology and composition of FeCoS via customized synthesis. rGO increases conductivity and cushions volume changes, enhancing stability [9]. These composites hold potential in supercapacitors, lithium ion, and sodium ion batteries. Future work is aimed at Fe/Co ratio optimization, rGO content, and new synthesis routes to further improve performance [1]. To further improve the electrochemical property of FeCoS, especially for supercapacitor performance, the incorporation of reduced graphene oxide (rGO) is investigated. rGO, with high surface area, good electrical conductivity, and mechanical flexibility, achieves a synergistic effect when combined with FeCoS [9]. This composite material exhibited superior charge storage properties, enhanced rate capability, and better cycle durability, making it a good material for advanced energy storage devices [10].

2. Materials and Methods

All the chemicals were bought from Sigma-Aldrich (Saint Louis, MO, USA): ferric chloride hexahydrate ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$; $\geq 76\%$), cobalt chloride hexahydrate ($\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$; $\geq 99\%$), and sodium sulfide nonahydrate ($\text{Na}_2\text{S} \cdot 9\text{H}_2\text{O}$; $\geq 99.99\%$). For enhanced electrochemical performance, a composite was prepared with reduced graphene oxide (rGO). Deionized water and ethanol, sourced from Sigma Scientific (Islamabad, Pakistan), were used during the precursor dissolution and washing. In synthesizing the $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{rGO}$ composite, a mass ratio of 10:1 was used for $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ and rGO. $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$, amounting to 0.0223 moles, were dissolved in 100 mL deionized water under stirring. Subsequently, 5.52 g of $\text{Na}_2\text{S} \cdot 9\text{H}_2\text{O}$ was added slowly, allowing for the in situ formation of Fe-Co-S by the replacement of Na^+ with Fe^{+2} and Co^{+2} ions at 70 °C. For even dispersion and agglomeration prevention, the reaction product was ultrasonicated. The material was then purified by washing and drying at 100 °C. Lastly, the resulting powder was thermally annealed at 400 °C using a ramped heating rate of 2 °C/min for four hours to maximize its crystallinity and electrochemical performance.

After material synthesis, iron copper sulfide ($\text{Fe}_{0.8}\text{Cu}_{0.2}\text{S}$) and the nanocomposite slurries were made. For the A1 $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ electrode, 12 mg of active material was mixed with 30 μL of polyvinylidene fluoride (PVDF) and 40 μL of N-methyl-2-pyrrolidone (NMP) in a slurry tube. The mixture was stirred at 360 rpm for 8 h. In the case of the $\text{Fe}_{0.8}\text{Cu}_{0.2}\text{S}/\text{rGO}$ (10:1) electrode (A2), 10.8 mg of the active material and 1.2 mg of reduced graphene oxide (rGO) were put into a slurry tube consisting of 30 μL of PVDF and 40 μL of NMP and stirred in an identical manner at 360 rpm for 8 h. Every prepared slurry was then coated onto individual nickel foam substrates and dried next to an oven at 80 °C for 10 h. The particles synthesized were characterized by scanning electron microscopy (SEM) and energy-dispersive X-ray analysis (EDS) through SEM (Carl Zeiss Evo 15, Jena, Germany), X-ray diffraction (XRD) (AXRD LPD, Proto, UK), and electrochemical analysis via a potentiostat (Gamry Instruments, Reference 3000, Warminster, PA, USA).

3. Results and Discussion

3.1. XRD Analysis

X-ray diffraction analysis was used to verify the phases of the synthesized materials by correlating the diffraction peaks to their respective standard JCPDS card numbers. X-ray diffraction (XRD) analysis identified clear crystalline phases for the two synthesized materials. For the first phase, referring to the $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ sample, diffraction peaks at 2 θ

angles of 19, 28, 31, 33, 35, 45, 64, and 66 degrees were indicated. These peaks suggest the existence of certain crystallographic planes in the $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ structure. In the second phase, corresponding to the $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{rGO}$ composite, the XRD pattern revealed peaks at 2θ values of 19, 28, 31, 33, 35, 40, and 63 degrees, corresponding to the $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ phase. Notably, a new peak was observed at $2\theta = 24$ degrees, characteristic of reduced graphene oxide (rGO). This indicates a successful integration of the rGO into the composite structure. The minimal shifts and intensity change in the $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ peaks between the two phases could indicate minor changes in the crystalline structure or particle size because of the incorporation of the rGO. The identification of the rGO peak indicates the hybrid formation as shown in Figure 1.

