

Bimetallic synergy boosts selective electrocatalytic nitrate reduction on CuFe layered double hydroxide nanosheets

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Abstract

Electrochemical nitrate (NO_3^- -N) reduction to nitrogen gas (N_2) is a sustainable route for addressing nitrate pollution in water. However, this process is limited by low catalytic activity and poor N_2 selectivity. Herein, we have synthesized a highly efficient and stable CuFe layered double hydroxide (CuFe LDH) catalyst with a nanosheet structure on nickel foam through a one-step hydrothermal technique. The copper (Cu) and iron (Fe) establish strong electronic coupling through oxygen bridges in this catalyst. This electronic coupling optimizes the d-band center position and balances the adsorption and desorption energy barriers of nitrogen-containing intermediates. Experimental results demonstrate the CuFe LDH/NF electrode achieves NO_3^- -N removal efficiency of 95.03% and N_2 selectivity of 99.63%. Density functional theory (DFT) calculations confirm that bimetallic synergy lowers the activation energy. This work elucidates the synergy mechanism and offers a strategy for non-noble-metal denitrification catalysts.

1. Introduction

Nitrate (NO_3^- -N) pollution in surface water and groundwater has become a global environmental and public health concern. The primary causes of this contamination are industrial wastewater discharge, overuse of agricultural fertilizers and other human activities (Li et al., 2026b; Qu et al., 2026; Tang et al., 2019; Zou et al., 2025). Nitrate accumulation can cause eutrophication and disrupt ecosystem balance, while also being converted in the human body into carcinogenic nitrosamines (Bai et al., 2021; Fajardo et al., 2021; Hua et al., 2024; Teng et al., 2017). It is imperative to develop efficient and low-cost green technologies for the selective conversion of nitrate to nitrogen gas (N_2). However, conventional physical, biological and chemical nitrate removal methods suffer from low efficiency, high operating costs and the risk of secondary pollution (Meng et al., 2023; Song et al., 2022; Wang et al., 2025). By contrast, electrochemical denitrification can be powered by renewable energy. This method requires no bulk chemical reagents and can directly reduce NO_3^- -N to N_2 . These advantages make electrochemical denitrification a promising alternative. Nevertheless, its practical application is still limited by insufficient catalyst activity, poor N_2 selectivity and the high costs of precious metal catalysts (Kim et al., 2023; Sanchis et al., 2022; Shen et al., 2020).

To address these challenges, transition-metal-based catalysts, especially iron (Fe) and copper (Cu), have drawn wide attention. These metals are earth-abundant, environmentally benign, and possess favorable redox properties (Mahidashti and Rezaei, 2026; Yuan et al., 2023; Zhang et al., 2025b). Studies show that Cu sites preferentially catalyze the conversion of NO_3^- to NO_2^- , a critical initiation step in nitrate reduction. Despite this advantage, single Cu-based catalysts often face insufficient activity in the subsequent hydrogenation steps and accumulation of intermediates (Hald et al., 2026; Wu et al., 2023a; Zhao et al., 2021). This limitation restricts overall performance. In response, introducing a second metal to form a bimetallic system can effectively enhance catalytic performance. Among the candidates, iron, an earth-abundant transition metal, offers distinct advantages in electrocatalytic nitrate reduction (Duan

et al., 2025; Fan et al., 2026; Liu et al., 2020; Zhang et al., 2025a). Specifically, Fe sites show strong thermodynamic adsorption affinity for nitrite (NO_2^-). This affinity enables efficient subsequent conversion of NO_2^- to NH_3 or N_2 . Consequently, the synergy between Cu and Fe can establish a relay catalysis mechanism: Cu sites handle the initial reaction of NO_3^- to NO_2^- , while Fe sites undertake further hydrogenation of NO_2^- (Hua et al., 2024). However, in conventional supported catalysts, Cu and Fe sites are randomly distributed, and the relay efficiency is limited by the diffusion distance between bimetallic sites. Therefore, constructing a catalyst with atomically dispersed and intimately contacted Cu–Fe sites is essential for achieving efficient synergistic catalysis (Zhang et al., 2025a).

Layered double hydroxides (LDH) provide an ideal platform for this study due to their unique two-dimensional layered structure (Wu et al., 2025). On one hand, the Cu^{2+} and Fe^{3+} in the LDH layered structure are restricted by the lattice and achieve uniform dispersion at the atomic level, effectively avoiding phase separation and agglomeration. On the other hand, the Cu atoms and Fe atoms in adjacent layers form a co-axial bonding network through oxygen bridges, triggering charge rearrangement and electron coupling between the bimetallic sites. This oxygen-bridge-mediated synergistic effect can precisely regulate the d-band center position, enabling the surface to simultaneously possess an appropriate NO_3^- adsorption energy and a lower $^*\text{NO}_2$ to $^*\text{N}$ desorption energy barrier (Ding et al., 2025; Pei et al., 2024; Riaz et al., 2024). Therefore, the LDH in this system not only serves as a support, but also acts as a platform for the dispersion of dual-metal electronic coupling.

In this study, we fabricate CuFe LDH nanosheet electrodes on nickel foam (NF) via a hydrothermal one-step method. The Cu–Fe bimetallic interface optimizes the adsorption energies of nitrogen-containing intermediates. This optimized interface suppresses the formation of toxic byproducts such as nitrite and ammonia, thereby greatly improving N_2 selectivity. Additionally, the effects of various reaction conditions, including electrolyte composition, current density, initial pH, and initial NO_3^- -N concentration, are evaluated. Ultimately, the key active species, reaction pathway, and reaction mechanism for electrocatalytic nitrate reduction on CuFe LDH are identified.

2. Experimental section

2.1 Preparation of Ni foam

Firstly, the Ni foam (NF) was cut into 5 cm × 5 cm pieces and ultrasonically cleaned sequentially in hydrochloric solution, deionized (DI) water and ethanol. Then, the NF pieces were washed thoroughly with DI water and dried at 60°C for 6 h.

2.2 Synthesis of CuFe LDH

$\text{Cu}(\text{NO}_3)_2 \cdot 3\text{H}_2\text{O}$ (1.3529 g), $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (2.2624 g), urea (1.6968 g) and NH_4F (0.2074 g) were fully dissolved in 70 mL of DI water. Subsequently, the pre-treated NF was placed in an autoclave and

maintained at 120°C for 10 h. After natural cooling to room temperature, the NF was taken out, rinsed with DI water and dried under vacuum at 60°C for 10 h.

