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RESEARCH ARTICLE

Ni-doped Co-layered Double Hydroxides as High-performance Electrocatalyst for the Hydrogen Evolution Reaction

Ashim Chaudhari [1], Mani Ram Kandel [1*], Abid Ali [1], Dasu Ram Paudel [2], Bipeen Dahal [3], Subhangi Subedi [2]

[1]Department of Chemistry, Amrit Campus, Tribhuvan University, Lainchaur, Kathmandu, Nepal

[2]Department of Chemistry, Tri-Chandra Multiple Campus, Tribhuvan University, Ghantaghar, Kathmandu, Nepal

[3]Central Department of Chemistry, Tribhuvan University, Kirtipur, Kathmandu, Nepal

**Corresponding E-mail: mrk.shringa4@gmail.com*

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Abstract

The development of cost-effective and efficient electrocatalysts for the hydrogen evolution reaction (HER) is crucial for advancing sustainable hydrogen production. In this study, nickel-doped cobalt-layered double hydroxides (Ni-Co-LDH) were successfully synthesized using a simple room-temperature aging method. This one-step process allows for the uniform incorporation of nickel into the cobalt LDH structure, which significantly improves catalytic activity. The synthesized Ni-Co-LDH catalysts demonstrated excellent HER performance in alkaline conditions, with low overpotentials of 104 mV at 10 mA cm^{-2} and 204 mV at 50 mA cm^{-2}. The enhanced performance is attributed to the synergistic effect between nickel doping and the cobalt-LDH, which creates an optimized surface for hydrogen evolution and accelerates charge transfer. Scanning electron microscopy (SEM) confirmed a uniform and favorable surface morphology, while electrochemical impedance spectroscopy (EIS) showed improved charge transport kinetics. These results suggest that Ni-Co-LDH is a highly promising, low-cost material for high-performance electrocatalysis in HER. The ease of synthesis, combined with its high efficiency, makes this catalyst a strong candidate for large-scale hydrogen production, contributing to the development of renewable energy solutions.

Keywords: Electrocatalyst; Ni-Co-LDH; Catalytic activity; Linear sweep voltammetry; Water-splitting

Introduction

The escalating challenges of climate change, the global energy crisis, and overdependence on fossil fuels have catalyzed intensive research into alternative energy sources for a sustainable future [1]. Hydrogen, recognized for its clean energy potential and high energy density, is gaining significant attention as a promising alternative fuel [2]. Electrolysis, particularly when powered by renewable energy sources like solar and wind, offers a sustainable pathway for hydrogen production [3]. This process is governed by two essential electrochemical reactions: the hydrogen evolution reaction (HER) and the oxygen evolution reaction (OER) [2–5]. Currently, noble metals such as platinum (Pt) are the most efficient catalysts for HER due to their near-zero overpotential and optimal hydrogen adsorption energy (ΔG) [2–5]. However, the prohibitive cost of noble metals has driven extensive research toward discovering earth-abundant, non-noble metal electrocatalysts that can deliver high catalytic efficiency across a range of electrolyte solutions [2–7]. Developing such catalysts is crucial for advancing cost-effective and scalable hydrogen production technologies.

Similarly, the synthesized catalysts, Pt/dNiCo LDH (synthesized using electrodeposition method) and Pt/eNiCo LDH (synthesized using ZIF-67 etching), exhibit remarkable catalytic performance for HER under alkaline conditions. This enhanced performance is attributed to electronic modulation, the intrinsic activity of Pt, and the synergistic effect of Pt doped on NiCo-LDH [18]. Transition metal doping enhances electronic conductivity, optimizes active sites, stabilizes the layered framework, and increases surface defects for improved catalytic activity. Another TM-LDH serves as the core component, offering a high surface area, facilitating ion diffusion, supporting redox reactions, and synergizing with the dopant Transition metal for efficient charge transfer and reactant adsorption. This interplay creates a highly efficient, durable, and robust system, ideal for applications such as electrocatalysis and energy storage [19–23]. Various cobalt-based catalysts have been extensively studied for electrochemical water splitting [7,19]. However, the incorporation of nickel as a dopant into cobalt-based layered double hydroxides to achieve excellent performance through a simple synthesis method remains largely unexplored. This research stands out by employing a simple and scalable room-temperature aging method to synthesize Ni-Co-LDH as cost-effective and efficient electrocatalysts for the HER. Unlike prior studies that often rely on complex techniques like hydrothermal synthesis or expensive noble metal doping, this work focuses on enhancing the intrinsic properties of cobalt-based LDHs through nickel incorporation [12,13,18]. A Ni-doped Co-LDH system, with enhanced electrical conductivity and synergistic interactions between doped Ni and Co, holds significant potential for superior electrocatalytic performance in overall water splitting.

