



Thermodynamic Evaluation of Energy Storage and Recovery in Capacitive Deionization with Synergistic Faradaic Double Layer Electrodes

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Abstract: Capacitive deionization (CDI) is increasingly regarded as an energy-efficient desalination technology, and numerous studies have explored transition-metal oxide-carbon composites as pseudocapacitive CDI electrodes, typically reporting salt adsorption capacities of about 8–15 mg/g and energy-recovery efficiencies of 60–80%. However, most previous studies provide limited insight into how the supplied electrical energy is partitioned into recoverable and dissipated components during CDI operation, and a unified thermodynamic framework remains lacking. In this work, $\text{Mn}_3\text{O}_4@\text{C}$, $\text{Co}_3\text{O}_4@\text{C}$, and $\text{Fe}_3\text{O}_4@\text{C}$ core-shell nanoparticles were incorporated into activated carbon to fabricate $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$, $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$, and $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$ composite electrodes. Under identical CDI conditions of 3000 mg/L NaCl and an applied charging voltage of 0.6 V, AC, $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$, $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$, and $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$ exhibited salt adsorption capacities of 6.7, 12.5, 10.8, and 9.3 mg/g, respectively. Under the same operating conditions, the three composite electrodes exhibited energy storage efficiencies of approximately 84 to 89%, compared with 54% for AC, with $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ showing the highest energy storage performance. Representative GCD measurements of $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ and open-circuit voltage-retention measurements of $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ and $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$ provided supporting operational information, whereas the cross-electrode energy metrics were derived from a common CDI charging and discharging protocol applied to all electrodes. Energy analysis showed that increasing the charging voltage systematically decreased energy storage efficiency because a smaller fraction of the supplied electrical energy was recovered during discharge. These findings provide a thermodynamic basis for designing energy-recoverable CDI systems with potentially lower net electrical energy demand.

Keywords: energy recovery; capacitive deionization; pseudocapacitive; core-shell nanoparticle

1. Introduction

Ensuring reliable, affordable, and safe water supplies for domestic, agricultural, and industrial uses remains a pressing global challenge in the coming decades [1,2]. Although the planet is covered by water, only a small fraction is available as readily usable freshwater, while the overwhelming majority is present as saline seawater [3,4]. This imbalance has stimulated sustained interest in desalination technologies capable of converting saltwater into potable or process-grade water in an efficient and sustainable manner. Reverse osmosis (RO) has become the dominant technology for seawater and brackish-water desalination and is widely deployed at large scale [5–7]. Nevertheless, RO systems require substantial electrical energy to drive water transport across semipermeable membranes and to maintain high operating pressures, resulting in significant operating costs and limiting broader implementation in regions with constrained energy resources [7–10].

Capacitive deionization (CDI) is regarded as a suitable option for brackish and moderately saline waters, as it operates at relatively low cell voltages and offers the possibility of recovering a portion of the electrical energy used during operation [11–13]. In a typical CDI process, ions in the feed solution are attracted to and temporarily stored within electric double layers (EDLs) that form inside porous carbon electrodes when an external potential is applied [14,15]. Once the cell is short-circuited or the potential is reversed, the adsorbed ions are released back into the electrolyte, enabling electrode regeneration and allowing the treated water to be collected. In CDI, activated



carbon, carbon fibers, carbon nanotubes, and three-dimensional graphene-type carbons are commonly chosen as electrode materials, mainly due to their high specific surface area and favorable electrical conductivity [16–19]. Beyond high specific surface area and electrical conductivity, an effective carbon electrode for CDI should possess an accessible pore network, adequate electrolyte wettability, chemically stable surface functionalities, and low interparticle and electrode-current-collector contact resistance. Micropores provide a large number of ion-storage sites, whereas mesopores and macropores facilitate electrolyte penetration and shorten ion-transport pathways. Consequently, the balance among accessible surface area, hierarchical porosity, surface chemistry, wettability, and electrical conductivity strongly governs salt adsorption capacity, charge efficiency, and rate performance.

The charge-storage behavior of CDI electrodes is closely related to that of electrochemical capacitors [20,21]. These devices are commonly divided into electric double-layer capacitors, in which charge is accumulated electrostatically within the interfacial double layer, and pseudocapacitors, where surface or near-surface redox processes provide additional capacitance [22,23]. Owing to this combination of electrostatic and Faradaic storage mechanisms, electrochemical capacitors can deliver high power outputs, respond quickly to charge-discharge cycling and sustain a large number of operating cycles [24]. Although CDI typically operates at relatively low voltages, typically around 0.8–1.4 V, coupling CDI with energy-recovery concepts may reduce the net electrical energy demand and, depending on the electricity source, the associated carbon emissions [25]. For instance, in a membrane-CDI system, previous studies have reported that a substantial fraction of the input energy, reaching up to about 83%, can be recovered during the regeneration step [26].

In our previous work, the principal objective was to enhance CDI desalination performance using $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ electrodes. At 1000 mg/L NaCl and an applied voltage of 1.2 V, the $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ electrode achieved an electrosorption capacity of 25 mg/g, while a maximum value of 29 mg/g was obtained when polarity-reversal regeneration was applied. While our previous work focused primarily on maximizing CDI desalination performance, the present study investigates how electrode composition and operating conditions affect energy storage, recovery, and dissipation. Rather than maximizing salt adsorption capacity, the present study comparatively evaluates $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$, $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$, and $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$ electrodes and quantifies the electrical energy supplied during charging, the energy recovered during discharge, and the resulting energy storage efficiency under different operating conditions. The novelty of this work lies in its comparative energy-recovery analysis and its evaluation of the relationships among electrode composition, applied voltage, electrolyte concentration, salt removal, and energy storage efficiency [27].

2. Materials and Methods

2.1. Chemicals and Materials

All chemicals and materials were used as received unless otherwise stated. Manganese(II) nitrate tetrahydrate ($\text{Mn}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$, Sigma-Aldrich, 98%), cobalt(II) nitrate hexahydrate ($\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, Alfa Aesar, 98%), iron(III) nitrate nonahydrate ($\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, Sigma-Aldrich, St. Louis, MO, USA, ACS reagent, $\geq 98\%$), and β -cyclodextrin (β -CD, Sigma-Aldrich, St. Louis, MO, USA) were used for the synthesis of the core-shell nanoparticles. Activated carbon (AC, Goldstar Carbon Tech Inc., Taichung, Taiwan), poly(vinylidene fluoride) (PVDF, Sigma-Aldrich, St. Louis, MO, USA), and N-methyl-2-pyrrolidone (NMP, Sigma-Aldrich, St. Louis, MO, USA) were used for CDI electrode fabrication. Analytical-grade sodium chloride (NaCl, Sigma-Aldrich, St. Louis, MO, USA, ACS reagent, $\geq 99.0\%$) was used to prepare the CDI feed solutions.