2.3 Characterization

Morphology, elemental composition, and elemental distribution were characterized using a field-emission scanning electron microscope (SEM, ZEISS Sigma 360, Germany) equipped with energy-dispersive X-ray spectroscopy (EDS). X-ray diffraction (XRD) patterns were collected on an X-ray diffractometer (Rigaku Ultima IV, Japan) with Cu K α radiation to analyze the crystal structure. The surface chemical composition was analyzed by X-ray photoelectron spectroscopy (XPS, Thermo Scientific K-Alpha, USA). Finally, the functional groups on the material surface was examined using a Fourier transform infrared spectrometer (FTIR, Nicolet iS50, USA) in the range of 400–4000 cm⁻¹.

All electrochemical tests were performed in a CHI 660E electrochemical workstation (Shanghai Chenhua) with a typical three-electrode system. The electrolyte consisted of 100 mL of solution containing 100 ppm NO₃⁻-N and 0.1 M Na₂SO₄. To evaluate the electrocatalytic activity, linear sweep voltammetry (LSV) curves were recorded from 0.1 to -1.6 V vs. SCE at a scan rate of 5 mV s⁻¹. Additionally, cyclic voltammetry (CV) curves for electrochemical double-layer capacitance (Cdl) measurements were recorded in the non-Faradaic potential region. These measurements are used to evaluate electrochemically active surface areas (ECSA) of the catalysts. Furthermore, electrochemical impedance spectroscopy (EIS) was carried out in a frequency range of 10⁻² to 10⁶ Hz to investigate the charge transfer resistance (Rct) of the CuFe LDH cathode.

More details about electrochemical tests, ion concentration detection, data analysis and DFT calculation methods are provided in the Supporting Information (Text S2-S5).

3. Result and discussion

3.1 Synthesis and structural characterization

A simple strategy for preparing the CuFe LDH electrode is illustrated schematically in Fig. 1a. Under hydrothermal conditions at 120°C for 10 h, CuFe LDH nanoarrays are grown in situ on NF. Cu(NO₃)₂ and Fe(NO₃)₃ serve as the metal sources. Urea and NH₄F act as the pH regulator and morphology controller, respectively. SEM images (Fig. 1b) reveal a smooth and edge-curved nanosheet morphology. An open pore network forms between the nanosheets. EDS (Fig. 1c) further confirms that Cu atoms and Fe atoms are uniformly distributed on the NF skeleton surface at the micron scale. Such uniform distribution indicates atomic-level mixing of the two metal species.

To further elucidate the crystal phase of the catalysts, XRD analysis is conducted. For the CuFe LDH sample (Fig. 1d), the diffraction peaks appear at 11.66°, 23.38°, 34.36°, 38.86°, 46.58°, 59.86° and 61.10° are correspond to the (003), (006), (012), (015), (018), (110) and (113) planes of layered double hydroxide, respectively. These peaks match well with the typical LDH diffraction features given in the

standard card (JCPDS 50–0235)(He et al., 2023). Fe LDH and Cu LDH also exhibit the above characteristic diffraction peaks. Notably, the pattern of pure Cu LDH shows an additional set of sharp diffraction peaks at 43.50°, 50.54° and 74.12°. These positions correspond exactly to the standard card of metallic Cu⁰ (JCPDS 04-0836)(Long et al., 2025). In contrast, no obvious characteristic peaks of metallic copper are detected in the CuFe LDH sample. This comparison clearly reveals that the introduction of Fe³⁺ can significantly suppress the reduction tendency of Cu²⁺. The introduction of Fe³⁺ forms a more stable Cu²⁺–O–Fe³⁺ bonding network with Cu²⁺, which enhances the oxidative stability of copper in the layer(Hua et al., 2024).

The chemical composition of the CuFe LDH was further determined by XPS measurements. In the Cu 2p spectrum (Fig. 1e), the main peaks at 934.5 eV and 954.3 eV correspond to the Cu 2p_{3/2} and Cu 2p_{1/2} spin–orbit doublet, respectively(Shih et al., 2020). These main peaks are accompanied by a strong satellite peak at 942 eV. These spectral features are typical signatures of Cu²⁺. However, an additional shoulder peak is clearly resolved at 932.5 eV. This binding energy is assigned to Cu⁰. This observation indicates that part of Cu²⁺ was reduced to Cu⁰ during hydrothermal synthesis and subsequent sample handling, generating a mixed Cu⁰/Cu²⁺ valence state(Hald et al., 2026). Meanwhile, the Fe 2p spectrum displays main peaks at 711.2 eV (Fe 2p_{3/2}) and 724.7 eV (Fe 2p_{1/2}), characteristic of Fe³⁺. A distinguishable sub-peak appears at 709.5 eV, which belongs to the fingerprint signal of Fe²⁺(Ma et al., 2025). Fe²⁺ serves as an electron-buffer reservoir. This reservoir rapidly modulates the local electron density through the reversible Fe²⁺/Fe³⁺ redox couple, thereby stabilizing reaction intermediates.

Furthermore, the structural integrity and bonding environment of the as-prepared samples are characterized by FTIR spectroscopy. All samples (Fig. 2) exhibit a broad and strong absorption band at 3450 cm^{–1}. This band is assigned to the stretching vibrations of layer hydroxyl groups (O–H) and interlayer water molecules. The absorption peak at 1630 cm^{–1} corresponds to the H–O–H bending vibration of water molecules(Chen et al., 2026). These two sets of characteristic peaks together confirm the intrinsic hydrated environment of layered double hydroxides. A sharp absorption peak appears at 1380 cm^{–1}. This peak is attributed to the ν₃ asymmetric stretching vibration of interlayer carbonate ions (CO₃^{2–}). Carbonate ions intercalate as charge-compensating anions to maintain charge balance. In the low-frequency region (400–800 cm^{–1}), the spectra display a set of complex absorption bands. These bands are assigned to metal–oxygen (M–O) and metal–hydroxyl (M–OH) stretching and bending vibrations(Altalhi et al., 2024).

To compare the intrinsic reactivity of the catalysts, electrochemical measurements are carried out. The LSV curves in Fig. 3a are recorded in an electrolyte containing 100 mg-N L^{–1} nitrate. Cu LDH delivers the highest reduction current density, followed by CuFe LDH and Fe LDH. LSV curves of CuFe LDH exhibit significant current density increase upon the addition of nitrate to the electrolyte, and a notable reduction peak is observed at around – 0.6 V vs. SCE (Fig. 3b). Meanwhile, electrochemical impedance spectroscopy (Fig. 3c) provides different kinetic information. CuFe LDH exhibits the smallest Nyquist

semicircle diameter. Correspondingly, its charge transfer resistance (R_{ct}) is significantly lower than those of Cu LDH and Fe LDH. This lower R_{ct} indicates faster electron transfer from the electrode to nitrate ions at the CuFe LDH electrode/electrolyte interface. The electrochemical active surface area (ECSA) measurement is primarily used to assess the true active surface area of electrodes participating in electrochemical reactions (Ma et al., 2023). The ECSA values derived from double-layer capacitance measurements reveal that CuFe LDH possesses the largest electrochemically active surface area (15.610 cm^2).

