

In doped NiCo-LDH as bifunctional electrocatalysts for overall water splitting

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Abstract

The development of efficient, stable, and cost-effective bifunctional electrocatalysts for overall water splitting is crucial for green hydrogen production. Herein, we report a series of indium-doped nickel-cobalt layered double hydroxide (In-NiCo LDH) catalysts synthesized directly on nickel foam via a one-step electrodeposition method. By varying the In doping concentration, we identified the optimal In/(Ni + Co) molar ratio of 1.0% that delivers exceptional bifunctional activity. The optimized In_{1.0}-NiCo LDH electrode exhibits low overpotentials of 215 mV for the oxygen evolution reaction (OER) and 180 mV for the hydrogen evolution reaction (HER) at 100 mA cm⁻², with small Tafel slopes of 71.53 mV dec⁻¹ (OER) and 47.69 mV dec⁻¹ (HER). Structural characterization reveals that In³⁺ ions are incorporated as interstitial defects into the LDH lattice, causing lattice expansion and distorted electronic structure. This unique doping mode creates new active sites that serve as electron-donating sites for HER and electron-withdrawing sites for OER. Furthermore, the In_{1.0}-NiCo LDH demonstrates remarkable long-term stability, maintaining stable operation for 100 hours in both OER and HER tests. When assembled as a two-electrode electrolyzer, it requires only 1.73 V to achieve 100 mA cm⁻² for overall water splitting. This work presents a facile electrodeposition strategy for fabricating high-performance In-doped NiCo-LDH bifunctional catalysts and elucidates the structure-activity relationship governed by atomic doping engineering.

1 Introduction

The escalating global energy demand and environmental concerns associated with fossil fuel consumption have spurred intensive research into sustainable and renewable energy alternatives[1, 2, 3]. Hydrogen (H₂), with its high gravimetric energy density and zero-carbon emission upon utilization, has emerged as a promising energy carrier to facilitate the transition toward a clean energy future[4, 5, 6, 7]. Among various hydrogen production technologies, electrochemical water splitting powered by renewable energy sources stands out as an environmentally benign and scalable approach[8, 9, 10, 11]. The water splitting process comprises two half-reactions: the cathodic hydrogen evolution reaction (HER) and the anodic oxygen evolution reaction (OER)[12, 13, 14]. However, both reactions suffer from sluggish kinetics and require substantial overpotentials to achieve practical current densities, particularly the OER with its complex four-electron transfer process[15]. Therefore, the development of efficient electrocatalysts to accelerate these reactions is paramount for improving the overall energy conversion efficiency[16, 17, 18, 19].

Noble metal-based materials, such as Pt/C for HER and IrO₂/RuO₂ for OER, exhibit excellent catalytic activity but are hindered by their high cost, scarcity, and poor stability, which limits their large-scale commercialization[20, 21]. This has motivated extensive research into transition metal-based alternatives, particularly layered double hydroxides (LDHs), which have garnered significant attention due to their tunable composition, unique two-dimensional layered structure, abundant redox-active sites, and earth-abundant constituents[22]. Among various LDH compositions, nickel-cobalt layered double

hydroxides (NiCo LDHs) have been particularly studied for electrocatalysis owing to the synergistic interaction between Ni and Co species[23]. Nickel sites serve as primary active centers for oxygen evolution, while cobalt sites facilitate electron transfer and enhance overall conductivity[24, 25, 26]. Nevertheless, pristine NiCo LDHs still suffer from moderate conductivity, limited active site exposure, and sluggish reaction kinetics, which restrict their practical application in overall water splitting[27, 28]. To overcome these limitations, various modification strategies have been developed, including nanostructure engineering, defect creation, hybridization with conductive substrates, and heteroatom doping[29, 30, 31, 32]. Among these, cationic doping has proven particularly effective in modulating the local electronic environment, increasing active site density, and enhancing intrinsic catalytic activity. Recent studies have demonstrated that doping with transition metals such as Fe, Mn, Cu, and Zn can significantly improve the OER performance of NiCo LDHs[33, 34, 35, 36]. For instance, Fe-doped NiCo LDH achieves an overpotential of 234 mV at 50 mA cm⁻², substantially lower than the 339 mV required for undoped NiCo LDH[37]. Indium (In), as a post-transition metal with an ionic radius of 80 pm for In³⁺, presents intriguing possibilities for LDH doping. The relatively large ionic radius of In³⁺ compared to Ni²⁺ (69 pm) and Co³⁺ (74.5 pm) suggests that it may preferentially occupy substitutional sites within the LDH lattice, replacing original metal cations. This substitutional doping mode could induce lattice expansion, distort electron density distribution, and create unique active sites with optimized adsorption energies for reaction intermediates[38, 39]. Furthermore, theoretical calculations indicate that substitutional doping can introduce new energy levels within the band gap, facilitating charge transfer and improving catalytic kinetics[40, 41].

In this study, we report for the first time the one-step electrodeposition synthesis of In-doped NiCo LDH directly on nickel foam (NF) for overall water splitting. By systematically varying the In doping concentration from 0 to 2 at%, we identify the optimal doping level that maximizes bifunctional catalytic activity. The optimized In_{1.0}-NiCo LDH electrode exhibits low overpotentials of 215 mV for OER and 180 mV for HER at 100 mA cm⁻². The electrocatalytic performance for OER, HER, and overall water splitting is evaluated in alkaline medium. It requires only 1.73 V to achieve 100 mA cm⁻² and exhibits excellent stability over 100 h. This work aims to establish a facile electrodeposition route for In-doped NiCo LDH and elucidate the role of substitutional defect in enhancing bifunctional water splitting performance.

2 Experimental Section

2.1 Materials

Nickel nitrate hexahydrate (Ni(NO₃)₂·6H₂O, 99.9%), cobalt nitrate hexahydrate (Co(NO₃)₂·6H₂O, 99.9%), indium nitrate hydrate (In(NO₃)₃·4H₂O, 99.9%), potassium hydroxide (KOH, 99.5%), hydrochloric acid (HCl, 37%) and ethanol (C₂H₅OH, 99.5%) were purchased from Aladdin Chemical Reagent Co., Ltd.. All chemicals were used as received without further purification. Nickel foam (NF, 1.6 mm) was purchased from Suzhou sinero technology Co., Ltd.. Deionized water (DI water, 18.2 MΩ·cm) was used throughout the experiments.