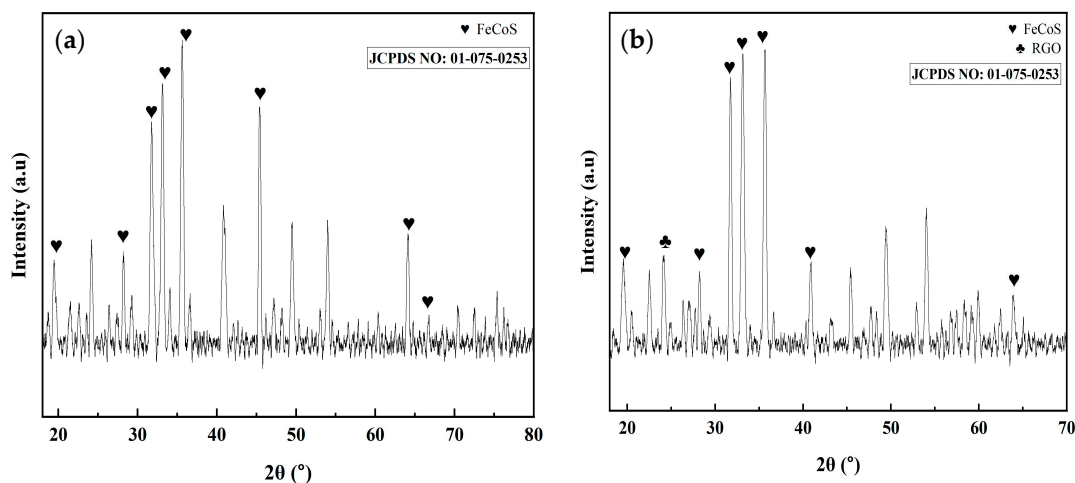


Figure 1. XRD analysis of (a) $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ and (b) $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{RGO}$ composite.

3.2. SEM

Scanning electron microscopy (SEM) characterization, as shown in Figure 2a–d, showed that $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ had an aggregated morphology of elongated and irregular structures. Upon the addition of reduced graphene oxide (RGO), a visible change in the morphology of the material was witnessed, as illustrated in Figure 2e,f. The successful formation of the composite material is substantiated by the presence of RGO sheets, upon which aggregated $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ particles are distributed. This structural modification is posited to enhance the feasibility of the $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{RGO}$ composite as a material for energy storage applications.

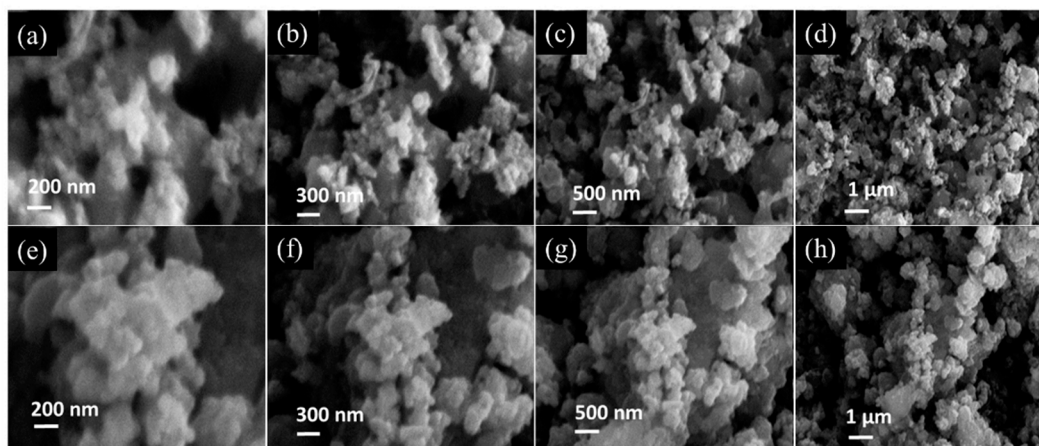


Figure 2. SEM analysis of $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ (a–d) and $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{RGO}$ (e–h).

