

Article

Confined Sulfate Radicals in Layered Double Hydroxide Nanoreactors for Efficient Defluorination Reactions

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Abstract

Controlling radical selectivity within nanoreactors remains a formidable challenge due to the inherent high reactivity and short half-lives of reactive species. Herein, we report a novel size-matched nanoconfinement strategy using a cobalt-nickel-layered double hydroxide (CoNi-LDH) nanoreactor for the highly selective generation and stabilization of sulfate radicals ($\text{SO}_4^{\cdot-}$) via piezoelectric activation of peroxymonosulfate (PMS). By precisely tailoring the LDH interlayer spacing to 5.27 Å to match the kinetic diameter of $\text{SO}_4^{\cdot-}$, the nanoreactor effectively suppresses non-selective side reactions and radical quenching. Consequently, the CoNi-LDH achieves an unprecedented reaction rate ($k_{\text{obs}} = 0.40 \text{ min}^{-1}$) and superior defluorination efficiency (78.9%) for fluoroquinolone antibiotics, significantly outperforming non-size-confined counterparts. Mechanistic insights reveal a synergistic pathway where piezo-generated hot electrons, mediated by Ni sites, accelerate the $\text{Co}^{2+}/\text{Co}^{3+}$ redox cycle to ensure long-term catalytic stability. The robustness of this nanoconfined system is further demonstrated by its exceptional tolerance to complex water matrices and its practical operability in a continuous-flow reactor. This study provides a pioneering approach for spatial radical control at the nanoscale to achieve efficient and targeted environmental remediation.

Keywords: nanoconfinement; sulfate radicals; fluoroquinolone antibiotics; Fenton-like reaction; defluorination reaction



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1. Introduction

Fluorinated organic contaminants, particularly fluoroquinolone antibiotics (FQs) in pharmaceutical wastewater, are emerging pollutants that pose significant threats to the environment and human health globally [1,2]. The strategic incorporation of fluorine atoms into these molecules, primarily as stable carbon–fluorine (C–F) bonds (~485 kJ/mol) [3], is critical in improving critical drug properties like bioavailability and target affinity. Paradoxically, the chemical stability of FQs renders them resistant to environmental degradation, resulting in their persistent accumulation in ecosystems and the accelerated spread of resistance genes [3–5], thereby exacerbating the global antibiotic resistance crisis and presenting a daunting challenge for their removal.

While physicochemical methods like adsorption and membrane filtration can sequester FQs, they merely transfer the pollutant from one phase to another, accompanied by secondary waste disposal issues [6]. In contrast, advanced oxidation processes (AOPs) offer a destructive solution by generating hydroxyl radical ($\cdot\text{OH}$), a potent oxidant

($E_{\text{red}} = 2.80 \text{ V vs. NHE}$) [7]. Unfortunately, the short lifespan and non-selective nature of $\cdot\text{OH}$ make it prone to be scavenged by background constituents in real water matrices, including inorganic species and organic matter [8]. Meanwhile, the electrophilic attack mediated by $\cdot\text{OH}$ fails to efficiently cleave the electron-deficient C–F bond, accumulating a complex mixture of intermediates retaining the C–F bond, which exhibit equal or even greater biotoxicity than the parent FQ molecules [9–12]. For comparison, the sulfate radical ($\text{SO}_4^{\cdot-}$) has emerged as a superior alternative, owing to its comparable oxidation power to that of $\cdot\text{OH}$, but with a significantly longer half-life [13], and capability to work across a broad operational pH range [14–19]. More importantly, $\text{SO}_4^{\cdot-}$ operates via a single-electron transfer process, enabling a targeted attack on electron-rich sites (–F) and tending to induce molecular rearrangement reactions to initiate oxidative defluorination [20,21]. However, practical efficiency of $\text{SO}_4^{\cdot-}$ is limited by physical mass-transfer barriers due to the slow, random movement of $\text{SO}_4^{\cdot-}$ and FQs through the bulk solution, leading to non-productive consumption of $\text{SO}_4^{\cdot-}$ before defluorination occurs. In light of this, engineering a reaction microenvironment that can steer the reaction toward selective defluorination and eliminating radical quenching is a prerequisite for safe, practical remediation of FQs in real wastewater.

Nanoconfinement is an emerging paradigm for engineering highly selective chemical transformations by enforcing molecular-scale proximity and providing a physical shield for reactive intermediates within well-defined nanospaces [22–26]. Despite successful demonstrations of generating confined radicals within nano-reactors, the subsequent confinement of these highly selective radicals for controlled reactions remains a significant challenge. Confinement at the nanoscale can modulate chemical reactivity and enhance selectivity, but maintaining high selectivity for radical species is difficult due to their inherent reactivity and tendency to undergo side reactions. By imposing strict geometric constraints, nanoconfinement offers the tantalizing prospect of harnessing the high reactivity of $\text{SO}_4^{\cdot-}$ and enriching target pollutants in close proximity, thereby enforcing the defluorination cascade governed by $\text{SO}_4^{\cdot-}$. Among various host materials, layered double hydroxides (LDHs) with tunable 2D nanochannels ($\sim 0.5\text{--}2 \text{ nm}$) stand out as a reliable platform to implement this strategy [27,28]. Their positively charged brucite-like layers offer an electrostatic driving force for the co-enrichment of both anionic persulfate precursors (e.g., peroxymonosulfate, PMS) and FQs of rich functional groups within their interlayers as a microenvironment reactor [29–31], and enable the piezoelectric catalytic performance in AOPs [27]. When catalytic transition metals (e.g., Co, Ni, Fe) are integrated into the LDH framework, these nanolayers become preferred sites for the in situ generation of shielded $\text{SO}_4^{\cdot-}$, which can preferentially direct the highly reactive $\text{SO}_4^{\cdot-}$ toward the target contaminant and increase the effective collision frequency, thus accelerating the defluorination kinetics.

Recently, significant efforts have been directed towards engineering the coordination environment and electronic structure of layered double hydroxides (LDHs) to boost their catalytic efficiency. For instance, the construction of coordinatively unsaturated Co–Cu dual active sites on ultra-thin LDHs has been shown to achieve ultrafast electron transfer for peroxymonosulfate (PMS) activation [32]. Meanwhile, the integration of high-entropy strategies into LDH frameworks has been reported to precisely steer inert sites, thereby facilitating the removal of complex pollutants like microplastics [33]. Beyond traditional environmental applications, advanced orbital coupling mechanisms, such as frustrated Lewis pair (FLP)-mediated f–p–d coupling, have demonstrated exceptional selective permeability and ion repulsion in seawater electrolysis, providing a biomimetic inspiration for achieving high selectivity in radical-based systems [34]. Furthermore, the hybrid design of Mg/Fe-LDH with biochar has been explored to enhance trichloroethylene remediation through synergistic radical and non-radical pathways [35].

However, despite these remarkable advancements in augmenting the total yield of reactive oxygen species (ROS), most existing LDH-based catalytic systems still suffer from the random diffusion and rapid self-quenching of radicals in bulk solutions. This lack of spatial control leads to suboptimal selectivity towards specific highly reactive $\text{SO}_4^{\cdot-}$, especially when facing the interference of complex water matrices. To realize the nanoconfinement of $\text{SO}_4^{\cdot-}$, in the study, we successfully engineered a unique nanorod-structured CoNi-LDH with (020) exposed surface that possessed a precisely tailored interlayer spacing of 5.27 Å, matching the kinetic diameter of the $\text{SO}_4^{\cdot-}$, estimated to be approximately 4.5–5.0 Å. Compared with the (001) exposed surface CoNi-LDH nanodisk counterpart with a wider 8.86 Å spacing [28], CoNi-LDH nanorod could effectively confine the $\text{SO}_4^{\cdot-}$ selectively highly generated from the piezoelectric activation of peroxymonosulfate (PMS) within its interlayer, boosting the ciprofloxacin defluorination rate from 43.9% to 78.9% and elevating the mineralization efficiency from 30.5% to 80.7%, meanwhile achieving the highest observed rate constant of 0.40 min^{-1} . We demonstrated that Co^{2+} within CoNi-LDH nanorod initiates a Fenton-like activation of PMS, while the piezoelectric process generates hot electrons under ultrasonic vibration, which are efficiently shuttled by the Ni sites as an electron mediator to accelerate the $\text{Co}^{2+}/\text{Co}^{3+}$ redox cycle, thereby ensuring long-term stability and practical operability within a continuous-flow piezoelectric-catalytic filter reactor. Furthermore, the robustness of the nanoconfined $\text{SO}_4^{\cdot-}$ was validated by its strong tolerance to coexisting ions and broad pH adaptability, displaying excellent degradation performance in authentic lake/river water matrices. Our findings offer new insights into bond-cleavage-targeted antibiotic degradation and pave the way for efficient wastewater remediation technologies through size-matched confinement.