3.2 Electrocatalytic nitrate reduction performance

To investigate how catalyst composition affects NO_3^- -N removal, CuFe LDH catalysts with different Cu/Fe ratios are synthesized via the hydrothermal method. The catalytic performance is shown in Fig. 4a-b. Cu LDH alone delivers a NO_3^- -N removal efficiency of about 90.70%. With the introduction of Fe, the Cu_1Fe_1 LDH catalyst achieves a removal efficiency of 95.03%. Cu_2Fe_1 LDH and Cu_3Fe_1 LDH also exhibit efficiencies above 92%. These results demonstrate that incorporating a suitable amount of Fe plays a critical role in electrocatalytic denitrification. To further probe the effect of Fe incorporation, the density of states for Fe LDH, Cu LDH, and CuFe LDH is calculated using Materials Studio. Partial density of states (PDOS) analysis shows that the d-band center of CuFe LDH is -1.37 eV . This value is higher than that of Cu LDH (-2.06 eV) (Fig S3). Consequently, the d-band center shifts closer to the Fermi level. Upon Fe addition, the Cu 2p electrons redistribute. Specifically, the 3d orbitals form a new hybridization, which shifts the 3d band toward the Fermi level. This upward shift indicates reduced antibonding occupation and also implies enhanced adsorbate binding strength (Yan et al., 2026).

Achieving high nitrate removal efficiency at low current densities is a key metric for evaluating catalyst performance. Figure 5a shows the product distribution and NO_3^- -N removal efficiency under different current densities ($15\text{--}35 \text{ mA cm}^{-2}$). Within this range, a higher current density leads to greater NO_3^- -N removal efficiency. Specifically, as the current density increases from 15 to 30 mA cm^{-2} , the NO_3^- -N removal efficiency rises from 82.93% to 95.03%, and the N_2 selectivity improves from 50.75% to 99.63%. This improvement occurs because a higher current density strengthens the electric field effect in the reaction system, allowing nitrate to acquire more electrons at the cathode. Electron transfer is thus promoted, accelerating the reduction of NO_3^- -N to N_2 . However, an excessively high current density actually decreases both the NO_3^- -N reduction efficiency and N_2 selectivity. Under high current densities, the competing hydrogen evolution reaction (HER) generates abundant H_2 bubbles. These bubbles adhere to the catalyst surface, disrupting electron transfer and nitrate diffusion (Zheng et al., 2025). High current densities also promote nitrification side reactions. Therefore, 30 mA cm^{-2} was selected as the optimal current density for subsequent tests unless otherwise specified.

Furthermore, the influence of initial NO_3^- -N concentration ($30\text{--}200 \text{ mg L}^{-1}$) is examined. The experimental results are shown in Fig. 5b. At an initial NO_3^- -N concentration of 30 mg L^{-1} , the removal

efficiency reaches 99.29%. As the initial concentration increases, the NO_3^- -N removal efficiency gradually decreases. This decline occurs because the number of active sites available for NO_3^- -N adsorption becomes insufficient at higher concentrations. Nevertheless, for initial concentrations ranging from 30 to 200 mg L^{-1} , the removal efficiency exceeds 85% after 120 min. For initial concentrations from 30 to 100 mg L^{-1} , N_2 selectivity surpasses 99%. When the initial NO_3^- -N concentration exceeds 100 mg L^{-1} , N_2 selectivity at 120 min drops significantly. However, when the reaction time is extended to 180 min, both the removal efficiency and N_2 selectivity exceed 90% (Fig S4). Therefore, the as-prepared catalyst demonstrates good reduction capability over an initial NO_3^- -N concentration range of 30 to 200 mg L^{-1} .

In addition, the effect of pH on nitrate reduction is investigated. The results show that neutral conditions are more favorable for electrocatalytic NO_3^- reduction (Fig. 5c). At pH 7.0, the NO_3^- -N removal efficiency reaches 97.24%, compared to only 90.49% at pH 3.0. The initial pH does not affect the final product selectivity profile; N_2 selectivity remains above 95% over a wide pH range. The enhanced performance under neutral pH is attributed to the consumption of H^+ during NO_3^- reduction. The electrolyte is rich in H^+ at neutral pH, which promotes the reaction. In acidic environments, the competing HER is intensified. Consequently, the H_2 bubbles formed cover the electrode surface, blocking contact between active sites and the electrolyte (Kang et al., 2025). Under alkaline conditions, negatively charged OH^- ions compete with NO_3^- for adsorption sites on the catalyst surface, inhibiting the nitrate reduction reaction and leading to performance degradation (Du et al., 2024).

Finally, the effect of chloride ion concentration on electrocatalytic denitrification is explored. Prior studies have shown that the concentration of chloride ions (Cl^-) in the electrolyte is crucial for achieving high N_2 selectivity in electrocatalytic denitrification (Luo et al., 2023; Wu et al., 2023b; Yu et al., 2022). Accordingly, electrocatalytic NO_3^- -N reduction experiments are conducted in electrolytes with different NaCl concentrations. As shown in Fig. 5d, N_2 selectivity rises markedly with increasing NaCl concentration. At 0.02 M NaCl, the NO_3^- -N removal efficiency is 95.03% and the N_2 selectivity reaches 99.63%. Further increasing the NaCl concentration to 0.04 M produces no significant change in N_2 selectivity but leads to a pronounced drop in NO_3^- -N removal efficiency. These results confirm the positive effect of Cl^- on N_2 selectivity, while also indicating that an excessively high Cl^- concentration reduces NO_3^- -N removal efficiency. This likely arises from competitive adsorption between Cl^- and NO_3^- near the cathode. Moreover, excessive ClO^- can re-oxidize the intermediate NO_2^- back to NO_3^- , thereby lowering the overall NO_3^- -N removal efficiency (Wang et al., 2023).