2.2 Synthesis of In-Doped NiCo LDH

The In-doped NiCo LDH catalysts were synthesized via a one-step cathodic electrodeposition method using a three-electrode configuration on an electrochemical workstation (CHI 660E). The pretreated NF (The NF was sequentially ultrasonicated in acetone, ethanol, and 1 M HCl to remove organic contaminants and surface oxide, then thoroughly rinsed with DI water, vacuum-dried at 60°C for 2 h, and stored in a desiccator.) served as the working electrode, a platinum plate (1 cm × 1 cm) served as the counter electrode, and Ag/AgCl served as the reference electrode. The electrodeposition bath was an aqueous solution containing 10 mM $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ and 10 mM $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ as a supporting electrolyte. To prepare In-doped samples, varying amounts of $\text{In}(\text{NO}_3)_3 \cdot 4\text{H}_2\text{O}$ were added to achieve In/(Ni + Co) molar ratios of 0% (undoped), 0.5%, 1.0%, 1.5%, and 2.0%. Electrodeposition was performed at a constant potential of -1.0 V vs. RHE for 300 s at room temperature without stirring. After deposition, the electrodes were immediately removed, gently rinsed with DI water to remove loosely adsorbed ions, and dried under vacuum at 40 °C for 2 h. The as-prepared samples are denoted as NiCo LDH (undoped), $\text{In}_{0.5}$ -NiCo LDH, $\text{In}_{1.0}$ -NiCo LDH, $\text{In}_{2.0}$ -NiCo LDH, and $\text{In}_{5.0}$ -NiCo LDH.

2.3 Material Characterization

The phase analysis of the catalysts was conducted using the D8 Advance model from Bruker. The morphology characterization of the materials was performed using Hitachi's S-4800 Field mission scanning electron microscopy. The instrument used for high-resolution mapping is FEI-TALOS-F200X. XPS data was obtained through Thermo Kalpha.

2.4 Electrochemical Measurements

All electrochemical measurements were performed on a CHI 660E electrochemical workstation at room temperature using a three-electrode configuration in 1.0 M KOH electrolyte. The as-prepared In-NiCo LDH/NF electrodes were used as the working electrode. A graphite rod and an Ag/AgCl electrode served as the counter electrode and reference electrode, respectively. All measured potentials were converted to the RHE scale using the equation: $E(\text{RHE}) = E(\text{Ag}/\text{AgCl}) + 0.196 \text{ V} + 0.059 \times \text{pH}$. Prior to electrochemical testing, the working electrode was subjected to cyclic voltammetry (CV) cycling in the potential range of 0-0.6 V vs Ag/AgCl at 50 mV s^{-1} for 20 cycles to achieve stable surface states. LSV was performed at a scan rate of 5 mV s^{-1} with 85% iR compensation to evaluate the OER and HER activities. The ECSA was estimated from the electrochemical Cdl by CV measurements in the non-Faradaic region at various scan rates (20, 40, 60, 80, 100 mV s^{-1}). EIS was conducted at open circuit potential with an AC amplitude of 5 mV over the frequency range from 100 kHz to 0.01 Hz. The long-term stability of the optimized $\text{In}_{1.0}$ -NiCo LDH electrode was evaluated by chronopotentiometry at constant current densities of 100 mA cm^{-2} for both OER and HER over 100 h. For overall water splitting, a two-electrode configuration was assembled using $\text{In}_{1.0}$ -NiCo LDH/NF as both the anode and cathode. LSV curves were recorded at a scan rate of 5 mV s^{-1} , and long-term stability was assessed at a constant cell current corresponding to 100 mA cm^{-2} .

3 Result and discussion

Scheme 1 Schematic of the synthetic method of In-NiCo LDH/NF

Figure 1 presents the XRD patterns of undoped NiCo LDH and In-doped samples with varying In concentrations. All samples exhibit characteristic diffraction peaks corresponding to NiCo LDH structure (JCPDS No. 40–0216). Due to the high diffraction peak intensity of foam nickel, the catalyst peak is relatively weak, but the diffraction peak of NiCo LDH can still be clearly identified. The peaks at approximately 11.5° and 23.2° are indexed to the (003) and (006) planes of the NiCo LDH phase, respectively. No additional peaks corresponding to $\text{In}(\text{OH})_3$ or other impurity phases are detected in the In-doped samples, indicating that In^{3+} is successfully incorporated into the LDH structure without forming separate phases.

The surface morphology of the synthesized electrodes was examined by SEM. Figure S1 shows the SEM image of undoped NiCo LDH, which exhibits a typical interconnected nanosheet array morphology aligned on the nickel foam substrate. These nanosheets are relatively uniform with thicknesses of approximately 20–30 nm and interwoven together. A larger specific surface area is beneficial for mass transfer and gas bubble release during electrocatalysis. Figure 2 shows the morphology of NiCo LDH doped with different ratios of In. It can be clearly seen that with the increase of In doping, the formation of nanosheet structures becomes more difficult, and instead transitions from ordered nanosheets to irregularly shaped particle stacking. When further increasing the In concentration to 1.5% and 2.0% leads to partial aggregation and thickening of nanosheets corresponding to excess In species. This morphological deterioration at high doping levels likely results from excessive atomic doping.

TEM analysis provides further insights into the microstructure of $\text{In}_{1.0}\text{-NiCo LDH}$. Figure 3a shows a TEM image of $\text{In}_{1.0}\text{-NiCo LDH}$ nanosheets exfoliated from the substrate, confirming that the nanosheets are very thin. The HRTEM image (Fig. 3b) reveals well-resolved lattice fringes with an interplanar spacing of 0.200 nm, corresponding to the (110) plane of LDH. The energy-dispersive X-ray spectroscopy (EDS) mapping of $\text{In}_{1.0}\text{-NiCo LDH}$ (Fig. 2c-g) confirms the uniform distribution of Ni, Co, and In elements throughout the nanosheet structure, indicating homogeneous In doping without elemental segregation.