2. Results and Discussion

2.1. Characterizations of CoNi-LDH

To identify the size-matched confinement for enhancing the ciprofloxacin (CIP) degradation, we characterized the interlayer spacings of CoNi-LDHs with different morphologies of nanorod and nanodisk-shaped structures synthesized via varying Co/Ni ratios, as shown in Figure 1a. Transmission electron microscopy (TEM) analysis (Figure 1b) divulged that CoNi-LDH possesses a nanorod morphology with diameters of ~50 nm and lengths extending to several micrometers. Tuning the Co/Ni ratio induced a morphological transition from nanorods to nanodisks in other LDHs (Figure S1). High-resolution TEM (HRTEM) images of CoNi-LDH (Figure 1c–e) showed lattice fringes with a spacing of 0.527 nm, corresponding to the (020) planes of LDH. This interlayer distance (0.527 nm) is comparable to the diameter of the sulfate radical ($\text{SO}_4^{\cdot-}$, ~0.5 nm) [36], suggesting potential radical confinement within the layered structure. However, the interlayer spacing of the CoNi-LDH (1:4) with the nanodisk morphology was about 0.886 nm, corresponding to the (001) crystal plane [23], which was not fit for the size confinement of the $\text{SO}_4^{\cdot-}$ due to the large size. Furthermore, STEM-energy dispersive X-ray spectroscopy (EDS) mapping (Figure 1f–i) confirmed the homogeneous distribution of Co, Ni, and O elements throughout the CoNi-LDH structure. Powder X-ray diffraction (XRD) analysis (Figure S2) corroborated that CoNi-LDH exhibits the characteristic pattern of a typical LDH structure (JCPDS PDF# 048-0083). The macroscopic crystalline structure was further cross-verified by comparing XRD and HRTEM data. The diffraction feature corresponding to the (020) plane aligns well with the periodic lattice fringes observed in the HRTEM images (Figure 1c–e). This multi-scale consistency confirms that the nanorods possess a highly ordered layered structure with a precisely tailored 5.27 Å interlayer environment. Inductively coupled plasma optical emission spectroscopy (ICP-OES) quantification determined the Co and Ni content in CoNi-LDH to be 38.5 wt% and 9.26 wt%, respectively,

consistent with the target Co/Ni molar ratio of 4:1. Therefore, the XRD pattern, elemental composition, and observed nanorod morphology validate the successful synthesis of CoNi-LDH. Furthermore, to unveil the piezoelectric catalytic performance of CoNi-LDH for driving the degradation processes, we used the atomic force microscopy (AFM) and piezo-response force microscopy (PFM) to characterize surface and piezoelectric potential. The surface potential of CoNi-LDH (Figure S3) was measured at +58.5 mV, a value beneficial for accelerating the separation of piezoelectric-induced electrons and holes. The piezoelectric properties of CoNi-LDH were further examined using piezo-response force microscopy (PFM). Figure 1j shows the characteristic PFM response: a butterfly-shaped amplitude loop and a 180° phase hysteresis loop, probed by applying an alternating voltage (± 10 V) across the sample surface. This behavior substantiates a significant piezoelectric response and polarization reversal in CoNi-LDH [27]. The piezoelectric amplitude exhibited a positive correlation with increasing AC drive voltage, further verifying the intrinsic piezoelectricity of the CoNi-LDH. Specifically, the CoNi-LDH-nanorod displayed a larger butterfly-loop amplitude compared to the CoNi-LDH-nanodisk (Figure S4). The piezoelectric coefficient (d_{33}) is another parameter to measure the piezoelectric properties of the samples [37]. The maximum effective d_{33} values were 51.7 pm V^{-1} for CoNi-LDH-nanorod and 27.5 pm V^{-1} for the nanodisk sample (Figure 1j and Figure S4). This substantial difference validates the superior piezoelectric properties of CoNi-LDH, implying its greater propensity to generate stronger piezoelectric polarization and enhance charge carrier transfer.

To further confirm the piezoelectric properties of CoNi-LDH, we measured its transient piezoelectric current density. As shown in Figure 1k, the piezoelectric current density of CoNi-LDH parallels with the elevation of the ultrasonic power (Figure S5) and demonstrates that the current density is significantly dependent on the Co/Ni ratio, where the nanorod-shaped CoNi-LDH was higher than that of nanodisk-shaped CoNi-LDH. Furthermore, by combining the bandgap from the diffuse reflectance spectroscopy spectra, Tauc plot and Mott–Schottky analysis (Figure S6), we constructed the band structure diagram for CoNi-LDH (Figure 1l). It indicated piezoelectric-induced band bending could generate the electrons and holes for the charge transfer to improve the degradation efficiency of targeted pollutants.

2.2. Catalytic Degradation of Fluoroquinolone Antibiotics

To evaluate the degradation efficiency of the newly developed nanorod CoNi-LDHs for challenging fluorinated organic contaminants like fluoroquinolone antibiotic ciprofloxacin (CIP), we constructed the size-matched confinement nanoreactor. In parallel, nanodisk CoNi-LDHs were assembled for comparison studies. To our delight, the nanorod-shaped CoNi-LDH showed exceptional activity (Figure 2a), achieving near-complete ($\sim 100\%$) CIP decomposition within 15 min via peroxymonosulfate (PMS) activation under ultrasonication (US). Control experiments ruled out the potential contributions from non-piezoelectric catalytic processes, adsorption, or direct sonolytic oxidation during ultrasonic irradiation. Only the system combining CoNi-LDH, PMS, and ultrasonication exhibited high degradation activity. Furthermore, in addition to CIP, CoNi-LDH can completely remove other FQs including levofloxacin (LEV) and norfloxacin (NOR) within 15 min (Figure 2b), indicating its generality and broad applicability for antibiotic treatment. The studies clearly show that the morphologies of CoNi-LDH significantly influenced the activity in the degradation of the FQs. As displayed in Figure 2c and Figure S7, CoNi-LDH with the nanorod structure exhibited the highest activity in degrading CIP with an observed rate constant (k_{obs}) of 0.40 min^{-1} , surpassing the nanodisk-shaped CoNi-LDHs (0.18 min^{-1}), and represents the highest k_{obs} value for CIP degradation among the values reported so far (Figure 2d and Table S1).