3.3. EDS Elemental Mapping

Elemental composition was determined via energy-dispersive X-ray spectroscopy (EDX). The EDX spectrum of $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ is displayed in Figure 3, validating the presence of iron, cobalt, and sulfur, confirming the successful synthesis. Minor peaks, likely artifacts of the coating, were also detected. Conversely, the EDX spectrum of $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{RGO}$, presented in Figure 3, exhibited additional peaks corresponding to carbon. This observation confirms the successful integration of reduced graphene oxide within the composite, a factor anticipated to positively influence its electrochemical performance.

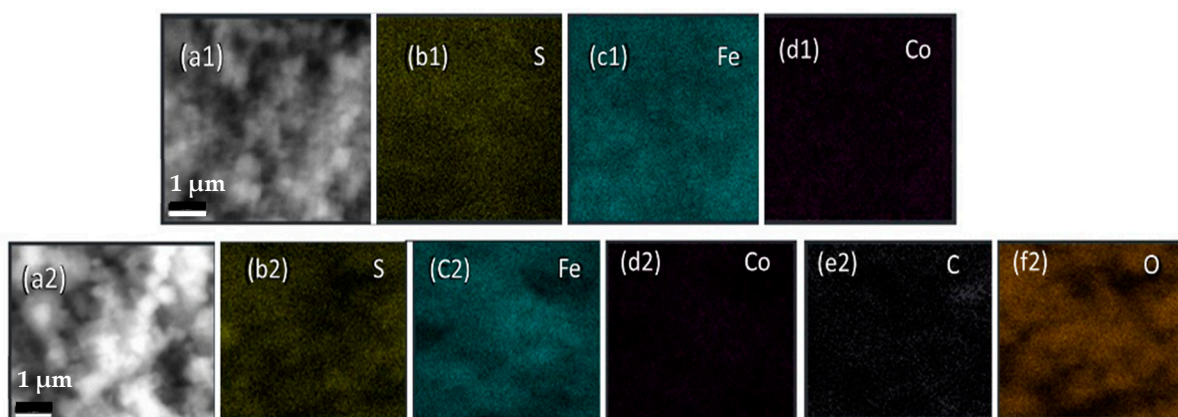


Figure 3. EDS mapping of $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ (a1–d1) and $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{RGO}$ (a2–f2).

3.4. EDX Spectra

The elemental composition of the samples was analyzed using EDX. The $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ sample (Figure 4) exhibited characteristic peaks for iron, cobalt, and sulfur, confirming its formation. Additional peaks were noted, likely due to the coating. The EDX spectrum of $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{RGO}$ (Figure 5) further confirmed the successful synthesis of the composite by showing the additional peak of carbon, indicative of the presence of RGO, which is expected to improve electrochemical properties.