XPS analysis was conducted to investigate the surface chemical composition and electronic states of the $\text{In}_{1.0}\text{-NiCo LDH}$ samples. Figure S2 presents the survey spectra, confirming the presence of Ni, Co, O, In, and C elements. Figure 4a displays the high-resolution Ni 2p spectra. The Ni 2p_{3/2} spectrum can be deconvoluted into two peaks at 855.6 eV and 857.3 eV, corresponding to Ni^{2+} and Ni^{3+} species, respectively, accompanied by satellite peaks at 861.5 eV. Similarly, the Co 2p spectra (Fig. 4b) show two main peaks for Co 2p_{3/2} at 780.8 eV (Co^{3+}) and 782.6 eV (Co^{2+}) with satellite features. The In 3d spectrum for $\text{In}_{1.0}\text{-NiCo LDH}$ in Fig. 4c shows two peaks at 444.8 eV and 452.4 eV, corresponding to In 3d_{5/2} and In 3d_{3/2}, respectively. The O 1s spectra (Fig. 4d) can be fitted with three components: lattice oxygen (O^{2-}) in metal-oxygen bonds at 529.8 eV, hydroxyl groups (OH^-) at 531.2 eV, and adsorbed water

molecules at 532.5 eV. The XPS results collectively demonstrate that In^{3+} effectively modulates the electronic structure of NiCo LDH by inducing charge redistribution and increasing the proportion of high-valence metal species. These electronic modifications are expected to optimize the adsorption energies of reaction intermediates and enhance catalytic activity.

Electrocatalytic Performance

The OER activity of the synthesized electrodes was evaluated in 1.0 M KOH using a three-electrode configuration. Figure 5a shows the LSV curves of undoped NiCo LDH and In-doped samples with various In concentrations. The undoped NiCo LDH exhibits an overpotential of 265 mV at 10 mA cm^{-2} and 425 mV at 100 mA cm^{-2} . Upon In doping, the OER performance improves significantly, with the $\text{In}_{1.0}$ -NiCo LDH electrode demonstrating the lowest overpotentials: 125 mV at 10 mA cm^{-2} and 215 mV at 100 mA cm^{-2} . The OER kinetics were further evaluated by Tafel plots derived from the LSV curves (Fig. 5b). The undoped NiCo LDH shows a Tafel slope of $130.94 \text{ mV dec}^{-1}$, indicating relatively sluggish reaction kinetics. In contrast, most In-doped samples exhibit reduced Tafel slopes, with $\text{In}_{1.0}$ -NiCo LDH achieving the lowest value of $71.53 \text{ mV dec}^{-1}$. This low Tafel slope suggests a favorable reaction pathway with more efficient charge transfer kinetics. Figure 5c summarizes the potential values at various current densities for all samples. The trend of overpotential and Tafel slope as a function of In doping concentration is visualized, clearly showing the optimal performance at 1.0% In doping. To evaluate the charge transport kinetics, electrochemical impedance spectroscopy (EIS) was conducted. The Nyquist plots for all samples exhibit a characteristic semicircle in the high-frequency region, with the diameter of this semicircle corresponding to the charge transfer resistance. Among the series, the pristine NiCo LDH displays the larger semicircle, indicating a relatively high charge transfer resistance. While In-doped samples show reduced semicircle diameters compared to the pristine material, a clear trend is observed with varying indium content. Notably, the $\text{In}_{1.0}$ -NiCo LDH sample possesses the smallest semicircle radius, signifying the lowest charge transfer resistance. This result suggests that the incorporation of indium at an optimal ratio of 1.0 effectively modulates the electronic structure and enhances the interfacial charge transport kinetics, outperforming the other doping concentrations and the undoped sample. In Fig. 5e, compared with other catalysts reported in the literature, our as-prepared catalyst exhibits significantly lower overpotential and Tafel slope. Specifically, at a current density of 100 mA cm^{-2} , the overpotential of this catalyst is considerably smaller than that of most reported electrocatalysts, reflecting its superior kinetic capability for catalytic initiation. Meanwhile, the lower Tafel slope indicates that this catalyst can increase the current density more rapidly during the reaction process, responding more sensitively to applied potential changes, and facilitating a more favorable catalytic reaction pathway. Long-term stability is a critical parameter for practical applications. Figure 5f shows the chronopotentiometric response of $\text{In}_{1.0}$ -NiCo LDH at a constant current density of 100 mA cm^{-2} . The potential remains remarkably stable over 100 h of continuous operation, with negligible degradation.

The HER activity of the synthesized electrodes was evaluated in the same 1.0 M KOH electrolyte. Figure 6a displays the LSV curves for HER. The undoped NiCo LDH shows an overpotential of 121 mV at -10 mA cm^{-2} and 284 mV at -100 mA cm^{-2} . Upon In doping, the HER performance improves substantially, following the same trend as OER. The $\text{In}_{1.0}\text{-NiCo}$ LDH electrode achieves the lowest overpotentials: 73 mV at -10 mA cm^{-2} and 180 mV at -100 mA cm^{-2} . The HER kinetics were assessed using Tafel plots derived from the LSV curves. The pristine NiCo LDH exhibits a Tafel slope of $58.58 \text{ mV dec}^{-1}$, reflecting relatively sluggish reaction kinetics. In contrast, the incorporation of indium leads to notably reduced Tafel slopes across doped samples, with $\text{In}_{1.0}\text{-NiCo}$ LDH delivering the smallest value of $47.69 \text{ mV dec}^{-1}$. Such a low Tafel slope indicates a more favorable reaction pathway accompanied by enhanced charge transfer efficiency. The charge transport behavior was further investigated through EIS. In the Nyquist plots, each sample presents a semicircle in the high-frequency region, where the diameter corresponds to the charge transfer resistance. The undoped NiCo LDH shows the larger semicircle, suggesting the highest charge transfer resistance. Although In-doped variants exhibit diminished semicircle diameters relative to the pristine material, the extent of reduction varies with indium content. Impressively, $\text{In}_{1.0}\text{-NiCo}$ LDH displays the smallest semicircle radius, pointing to the lowest charge transfer resistance. This result implies that doping with an optimal indium ratio of 1.0 effectively tunes the electronic structure and facilitates interfacial charge transport, outperforming both the undoped sample and those with other doping concentration. The ECSA was determined by measuring the electrochemical Cdl via CV in the non-Faradaic region. As depicted in Fig. 6e, $\text{In}_{1.0}\text{-NiCo}$ LDH achieves the highest Cdl of 20.28 mF cm^{-2} , approximately 2 times that of undoped NiCo LDH (10.78 mF cm^{-2}). This markedly enlarged ECSA aligns with the morphological features of thinner and more dispersed nanosheets, offering a greater number of accessible active sites for HER. Stability testing for HER (Fig. 6f) demonstrates that $\text{In}_{1.0}\text{-NiCo}$ LDH maintains stable operation at -100 mA cm^{-2} for 100 h with negligible potential increase, confirming its excellent durability under cathodic conditions.