2.2. Preparation of Core-Shell Nanoparticles

Manganese(II) nitrate tetrahydrate, cobalt(II) nitrate hexahydrate, and iron(III) nitrate nonahydrate were used as the metal precursors for the separate preparation of $\text{Mn}_3\text{O}_4@\text{C}$, $\text{Co}_3\text{O}_4@\text{C}$, and $\text{Fe}_3\text{O}_4@\text{C}$, respectively. Each metal precursor was separately complexed with β -cyclodextrin in 200 mL of deionized water at a carbon-to-metal molar ratio of 6. Each mixture was stirred at 298 K for 2 h and subsequently dried at 333 K for 8 h to obtain a solid intermediate. The resulting solids were heat-treated separately in a tubular furnace at 673 K for 4 h under a continuous flow of high-purity nitrogen at 100 mL/min. During the heat treatment, β -cyclodextrin was carbonized to form the carbon shell, while the metal precursors were converted into the corresponding metal oxide cores. The resulting products were denoted as $\text{Mn}_3\text{O}_4@\text{C}$, $\text{Co}_3\text{O}_4@\text{C}$, and $\text{Fe}_3\text{O}_4@\text{C}$ core-shell nanoparticles, respectively, and were subsequently incorporated into activated carbon electrodes as pseudocapacitive components, as schematically illustrated in Figure 1.

2.3. Fabrication of CDI Electrodes

The CDI electrodes were fabricated by blending activated carbon with the as-prepared $\text{Mn}_3\text{O}_4@\text{C}$ nanoparticles. The active-material mixtures were prepared by replacing AC with 5, 10, or 15 wt.% $\text{M}_3\text{O}_4@\text{C}$ while maintaining a constant combined mass of AC and $\text{M}_3\text{O}_4@\text{C}$. The $\text{M}_3\text{O}_4@\text{C}$ content was defined as $m_{\text{M}_3\text{O}_4@\text{C}}/(m_{\text{AC}} + m_{\text{M}_3\text{O}_4@\text{C}}) \times 100\%$. PVDF and NMP were subsequently added at 10 and 20 wt.%, respectively, relative to the combined mass of AC and $\text{M}_3\text{O}_4@\text{C}$. Accordingly, the AC: $\text{M}_3\text{O}_4@\text{C}$:PVDF:NMP mass ratios were 95:5:10:20, 90:10:10:20, and 85:15:10:20 for the 5, 10, and 15 wt.% $\text{M}_3\text{O}_4@\text{C}$ formulations, respectively. The mixture was stirred at 298 K for 3 h without sonication to form a homogeneous slurry. The graphite current collectors were sequentially rinsed with deionized water and ethanol, dried, and weighed before coating. The slurry was drop-cast onto each graphite current collector over an exposed geometric area of $2.5 \times 2.5 \text{ cm}^2$. The coated electrodes were dried at 353 K for 6 h to remove residual NMP. After cooling to room temperature, the electrodes were weighed to verify the dry coating mass. The amount of slurry applied was controlled to obtain a total dry coating mass of 0.500 g per electrode, including AC, $\text{M}_3\text{O}_4@\text{C}$, and PVDF but excluding the graphite current collector. This corresponded to a total dry coating loading of 80.0 mg/cm^2 . Based on the dry AC: $\text{M}_3\text{O}_4@\text{C}$:PVDF mass ratios of 95:5:10, 90:10:10, and 85:15:10, the $\text{M}_3\text{O}_4@\text{C}$ loadings were 22.7, 45.5, and 68.2 mg per electrode for the 5, 10, and 15 wt.% $\text{M}_3\text{O}_4@\text{C}$ formulations, respectively. These values corresponded to $\text{M}_3\text{O}_4@\text{C}$ areal loadings of 3.64, 7.27, and 10.91 mg/cm^2 , respectively.

2.4. Electrochemical and CDI Operating Protocols

Three electrochemical operating protocols were used. First, the common protocol used for cross-electrode comparison consisted of potentiostatic charging for 1 h at applied cell voltages of 0.4, 0.6, 0.8, and 1.2 V, followed by galvanostatic discharge at a constant areal current density of 1 mA/cm^2 until the cell voltage reached 0 V. Comparisons of electrode composition and NaCl concentration were conducted at 0.6 V, whereas the effect of applied voltage on energy recovery was evaluated at 0.6, 0.8, and 1.2 V. The energy storage efficiency and recoverable stored energy density reported for AC, $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$, $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$, and $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$ were calculated exclusively from this common protocol. Second, separate GCD measurements were performed only for the $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ cells at constant charging and discharging currents of 1 or 10 mA in 1000 or 3000 mg/L NaCl solutions. These GCD profiles were collected to illustrate the operational voltage response and energy hysteresis of $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ under different current and salinity conditions; they were not used for cross-electrode comparison or for comparison of intrinsic capacitance. Third, open-circuit voltage-retention experiments were conducted by first charging the cells potentiostatically to 0.6 or 0.8 V. The cells were then disconnected from the power supply and maintained under open-circuit conditions for 500 s. The cell voltage was recorded immediately after disconnection and at the end of the 500 s retention period.

2.5. Characterization

The morphology and microstructure of the core-shell nanoparticles were examined using high-resolution transmission electron microscopy (TEM, JEOL JEM-3010 and JEM-2100F) and scanning electron microscopy (SEM, JEOL JSM-6700). Crystal structures were examined by X-ray diffraction (XRD, Bruker D8 Advance) using Cu $K\alpha$ radiation at 40 kV and 40 mA with a scan rate of $5^\circ/\text{min}$. The apparent crystallite size was estimated using the Scherrer Equation (1):

$$D = \frac{K\lambda}{\beta \cos \theta} \quad (1)$$

where D is the apparent crystallite size in nm, K is the shape factor of 0.9, λ is the wavelength of Cu $K\alpha$ radiation of 0.15406 nm, β is the full width at half maximum of the selected diffraction peak in radians, and θ is the corresponding Bragg angle. The Mn_3O_4 (211), Co_3O_4 (311), and Fe_3O_4 (311) reflections were used for the crystallite-size calculations. Peak positions and FWHM values were obtained by peak fitting using Jade 5.0 software. No correction for instrumental broadening was applied.

Nitrogen adsorption and desorption isotherms were measured using a 3Flex Physisorption Analyzer (Micromeritics). Before analysis, the $\text{Fe}_3\text{O}_4@\text{C}$ core-shell nanoparticle powder was degassed under vacuum at 378 K for 12 h. The BET specific surface area was determined from the nitrogen adsorption isotherm.

The impedance characteristics of the AC, $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$, $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$, and $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$ electrodes were evaluated by electrochemical impedance spectroscopy using an Autolab potentiostat. A conventional three-electrode configuration was used, in which the AC or $\text{M}_3\text{O}_4@\text{C}/\text{AC}$ composite electrode served as the working electrode, a platinum electrode served as the counter electrode, and an Ag/AgCl electrode served as the reference

electrode. EIS measurements were conducted at the open-circuit potential in 1.0 M NaCl using the same three-electrode configuration over a frequency range of 100 kHz to 0.01 Hz with a sinusoidal potential perturbation amplitude of 10 mV. The GCD operating conditions for the $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ cells are described in Section 2.4.