3.3 Long-term stability

The stability and reusability of the CuFe LDH electrode are evaluated through cycling tests. CuFe LDH exhibits an outstanding stability and high performance over 10 cycles with the NO_3^- -N removal efficiency maintaining $94.24 \pm 1.25\%$ and the N_2 selectivity remaining $99.06 \pm 0.76\%$ (Fig. 6a). The bimetallic Pd-Cu

system is commonly regarded as a typical catalyst for denitrification, owing to the synergistic actions of the two metals(Xu et al., 2019). Table S1 shows that the nitrate reduction performance of CuFe LDH surpassed that of previously reported Pd-Cu electrocatalysts, suggesting the viability of using iron as an effective alternative to noble metals.

To assess the structural durability of the catalyst, XRD analysis is performed before and after the reaction. After the reaction, the sample still retains the characteristic LDH peaks and closely match the pristine sample. This retention confirms that the main LDH framework of CuFe-LDH does not collapse under electrolytic conditions. Consequently, the catalyst exhibits excellent structural stability. Notably, new diffraction peaks are detected in the post-reaction pattern 43.3° , 50.4° and 74.1° . These new peaks can be indexed to metallic copper (JCPDS 04-0836) and correspond to the (111), (200) and (220) planes. This observation indicates that under cathodic polarization, a fraction of the Cu species are electrochemically reduced to metallic Cu⁰. However, the original LDH layered host structure remains intact.

Furthermore, as shown in Fig. 6c, the high-resolution Fe 2p spectra before and after the reaction are nearly identical. The main peaks of Fe 2p_{3/2} and Fe 2p_{1/2} and their corresponding satellite peaks exhibit no obvious shift. The mixed Fe²⁺/Fe³⁺ valence state is maintained. This stability indicates that Fe is chemically very stable during the reaction and undergoes no significant valence change. Thus, its primary role likely lies in stabilizing the LDH layer structure as a framework element(Altalhi et al., 2024). Figure 6d illustrates the evolution of the Cu 2p chemical state. Before the reaction, Cu exists predominantly as Cu²⁺, accompanied by strong satellite peaks. After ten cycles, the peak at 932.5 eV, assigned to Cu⁰ intensifies significantly. Simultaneously, the intensity of the Cu²⁺ satellite peaks weakens relatively. This result perfectly agrees with the appearance of metallic copper peaks in XRD and confirms that a portion of the Cu²⁺ on the catalyst surface is in-situ reduced to metallic Cu⁰ under electrocatalytic reduction conditions. Such dynamic valence-state reconstruction of surface Cu species is generally considered a crucial process for creating highly active catalytic sites(Zhou et al., 2022).

3.4 Electrocatalytic reduction mechanism

Prior studies indicate that electrocatalytic nitrate reduction proceeds mainly through two pathways: indirect hydrogenation and direct electron transfer(Li et al., 2026a). To investigate the role of *H, radical quenching experiments are performed using tert-butyl alcohol (TBA). Upon TBA addition, the nitrate removal efficiency shows only a relatively small decrease (Fig. 7a). When the TBA dosage reaches 0.05 mol L⁻¹, the nitrate reduction rate constant drops from 0.025 to 0.019 h⁻¹ (Fig. 7b). However, the final nitrate removal efficiency still remains at 90%. These results demonstrate that *H plays a certain role in nitrate reduction, but this pathway is not the primary pathway on CuFe LDH.

To further elucidate the reaction pathway of nitrate reduction on CuFe LDH, first-principles calculations are performed. The resulting Gibbs free energy diagram is shown in Fig. 7(c). Free nitrate ions in the electrolyte are first adsorbed onto the electrode surface, forming *NO₃. Subsequently, *NO₃ gains an

electron and cleaves an N–O bond to produce $^*\text{NO}_2$. This step is considered the rate-determining step of the overall reduction mechanism (Du et al., 2022; Zhou et al., 2024). Subsequently, $^*\text{NO}_2$ is gradually reduced to $^*\text{N}$ through successive N–O bond cleavage. The resulting $^*\text{N}$ then undergoes three hydrogenation steps involving $^*\text{NH}$, $^*\text{NH}_2$ and $^*\text{NH}_3$ to form $^*\text{NH}_3$. Finally, the generated ammonia reacts with hypochlorite produced at the anode and is ultimately oxidized to N_2 (Liu et al., 2025).

4. Conclusions

In this study, we have successfully synthesized self-supporting CuFe LDH nanosheet electrodes on nickel foam via a facile hydrothermal process. According to characterization analysis, the CuFe LDH has a homogeneous elemental distribution, a characteristic LDH layered structure, and distinct mixed $\text{Cu}^0/\text{Cu}^{2+}$ and $\text{Fe}^{2+}/\text{Fe}^{3+}$ valence states. This layered double hydroxide nanosheets structure electrode (CuFe LDH) achieved high nitrate removal efficiency (> 95.03%) and precise conversion of nitrate to dinitrogen (99.63%). Furthermore, DFT calculations and radical quenching experiments together elucidate the bimetallic synergistic effect. This synergy optimizes the d-band center position and balances the adsorption and desorption of nitrogen-containing intermediates. This work can provide a reference for the electrochemical reduction of nitrate via bimetallic catalysts. By adding appropriate metals, the local electronic environment of the metal cations can be significantly altered, providing a way to the development of sophisticated electrocatalytic denitification catalysts in the future.

Declarations

Declaration of Competing Interest:

The authors declare no conflict of interest.

Author Contribution

J.L. designed the experiments, collected the data, and wrote the original draft of the manuscript. N.L. performed the formal analysis and prepared the figures. L.C. supervised the project and was responsible for reviewing and editing the manuscript. J.B. contributed to the interpretation of the results and acquired the funding. All authors reviewed and approved the final manuscript.

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Data Availability Statement:

The data that supports the findings of this study is available from the corresponding authors upon reasonable request.

