

## Article

# NiO/AC Active Electrode for the Electrosorption of Rhodamine B: Structural Characterizations and Kinetic Study

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**Abstract:** In this work, the aim was to enhance the performance of activated carbon (AC) as an electroadsorbent by incorporating NiO particles, thereby increasing its electrochemical capacity and its ability to adsorb Rhodamine B (RhB) dye. The prepared NiO/AC material was characterized using X-ray diffraction, scanning electron microscopy, BET surface area analysis, and infrared spectroscopy. The study involved the preparation of AC from almond shell biomass and the subsequent formation of a composite structure with NiO. The objective was to investigate the electrochemical adsorption capacity of the NiO/AC composite for RhB dye removal from simulated wastewaters. The experimental results demonstrated that the removal efficiency of RhB dye increased with an increase in the applied cell voltage. At a voltage of 1.4 V, a remarkable 100% removal efficiency was achieved. The electroadsorption process was well described by fitting the experimental data to the Freundlich isotherm model. The maximum adsorption capacities for RhB dye at concentrations of 7, 8, and 9 ppm were determined to be 149, 150, and 168 mg/g, respectively. Based on the obtained results, an electroadsorption mechanism was proposed to explain the observed behavior of the NiO/AC composite in adsorbing RhB dye. Overall, this study highlights the potential of the NiO/AC composite as an effective electroadsorbent for the removal of Rhodamine B dye from wastewater. The improved electrochemical capacity, coupled with the high adsorption capacity of the composite, makes it a promising material for wastewater treatment applications.

**Keywords:** electroadsorption; Rhodamine B; activated carbon; wastewater treatment



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## 1. Introduction

The issue of wastewater containing dyes is a major problem that requires immediate attention. According to sources, around 7.105 tons of 100,00 different types of dyes and pigments are produced globally on an annual basis [1]. These synthetic dyes are widely utilized by various industries worldwide, including those involved in textile, leather, food, plastics, and printing. However, these dyes are notorious for their significant contribution to environmental pollution due to the large volume of dye discharged and the composition of the effluent [1,2]. Therefore, it has become crucial to eliminate these dyes from colored effluents before releasing them into aquatic environments. Furthermore, approximately 10–15% of dye materials are lost as industrial effluents during processing operations [2,3]. The discharge of these wastewater containing dyes into the environment leads to a reduction in light penetration, which in turn results in decreased photosynthetic efficiency of aquatic diversity, as sunlight is blocked [4,5]. Advanced oxidation processes (AOPs) are a group of techniques that use highly reactive species, such as hydroxyl radicals ( $\bullet\text{OH}$ ) to degrade

pollutants into less harmful substances [6,7]. AOPs can effectively remove a wide range of pollutants, including persistent organic pollutants, pharmaceuticals, and dyes, and have been extensively studied in recent years [8,9]. AOPs work by generating highly reactive species through various means, such as light, ozonation, and electricity [10–12]. These reactive species then react with the pollutants in the water, breaking them down into less harmful substances.

Rhodamine B (RhB) is an organic dye which falls under the xanthene family known for their vivid reddish-violet color, limited biodegradability, and remarkable chemical stability [13]. It is widely employed as a colorant in industries, such as textiles, leather, and food [14,15]. When present in water bodies, these dyes create a substantial chemical oxygen demand and impede light transmission in photosynthetic processes, resulting in hindered growth of aquatic organisms [16]. RhB can also cause skin and eye irritation, as well as respiratory and gastrointestinal tract discomfort when it enters the body. Furthermore, it has been identified as carcinogenic and mutagenic for living organisms [17].

Electroadsorption, also known as capacitive deionization, is a process that uses an electric field to remove charged pollutants from water [18]. This process is highly efficient and can remove a wide range of pollutants, including heavy metals, organic compounds, and salts [19–21]. An electric field is generated between two electrodes in this technology, which attracts charged pollutants toward the electrode surface. Once adsorbed onto the electrode surface, the pollutants can be conveniently extracted from the water [22]. Rhodamine B, a hazardous dye utilized in numerous industrial processes, such as paper dyeing and laser dye production [23,24], belongs to the family of xanthene dyes and is vivid pink in color. It has a molecular formula of  $C_{28}H_{31}ClN_2O_3$  and a molecular weight of 479.01 g/mol. Over the years, Rhodamine B has been the subject of extensive studies due to its hazardous nature [25–27]. Several treatment methods, including adsorption, membrane filtration, ion-exchange, reverse osmosis, precipitation, advanced oxidation processes, and biological methods, have been employed to eliminate Rhodamine B from aqueous solutions [28–33]. Despite its widespread use, Rhodamine B is considered to be a potential health hazard and has been banned for use in food and cosmetic products due to its potential toxicity [34].

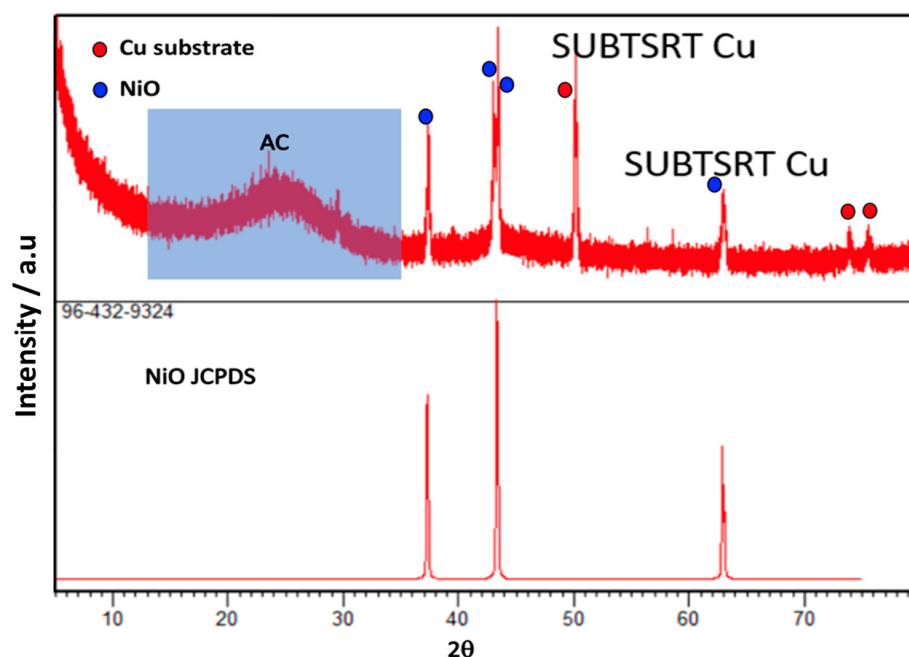
Nowadays, researchers have strengthened the process of adsorption by activated carbon using a controlled electrical potential in order to improve the efficiency of the electroadsorption process [35,36]. In this sense, electrodes made from activated carbon (AC) and its modified derivatives have been found to be suitable for electroadsorption [30]. Currently, environmentalists are looking for new precursors for the production of activated carbon and certain wastes have been used as precursors for the preparation of activated carbons [37–40]. These experimental results demonstrate that carbon materials and their composites are effective in removing pollutants from water due to its cost effectiveness, wide availability, and large variety of precursors. Therefore, to enhance the adsorption capacity, researchers are attempting to introduce new functionalities on AC into electroadsorption properties.

