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Rapid Liquid-Phase Plasma Synthesis of Surfactant-Free Colloidal Ni-Fe Nanoparticles for Highly Efficient Oxygen Evolution Reaction

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ABSTRACT

The oxygen evolution reaction (OER) is a fundamental bottleneck in overall water electrolysis due to its sluggish kinetics, necessitating the development of highly active and earth-abundant electrocatalysts. While nickel-iron (Ni-Fe)-based materials are highly promising, conventional wet-chemical syntheses often require hazardous precursors and surfactants that block catalytic active sites. In this study, we demonstrate a facile, rapid, and single-step liquid-phase plasma (LPP) discharge process to synthesize highly pure and surfactant-free colloidal Ni-Fe-based nanoparticles directly in ethyl alcohol. The LPP process utilizes localized Joule heating to overcome the dielectric barrier, enabling ultrafast nucleation of bare nanoparticles. When electrodeposited onto a carbon cloth substrate (Col-NiFe-C.C.), the resulting electrode exhibits exceptional intrinsic OER electrocatalytic performance in 1.0 M KOH. It achieves a benchmark current density of 100 mA cm^{-2} at a low overpotential of 400 mV and a favorable Tafel slope of 42.2 mV dec^{-1} , significantly outperforming commercial IrO_2 catalysts. Furthermore, the Col-NiFe-C.C. catalyst demonstrates robust long-term durability, maintaining stable continuous electrolysis at 10 mA cm^{-2} for 100 h with negligible degradation. This work presents a scalable and environmentally benign process for the design and fabrication of advanced transition metal nanocatalysts for alkaline water electrolysis.

1 | Introduction

The escalating global energy crisis and the urgent need to mitigate climate change have dramatically accelerated the search for sustainable and clean energy architectures [1–3]. Electrochemical water splitting stands out as a highly promising technology for the mass production of green hydrogen, a vital zero-emission energy carrier for the future [4–6]. However, the overall efficiency of water electrolysis is fundamentally bottlenecked by the anodic oxygen evolution reaction (OER) [7–9]. The OER is a kinetically sluggish, thermodynamically uphill four-electron transfer process that requires an excessive overpotential to proceed. While noble-metal-based materials, such as RuO_2 and IrO_2 ,

exhibit state-of-the-art OER performance, their high cost, elemental scarcity, and poor long-term stability severely impede their large-scale industrial deployment [10–13].

Consequently, the development of highly efficient, robust, and earth-abundant electrocatalysts has become a critical issue in energy research. Among various nonprecious alternatives, nickel-iron (Ni-Fe)-based materials have emerged as arguably the most active catalysts for the OER in alkaline media [14, 15]. The strong synergistic electronic interactions between Ni and Fe finely tune the binding energies of oxygenated intermediates (e.g., $^*\text{OH}$, $^*\text{O}$, and $^*\text{OOH}$), thereby significantly lowering the activation energy barrier [16–18]. To maximize this intrinsic

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or extrinsic catalytic activity, several previous researches has been performed based on nanostructure, defect, and interface engineering strategies [19–22]. Further, the Ni-Fe-based electrocatalysts in nanoscale can improve their number of catalytic active sites while exhibiting enhanced intrinsic catalytic activity [23, 24]. In this regard, colloidal Ni-Fe nanoparticles can be a strong potential candidate for effective OER electrolysis as it can offer a dramatically increased surface-to-volume ratio, ensuring an abundance of exposed, highly active edge sites for catalytic turnover [25].

Despite these advantages, the scalable synthesis of pristine, highly active Ni-Fe colloidal nanoparticles remains a persistent challenge. Conventional wet-chemical approaches, such as hydrothermal synthesis, chemical reduction, or thermal decomposition, often involve complex, multistep procedures with prolonged reaction times [26, 27]. More critically, these traditional routes necessitate the use of hazardous chemical precursors, reducing agents, and organic surfactants [28–30]. These capping agents invariably remain adsorbed onto the nanoparticle surface, inadvertently blocking the active catalytic sites required for the OER and thereby suppressing electrochemical performance. Therefore, developing a rapid, one-step, and chemically clean synthetic methodology is highly desirable but challenging.

Liquid-phase plasma (LPP) discharge has recently attracted profound attention as a disruptive, nonequilibrium technique for nanomaterial synthesis [31–34]. Unlike conventional bulk chemistry, submerged electrical discharges generate highly localized, extreme thermodynamic conditions, featuring transient high temperatures, extreme pressures, and intense UV irradiation within microseconds. This environment forces rapid erosion, atomization, and subsequent nucleation of electrode materials, enabling the ultrafast synthesis of bare, surfactant-free nanoparticles directly into a solvent medium. In this study, we demonstrate a novel, facile, and rapid plasma-assisted process for the synthesis of colloidal Ni-Fe alloy nanoparticles directly in ethyl alcohol. Utilizing a custom-designed high-voltage alternating current discharge apparatus, we initiate a unique localized Joule heating process at the tips of submerged Ni-Fe alloy electrodes, overcoming the high dielectric barrier of the liquid to ignite a sustained plasma. This single-step process yields highly pure, surfactant-free Ni-Fe nanoparticles without the need for external chemical reducing agents. We systematically investigate the distinct electro-physical characteristics of the plasma generation and demonstrate that the directly synthesized nanoparticles exhibit exceptional electrocatalytic performance for the OER. This work provides a highly scalable, “green” pathway for the fabrication of transition metal nanocatalysts, offering a new paradigm for advanced energy material synthesis [35].

2 | Experimental Procedure

2.1 | Synthesis of Col-NiFe-C.C.