For CDI tests, the cell voltage and current were monitored using a potentiostat (Autolab) during charging and discharging steps. The electrosorption performance was evaluated from conductivity changes of NaCl solutions (100, 1000, and 3000 mg/L), and the electrosorption efficiency was calculated as

$$(C_i - C_f)/C_i \times 100\% \quad (2)$$

where C_i and C_f are the initial and final NaCl concentrations in mg/L, respectively. The salt adsorption capacity (SAC) was calculated as

$$\text{SAC} = \frac{(C_i - C_f)V}{1000 m} \quad (3)$$

where V is the solution volume in mL, and m is the total dry coating mass of both electrodes in g, excluding the graphite current collectors. The calculated SAC is expressed in mg/g. The charge efficiency Λ was defined as the ratio of the charge associated with salt removal to the total charge delivered to the electrodes during CDI. It was calculated using Equation (4)

$$\Lambda = FV(C_{\text{ch}} - C_{\text{dis}})/(10^6 M \int |I(t)| dt) \quad (4)$$

where V refers to the solution volume (mL), C_{ch} and C_{dis} refer to the influent and effluent concentrations of NaCl solution (mg/L), respectively, I is the absolute value of current during CDI, and F and M refer to the Faraday constant (96,485 C/mol) and molar mass of NaCl (58.5 g/mol), respectively. Energy storage efficiency was obtained by comparing the energy recovered during discharge with the energy supplied during charging, whereas charge efficiency Λ was calculated as the fraction of the total charge passed through the electrode that corresponded to net salt removal.

2.6. Energy Recovery

Energy storage efficiency was defined as the ratio of the electrical energy recovered during discharge to the electrical energy supplied during charging. To accommodate the different charging and discharging control modes used in this study, the supplied and recovered energies were calculated using general time integrated expressions. The electrical energy supplied during charging, E_{ch} , was calculated as:

$$E_{\text{ch}} = \int_{t_{\text{ch},0}}^{t_{\text{ch},f}} V_{\text{cell}}(t) I_{\text{ch}}(t) dt \quad (5)$$

The electrical energy recovered during discharge, E_{dis} , was calculated as:

$$E_{\text{dis}} = \int_{t_{\text{dis},0}}^{t_{\text{dis},f}} V_{\text{cell}}(t) |I_{\text{dis}}(t)| dt \quad (6)$$

The energy storage efficiency, η_{ES} , was calculated as:

$$\eta_{\text{ES}} = \frac{E_{\text{dis}}}{E_{\text{ch}}} \times 100\% \quad (7)$$

The recoverable stored energy density, e_{st} , was calculated as:

$$e_{\text{st}} = \frac{1000 E_{\text{dis}}}{A} \quad (8)$$

where $V_{\text{cell}}(t)$ is the measured cell voltage in V, $I_{\text{ch}}(t)$ and $I_{\text{dis}}(t)$ are the charging and discharging currents in A, respectively, t is time in s, and A is the exposed geometric electrode area of 6.25 cm². The calculated values of E_{ch} and E_{dis} are expressed in J, and e_{st} is expressed in mJ/cm². During potentiostatic charging, V_{cell} was maintained at the specified applied voltage while I_{ch} varied with time. During galvanostatic discharge, I_{dis} was maintained constant while V_{cell} decreased with time.

The total energy that was not recovered under the specified discharge protocol was calculated as the difference between the energy supplied during charging and the energy recovered during discharge:

$$E_{\text{unrecovered}} = E_{\text{ch}} - E_{\text{dis}} \quad (9)$$

This difference was reported as the total unrecovered energy under the specified charging and discharging protocol. The scope and limitations of this aggregate term are discussed in Section 3.4.

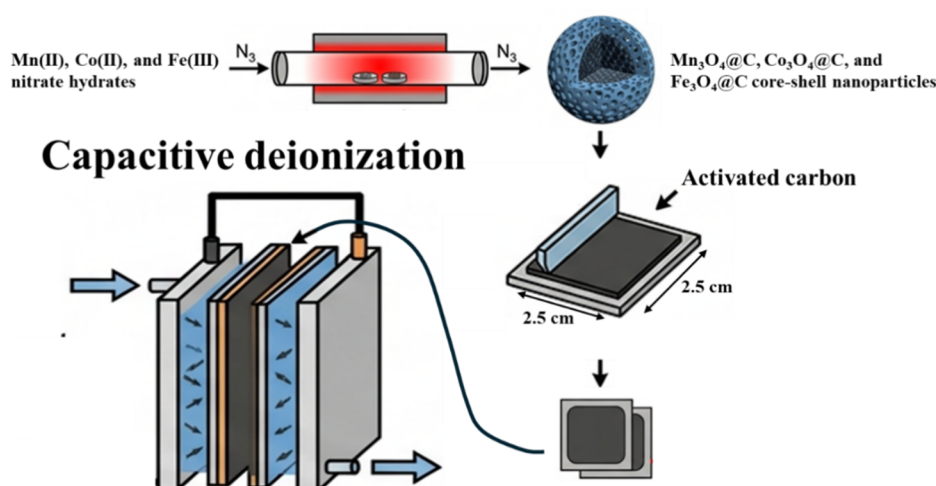


Figure 1. Schematic illustration of the preparation and CDI application of the $\text{Mn}_3\text{O}_4\text{@C/AC}$ composite electrodes.

3. Results and Discussion

3.1. Morphology and Structure of $\text{M}_3\text{O}_4\text{@C}$ Core-Shell Nanoparticles

Figure 2 compares the experimental XRD patterns of the three $\text{M}_3\text{O}_4\text{@C}$ nanoparticles with the corresponding reference patterns. The black, red, and blue vertical bars represent the reference patterns of Co_3O_4 (PDF No. 42-1467), Mn_3O_4 (PDF No. 24-0734), and Fe_3O_4 (PDF No. 96-900-7645), respectively. The diffraction reflections of $\text{Co}_3\text{O}_4\text{@C}$ are consistent with the (111), (220), (311), (222), (400), (422), (511), and (440) planes of spinel Co_3O_4 . The $\text{Mn}_3\text{O}_4\text{@C}$ pattern contains the characteristic reflections of tetragonal hausmannite Mn_3O_4 , which are indexed to the (101), (112), (200), (103), (211), (004), (220), (105), (312), (303), (321), (224), and (400) planes. The $\text{Fe}_3\text{O}_4\text{@C}$ diffraction pattern is consistent with the (220), (311), (222), (400), (422), (511), and (440) planes of spinel Fe_3O_4 . No additional crystalline impurity peaks were evident within the detection limit of the XRD measurement. The broad diffraction peaks indicate nanoscale crystalline domains. The apparent crystallite sizes estimated using the Scherrer equation were approximately 13 to 14 nm for $\text{Mn}_3\text{O}_4\text{@C}$, 10 to 11 nm for $\text{Co}_3\text{O}_4\text{@C}$, and 34 to 36 nm for $\text{Fe}_3\text{O}_4\text{@C}$.