In the present work, nickel oxide was combined with a high surface area activated carbon prepared from agricultural waste almond shell to prepare a novel NiO/AC electrode for Rhodamine B removal. The main objective of this short paper is to investigate the RhB electroadsorption using NiO/AC electrode as well as its kinetic and isotherm models.

## 2. Results and Discussion

The NiO/AC composite's X-ray diffraction patterns are shown in Figure 1. Broad peaks in the 20–30° and 40–50° ranges on the diffractogram show the existence of amorphous activated carbon with different functional groups. The main peaks are indexed at (2θ) 37.37° (1 1 1), 43.6° (2 0 0), and 62.94° (2 2 0) reflections corresponding to the cubic NiO structure according to the JCPDS file 96-432-9324. The Scherrer formula,  $D = (0.9\lambda / \beta \cos\theta)$ , was used to determine the NiO crystallite size, where  $\lambda$  represents the wavelength of the

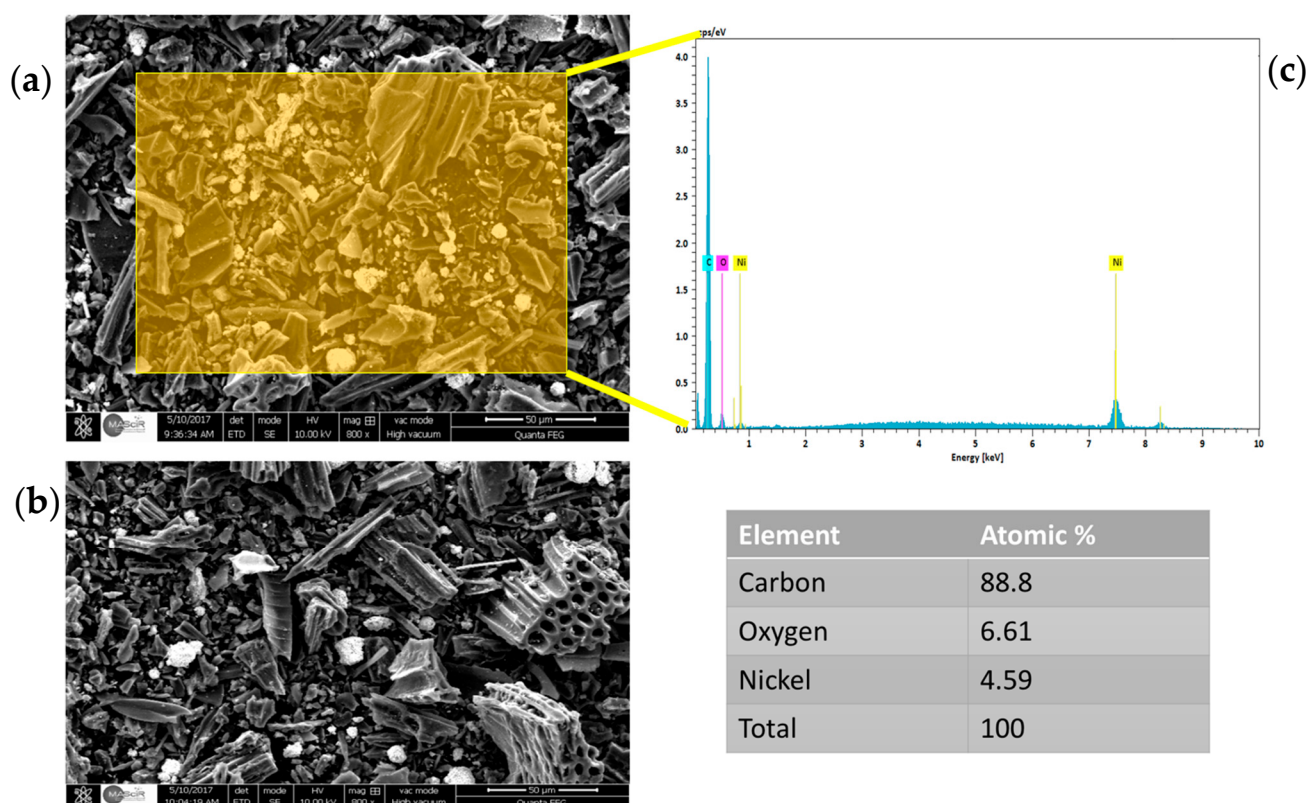
X-ray source, 0.9 is the value for Gaussian profiles,  $\beta$  indicates the broadening of Bragg, and  $\theta$  represents the Bragg angle. The calculated crystallite size is 47 nm.



**Figure 1.** XRD profiles of the NiO/AC sample compared to the JCPDS card 96-432-9324.

The prepared NiO/AC morphology is displayed in Figure 2. The NiO/AC substance has a microporous structure with well-established pores and an uneven surface, as observed. Therefore, the morphology of the NiO/AC material is suitable for adsorbing Rhodamine B. Based on the elemental analysis (EDS), three primary atoms were detected: Carbon (88.8%), Oxygen (6.61%), and Nickel (4.59%). On the other hand, the morphology of the NiO/AC material is an important factor in determining its electroadsorption properties. The microporous structure with well-established pores and an uneven surface allows for a greater surface area, which in turn increases the number of active sites available for adsorption. This is important for the effective removal of Rhodamine B dye from wastewater, as it requires a high degree of surface area for efficient adsorption. The elemental analysis (EDS) also provides valuable information regarding the composition of the NiO/AC material. The high percentage of carbon indicates a high degree of activated carbon, which is known to be an effective adsorbent for various pollutants. The presence of nickel suggests that the NiO particles are well dispersed and well anchored onto the surface of the activated carbon. This is important for the electroadsorption process, as it ensures that the active sites are evenly distributed throughout the material and readily accessible for adsorption. Overall, the morphology and composition of the NiO/AC material are important factors in determining its electroadsorption properties for Rhodamine B dye removal. The microporous structure with a high surface area and the presence of nickel ensure efficient adsorption of the dye, making it a promising material for use in wastewater treatment applications.