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Figures

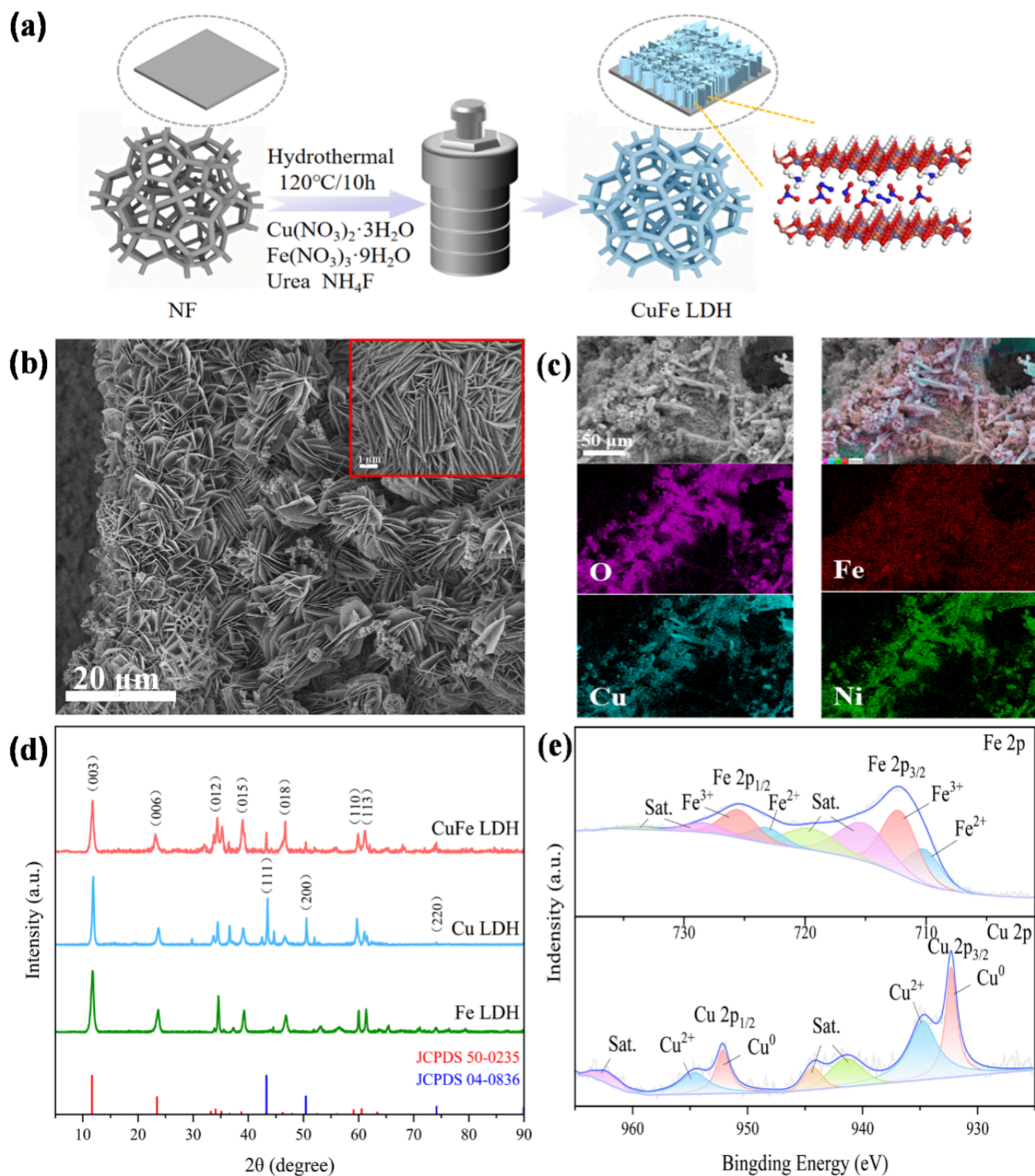


Figure 1

Characterization of the structure and composition of catalysts. (a) Schematic illustration of the preparation of CuFe LDH, (b) SEM image, (c) EDS mapping, (d) XRD patterns of Cu LDH, Fe LDH and CuFe LDH, (e) high resolution Fe 2p and Cu 2p spectra of CuFe LDH.

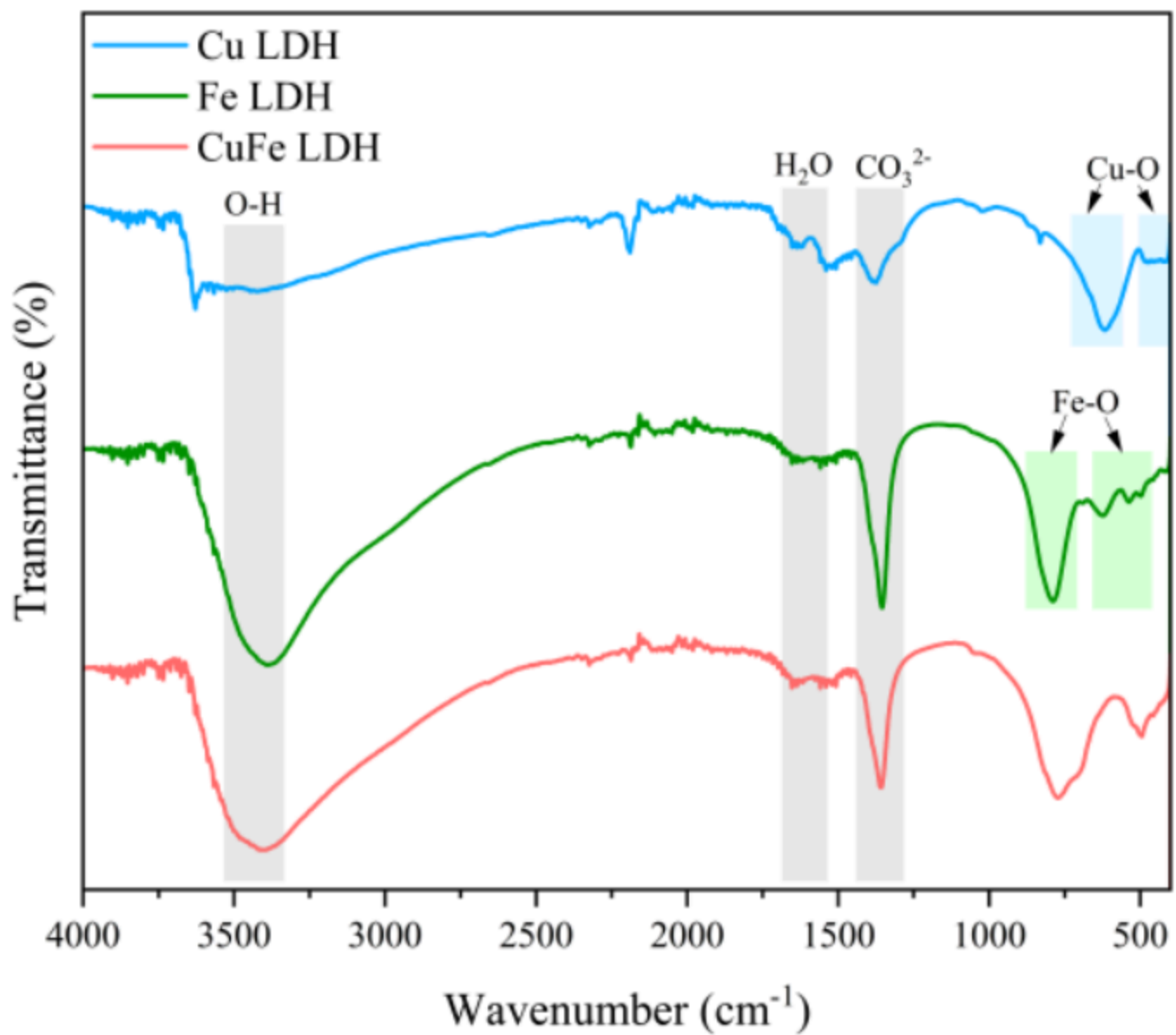


Figure 2

FTIR spectra of Cu LDH, Fe LDH and CuFe LDH.