To synthesize Col-NiFe-C.C., a colloidal solution (45 mL) was used for electrodeposition. The electrodeposition was performed in a conventional three-electrode system consisting of an Ag/AgCl reference electrode, a Pt sheet counter electrode, and carbon cloth as the working electrode substrate. Prior to the deposition process, the carbon cloth was pretreated in 4M

HNO₃ at 353K for 1 h under stirring. The treated carbon cloth was subsequently rinsed thoroughly with deionized (DI) water and used as the working electrode. Electrodeposition was carried out at an applied potential of −0.3 V (vs. reversible hydrogen electrode [RHE]) for 60 min at room temperature. After deposition, the samples were repeatedly washed with DI water and dried in an oven at 333 K overnight.

2.2 | Synthesis of IrO₂-C.C.

A commercial IrO₂ catalyst (Alfa Aesar, Thermo Fisher Scientific) was used as the reference catalyst for electrochemical performance evaluation. The catalyst ink was prepared by dispersing 5 mg of the catalyst in a mixture of 750 μL of DI water, 250 μL of ethyl alcohol (99.9%, Daejung Chemicals & Metals, Republic of Korea), and 20 μL of Nafion solution (5 wt%). The suspension was ultrasonicated for 30 min to obtain homogeneous ink. Subsequently, 17.88 μL of the catalyst ink was drop-cast onto a pretreated carbon cloth substrate. The carbon cloth (0.5 × 0.5 cm²) was used as the working electrode in the three-electrode electrochemical system.

2.3 | Material Characterization

The morphology of the samples was characterized by field-emission scanning electron microscopy (FE-SEM, SU-8010, Hitachi). The crystal structure was determined by X-ray diffraction (XRD, Smartlab, Rigaku) using Cu Kα radiation (40 kV, 100 mA). High-resolution structural analysis of the colloidal particles was conducted using Cs-corrector transmission electron microscope (Cs-TEM, JEM-ARM200F, JEOL). Elemental distributions were examined by energy-dispersive X-ray spectroscopy (EDX) integrated with the Cs-TEM system. Surface chemical states and electronic structures were analyzed by X-ray photoelectron spectroscopy (XPS, K-Alpha+, Thermo Fisher Scientific), and the binding energies were calibrated to the C 1s peak at 284.8 eV.

2.4 | Electrochemical Measurement

The electrochemical performance was carried out using an Autolab PGSTAT potentiostat (Metrohm) in 1 M KOH electrolyte. Prior to the measurements, the electrolyte was purged with Ar gas for 30 min to remove the dissolved gases. A conventional three-electrode configuration was employed, consisting of carbon cloth as the working electrode, Hg/HgO as the reference electrode, and a Pt sheet as the counter electrode. All measured potentials were converted to the RHE scale according to the following equation: $E_{\text{RHE}} = E_{\text{Hg/HgO}} + 0.059 \text{ pH} + 0.098$. Linear sweep voltammetry (LSV) was performed at a scan rate of 5 mV s^{−1}. Electrochemical impedance spectroscopy (EIS) measurements were carried out over a frequency range from 0.1 to 100 kHz with an AC amplitude of 5 mV.

3 | Results and Discussion

Figure 1a shows the schematic of the plasma generation apparatus in ethyl alcohol using two circular rods facing each other. A circular rod is made of Ni-Fe alloy (Ni52/Fe48, GoodFellow, UK), the diameter of which is 2 mm. The ethyl alcohol (Duksan Pure Chemicals, Republic of Korea) has a minimum assay of 99.9%