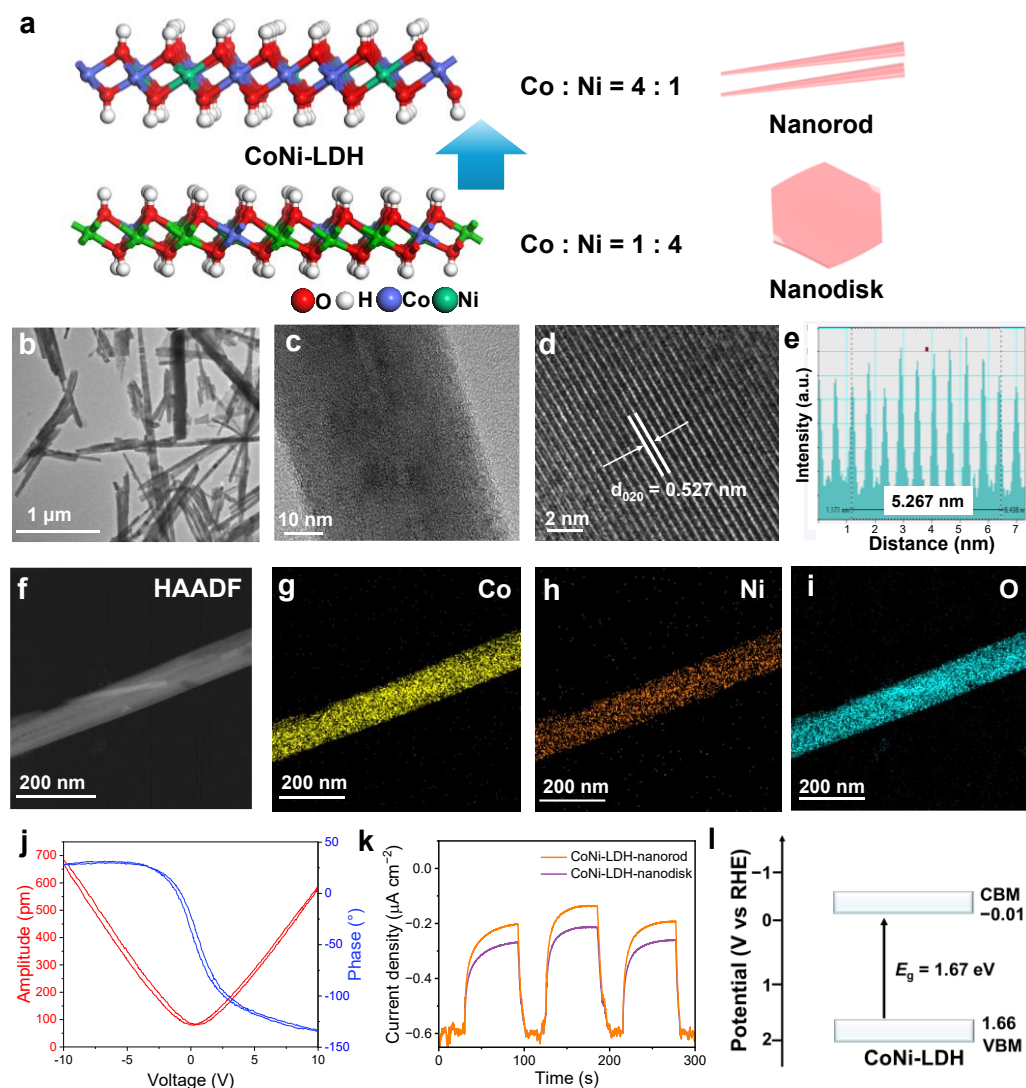


Figure 1. (a) Schematic illustration of CoNi-LDH with varying Co/Ni ratios to control nanorod and nanodisk morphologies. (b) TEM image showing the nanorod morphology of CoNi-LDH. (c) HRTEM image of CoNi-LDH. (d) Magnified region of (c) highlighting the layered structure and corresponding lattice spacings. (e) Measurement of interlayer distances from (d). (f–i) STEM-EDS elemental mapping confirming homogeneous distribution of Co, Ni, and O in CoNi-LDH. (j) Characteristic PFM hysteresis loops: butterfly-shaped amplitude and 180° phase response under ± 10 V bias of CoNi-LDH. (k) Compositional and morphologies dependence (Co/Ni ratio, nanorod and nanodisk) of piezoelectric current density. (l) Band energy diagram of CoNi-LDH (CBM: conduction band minimum, VBM: valence band maximum).

Since pH can affect the degradation activity significantly, we assessed the effect of various pHs on the PMS-mediated piezoelectric catalytic Fenton-like process for the treatment of FQ contaminated wastewater (Figure 2e). Notably, the CoNi-LDH maintained high CIP degradation efficiency across a wide pH range (4.0–10.0) and overcame the complexity of real water bodies in wide pH applicability. In addition, degradation efficiency of CIP for CoNi-LDH increased with US power and plateaued above 200 W, as shown in Figure S8a. Reaction temperature had no significant effect, demonstrating that piezoelectric catalytic properties are not thermally induced (Figure S8b). Furthermore, to substantiate the piezoelectric catalytic mechanism of CoNi-LDH under different driving modes, we equated the mechanical stirring and ultrasonication methods. Degradation efficiency of CIP for CoNi-LDH increased with stirring speed, reaching levels equivalent to 250 W ultrasonication at 1200 rpm (Figure S8c). This corroborates that mechanical vibration induced by water flow

can also effectively drive the piezoelectric catalytic reaction. It denoted that the CoNi-LDH exhibited high activity in the degradation of CIP over a wide pH range and driven in the water flow for a practical application.

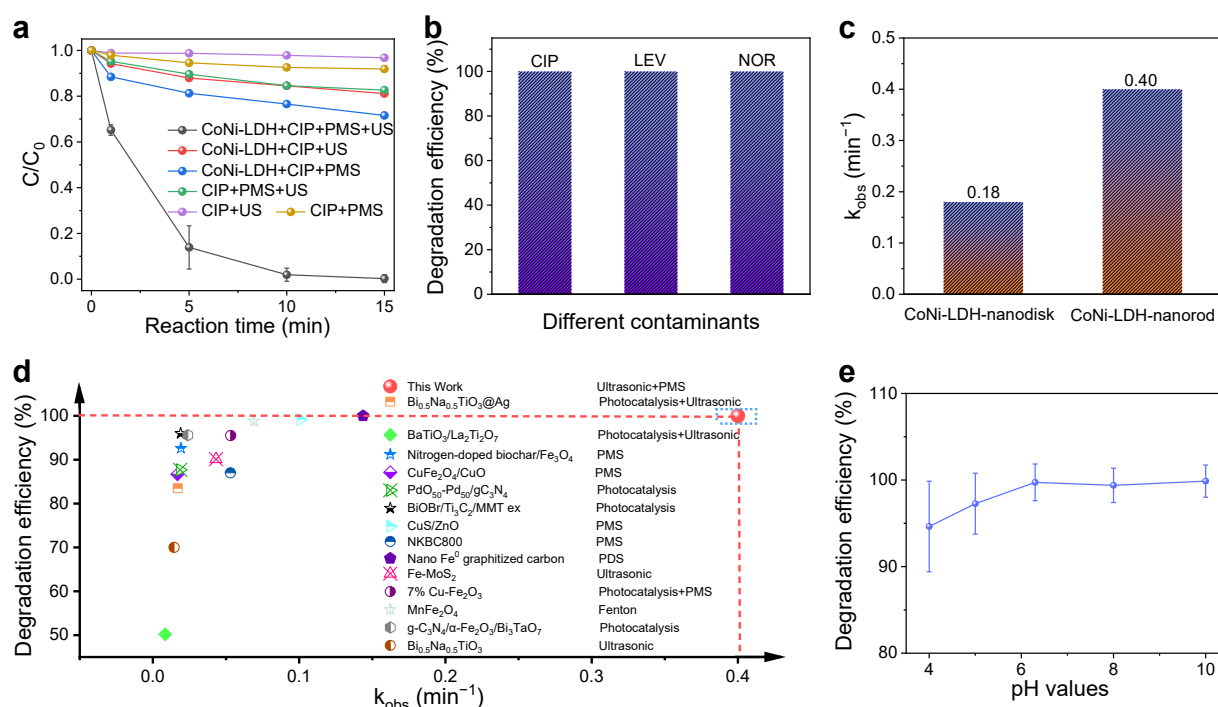


Figure 2. (a) Control experiments confirming the piezoelectric catalytic Fenton-like reaction mechanism for CIP degradation using CoNi-LDH with peroxymonosulfate (PMS) activation. (b) Degradation efficiency of CoNi-LDH for CIP and other fluoroquinolone antibiotics: levofloxacin (LEV) and norfloxacin (NOR) within 15 min. (c) The observed rate constants (k_{obs}) of the CIP for CoNi-LDHs with the morphologies of nanorod and nanodisk. (d) Comparison of CoNi-LDH with reported referenced catalysts and driven methods for the degradation of CIP. (e) pH dependence of CIP degradation efficiency for CoNi-LDH within 15 min.

2.3. Catalytic Mechanism and Reactive Species Trapping Experiments

To elucidate the mechanism of peroxymonosulfate (PMS) activation, electron transfer, and radical effects in the CoNi-LDH piezoelectric catalytic Fenton-like system—specifically the confined $SO_4^{\cdot-}$ radical into the interlayer spacings (5.27 Å) for FQ degradation—we performed radical scavenging experiments to identify the dominant reactive oxygen species (ROS). As shown in Figure 3a,b, methanol (MeOH), a scavenger for both $\cdot OH$ and $SO_4^{\cdot-}$, significantly suppressed CIP degradation efficiency in the CoNi-LDH/PMS/US system. Similarly, *tert*-butanol (TBA) and isopropyl alcohol (IPA), which preferentially scavenge $\cdot OH$ radical, caused substantial inhibition. These results demonstrate that $\cdot OH$ and $SO_4^{\cdot-}$ are the primary ROS responsible for CIP degradation. In contrast, the addition of furfuryl alcohol (FFA) (a scavenger for singlet oxygen, $^1O_2$) or dimethyl sulfoxide (DMSO) (a scavenger for high-valent cobalt, $Co(IV)=O$) resulted in only minor inhibition. This implies that $^1O_2$ and $Co(IV)=O$ play negligible roles in the degradation process. In addition, the degradation efficiency remained unaffected when adding scavengers for charge carriers: $AgNO_3$ (electron, e^- scavenger) or ammonium oxalate (AO) (hole, h^+ scavenger). This suggests that piezoelectric-induced electrons and holes do not directly prompt the primary degradation reactions. N_2 purging (to eliminate dissolved O_2 and potential superoxide anion radicals, $O_2^{\cdot-}$) also showed no significant effect, indicating $O_2^{\cdot-}$ does not play a major role in the CoNi-LDH/PMS/US system.