Overall Water Splitting Performance

To evaluate the applicability of the $\text{In}_{1.0}\text{-NiCo}$ LDH catalyst, a two-electrode system was assembled using $\text{In}_{1.0}\text{-NiCo}$ LDH as both the anode and cathode to test the overall electrochemical water splitting performance in 1 M KOH electrolysis cell (Fig. 7a). The linear sweep voltammetry (LSV) curve in Fig. 7b demonstrates that a current density of 100 mA cm^{-2} is achieved at a low cell voltage of 1.73 V, indicating that the system possesses low resistance and facilitates rapid and efficient charge transfer. Figure. 7c presents a comparison of the cell potentials required to deliver various current densities. Notably, the $\text{In}_{1.0}\text{-NiCo}$ LDH catalyst exhibits superior performance, requiring substantially lower potentials than the pristine NiCo LDH and NF counterparts under the same conditions. Furthermore, long-term durability was assessed through chronopotentiometric measurement. As shown in Figure. 7d, the $\text{In}_{1.0}\text{-NiCo}$ LDH catalyst operates stably for 100 h at a constant current density of 100 mA cm^{-2} with negligible potential decay, demonstrating excellent operational stability. The remarkable overall water

splitting activity and robust stability achieved position $\text{In}_{1.0}\text{-NiCo}$ LDH as a promising candidate for practical water electrolysis applications, offering valuable insights for the development of high-performance electrocatalysts.

4 Conclusion

In summary, the bifunctional $\text{In}_{1.0}\text{-NiCo}$ LDH catalyst is successfully synthesized via a one-step electrodeposition method, and this bifunctional catalyst exhibits excellent overall performance in water electrolysis. Experimental results show that the $\text{In}_{1.0}\text{-NiCo}$ LDH heterostructure performs superiorly in alkaline environments. This material possesses an optimized nanosheet array architecture with In^{3+} homogeneously incorporated into the NiCo LDH lattice, effectively modulating the electronic structure and exposing abundant active sites on its surface. This facilitates accelerated catalytic reaction kinetics for HER and OER, exhibiting excellent bifunctional catalytic activity. When used as an OER electrocatalyst, an overpotential of only 215 mV is achieved at a current density of 100 mA cm^{-2} with a Tafel slope of $71.53 \text{ mV dec}^{-1}$. When the HER occurs, a current density of 100 mA cm^{-2} can be achieved with an overpotential of 180 mV, and it can operate stably for 100 h under these conditions. $\text{In}_{1.0}\text{-NiCo}$ LDH was assembled as both the positive and negative electrodes in a two-electrode full decomposition electrolysis cell to test the performance of water splitting. A small voltage of 1.73 V is sufficient to achieve a current density of 100 mA cm^{-2} , and it can operate stably for 100 h under these conditions. This work presents promising application prospects and provides an alternative approach for the development of transition-metal based catalysts for water splitting.

Declarations

Conflicts of interest

The authors declare no competing interests.

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Author Contribution

Zicheng Huang was responsible for experimental design, experimental operations, and data testing, and wrote the main manuscript text. Mengwei Yu and Yuedong Guo participated in experimental operations and data analysis. Yulun Xie served as the corresponding author and was responsible for experimental guidance. All authors conducted data analysis and reviewed the manuscript.

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Scheme

Scheme 1 is available in the Supplementary Files section.

Figures

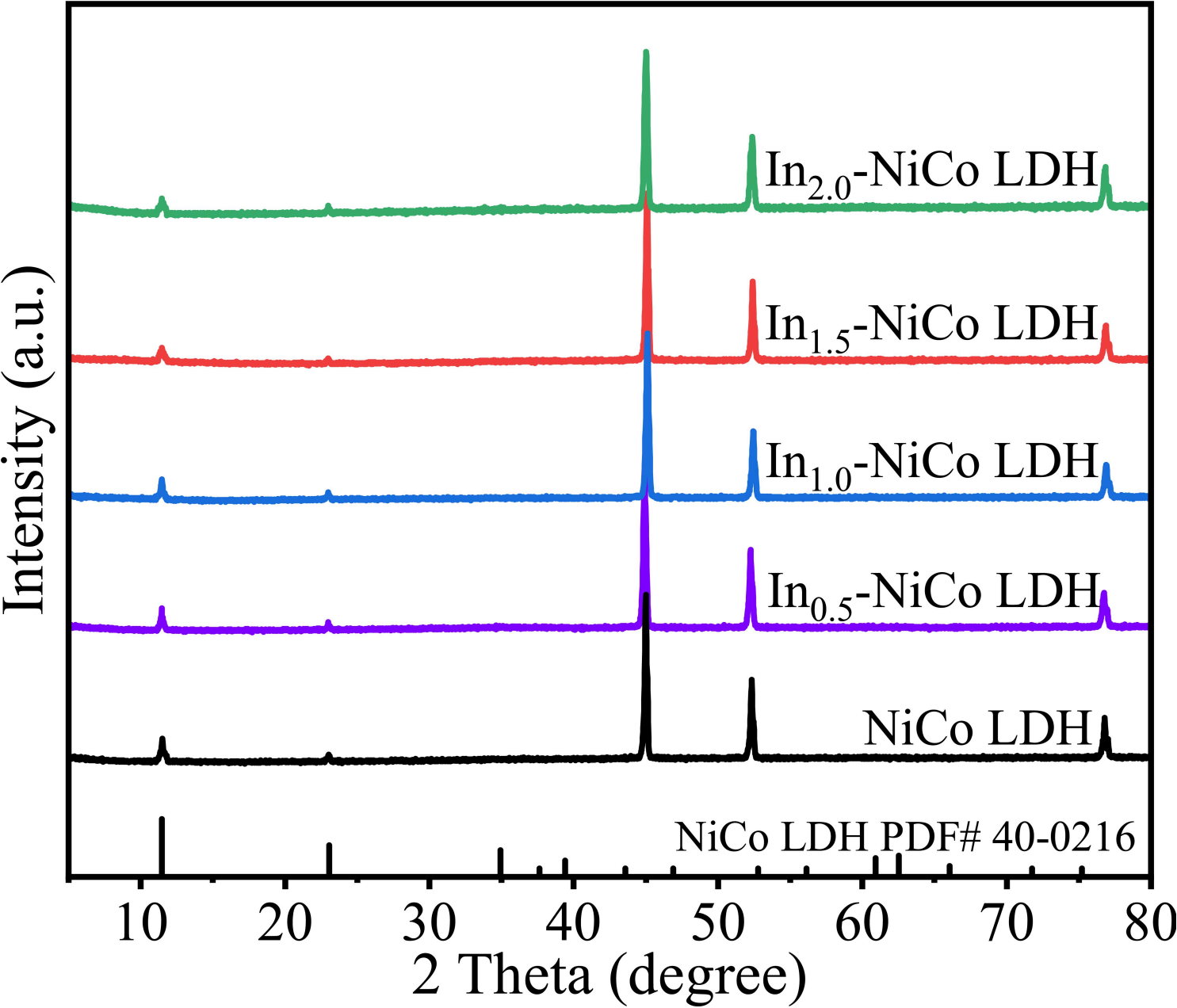


Figure 1

XRD patterns of undoped NiCo LDH and In-doped samples with varying In concentrations