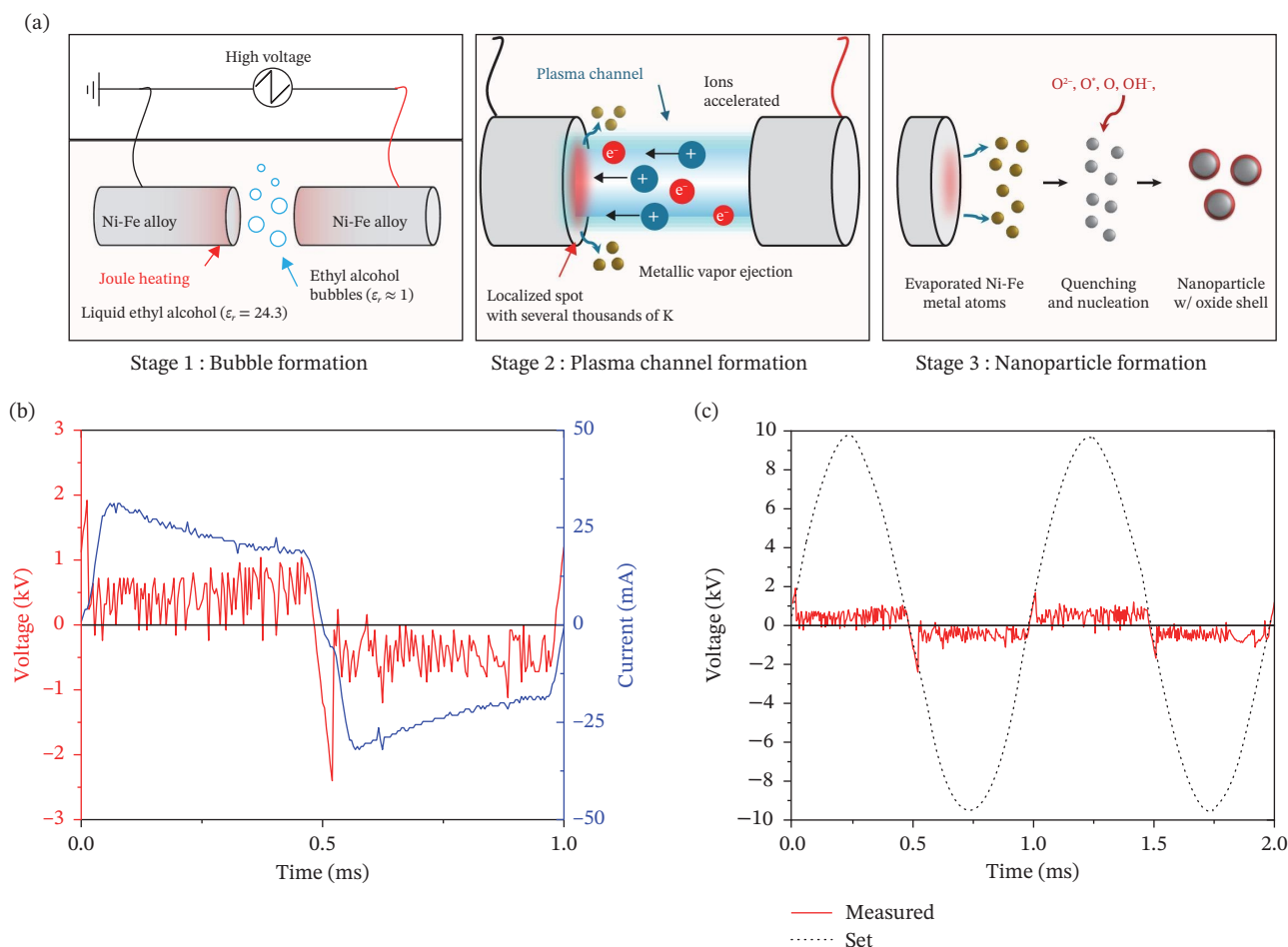


FIGURE 1 | (a) Schematic illustration for plasma-based NiFe colloidal particle synthesis. (b) Voltage and current profile. (c) Set and measured voltage profile during plasma process.

and contains a crystallizing dish of 100 mL. The two rods were fully submerged in the ethyl alcohol. The gap between the two rods was 0.5 mm. One rod was connected to a high-voltage power amplifier (10/10B-HS, Trek, USA) supplying a 1 kHz sinusoidal wave with an amplitude of 9.75 kV, while the other was grounded. Figure 1b also shows the voltage applied between two rods and the current flowing through plasma. The output voltage and current were monitored using the internal sensors of the amplifier. The measured voltage was significantly lower than the set point (Figure 1c). When the plasma was ignited, the impedance between the rods dropped sharply, leading to a high current demand that activates the protective circuit within the amplifier. This resulted in limiting the output voltage. Conversely, when the plasma is momentarily extinguished, the impedance increases, allowing the amplifier to recover and provide the high voltage necessary for subsequent plasma ignition. Such a process occurs at irregular intervals. While the overall current changes monotonically, irregular small peaks are also observed, corresponding to individual plasma discharge events.

The electrical breakdown in liquid ethyl alcohol is initiated with vaporized bubbles created by high voltage [36]. Due to the high dielectric constant of ethyl alcohol (24.3), an electrical breakdown is hard to occur within the liquid phase. Accordingly, the initial energy is dissipated through Joule heating at the opposing tips of the rods, where they face each other. This localized

thermal energy causes the liquid to reach its boiling point, forming ethyl alcohol bubbles within the narrow gap before the actual breakdown occurs. These bubbles have a much lower dielectric permittivity than the surrounding liquid and act as a gaseous medium where electrical breakdown can easily start. After electrical breakdown, a conductive plasma channel was formed between the two metal rods.

Positive ions in the plasma channel accelerated toward the cathode and collided with the surface of the metal rod. These ions instantly created localized high-temperature spots. At this moment, the gas temperature in plasma reaches up to approximately several thousands of K instantaneously [36]. This can cause rapid thermal evaporation of the Ni-Fe alloy, ejecting metallic vapor into the liquid. Consequently, these vapors undergo rapid quenching and nucleation, forming the final Ni-Fe nanoparticles [37]. The nanoparticle generation process was carried out for 20 min. It is expected that the oxidation occurs by interacting with these metallic vapors and the species containing oxygen generated from the dissociation of ethyl alcohol molecules within the plasma channel. To evaluate the energy efficiency of the LPP process, power consumption was determined by integrating time-resolved voltage and current measurements over the reaction duration, yielding an average of 11.46 W. This low power demand, coupled with rapid synthesis and the absence of secondary reducing agents or high-temperature treatments,

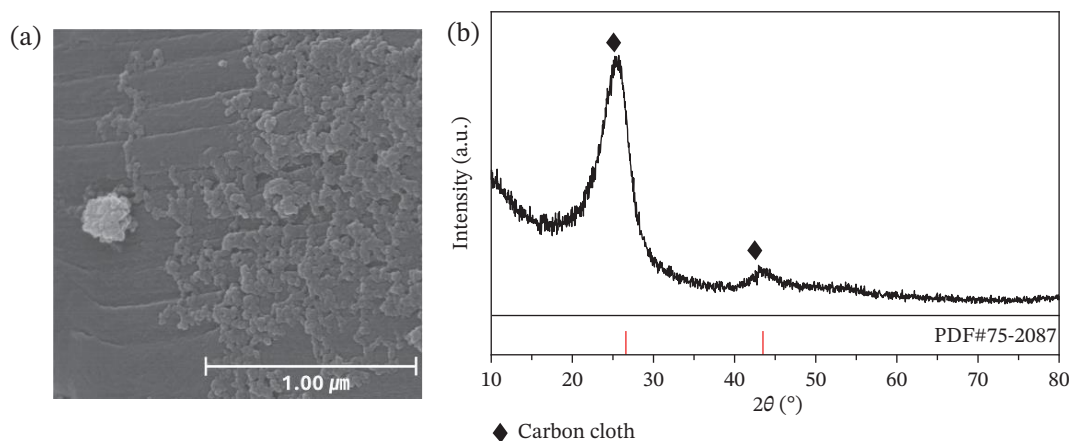


FIGURE 2 | (a) FESEM image of colloidal particles electrodeposited on carbon cloth. (b) XRD pattern of the colloidal catalyst deposited on carbon cloth.

underscores the operational simplicity and environmental sustainability of the LPP method.