Figure 3a–c present SEM images of the $\text{Mn}_3\text{O}_4\text{@C}$, $\text{Co}_3\text{O}_4\text{@C}$, and $\text{Fe}_3\text{O}_4\text{@C}$ nanoparticle powders. The three samples exhibit granular morphologies composed of closely packed nanoparticles with observable interparticle voids. No large micron-scale aggregates are evident within the examined regions. These morphological observations suggest the presence of interparticle voids that may provide electrolyte-accessible pathways, although SEM alone does not quantify accessible surface area or pore-size distribution. The BET specific surface area of the $\text{Fe}_3\text{O}_4\text{@C}$ core-shell nanoparticle powder measured in the present study was $515 \text{ m}^2/\text{g}$. For structural context, our previous study reported BET specific surface areas of 715 and $645 \text{ m}^2/\text{g}$ for similarly prepared $\text{Mn}_3\text{O}_4\text{@C}$ and $\text{Co}_3\text{O}_4\text{@C}$ core-shell nanoparticle powders, respectively [27]. All three values refer to the core-shell nanoparticle powders rather than the completed $\text{M}_3\text{O}_4\text{@C/AC}$ composite electrodes. Because the $\text{Mn}_3\text{O}_4\text{@C}$ and $\text{Co}_3\text{O}_4\text{@C}$ measurements were obtained in the previous study rather than in the same analytical batch as $\text{Fe}_3\text{O}_4\text{@C}$, the numerical differences are interpreted cautiously and are not used to attribute the CDI performance quantitatively to BET surface area alone.

Figure 3d–f show high resolution TEM images of the $\text{Mn}_3\text{O}_4\text{@C}$, $\text{Co}_3\text{O}_4\text{@C}$, and $\text{Fe}_3\text{O}_4\text{@C}$ nanoparticles. In each sample, a darker crystalline oxide core containing visible lattice fringes is surrounded by a lower contrast amorphous region. Based on the synthesis route and the observed contrast, this outer region is attributed to the carbon shell derived from β -CD. The carbon shell thickness estimated from the representative high resolution TEM images was approximately 3 to 6 nm. The visible lattice fringes support the crystalline nature of the oxide cores, whereas phase identification and plane assignment are based primarily on the XRD results. These images support the formation of a core shell architecture but do not independently establish the electrical conductivity or redox activity of the carbon shell. The electrochemical implications of this architecture are therefore discussed using the electrochemical measurements available in the present study together with the previously reported evidence in Ref. [27].

The structural and textural parameters of the $\text{M}_3\text{O}_4\text{@C}$ nanoparticles, including the apparent crystallite sizes estimated using the Scherrer equation, typical carbon-shell thicknesses observed by high-resolution TEM, and BET specific surface areas, are summarized in Table 1. Overall, the XRD and TEM results support the formation

of $\text{Mn}_3\text{O}_4@\text{C}$, $\text{Co}_3\text{O}_4@\text{C}$, and $\text{Fe}_3\text{O}_4@\text{C}$ nanoparticles with qualitatively similar core-shell architectures, although their apparent crystallite sizes and measured surface areas differ.

Table 1. Structural and textural parameters of the $\text{M}_3\text{O}_4@\text{C}$ core-shell nanoparticles [27].

Sample	Apparent Crystallite Size (nm)	Carbon-Shell Thickness (nm)	BET Specific Surface Area (m^2/g)
$\text{Mn}_3\text{O}_4@\text{C}$	13–14	3–6	715
$\text{Co}_3\text{O}_4@\text{C}$	10–11	3–6	645
$\text{Fe}_3\text{O}_4@\text{C}$	34–36	3–6	515

Apparent crystallite sizes were estimated from the Mn_3O_4 (211), Co_3O_4 (311), and Fe_3O_4 (311) reflections using the Scherrer equation without correction for instrumental broadening. Carbon-shell thickness ranges were estimated from representative high-resolution TEM images.

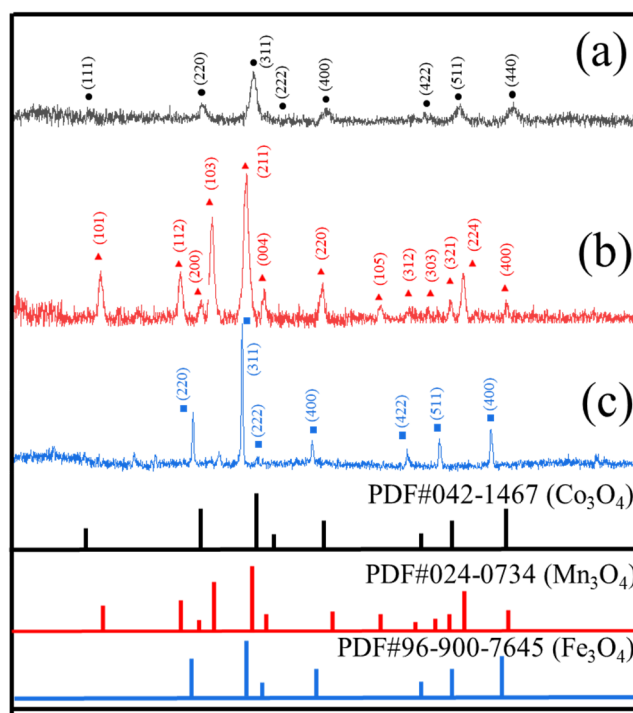


Figure 2. XRD patterns of the (a) $\text{Co}_3\text{O}_4@\text{C}$, (b) $\text{Mn}_3\text{O}_4@\text{C}$, and (c) $\text{Fe}_3\text{O}_4@\text{C}$ core-shell nanoparticles.

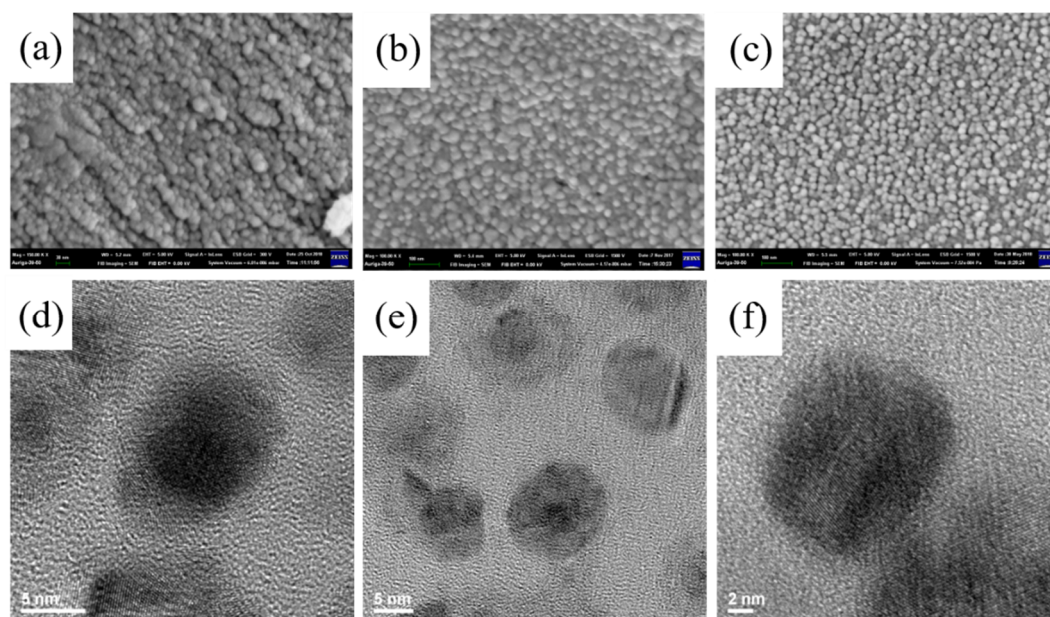


Figure 3. (a–c) SEM images and (d–f) high-resolution TEM images of $\text{Mn}_3\text{O}_4@\text{C}$, $\text{Co}_3\text{O}_4@\text{C}$, and $\text{Fe}_3\text{O}_4@\text{C}$ core-shell nanoparticles.