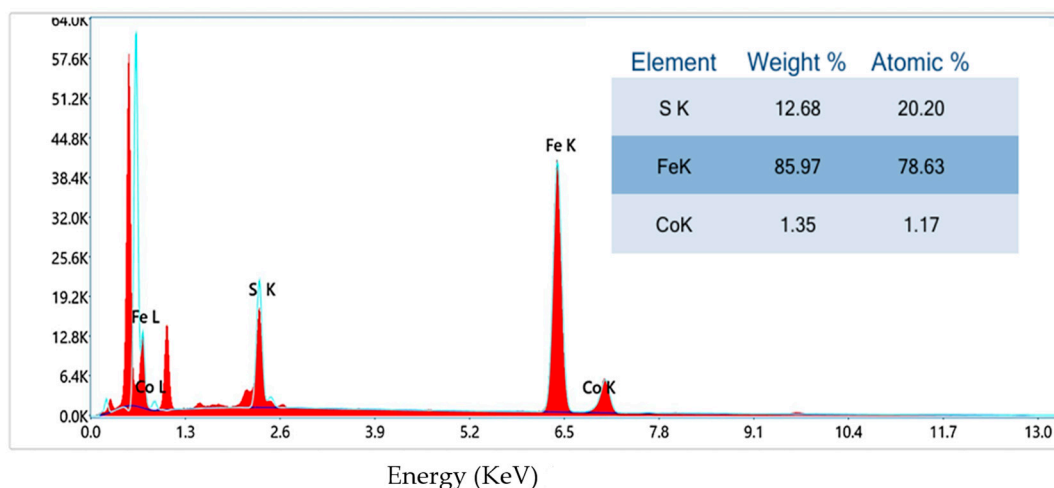


Figure 4. EDX graphs of $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$.

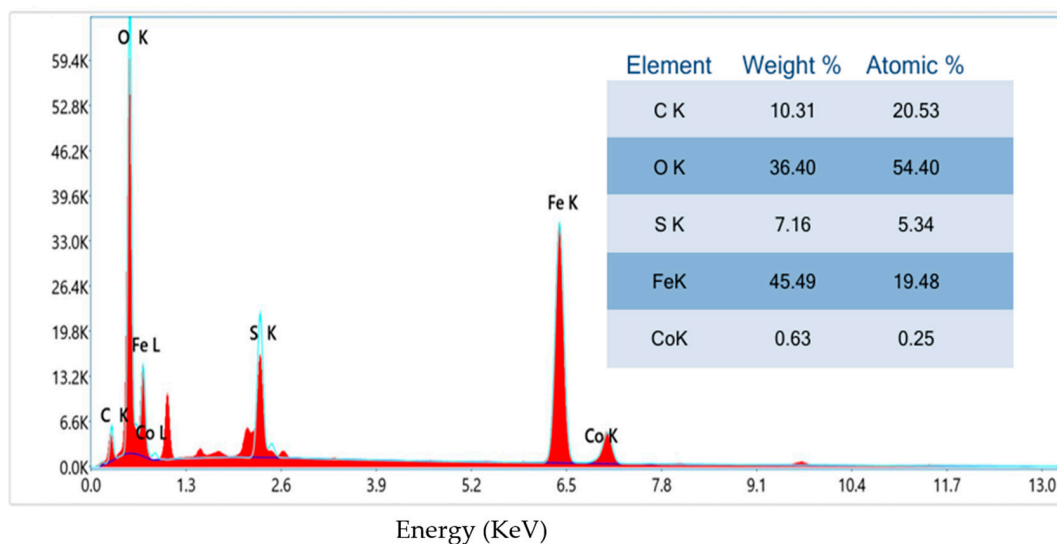


Figure 5. EDX graphs of $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{rGO}$.

3.5. Cyclic Voltammetry Analysis

Electrochemical performance was assessed via cyclic voltammetry across a range of scan rates, from 2 to 50 mV/s. The $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ electrode (P1), as shown in Figure 5, displayed distinct redox peaks, signifying Faradaic reactions. At 10 mV/s, P1 exhibited an anodic peak at +25 mA and a cathodic peak at −18 Ma, see Figure 6a. In contrast, the $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{RGO}$ electrode (P2), depicted in Figure 6b, showed an anodic peak of +49 mA and a cathodic peak of −39 mA at the same 10 mV/s scan rate. A clear enlargement of the area under the curve was observed for P2, indicating a substantial increase in capacitance. A comparative visualization of these specific capacitance values is provided in Figure 6c.