The plasma-driven nanoparticle synthesis was conducted for 20 min, yielding a colloidal suspension that maintained stable dispersion for several months. To prepare the electrode for direct application in water electrolysis, a three-electrode electrochemical deposition system was utilized to immobilize these colloidal nanoparticles onto a porous carbon cloth substrate. Scanning electron microscopy (SEM) reveals the random distribution of spherical nanoparticles across the carbon cloth surface (Figure 2a).

XRD was employed to assess the crystallinity of the deposited catalyst (Figure 2b). However, beyond the characteristic peaks of the carbon substrate, no distinct catalyst diffraction peaks were observed [38]. This absence is likely attributable to the small amount and nanoscale size of the deposited catalyst, which makes clear diffraction signals difficult to detect by conventional XRD. Consequently, transmission electron

microscopy (TEM) was utilized to further elucidate the structural morphology. TEM analysis confirmed the presence of spherical nanoparticles with diameters ranging from 10 to 100 nm (Figure 3a). A quantitative particle size analysis based on TEM images showed an average particle size of 50.88 ± 27.20 nm, with most particles distributed below 100 nm (Figure S1). Subsequent high-resolution TEM (HR-TEM) coupled with fast Fourier transform (FFT) analysis determined a lattice fringe spacing of 0.244 nm (Figure 3b). This spacing corresponds to the (111) plane of NiO, supporting the formation of a Ni-Fe oxide phase [39]. The observed diffraction features are assigned with reference to NiO-related planes (the inset of Figure 3b), such as (111), (220), and (311), based on the measured d-spacing values. This indicates the formation of Fe-incorporated NiO-like oxide lattice in the Col-NiFe-C.C. Thus, the indexed SAED features support the presence of crystalline Ni-Fe oxide domains. Furthermore, scanning TEM-energy-EDX (STEM-EDX) elemental mapping demonstrated a homogeneous spatial distribution of Ni, Fe, and O

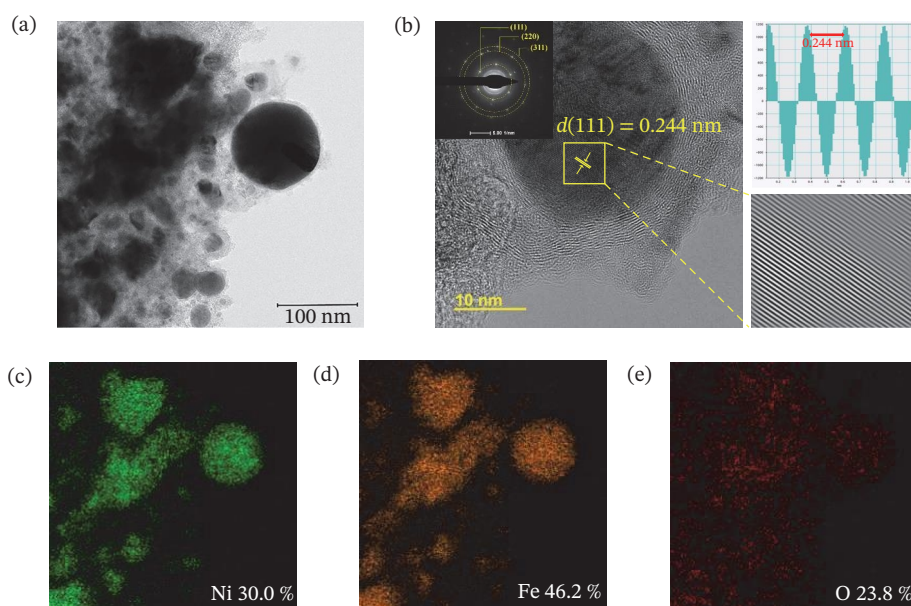


FIGURE 3 | (a) TEM image of the Col-NiFe. (b) HRTEM-derived d-spacing analysis of Col-NiFe nanoparticles, with the SAED pattern shown in the inset. (c-e) STEM-EDX elemental mapping images of Ni, Fe, and O in Col-NiFe.

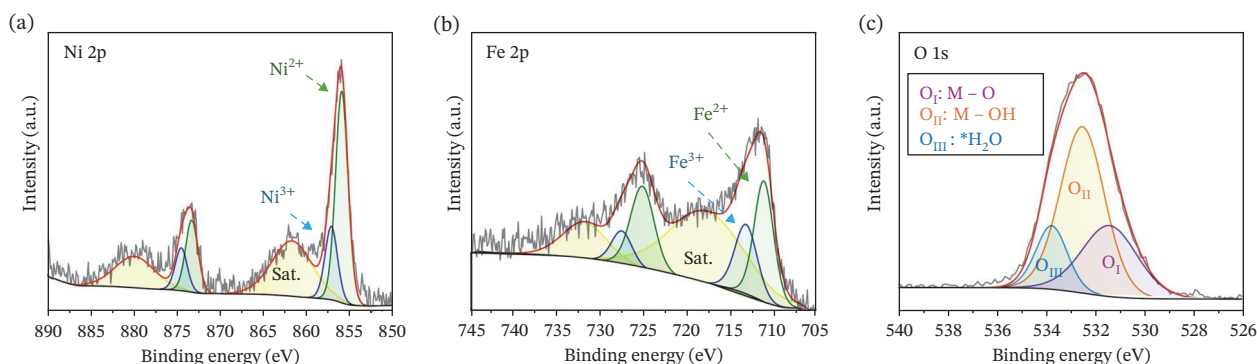


FIGURE 4 | (a) High-resolution XPS spectra of the (a) Ni 2p, (b) Fe 2p, and (c) O 1s present in Col-NiFe on carbon cloth.

throughout the nanoparticles (Figure 3c–e). Quantitative analysis revealed an atomic ratio of ~0.30:0.46:0.24 for Ni, Fe, and O, respectively, indicating thorough elemental integration. Hereafter, this prepared sample is denoted as Col-NiFe-C.C, unless otherwise specified.