3.2. Electrochemical Characterization

In Figure 4, the experimental Nyquist plots of the AC, $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$, $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$, and $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$ electrodes exhibit distinct frequency-dependent impedance responses. The higher-frequency responses may include contributions from uncompensated ohmic resistance and interfacial processes, whereas the inclined lower-frequency responses may reflect distributed ion transport and capacitive charge storage within the porous composite electrodes. Because statistically validated equivalent-circuit fitting was not available for all four electrodes, the spectra are interpreted qualitatively and individual resistance or capacitance values are not assigned. Previous CV evidence for $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ and $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$ was reported in Ref. [27], whereas directly comparable CV data were not available for $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$. Accordingly, Figure 4 is used as qualitative supporting electrochemical information, while the scope of the mechanistic interpretation is defined in the consolidated limitations paragraph at the end of Section 3.4.

The open circuit voltage retention behavior of the $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ and $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$ electrodes following normal and polarity reversal (vers) operating sequences is summarized in Table 2. The normal sequence consisted of $+V \rightarrow 0 \rightarrow +V$ operation, whereas the vers sequence consisted of $+V \rightarrow -V \rightarrow +V$ operation. After the final charging step, the cells were disconnected from the power supply, and V_{oc} was recorded immediately and after 500 s. Both electrodes retained a substantial fraction of their initial V_{oc} during the 500 s open circuit period. The calculated voltage retention ranged from 89.0 to 94.2% for $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ under the normal sequence and from 93.0 to 94.6% under the vers sequence. For $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$, the corresponding ranges were 83.9 to 93.9% and 91.8 to 94.4%, respectively [27]. These results indicate that the vers sequence generally resulted in slightly higher short term voltage retention. However, open circuit voltage retention is not equivalent to energy storage efficiency and is used only as supporting evidence of reversible charge retention.

Table 2. Open circuit voltage retention of the $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ and $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$ electrodes immediately after disconnection and after 500 s following normal and polarity reversal operating sequences.

Applied Voltage		$\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$				$\text{Co}_3\text{O}_4@\text{C}/\text{AC}$			
		0.8 V		0.6 V		0.8 V		0.6 V	
NaCl Conc (mg/L)	Open Circuit Time (sec)	Normal	Vers	Normal	Vers	Normal	Vers	Normal	Vers
3000	0	0.792	0.794	0.594	0.595	0.781	0.784	0.592	0.593
	500	0.746	0.751	0.546	0.560	0.711	0.733	0.556	0.560
100	0	0.729	0.751	0.582	0.587	0.757	0.780	0.578	0.576
	500	0.649	0.710	0.535	0.546	0.635	0.716	0.514	0.529

Normal: $+V \rightarrow 0 \rightarrow +V$; Vers: $+V \rightarrow -V \rightarrow +V$

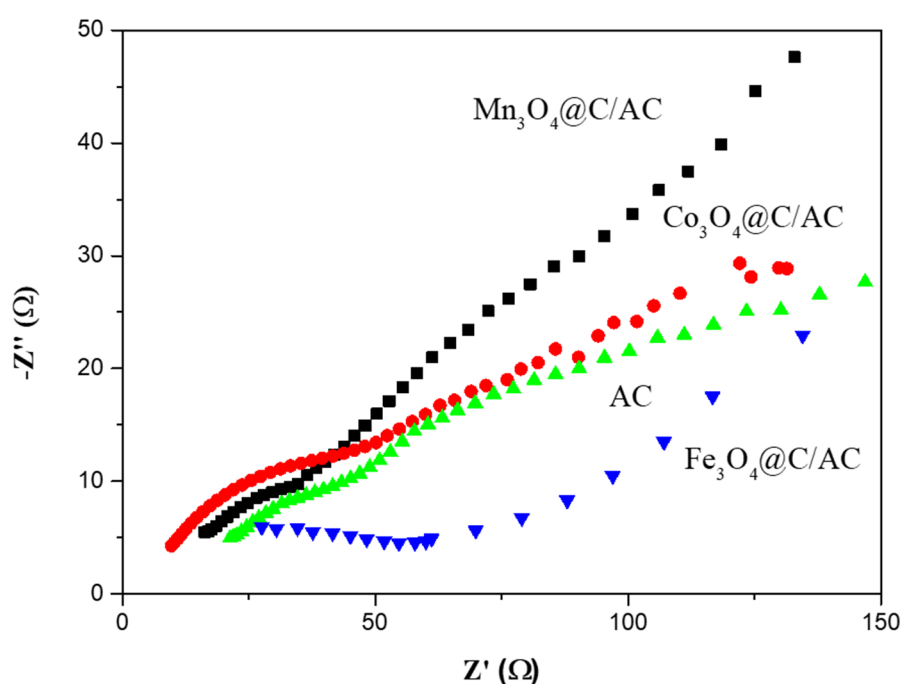


Figure 4. Experimental Nyquist plots of the AC, $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$, $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$, and $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$ electrodes measured at the open circuit potential in 1.0 M NaCl over a frequency range of 100 kHz to 0.01 Hz using a sinusoidal potential perturbation amplitude of 10 mV.

Under the same operating conditions used for the principal energy comparison, namely 3000 mg/L NaCl, potentiostatic charging at 0.6 V for 1 h, and subsequent galvanostatic discharge, the SAC values of AC, $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$, $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$, and $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$ were 6.7, 12.5, 10.8, and 9.3 mg/g, respectively. The values were normalized using the total dry coating mass of both electrodes, excluding the graphite current collectors. Figure 5 compares the charge efficiencies (Λ) of $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$, $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$, and $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$ in 100, 1000, and 3000 mg/L NaCl at applied voltages from 0.4 to 1.2 V. Charge efficiency quantifies the fraction of delivered charge associated with net salt removal and is distinct from energy storage efficiency. A high Λ indicates that a large fraction of delivered charge contributes to net salt removal, whereas a low Λ indicates a larger contribution from processes that do not produce net salt removal, including coion expulsion.

The relatively high Λ values of $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ indicate more effective coupling between delivered charge and net salt removal under the investigated conditions. The increase in Λ at higher NaCl concentrations may reflect improved ionic screening and a reduced relative contribution of coion expulsion or parasitic charge-transfer processes. $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$ exhibited lower charge efficiencies than $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ and $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$ under several investigated conditions, indicating that a smaller fraction of the delivered charge contributed to net salt removal. This behavior may involve coion redistribution, parasitic charge transfer, or reversible charge-storage processes that do not directly remove salt.

Importantly, the lower charge efficiency of $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$ does not necessarily imply a proportionally lower energy storage efficiency. Charge efficiency quantifies net salt removal per unit charge, whereas energy storage efficiency quantifies recovered electrical energy relative to the supplied energy. Electrical charge can be stored and subsequently recovered even when it is associated with coion redistribution or reversible processes that do not produce net salt removal. Consequently, a relatively high energy storage efficiency can coexist with a lower charge efficiency.