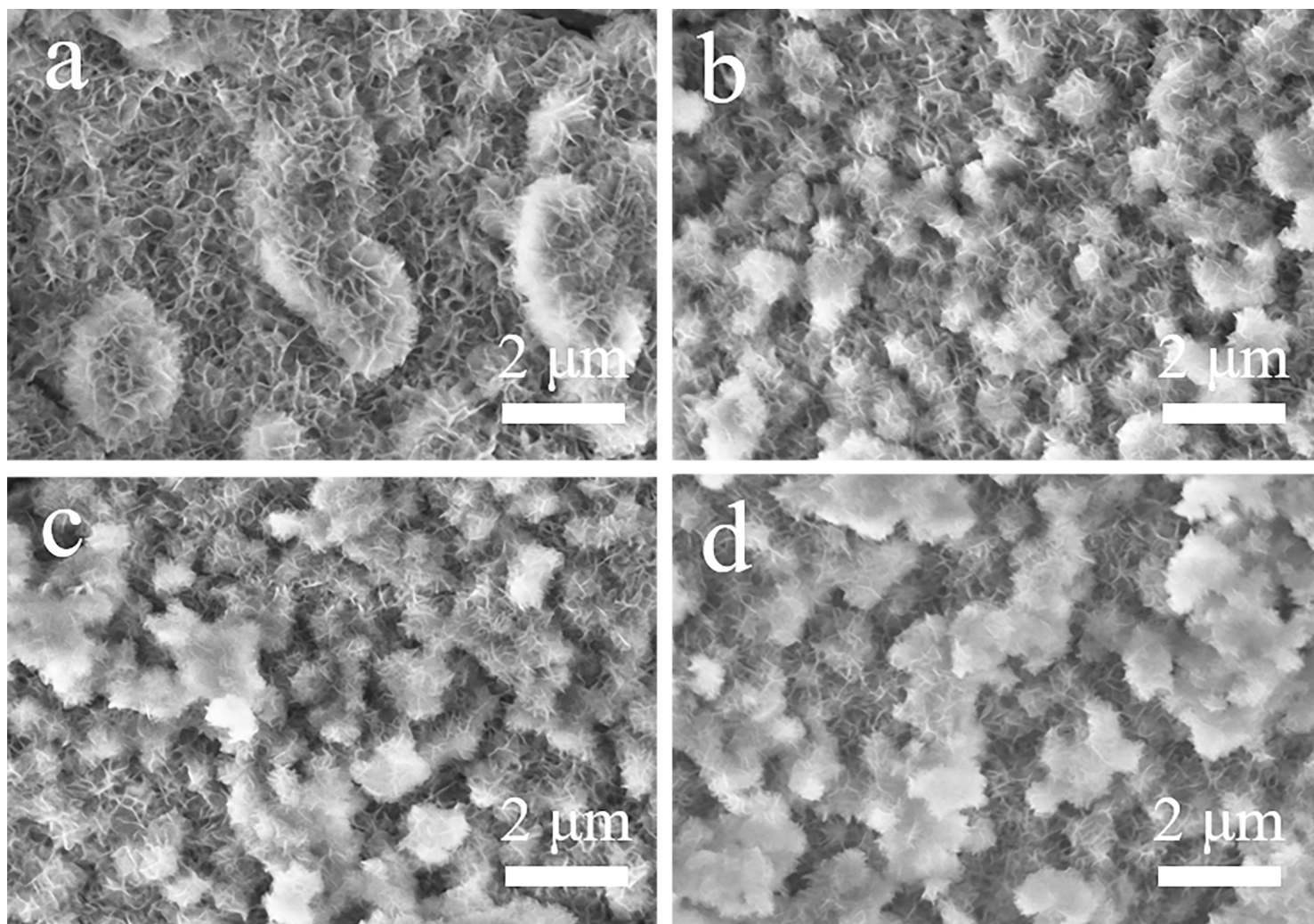


Figure 2

SEM images of (a) $\text{In}_{0.5}\text{-NiCo LDH}$, (b) $\text{In}_{1.0}\text{-NiCo LDH}$, (c) $\text{In}_{1.5}\text{-NiCo LDH}$, (d) $\text{In}_{2.0}\text{-NiCo LDH}$