Further, electronic valence states of Col-NiFe-C.C were investigated using XPS. The high-resolution Ni 2p XPS spectrum of Col-NiFe-C.C displayed characteristic Ni²⁺ and Ni³⁺ components (855.98 and 857.18 eV, respectively), along with a satellite peak in the Ni 2p_{3/2} region (861.78 eV), while similar features were

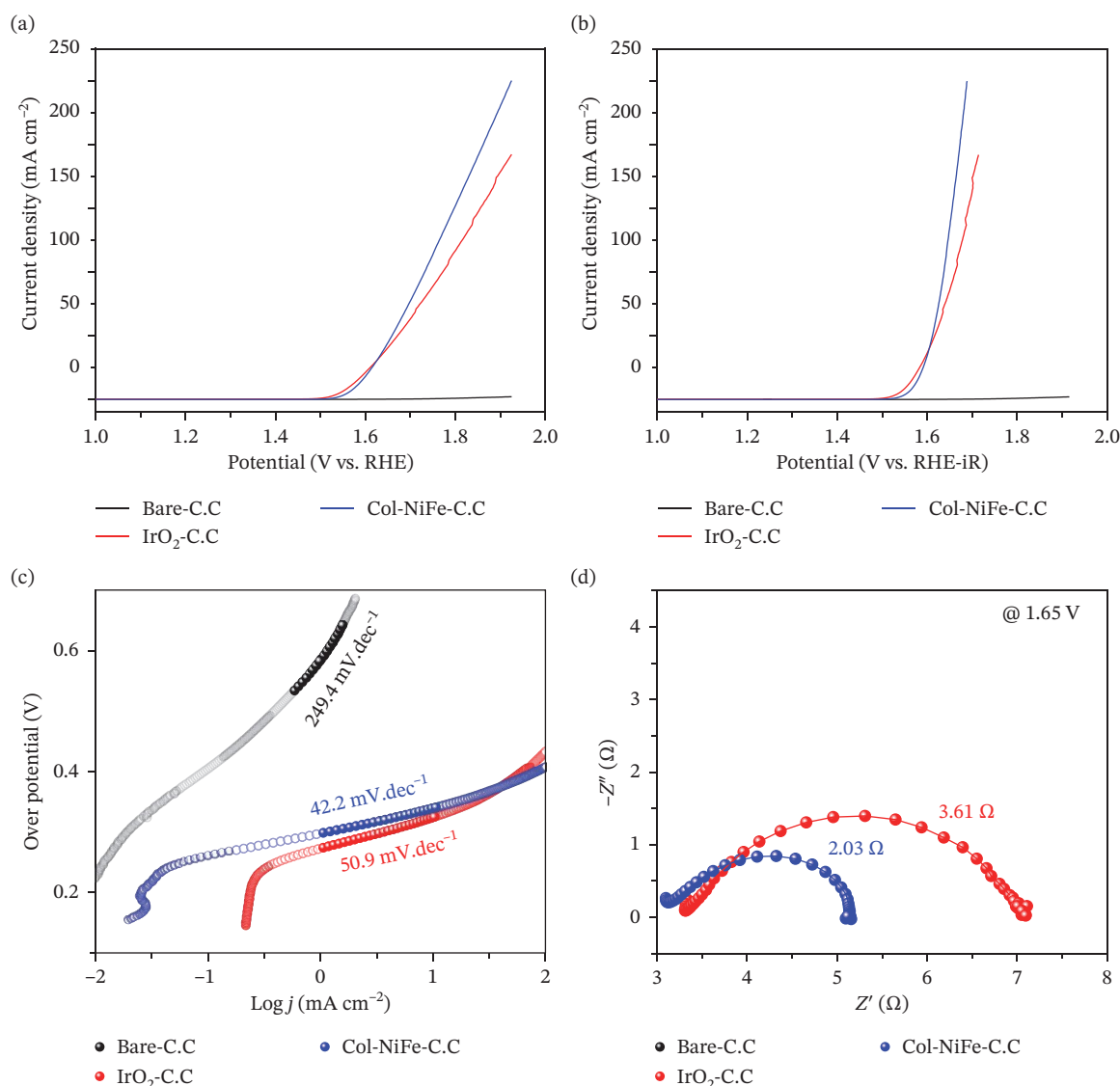


FIGURE 5 | (a) Polarization curves of the tested catalysts. (b) iR-corrected polarization curves. (c) Tafel plots derived from the polarization curves. (d) Nyquist plots of Bare-C.C., IrO₂-C.C., and Col-NiFe-C.C. measured at 1.65 V vs. RHE.

also observed in the Ni 2p_{1/2} region (Figure 4a) [40–42]. The Fe 2p spectrum (Figure 4b) showed characteristic peaks corresponding to Fe²⁺ and Fe³⁺ species (711.18 and 713.18 eV, respectively) [40, 43], suggesting the coexistence of multiple Fe oxidation states within the catalyst. Such mixed-valence Fe species are known to modulate the electronic structure of neighboring Ni sites, thereby facilitating the OER process. Furthermore, the O 1s spectrum (Figure 4c) can be deconvoluted into several components corresponding to M–O bonds (531.43 eV), M–OH species (532.58 eV), and adsorbed water species [44]. The presence of hydroxyl-related oxygen species suggests the formation of surface oxyhydroxide phases, which are widely recognized as active structures for alkaline OER.