After 15 cycles over 10 h at 0.6 V in 1000 mg/L NaCl, the $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ and $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$ electrodes retained approximately 94% and 93% of their initial electrosorption efficiencies, respectively. These results indicate short term repeatability under the investigated conditions but do not establish long term durability. Extended cycling tests, metal leaching measurements, and post cycle structural characterization would be required to evaluate practical long term stability.

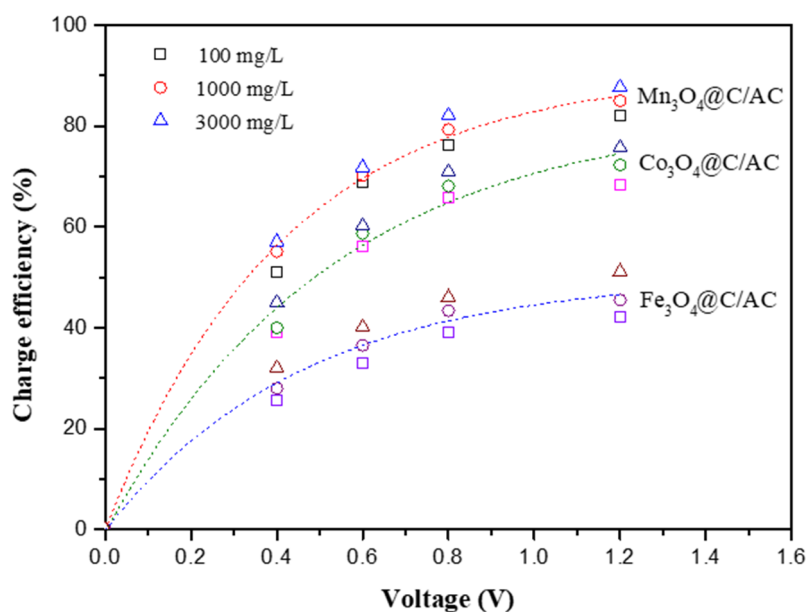


Figure 5. Charge efficiencies of the $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$, $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$, and $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$ electrodes during CDI charging in NaCl solutions with concentrations of 100, 1000, and 3000 mg/L at applied cell voltages ranging from 0.4 to 1.2 V. Charge efficiency represents the fraction of delivered charge associated with net salt removal and is distinct from energy storage efficiency.

3.3. Galvanostatic Charge-Discharge Behavior and Energy-Recovery Analysis

The electrical energy that can be recovered during CDI regeneration depends on the dynamic cell voltage and current responses during charging and discharging. Figure 6 presents representative galvanostatic charge and discharge profiles obtained only for $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ CDI cells at total current magnitudes of 1 and 10 mA in 1000 and 3000 mg/L NaCl. These separate GCD experiments were used to illustrate how current and salinity affected

the operational voltage response, polarization, and energy hysteresis of $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$. They were not the source of the cross-electrode energy comparison in Figures 7–9, which was calculated from the common CDI protocol applied to all four electrodes as described in Section 2.4. Upon application of the charging current, the cell voltage exhibited an initial rapid increase followed by a more gradual rise during charge accumulation. A larger initial voltage increase was observed at 10 mA than at 1 mA, consistent with greater resistive and polarization contributions at the higher current. Increasing the NaCl concentration from 1000 to 3000 mg/L reduced the initial voltage increase, consistent with the higher ionic conductivity of the more concentrated solution.

The electrical energy supplied during charging, E_{ch} , and the electrical energy recovered during discharge, E_{dis} , were calculated by numerical integration of the measured cell voltage and current responses using the expressions defined in Section 2.6. The calculations were based on the experimental voltage profiles shown by the black curves in Figure 6. The blue and yellow shaded regions schematically identify the charging and discharging intervals, respectively, whereas the sloping dashed lines are included only as visual guides and were not used in the energy calculations. The total unrecovered energy, E_{unrec} , was calculated as:

$$E_{\text{unrec}} = E_{\text{ch}} - E_{\text{dis}}$$

The difference between E_{ch} and E_{dis} represents the total energy that was not recovered under the specified discharge protocol.

Figure 6 shows a smaller initial voltage response and lower apparent polarization at 1 mA than at 10 mA. A smaller initial voltage response was also observed in 3000 mg/L NaCl than in 1000 mg/L NaCl. These observations are qualitatively consistent with lower resistive and polarization contributions under lower current and higher salinity conditions. Because the GCD profiles were obtained over different voltage windows under the tested current and salinity conditions, the raw curves were not used for direct comparison of intrinsic capacitance among the operating conditions. Instead, they were used to examine how current and electrolyte concentration affected the operational voltage response and energy hysteresis of the $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ CDI cells. The supplied and recovered energies were calculated by integration of the measured voltage and current responses. Stored energy density was normalized by the exposed electrode area, whereas energy storage efficiency was calculated as the ratio of recovered energy to supplied energy.

Previous CV and in situ XANES results reported for $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ indicated that reversible manganese valence changes contributed to charge storage during CDI [27]. In the present core-shell structure, Mn_3O_4 domains were surrounded by carbon shells with typical thicknesses of approximately 3 to 6 nm. These shells may improve electrical contact between the oxide domains and the surrounding AC matrix and may shorten local ion-transport pathways. The previous CV and in situ XANES results are used only as mechanistic context for $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$; the overall evidentiary scope of the present study is summarized once at the end of Section 3.4.

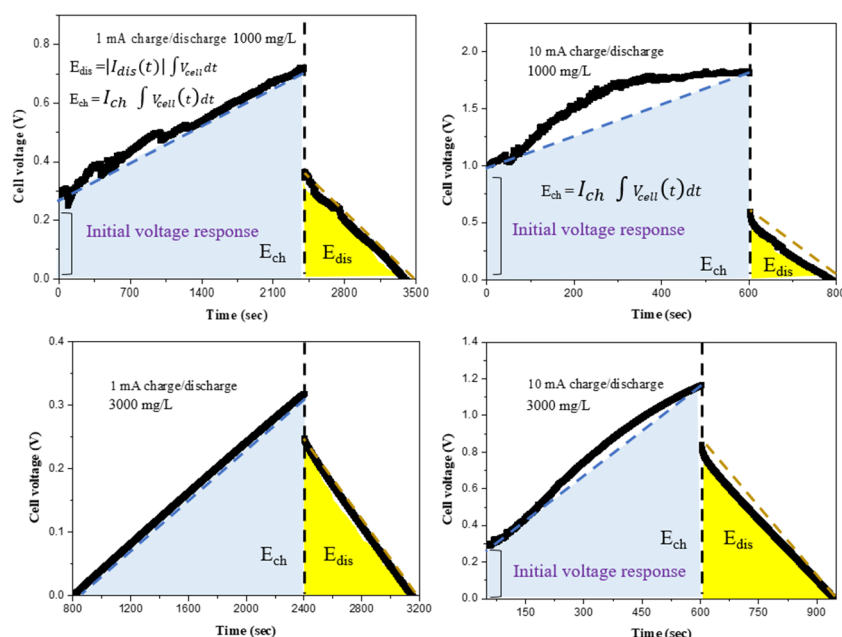


Figure 6. Representative galvanostatic charge and discharge profiles of the $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ CDI cells at total current magnitudes of (a,c) 1 mA and (b,d) 10 mA in (a,b) 1000 mg/L and (c,d) 3000 mg/L NaCl. The blue and yellow

regions indicate the charging and discharging intervals, respectively. The sloping dashed lines are visual guides only, and the vertical dashed lines indicate current reversal.