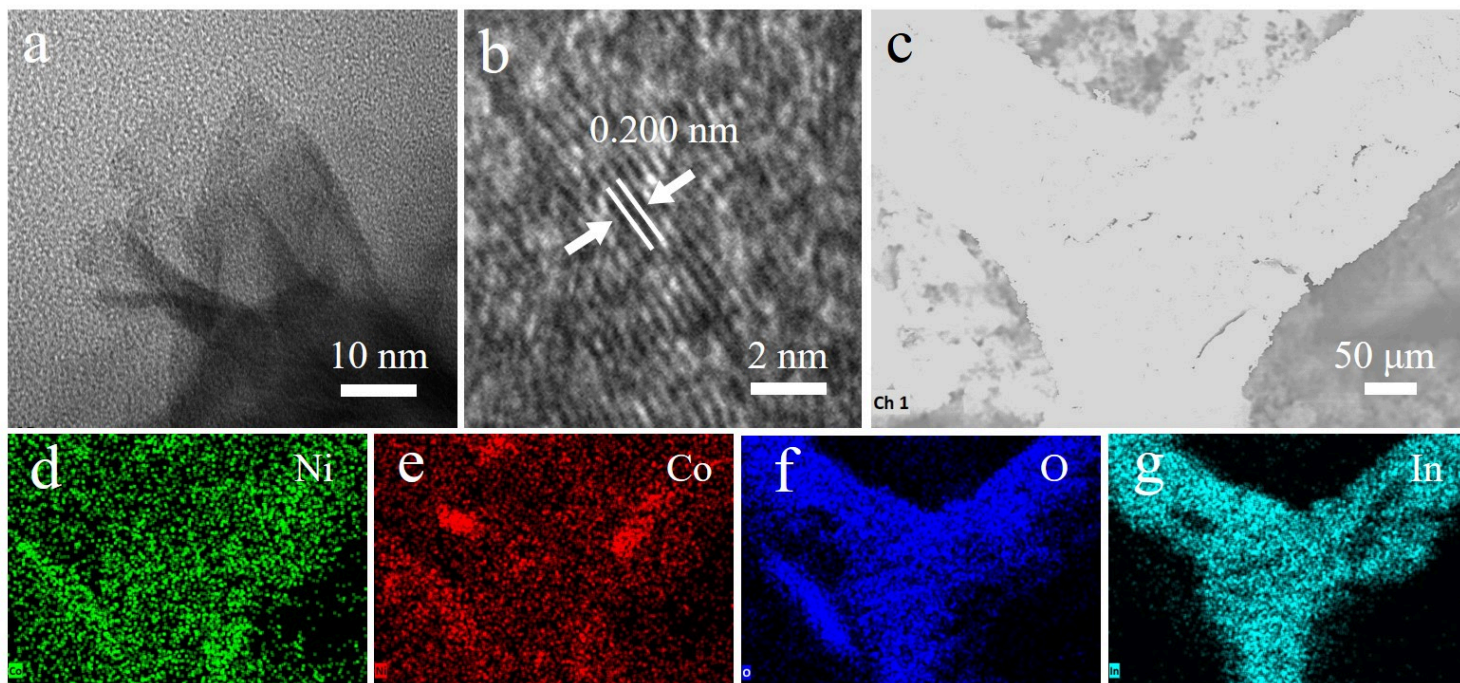


Figure 3

(a) TEM and (b) HRTEM image of $\text{In}_{1.0}\text{-NiCo}$ LDH nanosheets. (c-g) EDS elemental mapping of $\text{In}_{1.0}\text{-NiCo}$ LDH showing uniform distribution of Ni, Co, O and In

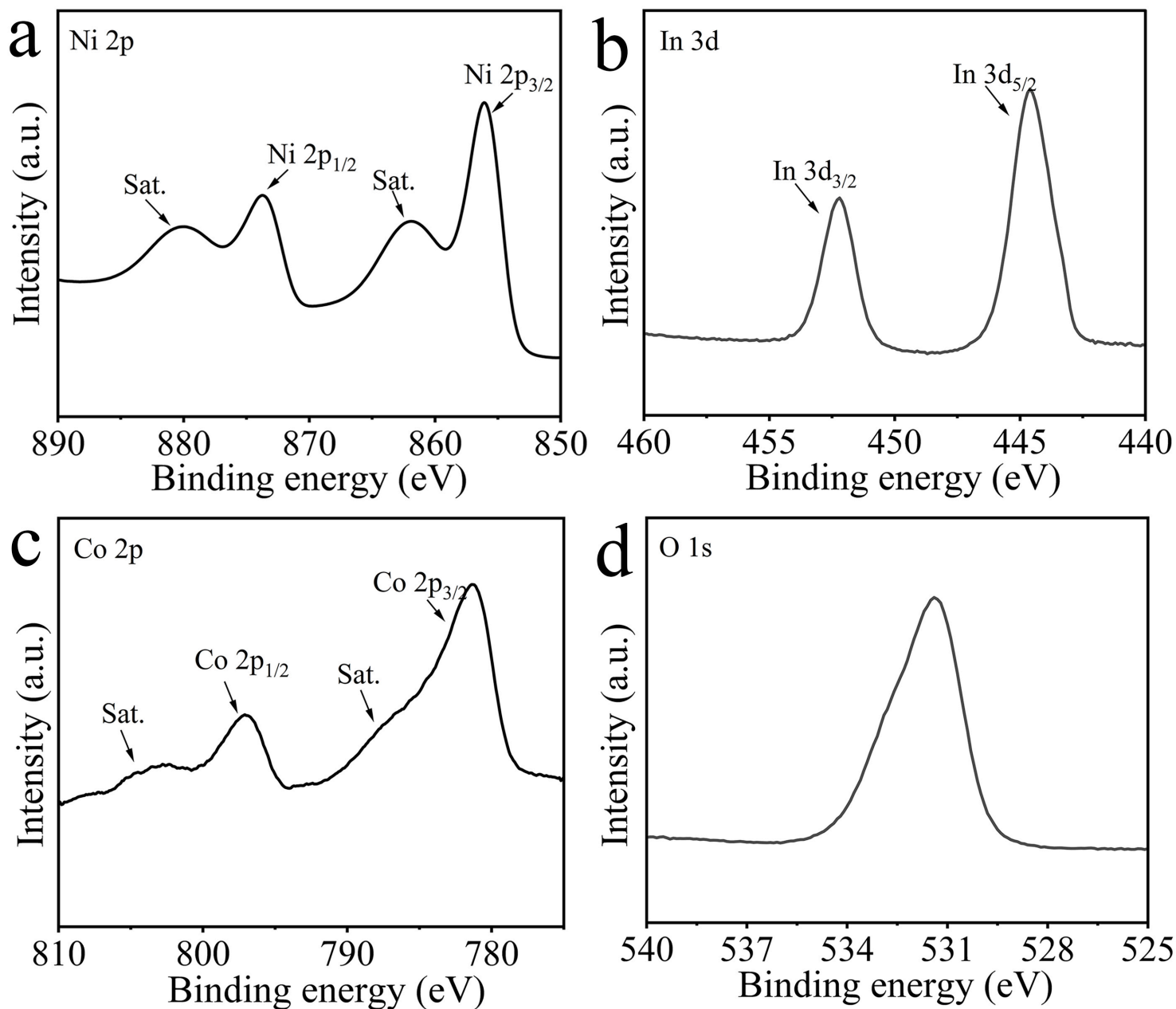


Figure 4

XPS spectra of In_{1.0}-NiCo LDH: (a) Ni 2p spectra, (b) Co 2p spectra, (c) In 3d spectrum, (d) O 1s spectra