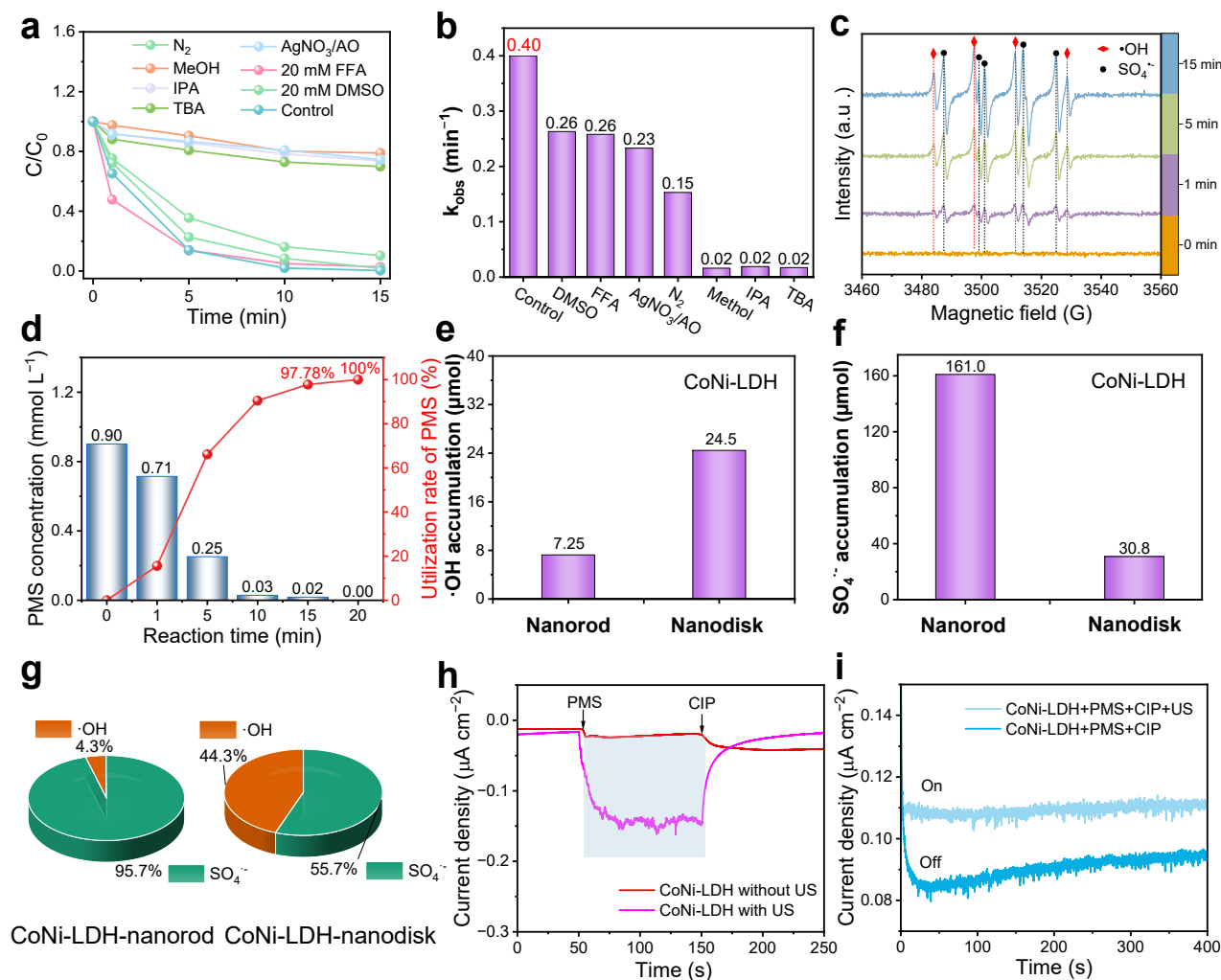


Figure 3. (a) The influence of different scavengers on the degradation characteristics of CIP for the CoNi-LDH. Other concentrations of the scavengers are 100 mM. (b) Comparison of the observed rate constant for the CIP removal using the different scavengers. (c) EPR spectra of DMPO-·OH and DMPO-SO₄·- with different reaction times. (d) The PMS concentration and utilization rate of PMS in the degradation processes of CIP. (e,f) Quantitative analysis and proportion of reactive oxygen species (ROS) produced by CoNi-LDH for ·OH (e) and SO₄·- (f). (g) Contribution of reactive oxygen species (ROS) in CoNi-LDHs with the nanorod and nanodisk. (h) Piezoelectric catalytic current density of CoNi-LDH with/without ultrasonication (US) upon sequential addition of PMS and CIP. (i) The piezoelectric current density of CoNi-LDH with/without US.

Furthermore, electron paramagnetic resonance (EPR) spectroscopy confirmed the generation of the dominant reactive oxygen species ·OH and SO₄·- within the confined CoNi-LDH/PMS/US system. Using 5,5-dimethyl-1-pyrroline N-oxide (DMPO) as the spin-trapping agent, characteristic signals for these ROS were clearly observed under ultrasonication with the increase of time (Figure 3c). The distinct peaks, exhibiting varying intensities, were absent under dark conditions. These results directly support the involvement of ·OH and SO₄·- radicals in the piezoelectric catalytic Fenton-like degradation process. Additionally, PMS utilization efficiency is a key parameter for evaluating PMS activation. The CoNi-LDH system achieved nearly 100% PMS consumption within 20 min (Figure 3d), demonstrating highly efficient oxidant usage that surpasses values reported for comparable catalysts.

To further elucidate the roles of ·OH and SO₄·- radicals in the piezoelectric catalytic Fenton-like destruction of FQs, we quantified their generation in CoNi-LDHs with the

morphologies of the nanorod and nanodisk (Figures S9 and S10). As shown in Figure 3e,f, it can be seen from the quantitative results that under the condition of ultrasonication for 15 min in the CoNi-LDH/PMS/US system, the generation yields of $\cdot\text{OH}$ in the CoNi-LDH-nanorod and CoNi-LDH-nanodisk were 7.25, and 24.5 μmol , respectively, while the concentrations of the $\text{SO}_4^{\cdot-}$ for LDHs were 161 and 30.8 μmol , respectively. To determine the percentages of the two free radicals, we found that the $\text{SO}_4^{\cdot-}$ in CoNi-LDHs was highly selectively generated and dominated at 95.7%, while the percentage of the $\cdot\text{OH}$ was only 4.3% (Figure 3g). This radical speciation aligns with the distinct catalytic roles of the metal sites: Co centers are primarily responsible for $\text{SO}_4^{\cdot-}$ generation for accelerating the $\text{Co}^{2+}/\text{Co}^{3+}$ cycling, while Ni sites mainly facilitate electron transfer for improving the performance of $\cdot\text{OH}$ generation rate. Consequently, $\text{SO}_4^{\cdot-}$ can act as the pivotal radical species driving CIP degradation within the CoNi-LDH system.

To elucidate the electron transfer pathways among FQs, CoNi-LDH, and PMS, we carried out transient current density measurements in a dual-cell setup connected via a salt bridge at open-circuit potential. As shown in Figure 3h, upon introducing PMS into the system, the piezoelectric catalytic current density of CoNi-LDH under US was $-0.15 \mu\text{A cm}^{-2}$ higher than that without US. This signifies that the piezoelectric effect generates electrons within CoNi-LDH, which are subsequently transferred to PMS for its activation, leading to the generation of the two key radicals ($\cdot\text{OH}$ and $\text{SO}_4^{\cdot-}$). The addition of FQ CIP to the system also significantly decreased the piezoelectric catalytic current density. This suggests that CIP donates electrons to CoNi-LDH, compensating for the consumption of piezoelectrically generated electrons. Collectively, these observations support an electron transfer pathway from CIP to CoNi-LDH, and then to PMS for radical generation. Furthermore, in the CoNi-LDH/PMS/US system, the piezoelectric catalytic current density was consistently higher than that without US (Figure 3i). Electrochemical impedance spectroscopy (EIS) measurements, fitted using a standard equivalent circuit (Figure S11), revealed resistances of 85.9 Ω and 111.1 Ω for CoNi-LDH films with and without US, respectively. These results show that under US, the CoNi-LDH/PMS system exhibits enhanced piezoelectric catalytic current density and reduced resistance. This facilitates the transfer of piezoelectrically generated electrons and improves the overall conductivity of the system.