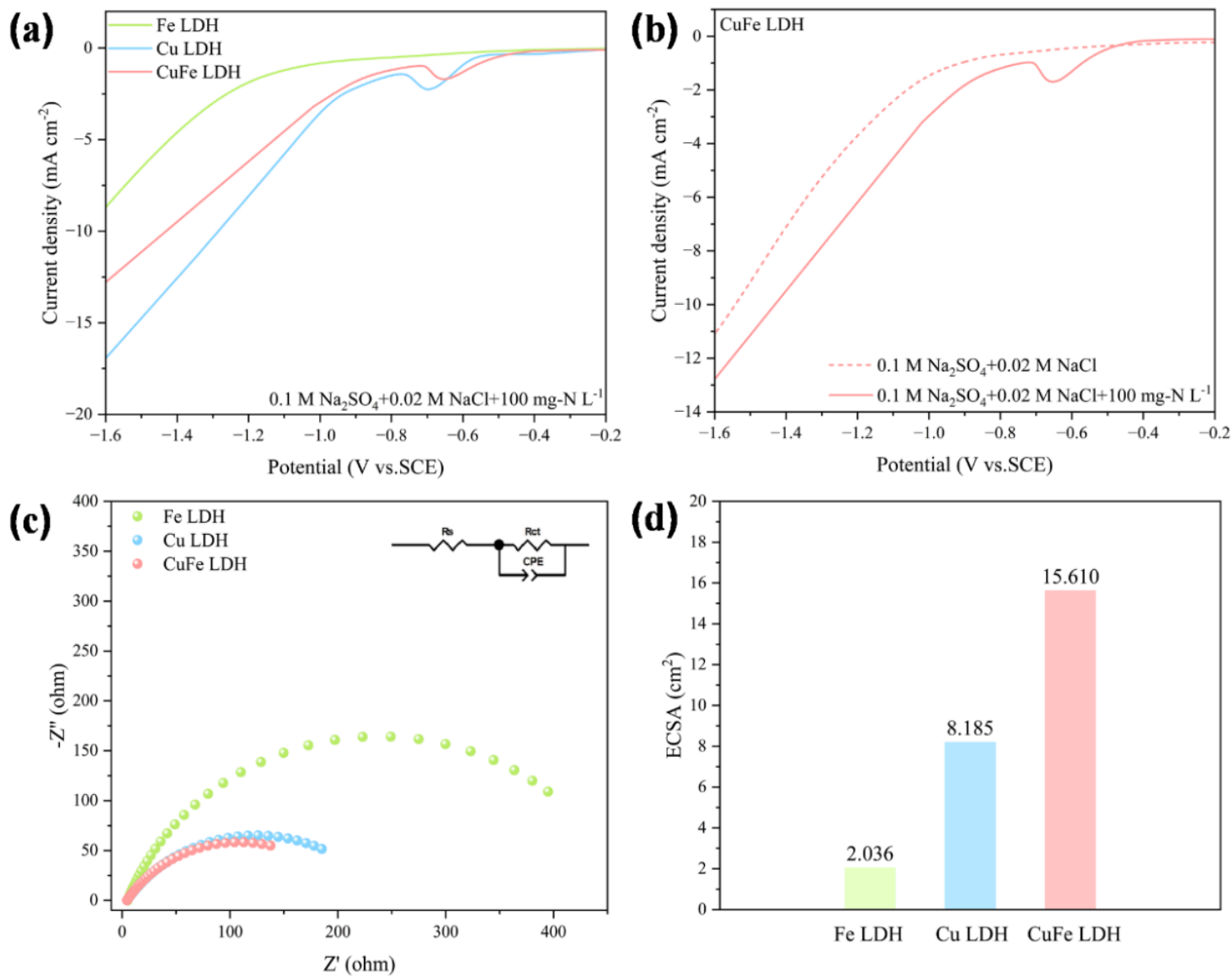


Figure 3

Electrochemical performance of catalysts. (a) LSV curve of Cu LDH, Fe LDH and CuFe LDH with nitrate, (b) LSV curve of CuFe LDH with and without nitrate, (c) EIS curve, (d) ECSA of catalysts.

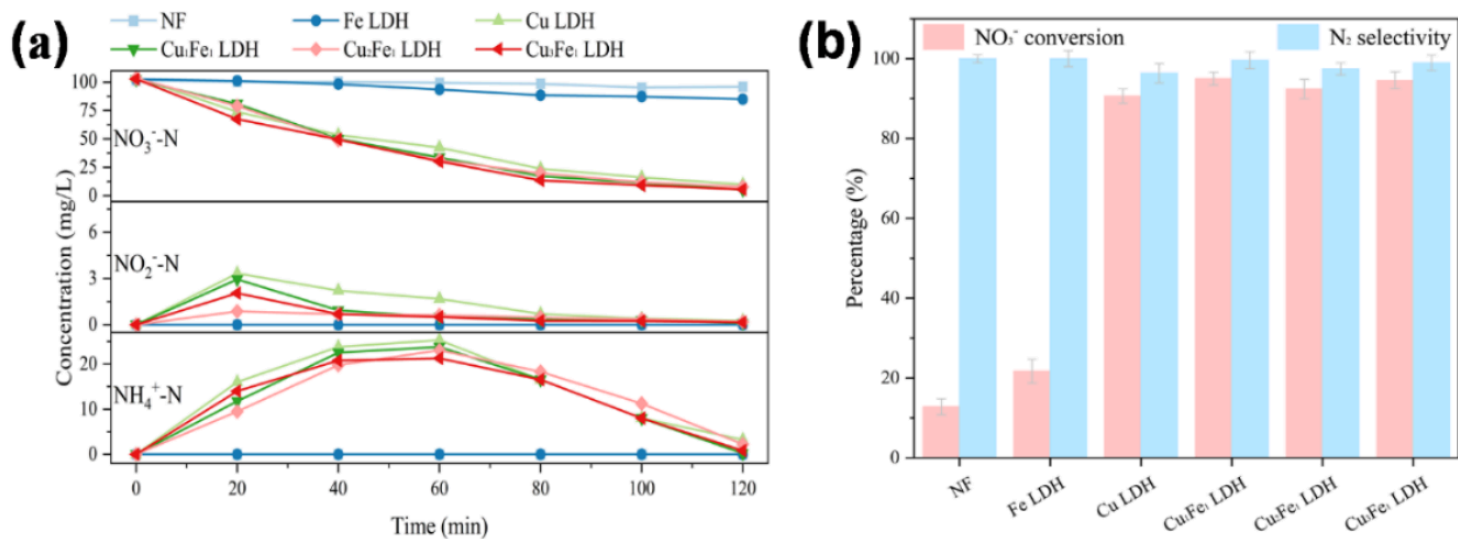


Figure 4

Electrochemical denitrification performance of CuFe LDH with different Cu-Fe ratios. (a) time-dependent NO_3^- -N, NO_2^- -N and NH_4^+ -N concentration, (b) nitrate reduction performance.