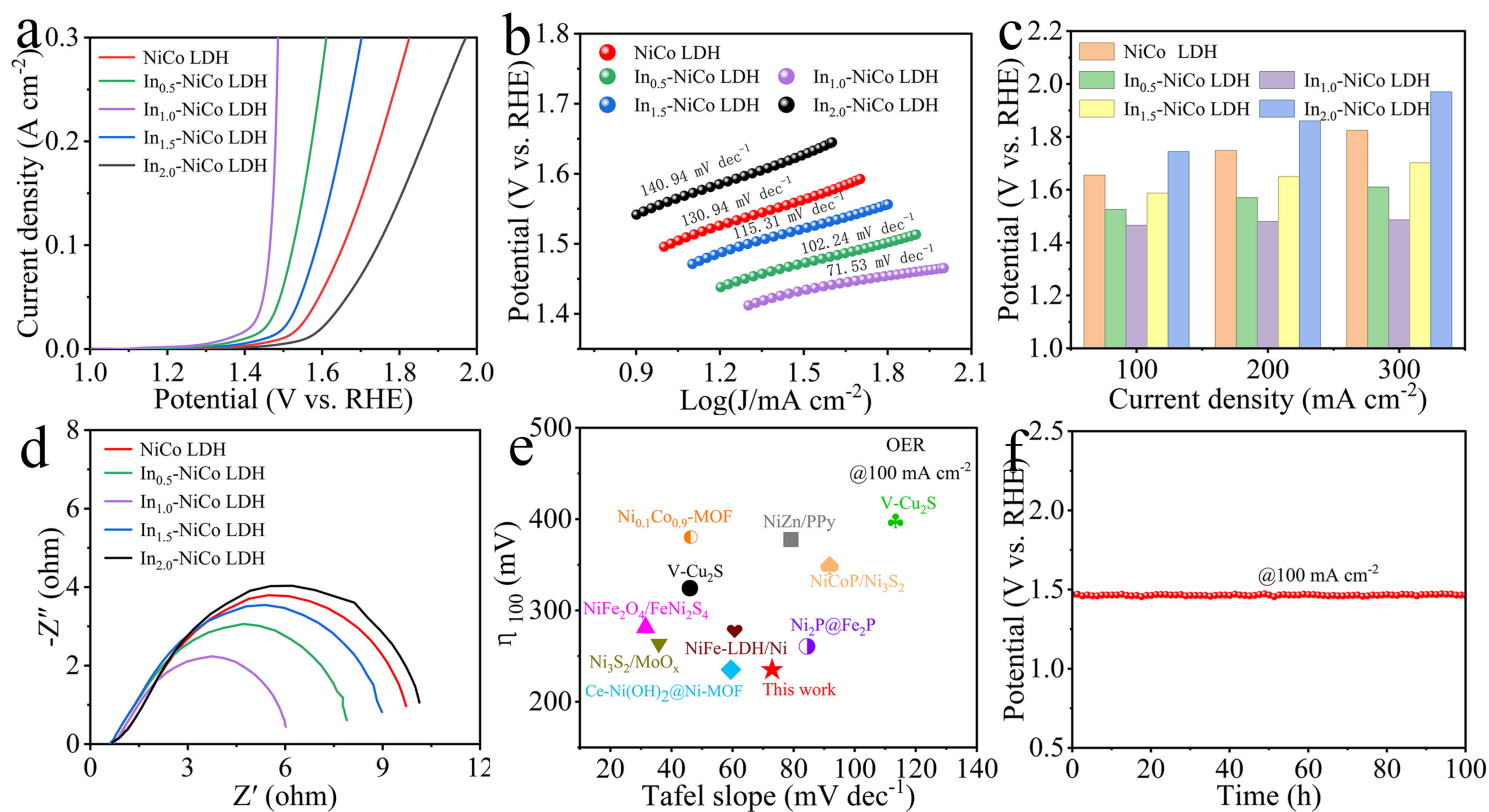


Figure 5

(a) OER LSV curves of undoped and In-doped NiCo LDH, (b) Corresponding Tafel plots, (c) Comparison of overpotential at 100, 200 and 300 mA cm^{-2} for different In doping concentrations, (d) EIS, (e) Comparison of OER activities among $\text{In}_{1.0}$ -NiCo LDH and other referenced electrocatalysts, (f) Chronopotentiometric stability test of $\text{In}_{1.0}$ -NiCo LDH