2.4. Defluorination Degradation Pathways and Size-Matched Confinement Mechanism

The degradation pathways of organic molecules are fundamentally governed by their structural properties. For ciprofloxacin (CIP), surface electrostatic potential (ESP) analysis (Figure 4a and Figure S12) reveals the electrophilic and nucleophilic characters of individual atoms, quantified in Table S2. Complementary analysis using the condensed Fukui function (specifically the condensed dual descriptor, CDD) further consolidates reactive sites. As listed in Table S3, the strongly negative CDD value at N37 indicates high nucleophilicity, rationalizing nucleophilic attack-driven cleavage of the piperazine ring. The positive CDD value at F27 corroborates electrophilic character, favoring electrophilic addition/substitution reactions. Furthermore, comparative analysis of CDD and ESP values for $\cdot\text{OH}$ and $\text{SO}_4^{\cdot-}$ radicals (Figure S13) suggest distinct reactivity preferences: the $\text{SO}_4^{\cdot-}$ exhibits pronounced nucleophilic character, predisposing it to attack electrophilic sites. While the $\cdot\text{OH}$ displays dual reactivity (nucleophilic/electrophilic), enabling broader reaction pathways.

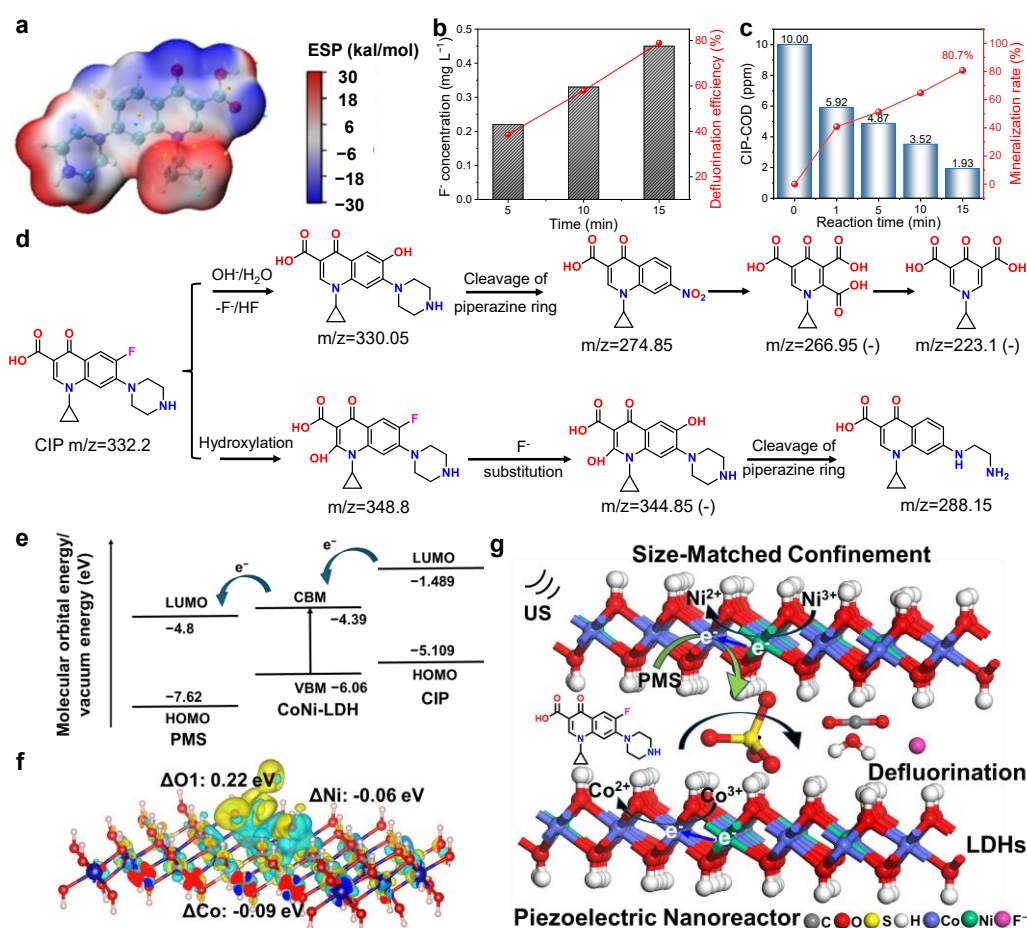


Figure 4. (a) Electrostatic potential surface of CIP molecule. (b) The F⁻ anion concentration and defluorination efficiency in the CIP degradation processes. (c) The chemical oxygen demand (COD) values of CoNi-LDH for degrading the CIP at different reaction times. (d) The proposed defluorination degradation pathways for CoNi-LDH in the piezoelectric catalytic Fenton-like reaction. (-) represents the negative ion mode; others are the positive ion mode. (e) The diagram of the electron transfer in the PMS, CoNi-LDH and CIP. LUMO: Lowest Unoccupied Molecular Orbital, HOMO: Highest Occupied Molecular Orbital. (f) Differential charge of the CoNi-LDH system after the adsorption of CIP on the Co site. The values of the atoms were the Bader charge and charge difference plot with the isosurface = 0.0016, with the yellow region indicating charge accumulation and the green region the charge depletion. (g) The proposed mechanism of the size-matched confinement for the defluorination reaction of CIP by ultrasonic-induced piezoelectric catalysis.

To assess defluorination and mineralization during CIP degradation by the CoNi-LDH, we monitored fluoride ion release and chemical oxygen demand (COD) over time (Figure 4b,c). Within 15 min, the system achieved 78.9% defluorination efficiency and a 80.7% mineralization rate, while the CoNi-LDH-nanodisk with the large size of anion layer (~0.886 nm) exhibited only 43.9% defluorination efficiency, as shown in Figure S14, and a 30.5% mineralization rate. Liquid chromatography–mass spectrometry (LC-MS) analysis identified degradation intermediates (Figure S15), enabling proposal of two dominant pathways in the CoNi-LDH/PMS/US system, as shown in Figure 4d. Firstly, the direct nucleophilic [•]OH radical substitution at the C6-F27 bond, releasing the F⁻, occurred through the defluorination processes [14,15]. Then the N37 atom was attacked by the nucleophilic reaction via the cleavage of the piperazine ring. The other way was that the C11 site was substituted by the [•]OH via oxidation. Then the F27 was nucleophilically replaced by the [•]OH for the defluorination reaction. Furthermore, breaking the ring of piperazine occurs at the C28 and C30. The results showed a high mineralization rate (80.7%) that enabled

the generation of harmless final products (CO_2 , H_2O and F^-), demonstrating effective and environmentally friendly detoxification. However, for the nanodisk-shaped CoNi-LDH without the size-matched confinement, the dominant degradation reactions were processed through the replacement of $\cdot\text{OH}$ substitution and the cleavage of the piperazine ring to still give the residual fluoro-containing products.

The interlayer spacing of LDH typically ranges from 0.7 to 0.9 nm, depending on the anion type and water content [28]. The observed lattice spacing of CoNi-LDH was 0.527 nm (Figure 2d), which was similar to the size of the $\text{SO}_4^{\cdot-}$ with the approximate size of 0.5–0.6 nm (Figure S16), while the hydroxyl radical ($\cdot\text{OH}$) has a smaller kinetic diameter of about 0.26 nm. It was difficult to restrict its movement due to its small size compared with the interlayer distances. Therefore, the interlayer of the CoNi-LDH can confine the $\text{SO}_4^{\cdot-}$, leading to a long lifetime by similar size confinement, improving the degradation efficiency. To further explore the reaction mechanism in the CoNi-LDH/PMS/US system, we analyzed the molecular orbital energy levels of CIP and PMS [38] along with the energy band structure of CoNi-LDH (estimated from Figure 1l), as shown in Figure 4e. The electron flow can be transferred via the $\text{CIP} \rightarrow \text{CoNi-LDH} \rightarrow \text{PMS}$. We also conducted the theoretical calculation about the adsorption energy of PMS on the Co and Ni sites of CoNi-LDH (Figure S17). The lowest adsorption energy of -4.11 eV indicated that the O1 in PMS was priority to adsorb on the Co site of CoNi-LDH. Furthermore, the Bader charge suggested that the electron transfer takes place over $\text{Ni} \rightarrow \text{Co} \rightarrow \text{O1}$ between CoNi-LDH and PMS (Figure 4f), where the Ni site reduced the electrons to Co site with accumulation of electrons, and then transferred to O1 in PMS.

The proposed mechanism involves CIP losing electrons through the oxidation. These electrons were then transferred to the conduction band of CoNi-LDH. Subsequently, the piezo-induced electrons generated within the CoNi-LDH combined with the above electrons migrated to the LUMO of PMS, activating it to produce $\text{SO}_4^{\cdot-}$ radicals. These free radicals ultimately oxidize and degrade CIP. The highly selective generation of $\text{SO}_4^{\cdot-}$ was confined in the anion layer in CoNi-LDH through the size-matched confinement for the degradation of CIP via the defluorination pathway (Figure 4g and Figure S18). The Ni as the electron mediator was employed by piezoelectric electron to transfer the Co sites for the acceleration of the $\text{Co}^{2+}/\text{Co}^{3+}$ cycling for efficient PMS activation through the Fenton-like reaction.