The nitrogen adsorption/desorption isotherms shown in Figure 3a provide insights into the pore characteristics of both the activated carbon (AC) and NiO/AC composite. The isotherms exhibit Type I and Type IV shapes, indicating the presence of micropores and mesopores, respectively. This suggests that both materials possess a combination of micropores and mesopores. The pore size distribution (PSD) analysis depicted in Figure 3b further confirms this observation, revealing bi-modal peaks at approximately 0.5 nm and 2.9 nm, corresponding to micropores and mesopores, respectively.



**Figure 2.** (a,b) SEM photographs of the NiO/AC and (c) elemental analysis (EDS).

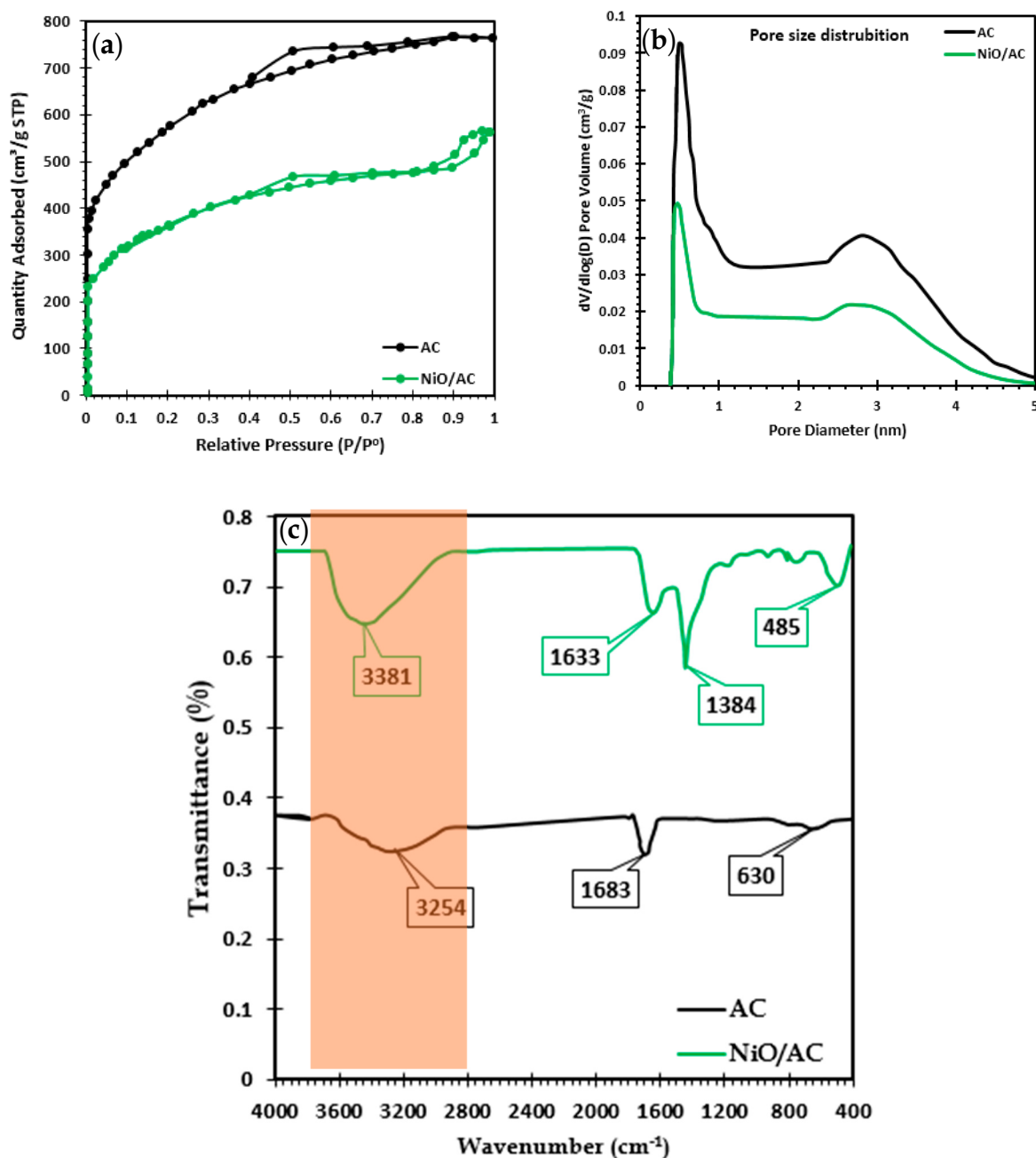
These findings are in agreement with previous studies [24,41] and indicate that the prepared materials exhibit a significant surface area, which is crucial for effective electroadsorption of dyes. The presence of mesopores enhances the diffusion of RhB molecules into the pores, thereby increasing the adsorption capacity.

However, it should be noted that the incorporation of NiO particles into the activated carbon matrix leads to a reduction in the surface area and pore volume of the NiO/AC composite. This is attributed to the blocking effect of NiO particles on some of the activated carbon pores. Despite this decrease, the surface area and pore volume of the NiO/AC composite remain higher than those of many other materials commonly used in adsorption and photocatalysis applications. The decreased surface area and pore volume in the NiO/AC composite could potentially impact its adsorption capacity for RhB. Nevertheless, the higher surface area and pore volume of the composite, compared to other materials, suggest that it still possesses favorable adsorption and electroadsorption activity. These properties highlight the potential of the NiO/AC composite for enhancing adsorption and photocatalytic performance in the removal of RhB from wastewater.

Overall, the nitrogen adsorption/desorption measurements indicate the presence of micropores and mesopores in both the AC and NiO/AC materials. The bi-modal peak distribution confirms the coexistence of micropores and mesopores. The incorporation of NiO particles results in a decrease in surface area and pore volume, but does not eliminate the favorable adsorption and electroadsorption properties of the composite. The surface properties and porosity of the NiO/AC composite are expected to play a significant role in its electroadsorption performance for RhB removal from wastewater.