3.4. Thermodynamic Analysis of Energy Recovery

In this study, the thermodynamic analysis refers to an electrical energy balance based on the measured charging and discharging power profiles. During CDI operation, a portion of the electrical energy supplied during charging was recovered during the subsequent discharge step, whereas the remaining portion was reported as the total unrecovered energy under the specified protocol. Energy storage efficiency was defined as the ratio of the electrical energy recovered during discharge to that supplied during charging. The supplied and recovered energies were calculated by integrating the measured cell voltage and current responses over time, as described in Section 2.6. This energy balance enables a system-level comparison of AC and the three $M_3O_4@C/AC$ composite electrodes under identical electrode area, electrolyte concentration, charging voltage, charging duration, and discharging protocol.

$Fe_3O_4@C/AC$ was retained in the comparative energy analysis because the voltage and current profiles required to calculate supplied energy, recovered energy, and energy storage efficiency were obtained using the same common CDI protocol as for the other electrodes. Its inclusion therefore supports comparison of operational energy recovery. The separate $Mn_3O_4@C/AC$ GCD dataset in Figure 6 was not used to infer the intrinsic capacitance or charge-storage mechanism of $Fe_3O_4@C/AC$.

Figure 7 compares the recoverable stored energy density and energy storage efficiency of AC, $Mn_3O_4@C/AC$, $Co_3O_4@C/AC$, and $Fe_3O_4@C/AC$ in 3000 mg/L NaCl at an applied charging voltage of 0.6 V. Under the specified operating conditions, the three composite electrodes exhibited higher recoverable stored energy densities than AC, with $Mn_3O_4@C/AC$ reaching approximately 220 mJ/cm². The energy storage efficiencies of the composite electrodes ranged from approximately 84 to 89%, compared with approximately 54% for AC. These results indicate that incorporation of the $M_3O_4@C$ components was associated with higher recovered energy per unit area and a larger recovered fraction of the supplied electrical energy.

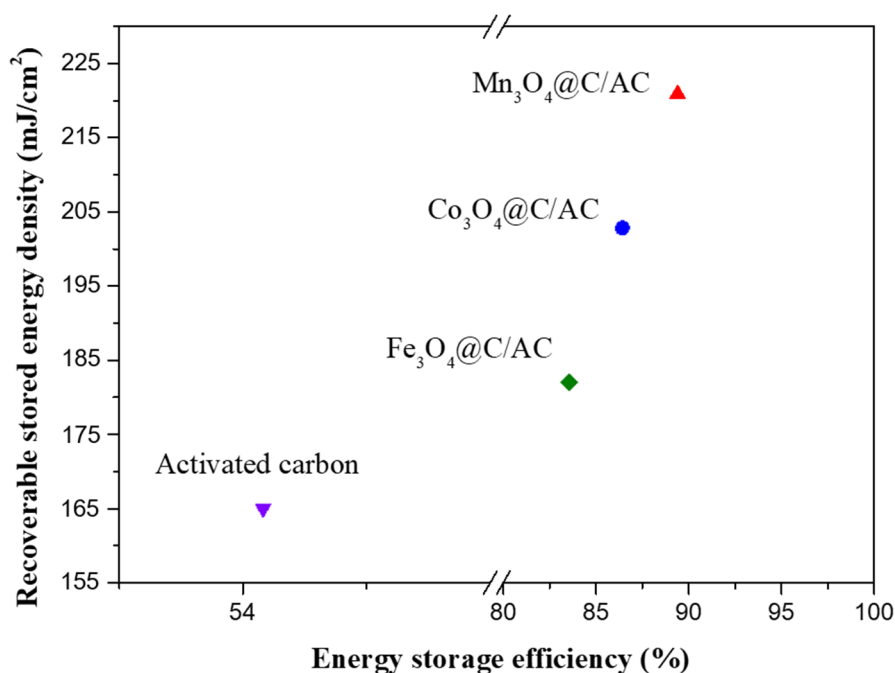


Figure 7. Relationship between recoverable stored energy density and energy storage efficiency for AC, $Mn_3O_4@C/AC$, $Co_3O_4@C/AC$, and $Fe_3O_4@C/AC$ in 3000 mg/L NaCl at an applied charging voltage of 0.6 V.

Figure 8 shows the effects of NaCl concentration on the recoverable stored energy density and energy storage efficiency of $Mn_3O_4@C/AC$, $Co_3O_4@C/AC$, and $Fe_3O_4@C/AC$ at an applied charging voltage of 0.6 V. As the NaCl concentration increased from 100 to 3000 mg/L, both the recoverable stored energy density and energy storage efficiency increased for all three composite electrodes. This trend is consistent with the higher ionic conductivity of the more concentrated NaCl solutions, which may reduce bulk-electrolyte resistance and overall cell polarization. Among the three composite electrodes, $Mn_3O_4@C/AC$ exhibited the highest recoverable stored energy density and energy storage efficiency over the investigated concentration range. At 3000 mg/L NaCl, the energy storage efficiencies of the three composite electrodes ranged from approximately 84 to 89%.

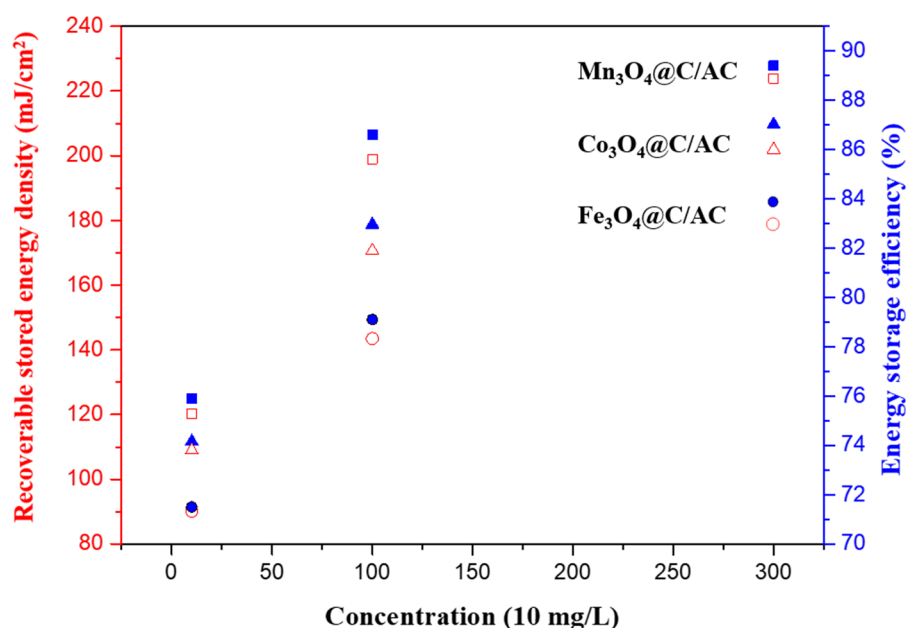


Figure 8. Effects of NaCl concentration on the recoverable stored energy density and energy storage efficiency of $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$, $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$, and $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$ at an applied charging voltage of 0.6 V. Open red and filled blue symbols represent recoverable stored energy density and energy storage efficiency, respectively.