2.5. Operational Stability Testing and Practical Application in Different Actual Water Systems

Furthermore, the stability and degradation efficiency of piezoelectric catalysts affected by the aqueous scavengers/pH interference are the critical factors for their practical application in organic pollutant treatment. As demonstrated in Figure 5a, the CoNi-LDH/PMS/US system maintained consistent CIP degradation efficiency over five consecutive cycles, indicating excellent operational stability. Post-stability characterizations by XPS and XRD (Figure S19) revealed no significant structural changes in the CoNi-LDH catalyst. Additionally, metal leaching (Co and Ni) was minimal, quantified at only <10 per billion (ppb), further supporting the catalyst's reusability. For real wastewater treatment, resilience to complex water matrices is essential to practically address persistent pollution. Real water bodies contain various inorganic anions and impurities that may compete with or react with target pollutants during oxidation processes. Figure 5b shows that common coexisting anions (HCO_3^- , NO_3^-) had negligible impact on CIP degradation efficiency in the CoNi-LDH/PMS/US system. However, Cl^- significantly suppressed efficiency ($\sim 30\%$ reduction), likely due to radical scavenging ($\text{SO}_4^{\cdot-} + \text{Cl}^- \rightarrow \cdot\text{Cl}$) [39]. Furthermore, as shown in Figure 5c, the CoNi-LDH exhibited remarkable CIP degradation efficiency in actual water (tap water, lake and river), leading to the complete removal of CIP within

20 min. Though the Cl^- could reduce the efficiency of CIP degradation, it was not affected in the actual water body, due to the multispecies contribution in the confined nanoreactor for alleviating this effect. However, they were significantly affected by the nanodisk-shaped CoNi-LDH without the size confinement, suggesting that the free radicals were remarkably quenched by the Cl^- (~80% reduction) and reacted with anions or other impurities without the shielding of the nanoconfinement, as shown in Figure S20. These results indicated that the size-matched confinement of $\text{SO}_4^{\cdot-}$ could achieve high reactivity, resulting in the extraordinary performance and excellent stability of the CoNi-LDH, leading to the practical applications for the degradation of the CIP.

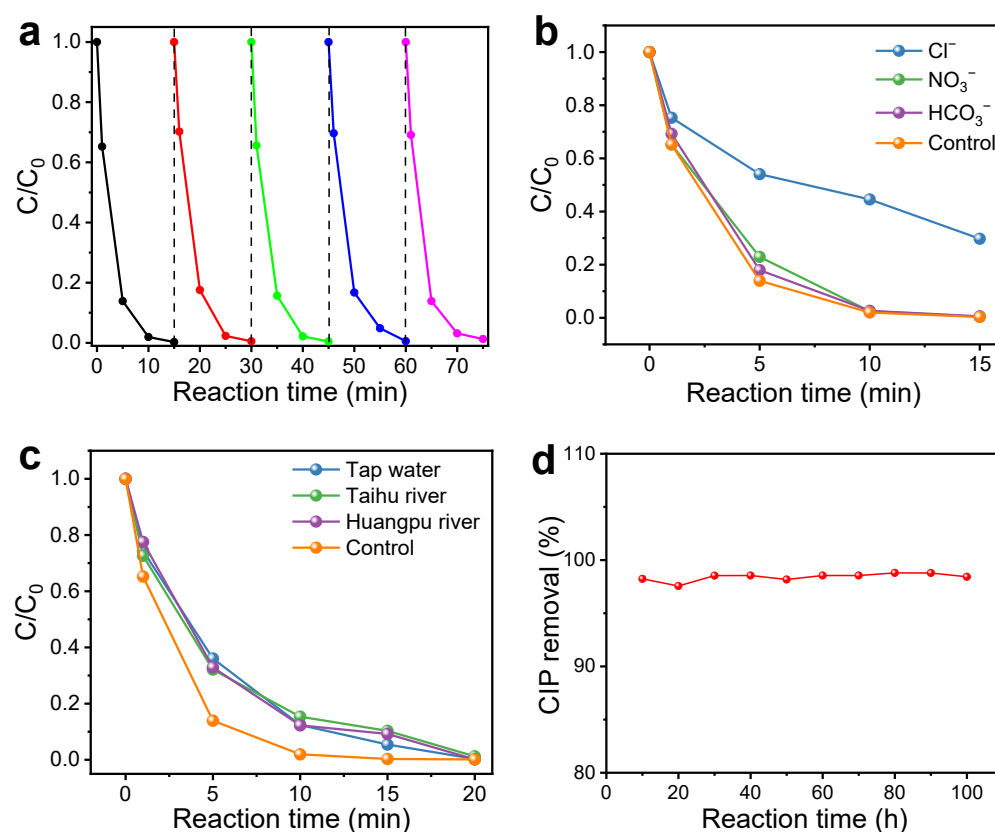


Figure 5. (a) Cycling stability of CIP degradation over five consecutive reuse cycles. (b) Effect of common inorganic anions (Cl^- , HCO_3^- , NO_3^-) on CIP degradation efficiency in simulated water matrices. (c) The CIP removal efficiency in actual water of CoNi-LDH. Samples of tap water, lake water, and river water were collected from the University of Shanghai for Science and Technology, Taihu Lake, and Huangpu River in Shanghai, respectively. (d) The CIP removal in the piezoelectric catalytic–Fenton filter (experiment conditions: the CoNi-LDH on the surface of the shredded chayote, $[\text{CIP}]_0 = 10$ ppm, $[\text{PMS}] = 100$ ppm, flow rate = 10 mL h^{-1}).

To enhance practical deployment, CoNi-LDH was thermally synthesized in situ on shredded chayote biochar to create a floating catalytic system (Figure S21). Remarkably, the degradation activity of this immobilized CoNi-LDH/chayote composite for CIP was comparable to that of the powdered CoNi-LDH catalyst (Figure S22). Notably, the quenching effect of the Cl^- could be significantly reduced. Building on this, we designed a continuous-flow reactor (Figure S23) as a proof-of-concept for practical implementation. The reactor employed the CoNi-LDH/chayote composite sandwiched between cotton layers (serving as a support matrix with negligible impact on degradation) as a piezoelectric, catalytic, Fenton-like filter. Operating at a flow rate of 10 mL h^{-1} , the system achieved a sustained CIP removal rate exceeding 98% over 100 h of continuous operation, as shown in Figure 5d. Collectively, these results demonstrate that the CoNi-LDH system exhibits high activity

towards recalcitrant CIP and possesses exceptional stability, making it highly suitable for practical wastewater treatment applications.

3. Materials and Methods

3.1. Chemical and Materials

Cobalt(II) nitrate hexahydrate ($\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), nickel(II) nitrate hexahydrate ($\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), hexamethylenetetramine (HMTA), methanol (MeOH), tert-butyl alcohol (TBA), isopropanol (IPA), furfuryl alcohol (FFA), silver nitrate (AgNO_3), ammonium oxalate (AO, $(\text{NH}_4)_2\text{C}_2\text{O}_4$), dimethyl sulfoxide, potassium peroxymonosulfate (PMS , $\text{KHSO}_5 \cdot 0.5\text{KHSO}_4 \cdot 0.5\text{K}_2\text{SO}_4$, AR > 42% KHSO_5), ciprofloxacin (CIP), levofloxacin (LEV) and norfloxacin (NOR) were purchased from Aladdin (Shanghai, China). The solutions in this experiment were prepared with deionized water. All chemicals were of analytical grade, and the materials were used without any further purification.

3.2. Synthesis of LDHs

A solution was prepared by dissolving 10 mmol of cobalt(II) nitrate hexahydrate ($\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), 2.5 mmol of nickel(II) nitrate hexahydrate ($\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), and 6.5 mmol of hexamethylenetetramine (HMTA) in 20 mL of deionized water. After stirring for 30 min, the mixture was transferred to a Teflon-lined autoclave and heated in an oven at 120 °C for 24 h. The resulting product was collected by centrifugation, washed repeatedly with deionized water and ethanol, and dried at 60 °C for 12 h, yielding CoNi-LDH with the molar ratio of Co:Ni = 4:1. The nanodisk CoNi-LDH with a Co:Ni ratio of 1:4 was synthesized in the same procedures. Unless otherwise specified, CoNi-LDH stands for nanorod-shaped LDH.