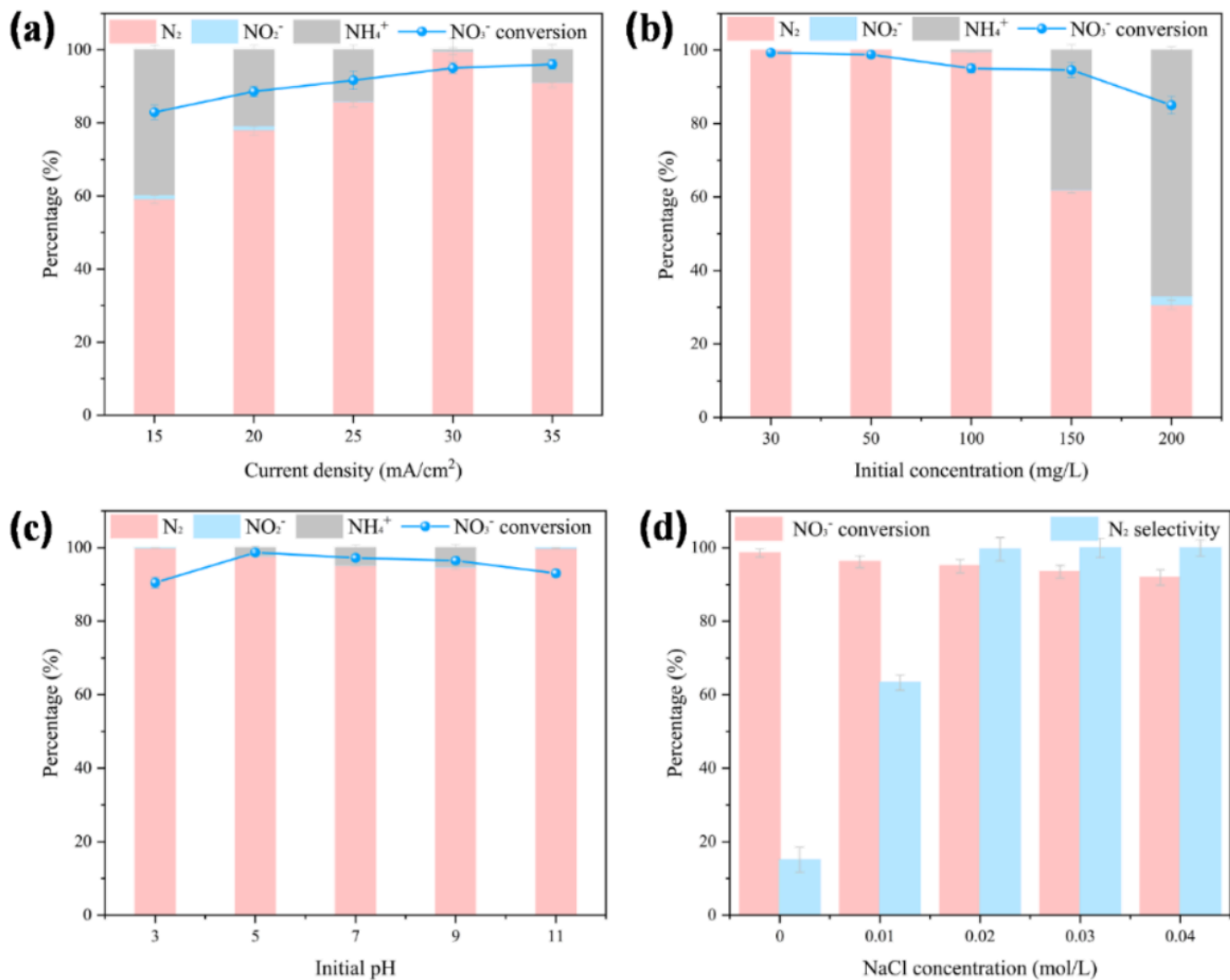


Figure 5

Electrocatalytic nitrate reduction performance of CuFe LDH under different conditions. (a) current density, (b) initial concentration, (c) initial pH, (d) electrolyte composition.