Fourier transform infrared spectroscopy was employed to determine the functional groups on the surface of the prepared NiO/AC. The FT-IR spectra of AC and NiO/AC are presented in Figure 3c. The AC spectra exhibit two peaks at  $3254\text{ cm}^{-1}$  and  $1683\text{ cm}^{-1}$ , which correspond to the  $\text{-OH}$  stretching vibration of water molecules on the activated carbon surface and the aromatic  $\text{C}=\text{C}$  rings, respectively [42]. Additionally, the IR band at  $630\text{ cm}^{-1}$  is attributed to the in-plane ring deformation [43]. The preparation of NiO/AC

from NiO and AC resulted in a decrease in the intensities of the peaks at 3254 and 1683  $\text{cm}^{-1}$ , and a shift to 3381 and 1633  $\text{cm}^{-1}$ , respectively. The peak at 1384  $\text{cm}^{-1}$  could be attributed to the  $\nu(\text{C}-\text{O})$  vibrations in carboxylate groups, while the characteristic peaks of NiO are observed at 485  $\text{cm}^{-1}$  [44], confirming the impregnation of NiO on the AC surface.

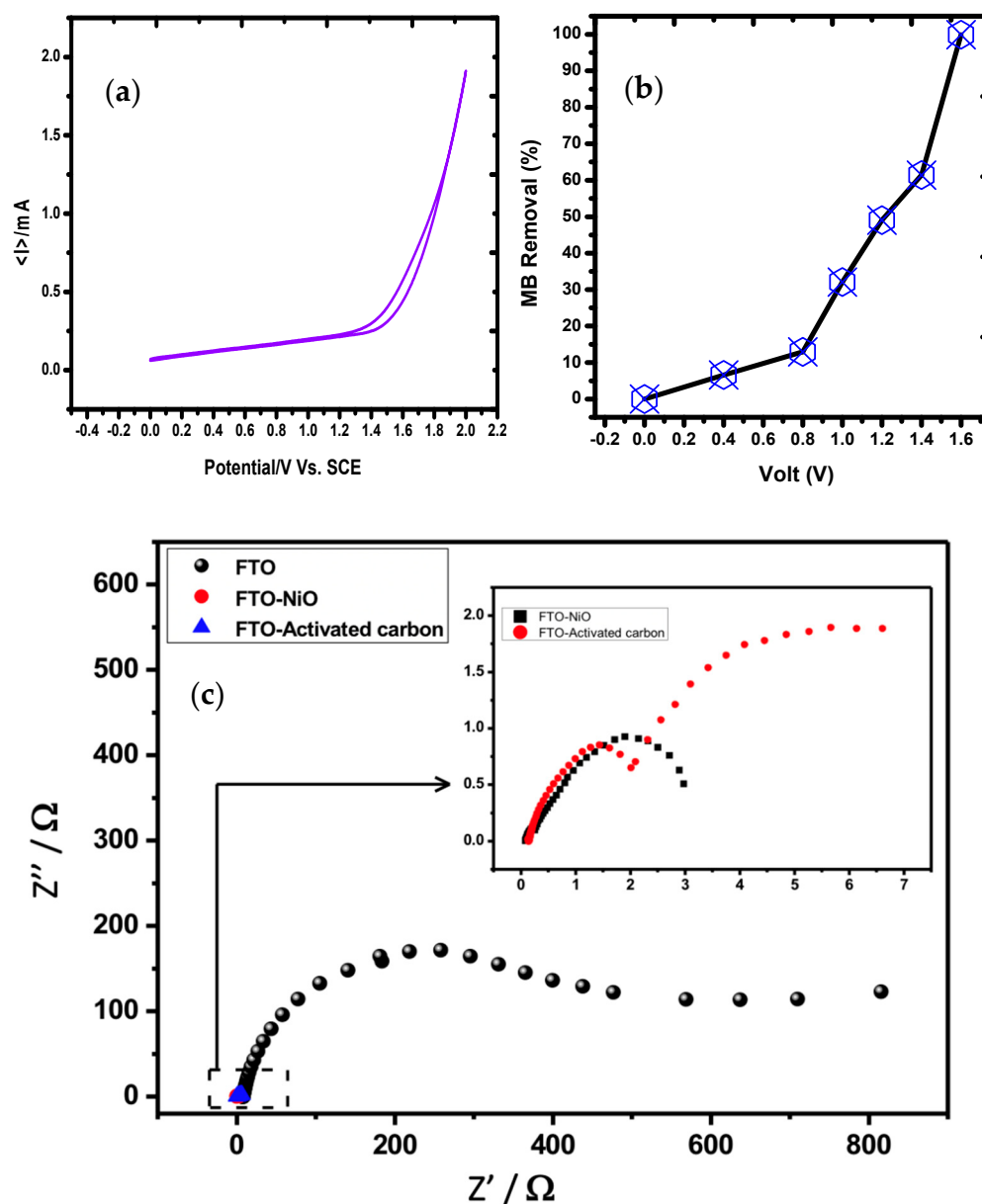


**Figure 3.** (a) Nitrogen physisorption isotherms of AC and NiO/AC; (b) pore size distribution curves of AC and NiO/AC; (c) FTIR spectra of AC, NiO, and NiO/AC.

### 3. Electrochemical Characterization

The electrochemical behavior of FTO/NiO/AC upon electroabsorption of RhB was studied by potential polarization of the working electrode in an electrolyte solution of RhB [45]. In order to demonstrate the degradation mechanism, cyclic voltammetry experiments were performed in an aqueous solution containing 0.5 M NaCl and 7 ppm M RhB (Figure 3a). The electrode polarization between  $-0.2$  and  $2$  V was initiated in the positive potential direction at a scan rate of 50 mV/s. The recorded voltammogram shows a slight

increase in cathodic current and a significant increase from 1.4 V to 2 V. We note the almost total absence of intense current (reduction or oxidation peaks) in this potential range, as shown in Figure 4a, which reflects the fact that no interfering electroactive impurities could be detected and shows the stability of the electrode and adhesion of the NiO/AC layer deposited on FTO [46].



**Figure 4.** (a) Cyclic voltammograms of the FTO/NiO/AC electrode in a RhB solution with 0.5 M NaCl (at a sweep rate of 50 mV/s); (b) electrosorption percentage at different applied potentials; (c) electrochemical impedance data recorded of FTO, FTO/NiO, and FTO/AC.