3.3. Catalyst Performance Evaluation

The degradation reactions of as-obtained samples were performed in a 150 mL glass beaker under ultrasonication to drive the reaction. In a typical experiment, the catalysts (20 mg) were mixed with the CIP solution (10 ppm, 100 mL). The mixture was first sonicated for 10 min to uniformly disperse the catalyst in the solution to achieve adsorption/desorption equilibrium. Then, the 20 mg PMS was added to the solution and the reaction time was kept at a certain interval. 3.0 mL of the reaction solution was removed at appropriate intervals and 20 mM $\text{Na}_2\text{S}_2\text{O}_4$ solution was added to quench the reaction. The solution containing the catalyst was then separated using a 0.22 μm membrane filter and added to a liquid phase vial or a glass bottle. The resulting solution was analyzed in a UV-Vis spectrophotometer at the peak of 270 nm. The steps for the trapping experiments were the same as described above, except those different concentrations of various trapping agents were added to the reaction system. In addition, the free radical capture experiments were performed by adding 10 mM of different capture agents to the beaker, and the remaining experimental procedures were the same as the above. To investigate the role of dissolved oxygen, N_2 saturated conditions were achieved by continuously sparging the solution with N_2 for 30 min prior to and during the reaction.

3.4. Analytic Method

The degradation efficiency of CIP by the catalyst was studied, and the corresponding pollutant degradation kinetics was established according to the pseudo-first-order reaction kinetics. The reaction kinetic equation is as follows:

$$\ln \frac{[\text{CIP}]}{[\text{CIP}]_0} = -k_{\text{obs}}t \quad (1)$$

where k_{obs} represents the pseudo-first-order reaction rate constant (min^{-1}); t represents the reaction time (min); $[\text{CIP}]$ denotes the pollutant concentration at time t ; $[\text{CIP}]_0$ represents the initial concentration of the pollutant.

3.5. COD Measurements

The 1.5 mL reaction solution was taken and filtered with a 0.22- μm PTFE syringe filter immediately in the degradation processes for the CoNi-LDH/PMS/US system. The above solution samples were mixed with the COD reagents comprising 1 mL of $\text{K}_2\text{Cr}_2\text{O}_7$ solution (0.16 M, in 10 $v/v\%$ $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$), 0.5 mL of HgSO_4 solution (100 g/L, in 10 $v/v\%$ $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$), and 4 mL of Ag_2SO_4 solution (10 g/L, in concentrated H_2SO_4). The mixtures were heated at 165 $^\circ\text{C}$ for 30 min and then cooled to ambient temperature. The obtained clear solutions were analyzed using a UV-visible spectrophotometer (Shimadzu 1900i, Kyoto, Japan) at 440 nm.

3.6. Characterization

Transmission electron microscopy (TEM) was performed on HT7800 (Hitachi, Tokyo, Japan) with an accelerating voltage of 120 kV. High-resolution TEM (HRTEM), high-angle annular dark-field scanning TEM, and energy-dispersive spectrometry mapping were performed with a TalosF200x (FEI, Waltham, CA, USA, equipped with Super-X EDS) electron microscope at an acceleration voltage of 200 kV. The three-dimensional morphology of all prepared samples was characterized using an Atomic Force Microscope (Bruker Dimension ICON, Santa Barbara, CA, USA). Samples were mounted on adhesive silicon plates and analyzed under different magnifications at an operating voltage of 15 kV. To characterize, the crystal structure and phase composition of the prepared CoNi-LDH were analyzed using a Bruker X-ray Powder Diffractometer (D8 Advance A25) with $\text{Cu-K}\alpha$ radiation ($\lambda = 1.5406 \text{ \AA}$), yielding X-ray diffraction patterns. The mass ratios of Ni and Co were determined using Inductively Coupled Plasma Optical Emission Spectrometry (ICP-OES, Agilent 5110, Santa Clara, CA, USA). X-ray Photoelectron Spectroscopy (XPS) measurements were conducted on a Thermo Scientific K-Alpha spectrometer (East Grinstead, UK) using 300 W Al $\text{K}\alpha$ radiation. The C 1s peak (284.6 eV) served as the reference for binding energy calibration. Elemental composition and chemical states were determined using Advantage software v5.9931 for spectral fitting. Testing conditions for ion chromatography to detect F ion to demonstrate the defluorination reaction, instrument model: Thermo IC1600 (Sunnyvale, CA, USA), mobile phase: potassium hydroxide solution, 20 mmol L^{-1} , mobile phase flow rate: 1.0 mL min^{-1} , column model: DionexTM IonPacTM AG11-H RFIC 4 $\times$ 50 mm, column temperature box temperature: 35 $^\circ\text{C}$, suppression current: 50 mA. Liquid chromatography was conducted using a Shimadzu LC-20AB (Kyoto, Japan) equipped with a Waters BEH C18 column (2.1 $\times$ 100 mm, 1.7 μm) and a mobile phase of 90% water containing 0.2% formic acid (A) and 10% acetonitrile (B) at 0.3 mL/min (injection: 5 μL). Mass spectrometric detection employed an Agilent 6550 Q-TOF (Santa Clara, CA, USA) in positive/negative ESI modes (capillary voltage: 3200 V; sheath gas: 350 $^\circ\text{C}$, 12 L/min), scanning m/z of 100–800.

3.7. Photoelectrochemical (PEC) Measurements

The PEC measurements were performed in a conventional three-electrode system using a single-chamber quartz cell on an electrochemical station (CHI-660E). A 0.5 M Na_2SO_4 aqueous solution (pH = 6.8) was used as the electrolyte for PEC measurement. A 150 μL solution was used with a mixture of Nafion solution (10 μL , 5%), catalysts (3 mg) and ethanol (500 μL), and slowly dropped onto an FTO conductive glass ($A = 1 \text{ cm}^2$). The initial pH of the electrolyte was precisely adjusted to 6.8 via the dropwise addition of 0.1 M NaOH and 0.1 M H_2SO_4 , and monitored by a digital pH meter to ensure stability throughout the

PEC measurement. The working electrode was then dried in a vacuum at 60 °C for 1 h to prepare a working electrode. A silver chloride electrode (Ag/AgCl) and Pt electrode were used as the reference electrode and counter electrode, respectively. The piezoelectric current was measured at a bias voltage of 0.5 V. The ultrasonication instrument (KQ-250DE, Kunshan, China) was purchased from Kunshan with tunable power from 0–250 W. The open circuit voltage of the impedance test is in the frequency range of $10^5\sim 10^{-2}$ Hz, and the AC voltage is 5 mV s^{-1} . The Mott–Schottky diagram was performed at fixed frequencies of 1000 Hz. It calculated the relationship between potential and RHE using Formula (2):

$$E_{\text{vs.RHE}} = E_{\text{vs. Ag/AgCl}} + 0.1976 + 0.0592 \times \text{pH} \quad (2)$$

A dual-cell setup connected via a salt bridge at open-circuit potential using the glassy carbon electrode coating on as-obtained samples was constructed to investigate the electron transfer among CIP, PMS and CoNi-LDH.

3.8. Quantitative Methods for Detecting $\cdot\text{OH}$ and $\cdot\text{SO}_4^-$

$\cdot\text{OH}$ quantification: The terephthalic acid (TA) used and $\cdot\text{OH}$ can react to form a highly fluorescent product (2-hydroxyterephthalic acid, 0.05 mM), which is used to determine the amount of $\cdot\text{OH}$. The wavelength at 315 nm was excited by a fluorescence spectrometer.

$\cdot\text{SO}_4^-$ quantification: Quantitation of p-benzoquinone (p-BQ), which is the major by-product of the reaction between $\cdot\text{SO}_4^-$ and p-hydroxybenzoic acid (p-HBA). Briefly, 20 mg catalyst, 1 mM p-HBA, and 20 mg PMS were prepared in a quartz reactor. After a defined ultrasound time interval, a 2.5 mL sample was taken out from the quartz reactor using a syringe and filtered through a $0.22\text{ }\mu\text{m}$ PTFE filter and quenched with $\text{Na}_2\text{S}_2\text{O}_4$ immediately. Then, the liquid sample was measured by HPLC for the quantitation of p-BQ. The concentration ratio of p-BQ and $\cdot\text{SO}_4^-$ is 1:1, which was measured by HPLC at 244 nm.

3.9. EPR Measurements

The EPR measurements at room temperature (Bruker EMXplus X-band CW EPR spectrometer, Bruker Corporation, MA, USA, microwave frequency: 9.83 GHz; the microwave power: 2.00 mW) were used to determine the type of reactive oxygen species (ROS), in which 5,5-dimethyl-1-pyrroline N-oxide (DMPO) was used as a free radical scavenger in water. The sample was dispersed into deionized water (10 mg sample + 80 mL H_2O) and stirred for different reaction times. Then the DMPO was added in the reaction system under the ultrasonication. A point was taken under dark conditions and tested in EPR equipment.