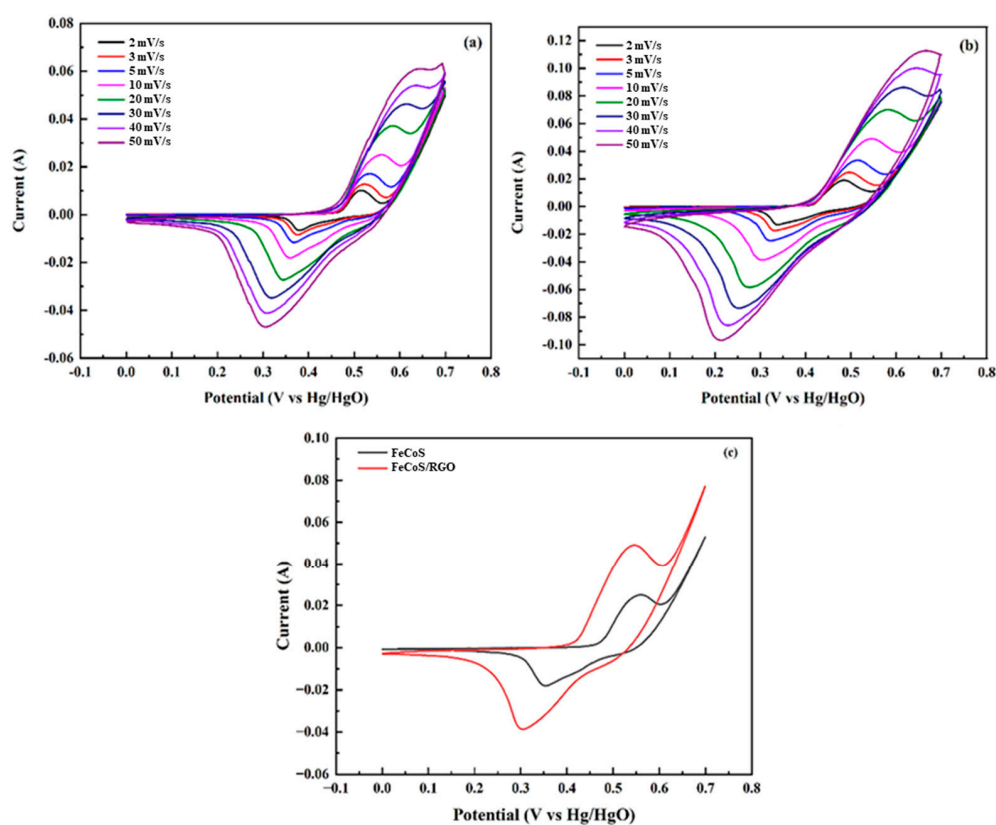


Figure 6. Cyclic voltammetry analysis from 2 to 50 mV/s for (a) $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ (P1) and (b) $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{RGO}$ (P2) and (c) comparative plot of P1 and P2.

3.6. GCD Analysis

The galvanostatic charge–discharge (GCD) profiles provide insights into the charge and discharge mechanisms of the electrode material. Equation (1) is used to determine the specific capacity during the discharge process. In this equation, I/m represents the current density and Δt denotes the time taken for discharge:

$$Q_s = (I \times \Delta t)/m \quad (1)$$

Subsequently, the specific capacitance and energy density was calculated by incorporating the value obtained from Equation (1) into the following expression:

$$C_s = Q_s/\Delta V \quad (2)$$

$$E = 0.139 \times C_s \times \Delta V^2 \quad (3)$$

The GCD profiles, illustrated in Figure 7a,b, demonstrate the electrochemical behavior of the electrodes across current densities from 0.5 to 4 A/g within a 0 to 0.5 V potential window. Notably, the discharge time and specific capacitance increased significantly from 78 s to 300 s and 74.3 F/g to 285.7 F/g (calculated using Equations (1) and (2)), respectively, representing a 283% improvement; see Figure 7c. Furthermore, the energy density rose from 2.84 Wh/kg to 10.9 Wh/kg—a 284% enhancement—demonstrating the composite electrode's superior energy storage performance.