Figure 9 compares the energy storage efficiencies of $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$, $\text{Co}_3\text{O}_4@\text{C}/\text{AC}$, and $\text{Fe}_3\text{O}_4@\text{C}/\text{AC}$ at applied charging voltages of 0.6, 0.8, and 1.2 V in NaCl solutions ranging from 100 to 3000 mg/L. In each panel, blue triangles, red circles, and black squares represent 3000, 1000, and 100 mg/L NaCl, respectively. The dashed connecting lines are visual guides only, whereas the vertical dashed lines separate the three applied-voltage conditions. Within each voltage group, higher NaCl concentration was generally associated with higher energy storage efficiency, consistent with Figure 8. In contrast, energy storage efficiency decreased markedly as the applied charging voltage increased. Because the charging duration was fixed at 1 h for all tested voltages, this decrease cannot be attributed to extended charging time. The smaller recovered fraction at higher voltage indicates greater aggregate unrecovered energy under the tested protocol. Similar voltage-dependent changes in energy recovery have been reported for CDI systems under different charging and discharging conditions [28].

The voltage dependent behavior shown in Figure 9 has important implications for the selection of CDI operating conditions. At an applied charging voltage of 0.6 V in 1000 to 3000 mg/L NaCl, the three composite electrodes exhibited recoverable stored energy densities of approximately 143 to 223 mJ/cm² and energy storage efficiencies of approximately 79 to 89%. Increasing the applied charging voltage to 1.2 V resulted in a pronounced decrease in energy storage efficiency for all three composite electrodes, indicating that a smaller fraction of the supplied electrical energy was recovered during discharge. Although a higher charging voltage may provide a stronger electrostatic driving force for ion electrosorption, Figure 9 alone does not quantify the corresponding change in salt adsorption capacity or establish the electrical energy consumption per unit mass of salt removed. Under the investigated charging and discharging protocol, applied charging voltages of 0.6 to 0.8 V resulted in higher energy storage efficiencies than 1.2 V. This voltage range therefore appears more favorable for energy recovery under the tested conditions, but it should not be interpreted as a universally optimal operating window.

Overall, the energy recovery analysis demonstrates that electrode composition, applied charging voltage, and electrolyte concentration jointly influence the fraction of supplied electrical energy recovered during discharge. Under the specified operating conditions, the $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ composite electrodes exhibited higher recoverable stored energy densities and energy storage efficiencies than AC, with $\text{Mn}_3\text{O}_4@\text{C}/\text{AC}$ showing the highest values. Moderate charging voltages and higher NaCl concentrations were generally associated with larger recovered fractions. The scope and limitations of the analysis are consolidated here. The energy balance quantifies the supplied, recovered, and total unrecovered energies but does not resolve the individual ohmic, interfacial, diffusion-related, leakage, or parasitic contributions to unrecovered energy. Directly comparable CV and GCD measurements were not available for all electrodes; consequently, the present results do not independently quantify the relative Faradaic and electric-double-layer contributions or support cross-electrode comparison of intrinsic capacitance. Future work should combine comparative CV and GCD measurements, quantitatively validated

impedance analysis, extended cycling, metal-leaching analysis, post-cycle structural characterization, and system-level energy evaluation to resolve these mechanisms and establish practical applicability.

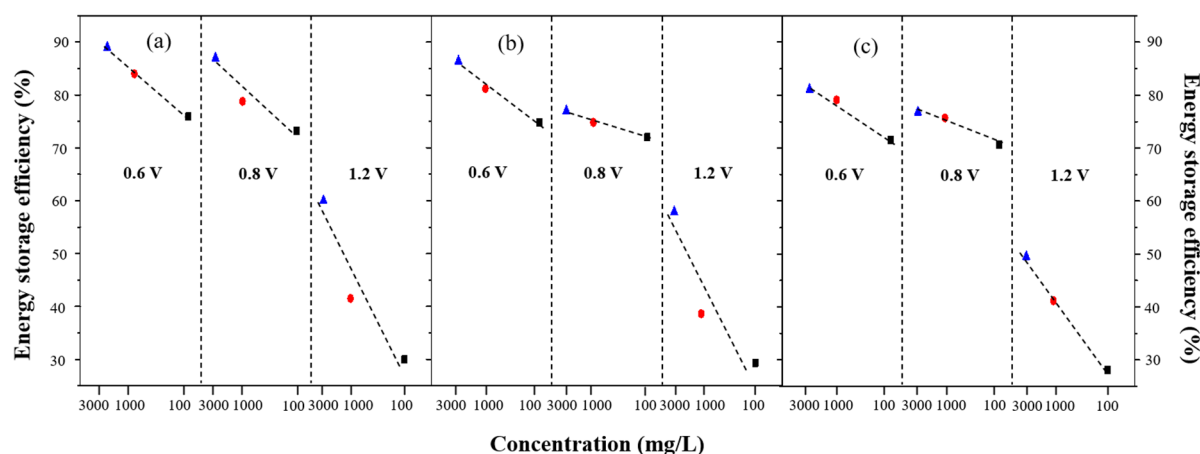


Figure 9. Energy storage efficiencies of (a) Mn₃O₄@C/AC, (b) Co₃O₄@C/AC, and (c) Fe₃O₄@C/AC at applied charging voltages of 0.6, 0.8, and 1.2 V in 100, 1000, and 3000 mg/L NaCl.

4. Conclusions

Mn₃O₄@C, Co₃O₄@C, and Fe₃O₄@C core-shell nanoparticles were incorporated into activated carbon to fabricate Mn₃O₄@C/AC, Co₃O₄@C/AC, and Fe₃O₄@C/AC composite electrodes for CDI. Under identical conditions of 3000 mg/L NaCl and an applied charging voltage of 0.6 V, AC, Mn₃O₄@C/AC, Co₃O₄@C/AC, and Fe₃O₄@C/AC exhibited salt adsorption capacities of 6.7, 12.5, 10.8, and 9.3 mg/g, respectively. Under the same operating conditions, the three composite electrodes exhibited energy storage efficiencies of approximately 84 to 89%, compared with approximately 54% for AC, and Mn₃O₄@C/AC achieved the highest recoverable stored energy density of approximately 220 mJ/cm². Energy storage efficiency generally increased with NaCl concentration but decreased markedly as the applied charging voltage increased from 0.6 to 1.2 V. These findings show that electrode composition and operating conditions influence salt removal, recoverable stored energy density, and energy storage efficiency. Effective reuse of recovered electrical energy may potentially reduce the net electrical energy demand of CDI. The 15-cycle test conducted over 10 h demonstrates short-term repeatability only. Further evaluation of long-term performance, operating cost, and carbon footprint will require the experimental and system-level studies identified in Section 3.4.

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