The OER performance was systematically evaluated using a standard three-electrode configuration in a 1.0 M KOH electrolyte. For comparison, bare carbon cloth (Bare-C.C.) and commercial IrO₂ supported on carbon cloth (IrO₂-C.C.) were also evaluated. LSV profiles before and after iR compensation are presented in Figure 5a,b, respectively. Among the evaluated electrodes, Col-NiFe-C.C. exhibited the highest OER activity, requiring an overpotential of 400 mV to reach a current density of 100 mA cm^{−2}. In comparison, Bare-C.C. showed negligible catalytic activity, while IrO₂-C.C. required a higher overpotential of 433 mV to achieve

the same current density. These findings demonstrate the superior OER electrocatalytic performance of Col-NiFe-C.C. To further benchmark the OER performance of Col-NiFe-C.C., representative Ni- and Fe-based benchmark electrodes, including Ni(OH)₂-C.C., NiFe-LDH-C.C., Fe₂O₃-C.C., and NiO-C.C., were additionally evaluated under identical electrochemical conditions (Figure S2). Col-NiFe-C.C. required 1.570 V to reach 10 mA cm^{−2}, which was lower than those of Ni(OH)₂-C.C. (1.720 V) and NiFe-LDH-C.C. (1.586 V). At 100 mA cm^{−2}, Col-NiFe-C.C. also showed a lower potential of 1.637 V than NiFe-LDH-C.C. (1.735 V). In contrast, Fe₂O₃-C.C. and NiO-C.C. exhibited much lower current responses within the investigated potential range. These results confirm the favorable OER activity of Col-NiFe-C.C. among the investigated Ni- and Fe-based benchmark electrodes.

To elucidate the intrinsic OER kinetics, Tafel plots were derived from the corresponding LSV polarization curves (Figure 5c) [45]. Notably, Col-NiFe-C.C. yielded the lowest Tafel slope of 42.2 mV dec^{−1}, indicative of highly favorable reaction kinetics compared to Bare-C.C. (249.4 mV dec^{−1}) and IrO₂-C.C. (50.9 mV dec^{−1}). A Tafel slope in the range of 40–60 mV dec^{−1} is generally indicative of favorable OER kinetics and can provide mechanistic insight into the kinetically relevant step. Col-NiFe-C.C. and IrO₂-C.C. showed comparable Tafel slopes of 42.2 and 50.9 mV dec^{−1},