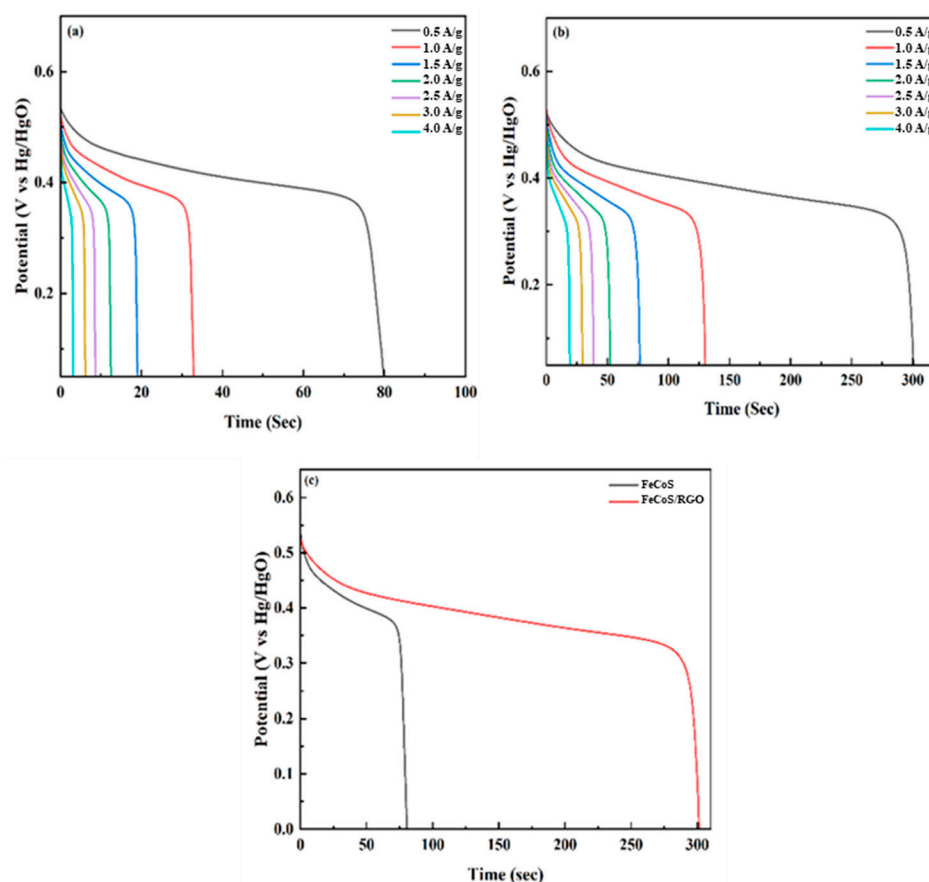


Figure 7. Discharge curves at various sweep rates of (a) Fe_{0.8}Co_{0.2}S and (b) Fe_{0.8}Co_{0.2}S/RGO and (c) comparative plot of Fe_{0.8}Co_{0.2}S vs. Fe_{0.8}Co_{0.2}S/RGO.

3.7. Nyquist Plots

Electrochemical impedance spectroscopy (EIS) was conducted on both $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ and $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{RGO}$ composites, see Figures 8a and 8b, respectively, in order to evaluate their charge storage behavior and interfacial kinetics. The Nyquist plot reveals enhanced EDLC behavior upon the incorporation of RGO, as evidenced by the upward shift in the low-frequency tail of the curve. Furthermore, the $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{RGO}$ composite exhibits a significantly reduced charge transfer resistance compared to the pristine material, as indicated by the smaller semicircle diameter in the high-frequency region as shown in Figure 8c. This enhanced behavior implies that RGO improves conductivity and provides a channel for quicker charge transfer, placing the composite in the running for high-performance energy storage applications.