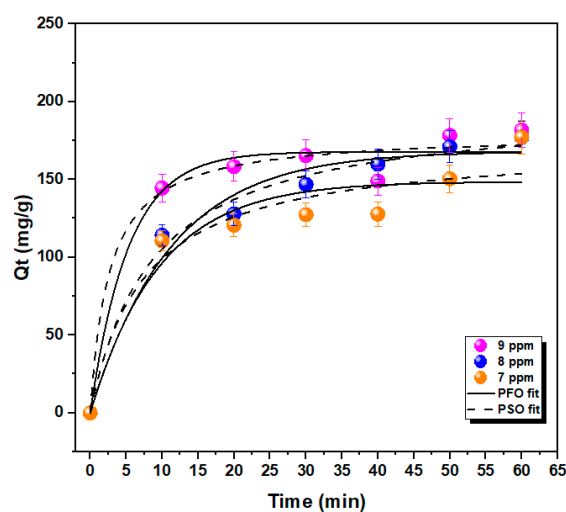
Generally, increasing the potential in a system that aims to remove pollutants is a crucial factor in enhancing the effectiveness of the process. By increasing the potential, we can provide more energy to the electrons in the system, which leads to higher oxidation reactions. In the case of pollutant removal, this indicates that we can oxidize the pollutant to a higher degree, making it easier to remove from the environment [11]. As one can see in Figure 3b, when the potential is increased, the system becomes more energetically favorable, and the reactions occur at a faster rate leading to 100% removal of RhB at 1.4 V, which is degraded at the interface of electrolyte–electrode [47]. This indicates that

more pollutant molecules can be removed per unit time, resulting in a higher overall removal rate. Moreover, an increase in potential can lead to the production of more reactive species, such as hydroxyl radicals, chlorine, and hypochlorite species, and thus promote electroremoval [48]. In summary, increasing the potential is a key factor in pollutant removal processes as it can lead to more reactive species production, and ultimately a higher removal rate.

The impedance spectra will be performed with the optimized characteristic parameters. Frequency range: 1 MHz to 50 MHz. Impedance measurement temperature: 25 °C. The typical impedance diagrams obtained from the analysis of FTO, FTO/NiO, and FTO/NiO/AC are shown in Figure 4c. The spectra are composed of a contribution in the form of a semi-circle whose diameter provides information on the resistance of the electrolyte  $R_e$  [49]. Figure 4c shows that  $R_e$  of the three electrodes FTO, FTO/NiO, and FTO/NiO/AC decreases, respectively, which shows that the electrical properties are improved. This indicates that the separation of charge carriers is easier for FTO/NiO/AC electrodes than its counterparts (FTO and FTO/NiO).

#### 4. Electroadsorption Kinetic Investigation

In this section, we have analyzed the electroadsorption properties of the Ni/AC electrode at a fixed 1.6 V. The electroadsorption kinetics of RhB onto NiO/AC at room temperature are depicted in Figure 4. Three different concentrations of RhB were used (7, 8, and 9 ppm). The findings showed that the adsorption capacities of NiO/AC increased with an increase in contact time up to 20 min, after which point they remained more or less stable. This suggests that there is a saturation point beyond which the adsorption capacity does not increase significantly. Moreover, the amount of RhB adsorbed onto NiO/AC at a time (measured in mg/g) increased noticeably as the RhB concentration increased (Figure 5). The increase in adsorption capacity can be attributed to the availability of active site on the surface of NiO/AC. As the concentration of RhB increases, more active sites become available for adsorption, resulting in a higher adsorption capacity. The non-linear equations of pseudo-first-order (PFO) and pseudo-second-order (PSO) models are used in order to investigate the adsorption kinetics of RhB onto NiO/AC (Table 1). The corresponding parameters for RhB adsorption at various concentrations on NiO/AC are summarized in Table 1, which can be found here. The coefficients of correlation ( $R^2$ ) show that the PSO model provides a more accurate description of the kinetics data for all of the studied RhB concentrations than the PFO model. This indicates that the adsorption process is better described by the PSO model, which suggests that the adsorption rate is proportional to the square of the number of available active sites.



**Figure 5.** Effect of contact time and concentration on the removal of RhB and fitting models (PFO and PSO).

**Table 1.** Highlights of the mathematical equations and models used in this study.

| Equations                                             | Name                | Description                                                                                                                        | References |
|-------------------------------------------------------|---------------------|------------------------------------------------------------------------------------------------------------------------------------|------------|
| $Q_t = Q_{cal}(1 - \exp^{-K_1 t})$                    | Pseudo-first-order  | $Q_e$ and $Q_t$ are the adsorbed amounts at equilibrium and at times $t$ , respectively.                                           | [50]       |
| $Q_t = \frac{(K_2 Q_{cal}^2 t)}{(1 + K_2 Q_{cal} t)}$ | Pseudo-second-order | $K_1$ : the rate constant; $K_2$ : rate constant.                                                                                  | [50]       |
| $Q_e = \frac{Q_m K_L C_e}{1 + K_L C_e}$               | Langmuir isotherm   | $K_L$ : direct measure of the intensity of the adsorption process; $Q_m$ : maximum adsorption capacity.                            | [51]       |
| $Q_e = K_f C_e^{\frac{1}{n}}$                         | Freundlich isotherm | $K_F$ : adsorption capacity; $n$ : intensity of adsorption; $1/n = 0$ irreversible; $1/n > 1$ unfavorable $0 < 1/n < 1$ favorable. | [52]       |

The standard deviation ( $\pm$ ) also provides information regarding the precision as well as the variability of the experimental results. When used in the context of RhB adsorption kinetics on NiO/AC, the standard deviation has the following interpretation:

Pseudo-first-order model: The steady-state adsorption capacities ( $Q_{e,cal}$ ) derived from the PFO model have a standard deviation between  $\pm 5.70$  mg/g and  $\pm 10.83$  mg/g.

The relatively small standard deviation values indicate that the experimental measurements of equilibrium adsorption capacities and rate constants are relatively consistent and accurate. However, the larger standard deviation values for  $Q_{e,cal}$  at higher concentrations (9 ppm) suggest slightly greater variability in the experimental data compared to other concentrations.

Pseudo-second-order model: The equilibrium adsorption capacities ( $Q_{e,cal}$ ) obtained from the PSO model have a standard deviation ranging from  $\pm 9.01$  mg/g to  $\pm 18.17$  mg/g. The standard deviation values for  $Q_{e,cal}$  in the PSO model indicate slightly greater variability than in the PFO model.