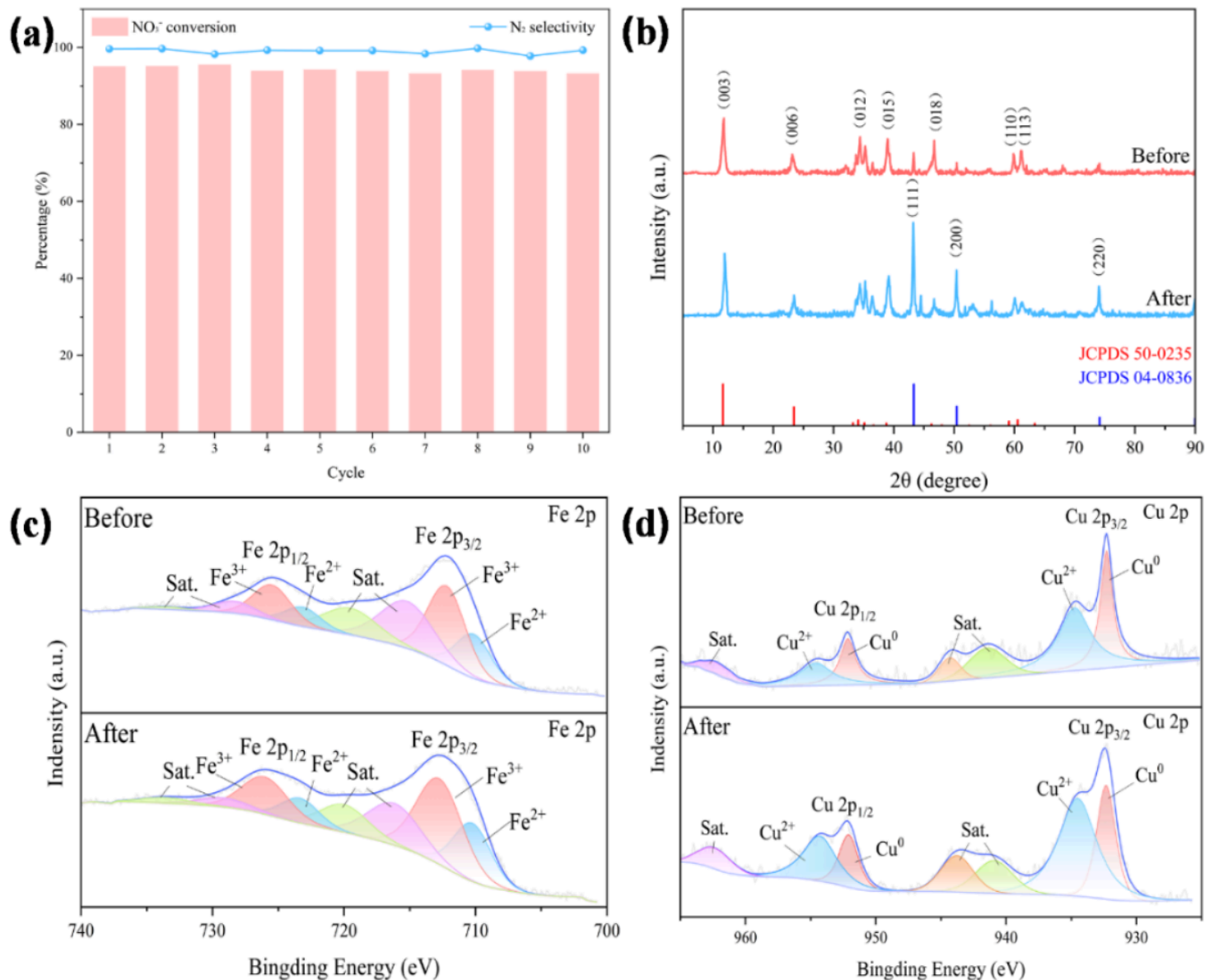


Figure 6

Long-term stability experiments of CuFe LDH. (a) Recycling tests, (b) XRD patterns before and after recycling tests, (c) high resolution Fe 2p spectra before and after recycling tests, (d) high resolution Cu 2p spectra before and after recycling tests.

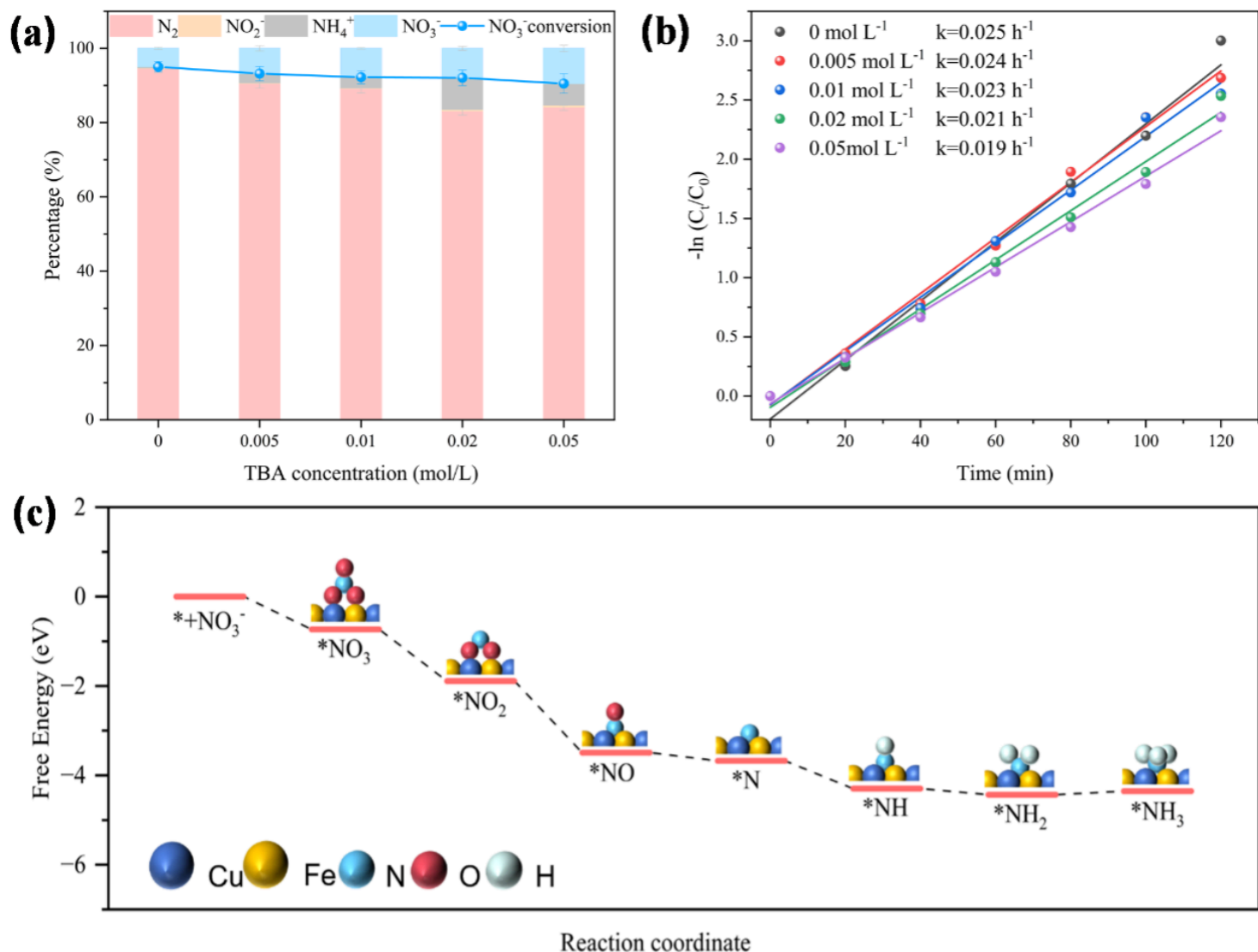


Figure 7

Effect of different concentrations of TBA on electrochemical denitrification performance of CuFe LDH and possible reaction pathways. (a) NO_3^- -N removal capacity, (b) k with different quenching reagent, (c) free energy diagram of NO_3^- reduction reaction.

Supplementary Files

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