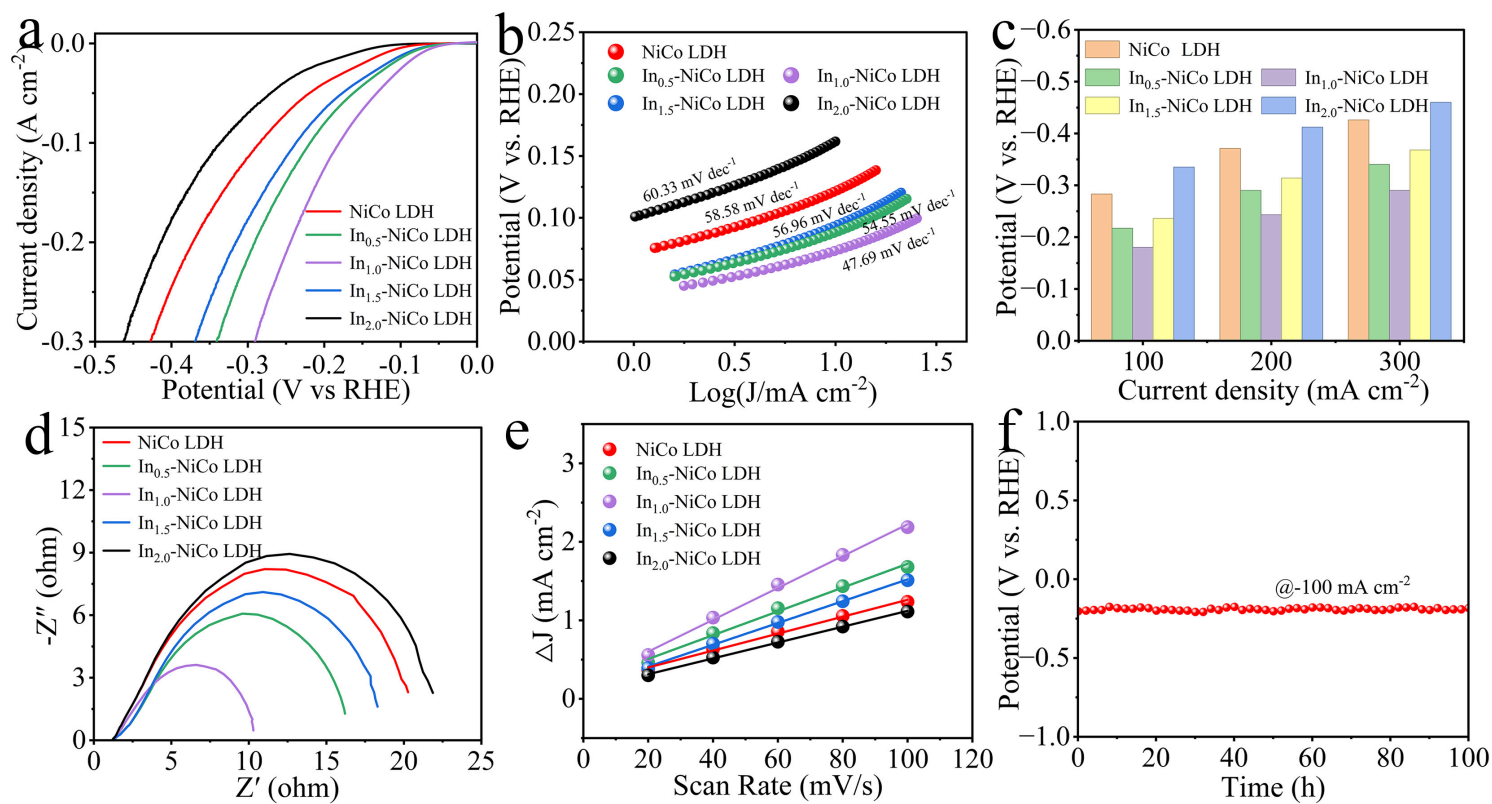


Figure 6

(a) HER LSV curves of undoped and In-doped NiCo LDH, (b) Corresponding Tafel plots, (c) Comparison of overpotential at 100, 200 and 300 mA cm^{-2} for different In doping concentrations, (d) EIS, (e) C_{dl} determination by CV at different scan rates, (f) Chronopotentiometric stability test of In_{1.0}-NiCo LDH

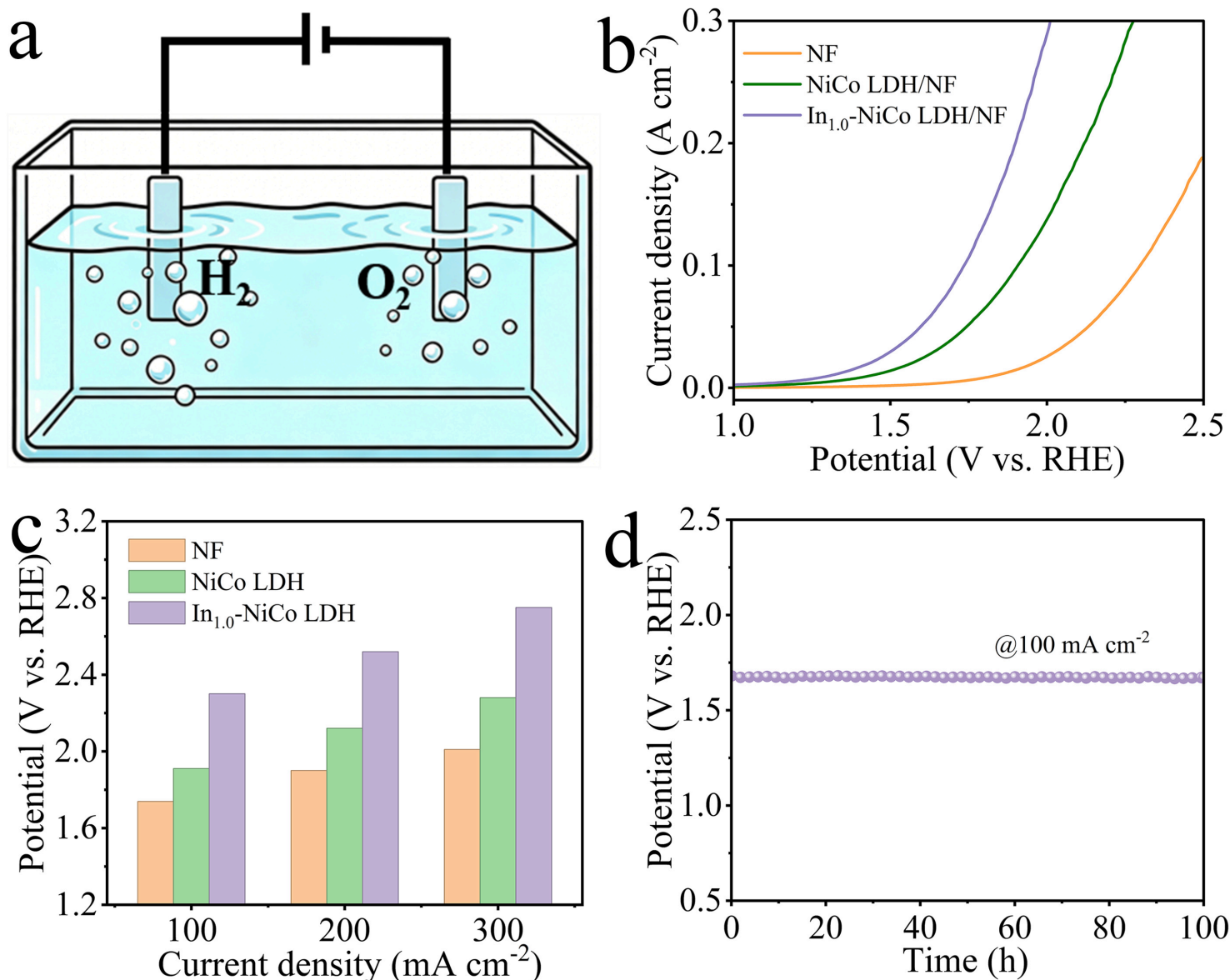


Figure 7

(a) Schematic illustration of the $\text{In}_{1.0}\text{-NiCo LDH}$ full-solution electrolyzer, (b) LSV curve of $\text{In}_{1.0}\text{-NiCo LDH/NF}$ in 1M KOH full-solution electrolyzer, (c) Performance comparison of different catalysts under different currents, (d) Stability test of $\text{In}_{1.0}\text{-NiCo LDH/NF}$

Supplementary Files

This is a list of supplementary files associated with this preprint. Click to download.

- [Supportinginformation.docx](#)
- [scheme1.png](#)