Overall, the standard deviation values provide important information regarding the accuracy and consistency of the experimental data. The higher standard deviation values observed for  $Q_{e,cal}$  in the PSO model, especially at higher concentrations, suggest some variability in the adsorption kinetics of RhB on NiO/AC. These variations could be attributed to experimental uncertainties, sample inhomogeneity, or other factors affecting the adsorption process.

The  $K_2$  values generated from the pseudo-second-order (PSO) model represent the adsorption rate constant in g/mg·min.  $K_2$  can be regarded in the context of the adsorption process as the rate at which the adsorbate (RhB) molecules are adsorbed onto the surface of the NiO/AC electrode. The  $K_2$  values observed in the current investigation for the different concentrations of RhB (7, 8, and 9 ppm) are rather tiny, implying that the adsorption rate is relatively slow. This is due to the fact that the NiO/AC electrode is a heterogeneous material with a limited number of active sites on its surface. Nevertheless, the low  $K_2$  values obtained by the PSO model can be viewed as a benefit. In practical applications, a low adsorption rate is frequently desired since it enables a more complete and increasingly efficient adsorption to occur over a longer period of time. In particular, a low adsorption rate might lead to a higher adsorption capacity in the long term since more adsorbate molecules can be accommodated on the adsorbent material's surface. In general, the  $K_2$  values derived from the PSO model in this study indicate that the NiO/AC electrode has the potential to be an effective adsorbent material for RhB removal; however, the adsorption process may be delayed due to a lack of active sites on its surface.

Modeling isotherms allow us to estimate various equilibrium parameters, the degree of affinity, and probable interactions between the adsorbent and adsorbate. The adsorption capacity of NiO/AC for RhB molecules was investigated utilizing Langmuir and Freundlich isotherms (Table 1).

The interpretation of the isotherm models in this section highlights the ability of these models to provide insights into the adsorption mechanism and behavior of the NiO/AC electrode as an adsorbent material for RhB molecules.

The Langmuir isotherm model [51] assumes that the adsorption occurs on a surface with a uniform level of energy, and that there is no interaction between adsorbate molecules. According to the results presented in Table 2, the Langmuir model was found to be a poor fit for the experimental data, with relatively low correlation coefficients ( $R^2$ ). This suggests that the assumption of uniform energy levels is not applicable to the adsorption of RhB molecules onto the NiO/AC electrode.

**Table 2.** Effect of RhB concentration on kinetic parameters for RhB adsorption onto NiO/AC.

|       | Pseudo-First-Order Model |                  |       | Pseudo-Second-Order Model |                     |       |
|-------|--------------------------|------------------|-------|---------------------------|---------------------|-------|
|       | $Q_{e,cal}$<br>(mg/g)    | $K_1$<br>(1/min) | $R^2$ | $Q_{e,cal}$<br>(mg/g)     | $K_2$<br>(g/mg·min) | $R^2$ |
| 7 ppm | $149.20 \pm 5.70$        | 0.10             | 0.912 | $173.29 \pm 9.01$         | 0.0007              | 0.941 |
| 8 ppm | $150.14 \pm 6.97$        | 0.08             | 0.969 | $199.25 \pm 8.84$         | 0.0005              | 0.987 |
| 9 ppm | $168.88 \pm 10.83$       | 0.18             | 0.974 | $180.88 \pm 18.17$        | 0.002               | 0.981 |

On the other hand, the Freundlich isotherm model [52] accounts for the heterogeneity of the adsorbent surface and the occurrence of interactions between adsorbate molecules. The computed parameters and correlation coefficients for the Freundlich model, as shown in Table 3, suggest that it provides a better fit to the experimental data, with higher correlation coefficients ( $R^2 > 0.93$ ). This indicates that the adsorption process is complex, involving interactions between RhB molecules and a surface with different energy levels.

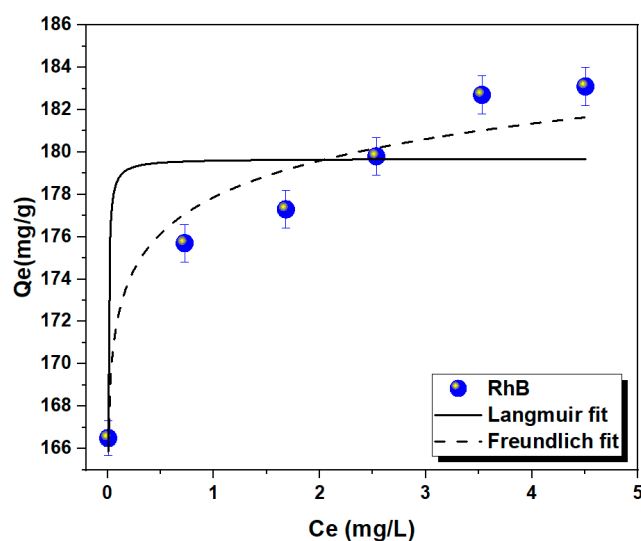
**Table 3.** Isotherm modeling parameters for RhB adsorption onto NiO/AC at 1.6 V.

| Models     | Parameters                         | RhB               |
|------------|------------------------------------|-------------------|
| Langmuir   | $Q_{max}$ (mg/g)                   | $180.03 \pm 1.44$ |
|            | $K_L$ (L/mg)                       | 2660.48           |
|            | $R^2$                              | 0.779             |
| Freundlich | $K_F$ (mg/g) (L/mg) <sup>1/n</sup> | $178.22 \pm 0.66$ |
|            | 1/n                                | 0.01              |
|            | $R^2$                              | 0.935             |

The fitting charts presented in Figure 6 also provide a visual representation of the fit of the two isotherm models to the experimental data. The Freundlich model appears to fit the experimental data more closely than the Langmuir model, further supporting the conclusion that the adsorption of RhB molecules onto the NiO/AC electrode is a multilayer process involving interactions between the adsorbate molecules and the heterogeneous surface of the electrode.

Overall, the results from the isotherm modeling suggest that the NiO/AC electrode has a high affinity for RhB molecules and has the potential to be an effective adsorbent material for RhB removal from wastewater. However, the complexity of the adsorption process highlights the need for further investigation to fully understand the underlying mechanisms and optimize the performance of the electrode as an adsorbent material.

Upon comparing the adsorption capacity of RhB determined from the Langmuir model (180.03 mg/g) with literature values, it is evident that NiO/AC shows a superior performance compared to various other adsorbents (Tables 3 and 4). The adsorption capacity of NiO/AC surpasses that of granules prepared from oil palm empty fruit bunch ( $Q_m = 69.86$  mg/g) [50], black tea leaves ( $Q_m = 53.12$  mg/g) [51], modified bentonite clay ( $Q_m = 155.10$  mg/g) [2], and phosphoric acid treated parthenium carbon ( $Q_m = 59.17$  mg/g) [52].