3.10. Continuous Flow Experiments

The peristaltic pump (Lange BT100-3J + YZ151515X, Baoding Longer Precision Pump Co., Ltd., Baoding, China) provides a flow rate of 10 mL h^{-1} for CIP and PMS solution with the concentration of 10 and 100 ppm, respectively. The liquid was taken every 10 h to measure the concentration of CIP.

3.11. Computational Details

Density functional theory (DFT) calculations on the performance and mechanism of the surface-confinement effect were performed by using the Vienna Ab initio Simulation Program (VASP). The generalized gradient approximation (GGA) in the Perdew–Burke–Ernzerhof (PBE) exchange–correlation potential was used to optimize the geometry and calculate the electronic and optical properties. A $3 \times 3 \times 1$ Monkhorst–Pack grid was used for sampling the Brillouin zones at structure optimization. The ion–electron interactions with a cutoff energy of 400 eV were described by the projector augmented wave (PAW)

method. The convergence criteria of structure optimization were chosen as the maximum force on each atom less than 0.02 eV/Å with an energy change less than 1×10^{-5} eV. The DFT-D3 semiempirical correction was described via Grimme's scheme method.

The adsorption energies (E_{ads}) were calculated as follows:

$$E_{\text{ads}} = E_{\text{ad/sub}} - E_{\text{ad}} - E_{\text{sub}} \quad (3)$$

The VASPKIT software package was utilized for post-processing the data obtained from VASP calculations.

3.12. Calculation of Fukui Functions

The Fukui function is defined as the partial differential of the electron density with respect to the number of electrons at a point in space as the following functions:

$$f(r) = \left(\frac{\partial \mu}{\partial v(r)} \right)_N = \left(\frac{\partial \rho(r)}{\partial N} \right)_{v(r)} \quad (4)$$

in which the r , μ , ρ , and N corresponded to the position vector, chemical potential, electron concentration and electron number.

$$f^+(r) = \lim_{\epsilon \rightarrow 0^+} \frac{\rho_{N+\epsilon}(r) - \rho_N(r)}{\epsilon} \quad (5)$$

$$f^-(r) = \lim_{\epsilon \rightarrow 0^-} \frac{\rho_N(r) - \rho_{N-\epsilon}(r)}{\epsilon} \quad (6)$$

$f^{(2)}(r)$: condensed dual descriptor (CDD):

$$f^{(2)}(r) = f^+(r) + f^-(r) \quad (7)$$

The more negative the value (CDD) is, the more vulnerable the place is to electrophilic attack. The more positive it is, the more vulnerable it is to nucleophilic attack.

4. Conclusions

This study demonstrates an innovative nanoconfinement strategy using a piezoelectric LDH nanoreactor with precisely tuned 5.27 Å interlayer spacing to enhance the reactivity of $\text{SO}_4^{\cdot-}$ for fluoroquinolone defluorination. By achieving a size-matched environment, the system reached a defluorination efficiency of 78.9% and a mineralization rate of 80.7% for ciprofloxacin, with the reaction kinetics (k_{obs}) and defluorination efficiency enhanced by 2.2 and 1.8 times, respectively, compared to non-confined systems. The LDH catalyst exhibited exceptional stability over 100 h and maintained high performance across a wide pH range (4–10) and in complex matrices, including lake and river water (>98% removal). Crucially, the nanoconfinement effect shielded radicals from aqueous scavengers, significantly suppressing toxic by-product formation. These quantitative results underscore the potential of this size-matched LDH platform as a robust and efficient advanced oxidation process (AOP) for the sustainable remediation of persistent fluorinated organic contaminants in diverse water treatment scenarios.

Supplementary Materials: The following supporting information can be downloaded at <https://www.mdpi.com/article/10.3390/catal16040336/s1>. References [40–59] are cited in Supplementary Materials. Figure S1: Representative TEM images of LDHs with different ratio of Co and Ni (a) 4:1 (nanorod-shaped CoNi-LDH in paper), (b) 2:1, (c) 1:1, (d) 1:2 and (e) 1:4 (nanodisk-shaped CoNi-LDH). (f) The schematic diagram of the synthesis of CoNi-LDH with different structures; Figure S2: The XRD patterns of LDHs with different ratio of Co and Ni.

Figure S3: (a) Atomic force microscopy (AFM) height image showing CoNi-LDH nanorod morphology. (b) Three-dimensional AFM rendering of CoNi-LDH nanorods. (c) Piezoelectric response force microscopy (PFM) surface potential map of CoNi-LDH. Figure S4: The butterfly-like amplitude loop and the phase lag loop with 180° polarization curves of nanodisk-shaped CoNi-LDH (1:4). Figure S5: Ultrasonication power dependence of transient piezoelectric current density of CoNi-LDH. Figure S6: (a) Diffuse reflectance spectra of CoNi-LDH-nanorod and CoNi-LDH-nanodisk. (b) Tauc plot estimating the optical bandgap (1.67 eV) of CoNi-LDH. (c) Mott–Schottky plot of CoNi-LDH. Figure S7: (a) CIP degradation efficiency and (b) kinetic profiles to estimate the observed rate constants (k_{obs}) for CoNi-LDHs with the morphologies of nanorod and nanodisk. Figure S8: (a) Ultrasonication power dependence and (b) temperature dependence of CIP degradation efficiency by CoNi-LDH. (c) Comparison of piezoelectric catalytic CIP degradation efficiency of CoNi-LDH driven by ultrasonication versus mechanical stirring. Figure S9: The quantification concentration of $SO_4^{\cdot-}$ in CoNi-LDH with the morphologies of nanorod (a) and nanodisk (b). (c) The standard curve for calculating the concentration of $SO_4^{\cdot-}$. Figure S10: The quantification concentration of $\cdot OH$ in CoNi-LDH with the morphologies of nanorod (a) and nanodisk (b). (c) The standard curve for calculating the concentration of $\cdot OH$. Figure S11: Electrochemical impedance spectroscopy (EIS) of CoNi-LDH with/without US. Figure S12: (a) The CIP molecular with the numbered formular. (b) The values of electrostatic potential surface for the Electrophilicity and Nucleophilicity. (c) The illustrated scheme of the molecular CIP orbital energy. The energies of HOMO and LUMO was calculated by the Multiwfn software. Figure S13: The ESP and dual descriptor values of each atom in $\cdot OH$ and $SO_4^{\cdot-}$. Figure S14: The F^- anion concentration and defluorination efficiency of nanodisk-shaped CoNi-LDH (1:4) in the CIP degradation processes. Figure S15: The intermediate products determined by the m/z values of the CIP degradation pathways. (a) The LC-MS spectra at positive ion mode (a), and negative ion mode (b–d). Figure S16: The size of $SO_4^{\cdot-}$ radicals and CIP (height). The anion layers' distances in CoNi-LDH are a similar size. Figure S17: Comparison of no and similar size confinement for $SO_4^{\cdot-}$. Figure S18: The adsorption energy of different reactive oxygen species of PMS in CoNi-LDH. Insert: PMS molecular and adsorption models. Simulated adsorption modeling of different PMS adsorption sites in CoNi-LDH. Figure S19: The XPS spectra of Co 2p orbitals (a) and Ni orbitals (b) in CoNi-LDH. (c) The XRD patterns of CoNi-LDH before and after reactions. Figure S20: (a) Effect of common inorganic anions (Cl^- , HCO_3^- , NO_3^-) on CIP degradation efficiency in simulated water matrices for nanodisk-shaped CoNi-LDH. (b) The CIP removal efficiency in actual water of nanodisk-shaped CoNi-LDH. Figure S21: (a) The digital picture of CoNi-LDH in-situ grown on the surface of the shredded chayote. (b) The carton picture of CoNi-LDH in-situ grown on the surface of the shredded chayote. Figure S22: The CIP degradation yield of CoNi-LDH on the surface of the shredded chayote at the practical water body containing different anion. Figure S23: The scheme of the continuous flow piezoelectric catalytic-Fenton filter in the degradation device for CIP. Table S1: Compared with the degradation yield of the reported photocatalysts.; Table S2: The numbered atoms in CIP for the electrophilicity and nucleophilicity determined by the values. Table S3: Hishfeld charges and condensed Fukui functions of the CIP.

Author Contributions: Y.Y. synthesized the samples and performed the reactions, and completed material characterization, electrochemical tests, and other characterizations, software and writing—original draft. L.W., H.X., D.L., X.H. and J.Z.: investigation, data curation, writing—reviewing and editing. Z.L. and W.W. conceived and designed the experiments and supervised the study, and completed writing—reviewing and editing. All authors have read and agreed to the published version of the manuscript.

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Data Availability Statement: The data that support the findings of this study are available on request from the corresponding author. The data are not publicly available due to privacy or ethical restrictions.

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