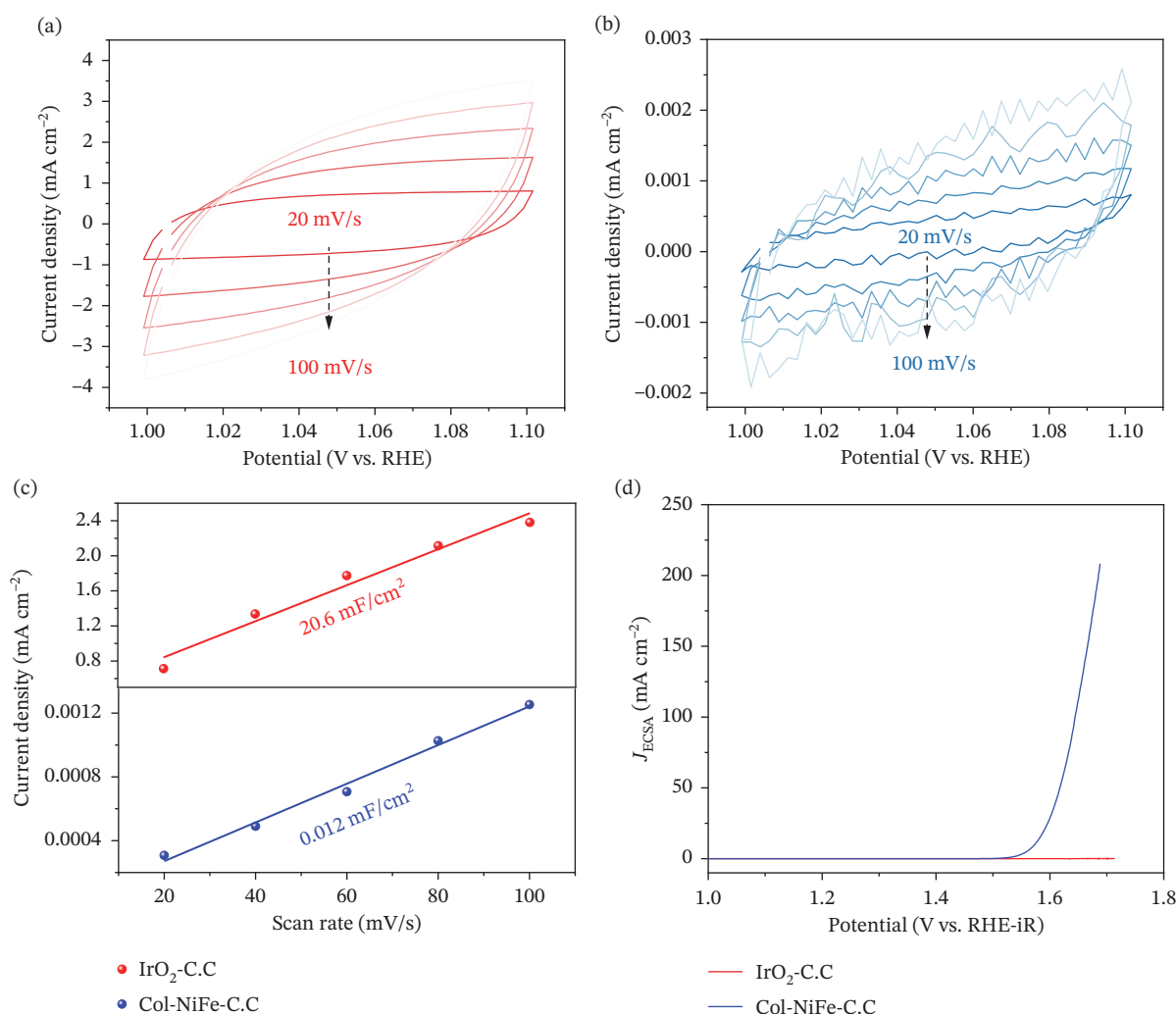


FIGURE 6 | Cycle voltammetry curves recorded at different scan rates (20–100 mV s^{−1}) for C_{dl} estimation of (a) IrO₂-C.C. and (b) Col-NiFe-C.C. (c) Determination of the double-layer capacitance (C_{dl}) at 1.05V (V vs. RHE). (d) ECSA-normalized polarization curves.

respectively, suggesting that both catalysts exhibit similar OER kinetic behavior in the low Tafel-slope range [46]. In addition, pH-dependent OER analysis showed positive kinetic slopes of 0.73 for Col-NiFe-C.C. and 0.63 for IrO₂-C.C., indicating that the OER rates of both catalysts depend on OH⁻ activity (Figure S3). In the adsorbate evolution mechanism, this combined Tafel and pH-dependent behavior is consistent with an OH⁻-coupled surface-intermediate transformation, such as the formation of M-OOH from M=O (M=O + OH⁻ → M-OOH + e⁻, M: active site) [47, 48]. Therefore, both catalysts likely follow a similar OH⁻-involved OER pathway, while the slightly lower Tafel slope of Col-NiFe-C.C. reflects faster apparent OER kinetics than IrO₂, consistent with its enhanced polarization performance.

Furthermore, EIS was employed to probe the interfacial charge-transfer dynamics behavior under OER conditions (Figure 5d). At 1.65 V vs. RHE, Col-NiFe-C.C. exhibited a lower charge-transfer resistance (R_{ct}) of 2.03 Ω than IrO₂-C.C. (3.61 Ω), indicating more favorable charge-transfer kinetics in the higher potential region. In contrast, Bare-C.C. did not display a discernible semicircular feature for reliable R_{ct} extraction. To verify that the anodic current originated from O₂ evolution, O₂ Faradaic efficiency was measured using gas chromatography in an H-type cell under chronopotentiometry operation at 100 mA cm⁻² for 1 h (Figure S4). The measured O₂ amount increased nearly linearly with electrolysis time and closely followed the theoretical O₂ production calculated from Faraday's law. The average O₂ Faradaic efficiency was determined to be 96.6%, confirming that the anodic current of Col-NiFe-C.C. predominantly corresponds to O₂ evolution.

To further investigate the electrocatalytic performance, the electrochemically active surface area (ECSA) was estimated via the electrochemical double-layer capacitance (C_{dl}) [49]. As C_{dl}

correlates directly with the density of accessible active sites, it serves as a critical metric for evaluating intrinsic electrode performance. C_{dl} values were extracted from cyclic voltammetry (CV) curves recorded at varying scan rates in the non-Faradaic region (Figure 6a,b) and are summarized in Figure 6c. Due to its negligible electrochemical response, a reliable C_{dl} could not be ascertained for Bare-C.C. The C_{dl} values were calculated as 20.6 mF cm⁻² for IrO₂-C.C. and 0.012 mF cm⁻² for Col-NiFe-C.C., yielding estimated ECSAs of 514.25 cm² and 0.30 cm², respectively [50]. These metrics suggest a substantially higher density of exposed active sites in the commercial IrO₂-C.C. Consequently, the LSV polarization curves were normalized against the ECSA to evaluate specific intrinsic catalytic activity (Figure 6d). The ECSA-normalized specific current density of Col-NiFe-C.C. greatly outperformed that of the benchmark IrO₂-C.C., unambiguously demonstrating that the plasma-driven colloidal Ni-Fe nanoparticles possess exceptional intrinsic electrocatalytic activity toward the alkaline OER.

The operational stability of Col-NiFe-C.C. was evaluated by chronopotentiometry under high current density OER conditions. At 50 mA cm⁻², Col-NiFe-C.C. showed a gradual potential increase from 1.61 to 1.82 V without an abrupt potential increase, indicating stable OER operation without severe deactivation at elevated current density (Figure 7a). In addition, long-term chronopotentiometry at 10 mA cm⁻² for 100 h showed a negligible overpotential increase of ~40 mV (Figure S5), further confirming its superior durability. Post-OER SEM and EDS elemental mapping further confirmed that Col-NiFe-C.C. well retained its spherical morphology with uniform distributions of Ni, Fe, and O after prolonged electrolysis. No severe aggregation or detachment of nanoparticles from the substrate was observed (Figure S6). To probe potential physicochemical transformations