**Figure 6.** Langmuir and Freundlich isotherm modeling of the RhB adsorption on NiO/AC at 1.6 V.

**Table 4.** Comparison results of different adsorbents used for RhB removal.

| Method     | Adsorbent                                                                         | pH  | $t_{eq}$<br>(min) | $q_m$<br>(mg/g) | Refs. |
|------------|-----------------------------------------------------------------------------------|-----|-------------------|-----------------|-------|
| Adsorption | Activated carbon prepared from the steel and fertilizer industries                | 5.5 | 30                | 91.1            | [53]  |
|            | Activated carbon prepared from walnut shell                                       | 9.0 | 300               | 18.70           | [54]  |
|            | Activated carbon prepared from an indigenous waste                                | 7.1 | 100               | 51.55           | [55]  |
|            | Cationic surfactant (hexadecyltrimethylammonium chloride) modified bentonite clay | 6.8 | 250               | 155.20          | [5]   |

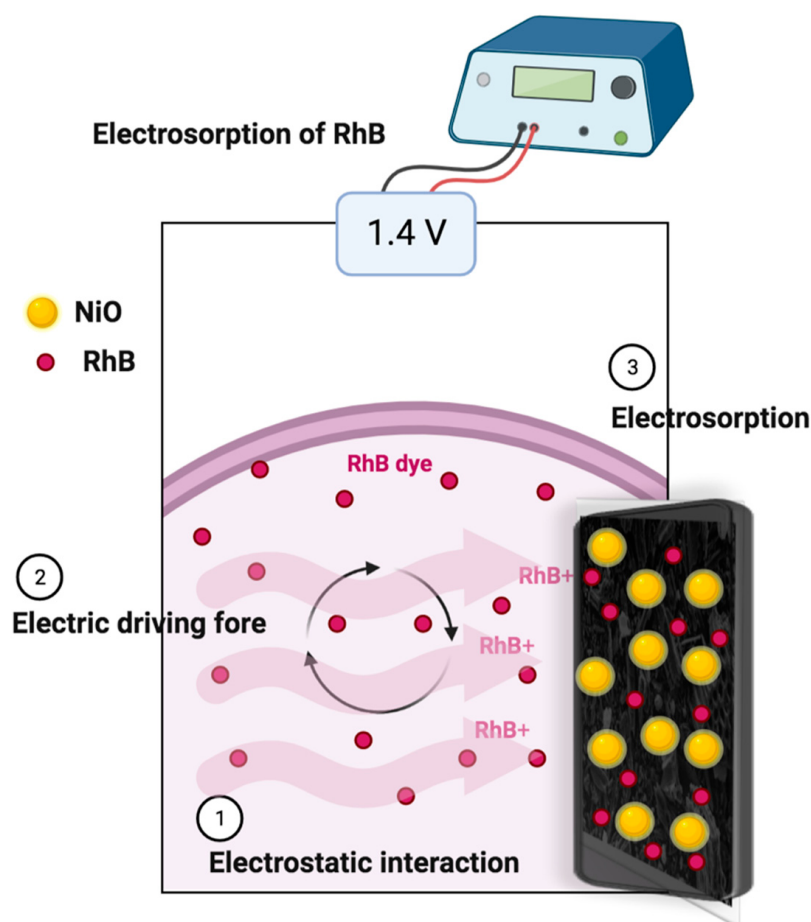
Furthermore, the comparison with other adsorbents provides additional insights into their respective adsorption capacities and properties (Table 4). For instance, activated carbon prepared from the steel and fertilizer industries exhibited an adsorption capacity of 91.1 mg/g at pH 5.5 [53]. Activated carbon derived from walnut shell demonstrated a lower adsorption capacity of 18.70 mg/g at pH 9.0 [54]. Activated carbon derived from an indigenous waste showcased an adsorption capacity of 51.55 mg/g at pH 7.1 [55]. Finally, cationic surfactant-modified bentonite clay achieved a higher adsorption capacity of 155.20 mg/g at pH 6.8 [5].

Comparing the results indicates that NiO/AC outperforms these adsorbents in terms of adsorption capacity, making it a highly promising material for the removal of RhB from wastewater. The incorporation of NiO particles into activated carbon enhances its adsorption capabilities, even with a reduction in surface area due to pore blocking. These findings highlight the potential of NiO/AC as an efficient and effective adsorbent for water treatment applications, particularly in the removal of RhB. Further studies can explore the optimization of NiO/AC synthesis and investigate its performance with other dyes and pollutants to fully understand its broader applications in water purification.

#### *Possible Mechanism of the Electrosorption*

Herein, a possible mechanism for the electrosorption of RhB on the NiO/AC electrode may be suggested according to the predicted steps below (Figure 7) [56–58]: The NiO/AC electrode is being prepared by deposition of a layer of nickel oxide (NiO) nanoparticles on the activated carbon (AC) surface. Its high surface area and porous structure of AC helps the NiO nanoparticles to disperse, thus creating a composite electrode with improved adsorptive properties. However, when the NiO/AC electrode is immersed in a solution that contains RhB, both electrostatic and chemical interactions take place between the RhB molecules and the surface of the NiO/AC electrode. RhB molecules with a positive

charge are attracted to the negatively charged NiO/AC electrode, which initiates the electroadsorption process. When an external electric potential is imposed, an electric field is created between the NiO/AC electrode and the solution which contains RhB. The driving force for RhB adsorption on the electrode surface is strengthened by means of this electric field [56]. RhB positively charged molecules are moving toward the electrode surface, where multilayers are formed as a consequence of the heterogeneous character of the NiO/AC composite. Therefore, the NiO nanoparticles serve as electroactive sites, aiding electron transfer and further boosting the electrostatic interactions between RhB molecules and the electrode surface. Briefly, the electroresorption of RhB on the NiO/AC electrode entails a combined electrostatic and chemical interaction between RhB molecules and the electrode surface, along with charge transfer phenomena mediated as a result of the electrical potential applied.