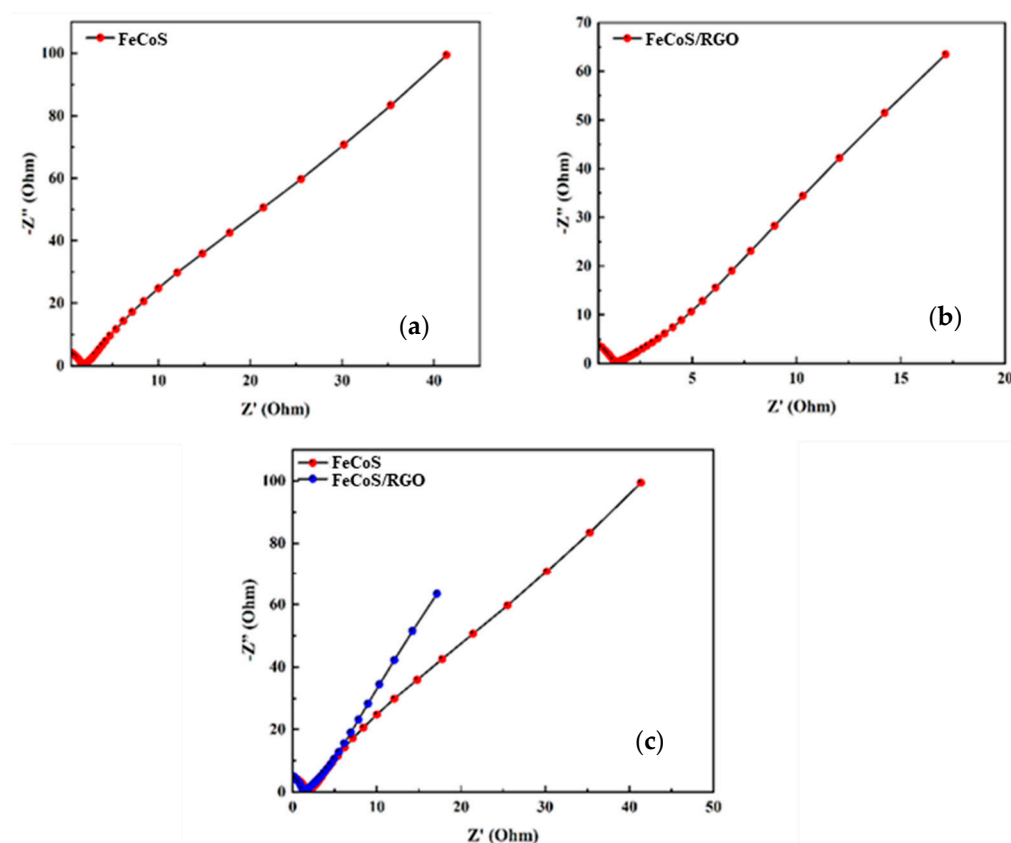


Figure 8. The Nyquist plots of (a) $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ and (b) $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{RGO}$ and (c) comparative plot of P1 vs. P2.

4. Conclusions

In this work, $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ and $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{rGO}$ composites were successfully synthesized, and their electrochemical performance as supercapacitor materials was assessed. The addition of rGO to the $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$ matrix greatly improved the specific capacitance, as indicated by a >283% increase compared to pure $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}$. Galvanostatic charge–discharge tests also verified this improvement, indicating a remarkable increase in discharge time. Characterization methods, such as XRD, SEM, and EDS, confirmed the successful formation of the composite and indicated the synergistic effect of rGO in enhancing the material's structural and electrochemical properties. These findings illustrate the potential of $\text{Fe}_{0.8}\text{Co}_{0.2}\text{S}/\text{rGO}$ composites as effective electrode materials for high-performance supercapacitors, emphasizing the significance of rGO integration for increased energy storage capacity. Additional optimization of the composition and the morphology of the

composite may provide enhanced electrochemical performance, enabling advanced energy storage devices.

Author Contributions: This study was designed, planned and supervised by M.A.M. The experimental work was performed, and initial draft was written by M.T. All authors have read and agreed to the published version of the manuscript.

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