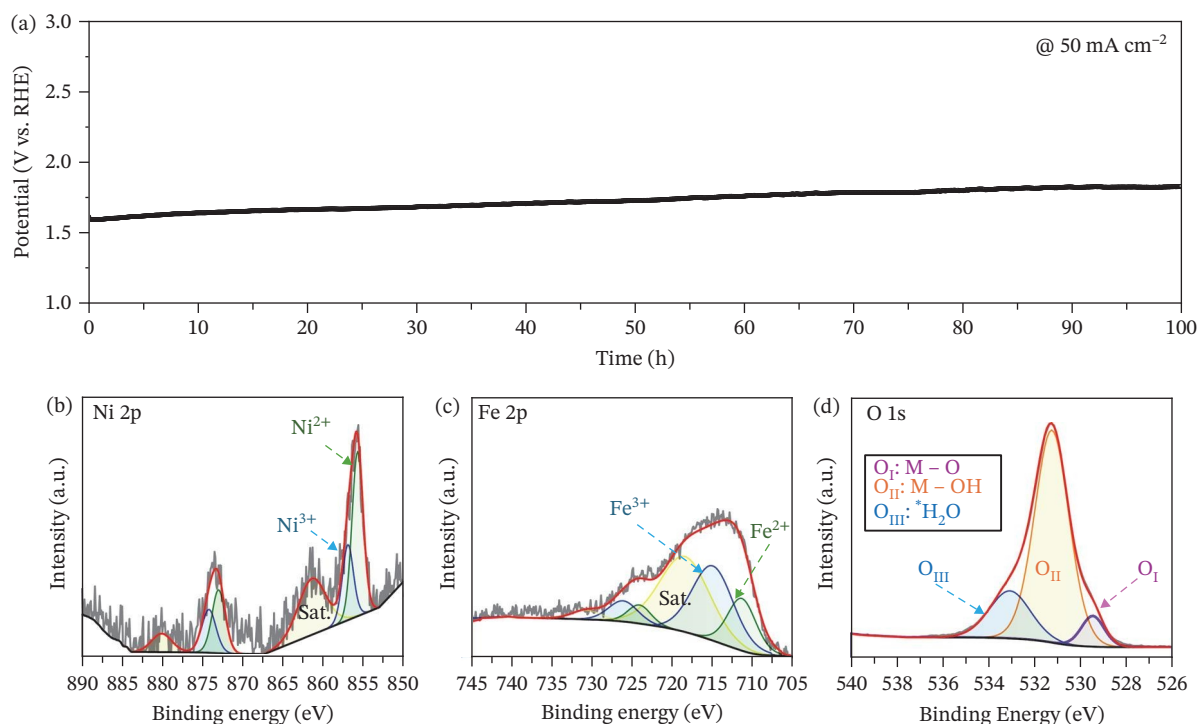


FIGURE 7 | (a) Chronopotentiometry stability test at 50 mA cm⁻² for 100 h. Postreaction high-resolution XPS spectra of (b) Ni 2p, (c) Fe 2p, and (d) O 1s for Col-NiFe-C.C. after the durability test.

during prolonged operation, postreaction XPS was conducted after the OER stability test (Figure 7b–d). Compared to the pristine state (Figure 4), the post-OER Ni 2p spectrum revealed an increased proportion of Ni³⁺ species, indicating the progressive oxidation of nickel components and the formation of NiOOH/NiFeOOH phases during the anodic process. The post-OER Fe 2p spectrum also showed a shift of the Fe-related components toward higher binding energies, suggesting changes in the local coordination and electronic environment of Fe species during OER-induced surface reconstruction. Furthermore, the deconvoluted O 1s spectrum exhibited a significantly enhanced relative intensity for M–OH species postelectrolysis, characteristic of surface reconstruction and hydroxide/oxyhydroxide formation under alkaline conditions [51]. These spectroscopic insights suggest that Col-NiFe-C.C. underwent surface reconstruction into Ni-Fe oxyhydroxide-like surface species. In this reconstructed surface, Ni can act as a redox-active component, while Fe modulates the local Ni-O coordination environment, contributing to favorable OER kinetics and long-term electrochemical stability [47, 52, 53].

4 | Conclusions

In summary, we have successfully developed a novel, rapid, and one-step LPP methodology for the synthesis of pristine and surfactant-free colloidal Ni-Fe alloy nanoparticles directly in an ethyl alcohol medium. By passing the limitations of conventional wet-chemical routes, this nonequilibrium plasma approach yields highly active catalyst surfaces devoid of capping agents. Structural and chemical analyses confirm the formation of ultrafine microstructures with mixed-valence Ni-Fe configurations that facilitate strong synergistic electronic interactions. Electrochemical evaluations demonstrate that the LPP-synthesized nanoparticles, when immobilized on carbon cloth (Col-NiFe-C.C.), exhibit remarkable OER activity in alkaline media, requiring a minimal overpotential of 400 mV to reach 100 mA cm^{−2} and achieving a low Tafel slope of 42.2 mV dec^{−1}. Additionally, the catalyst exhibits outstanding operational stability, enduring 100 h of continuous chronopotentiometry testing without significant performance loss. Postelectrolysis characterizations highlight the structural integrity and critical role of highly oxidized Ni redox species in driving the sustainable catalytic cycle. Ultimately, this study provides a highly scalable and green synthetic pathway for next-generation transition metal electrocatalysts, advancing the scalable implementation of efficient water-splitting technologies.

Author Contributions

JaeYeong Kim: investigation, formal analysis, data curation, validation, writing – original draft. **OhYung Kwon:** conceptualization, supervision, project administration, investigation, funding acquisition, writing – review and editing. **HyukSu Han:** conceptualization, supervision, project administration, investigation, funding acquisition, writing – review and editing.

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Conflicts of Interest

The authors declare no conflicts of interest.

Data Availability Statement

The data used to support the findings of this study are available from the corresponding author upon request.

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Supporting Information

Additional supporting information can be found online in the Supporting Information section. **Supporting Information.** Supporting information associated with this article includes particle size analysis (Figure S1), polarization curve analysis (Figure S2), pH-dependent OER analysis (Figure S3), Faradaic efficiency analysis (Figure S4), chronopotentiometry analysis (Figure S5), and post-OER SEM image of Co-NiFe-LDH (Figure S6).