**Figure 7.** Proposed adsorption mechanism of RhB on the NiO/AC electrode.

## 5. Experimental Section and Characterization

### 5.1. Materials Synthesis

The chemicals utilized in this investigation were of analytical reagent quality and were employed without any purification. The activated carbon made from almond shells was produced through chemical activation using KOH. The process involved combining 20 g of almond shell with 10 g of 90% KOH (obtained from Sigma-Aldrich, Darmstadt, Germany) and 100 mL of distilled water to form a solution. The solution was then heated to 60 °C for 12 h and dried at 110 °C. The dried sample was subsequently subjected to pyrolysis under N<sub>2</sub> flow (200 cm<sup>3</sup>/min) for 2 h at 300 °C, followed by 3 h at 800 °C, with a heating rate of 10 °C/min.

NiO nanoparticles were synthesized using the co-precipitation technique. The synthesis procedure involved the following steps:

**Preparation of Nickel Nitrate Solution:** A required quantity of nickel nitrate  $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  was added to 50 mL of milli-Q water. This solution served as the precursor for NiO nanoparticles. **Addition of Sodium Hydroxide (NaOH):** A 0.5 M NaOH solution was slowly added dropwise to the nickel nitrate solution while maintaining steady stirring. The addition of NaOH was carried out until the pH of the solution reached the range of 9–10. This pH range was chosen to facilitate the precipitation process. **Precipitation and Washing:** The addition of NaOH resulted in the formation of a precipitate containing nickel hydroxide. The resultant precipitate was washed multiple times with ethanol and milli-Q water to remove impurities and any remaining reactants. **Calcination:** The washed precipitate was then subjected to calcination at a temperature of 300 °C for a duration of 3 h. Calcination involves heating the material to a high temperature to induce chemical and structural transformations, leading to the formation of the desired NiO nanoparticles. To prepare the NiO/AC composite, 10 weight percent of the synthesized NiO nanoparticles was utilized. The composite formation involved the following steps: **Filtration and Rinsing:** The NiO nanoparticles were filtered to separate them from the remaining solution. The collected nanoparticles were then rinsed several times with distilled water and ethanol to remove any residual impurities or reactants. **Drying:** After the rinsing process, the NiO/AC composite was dried for a duration of 12 h at a temperature of 70 °C. This drying step aimed to remove any remaining moisture and solvent, resulting in a dry composite material ready for further characterization or application.

Overall, the synthesis of NiO nanoparticles involved the co-precipitation of nickel nitrate with sodium hydroxide, followed by washing, calcination, and subsequent formation of the NiO/AC composite by combining the synthesized NiO nanoparticles with activated carbon. These steps were designed to ensure the formation of high-quality NiO nanoparticles and the successful integration of NiO into the composite material.

## 5.2. Electroadsorption Study

For conducting the adsorption batch system tests, 150 mL of electrochemical cells were filled with 50 mL of RhB solutions and continuously stirred. Thereafter, several essential parameters were thoroughly investigated. The concentration of pollutants was measured at their specific maximum wavelengths of 554 nm using an UV-Vis spectrophotometer (UV 2300) after a defined duration. The amount of adsorption and percentage of removal under different conditions were calculated using the following formulas:

$$Q_{\text{ads}} = (C_0 - C_e) \times V/m \text{ (mg/g)}$$

$$\% \text{ Removal} = ((C_0 - C_e)/C_0) \times 100$$

The starting and balanced concentrations of dyes are indicated by  $C_0$  (mg/L) and  $C_e$  (mg/L), correspondingly. The weight of the adsorbent is denoted by  $m$  (g) and the adsorbate solution's volume is represented by  $V$  (L).

**X-ray diffraction:** X-ray diffraction (XRD) patterns were obtained utilizing an EMPYR EAN PANALYTICAL diffractometer (Malvern Panalytical, Malvern, UK). The instrument was operated at 45 kV/35 mA, employing  $\text{CuK}\alpha$  radiation with a Ni filter. The XRD measurements were conducted in continuous mode, and a step size of  $0.01^\circ$  was employed.

**Scanning electron microscopy:** The NiO/AC morphology and local composition were observed using scanning electron microscopy (SEM) analysis. The chemical composition was determined by scanning the sample surfaces with Energy Dispersive Spectroscopy (EDS) mode.

**Surface area:** To calculate the BET surface area ( $S_{\text{BET}}$ ), the nitrogen adsorption and desorption isotherm were utilized, and an AUTOSORB-1 surface area and pore size analyzer at 77 K was used to determine the specific surface area and pore size distribution.

Infrared spectroscopy: Using a Shimadzu 4800S (Shimadzu Corp., Kyoto, Japan), the Fourier Transform Infrared spectra of the AC sample were acquired in the mid-infrared range ( $400\text{--}4000\text{ cm}^{-1}$ ). The spectra were scanned with a resolution of  $2.0\text{ cm}^{-1}$  and 20 scanning passes.

## 6. Conclusions

In conclusion, this study investigated the incorporation of NiO particles into activated carbon (AC) and demonstrated its significant enhancement of electrochemical capacity and adsorption capability for Rhodamine B (RhB) dye. The NiO/AC composite was successfully prepared from almond shell biomass and thoroughly characterized using various analytical techniques. The electrochemical adsorption of RhB dye from simulated wastewaters using the NiO/AC composite exhibited a voltage-dependent behavior, with a remarkable 100% removal efficiency achieved at 1.4 V. These findings suggest a promising electroadsorption mechanism of NiO/AC for the effective removal of RhB dye from wastewater. The Freundlich isotherm model was found to best describe the electroadsorption data, providing insights into the adsorption capacity of the NiO/AC composite. The maximum adsorption capacities ranged from 149 to 168 mg/g, indicating the composite's high adsorption performance. As a proof of concept, this study confirmed the applicability of the NiO/AC composite strategy for the capacitive deionization of RhB dye. However, for future work, it is recommended to further investigate the electroadsorption mechanism of NiO/AC and optimize the process parameters. Additionally, the use of density functional theory (DFT) calculations could be employed to predict the effects of different parameters and enhance the adsorption capacity and efficiency. Furthermore, it is crucial to explore the application of NiO/AC on the removal of other dyes and pollutants from wastewater, expanding its potential in wastewater treatment. The innovative aspect of this composite strategy, utilizing biomass-derived AC, opens up possibilities for its utilization in battery systems (e.g., hard carbon electrode), supercapacitors, and electrocatalysis. Overall, the incorporation of NiO particles into AC demonstrated its efficacy in enhancing electrochemical capacity and adsorption performance for RhB dye. This study provides valuable insights into the potential of the NiO/AC composite for various applications and highlights avenues for future research and optimization.

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