This study aims to advance the efficiency and affordability of HER electrocatalyst by developing Ni-Co-LDH through a straightforward room-temperature aging process. The incorporation of Ni enhances catalytic activity by generating active sites, while Co contributes to structural stability and redux-driven activity. The Co-LDH framework provides a high surface area and the capacity to intercalate various anions, crucial for effective HER. Nickel foam is used as a conductive support, improving charge transfer and maintaining the structural integrity of the electrocatalyst. The Ni-Co-LDH catalysts demonstrate better HER performance in alkaline conditions, with low overpotential. This enhanced performance is attributed to the synergistic effects of nickel doping and the cobalt LDH matrix, which optimize the catalytic surface and facilitate efficient charge transfer. These findings underscore Ni-Co-LDH as a highly promising, economical material for high-performance HER, advancing the development of scalable hydrogen production technologies and supporting the transition to renewable energy solutions.

Materials and Methods

Chemicals

Cobalt (II) nitrate hexahydrate ($Co(NO_3)_2.6H_2O$, 97.0%, S. d. Fine-Chem Ltd.), nickel (II) nitrate hexahydrate ($Ni(NO_3)_2.6H_2O$, 98.0%, Sarabhai M. chemicals Ltd.), ammonium fluoride (NH_4F, 95.0%, Merck Specialities private Ltd.), urea (NH_2CONH_2, 99.5%, Blulux laboratories private Ltd.), Conc. hydrochloric acid (HCl), ethanol (C_2H_5OH, 99%) and distilled water. All the mentioned chemicals were received from Amrit Campus chemical lab. Nickel foam (NF) was also received from there. Those chemicals were used without any further purification.

Materials synthesis

Precleaning of nickel foam (NF)

There should be oxide on the surface of Ni foam due to environmental oxygen. So, it should be cleaned to remove the oxide layer. For cleaning, Ni foam of 2×1 cm^2 was cut and put into the beaker containing the solution of 1mL conc. HCl and 250 mL distilled water to remove the oxide layer. It was then sonicated for 15 min. After then Ni foam was washed with distilled water 7 times and again washed with ethanol. Finally, Ni foam was then dried in a vacuum at 60^oC for 4 hours.

Synthesis of Ni-Co-LDH electrocatalyst

For the synthesis Ni-Co-LDH, 0.5 g nickel nitrate (Ni $(NO_3)_2$, 5.0 g cobalt nitrate (Co $(NO_3)_2$, 1.0 g ammonium fluoride (NH_4F) and 2.0 g urea (NH_2CONH_2) were taken in a 200 mL beaker, and 50 mL distilled water was added to make a solution. The solution was sonicated for 1 hour. Afterward, pre-cleaned nickel foam was put in the solution, and the beaker was covered with aluminum foil. The solution is then put on a hot plate for 7 hours. at 60^oC. After cooling, the nickel foam was taken out and then kept in oven to dry. The remaining solution was filtered through filter paper, and the residue was collected. The collected residue was also dried in an oven. Finally, both dried nickel foam and residue were prepared for characterization. The control material, Co-LDH, was prepared using the same method outlined above, excluding the Ni dopant precursor.

Physicochemical and Electrochemical characterization

Cu Kα radiation (λ = 0.154 nm) was employed for X-ray diffraction (XRD) analysis to characterize the crystal structure of the catalyst, using a Bruker D8 Advance diffractometer. The morphology of the samples was investigated through Scanning Electron Microscopy (SEM) with a Zeiss 300 VP instrument. The hydrogen evolution reaction (HER) performance of the catalysts was investigated using an electrochemical workstation (CHI-660E electrochemical workstation (USA) in a 1 M KOH electrolyte. The electrochemical cell comprised graphite rods as the counter electrode, a saturated calomel electrode (SCE) as the reference electrode, and the prepared catalyst electrode with a geometric area of 1 × 1 cm^2 as the working electrode. Electrochemical impedance spectroscopy (EIS) was conducted over a frequency range of 0.1 to 100 kHz. The polarization curves for linear sweep voltammetry (LSV) were recorded at a scan rate of 2 mV s^{-1}. The potentials were converted to the reversible hydrogen electrode (RHE) scale using the Nernst equation:

($E_{RHE} = E_{Ag/AgCl} + 0.0591\ pH + E^0_{Ag/AgCl}$) [24]. Additionally, chronoamperometric measurements were carried out for 12 hours at a constant voltage to evaluate the long-term stability and performance of the working electrode. Nyquist plots were obtained using the EIS method, conducted over a frequency range of 10^5 to 10^2Hz.

Results and Discussion

The process to create the Ni-Co-LDH@NF heterostructure began with growing cobalt hydroxide nanosheets vertically on nickel foam (Ni-Co-LDH) by reacting specific precursors, as outlined in the experimental section. During the hot plate process, urea decomposed into ammonia and carbon dioxide. The ammonia then hydrolyzed under high temperature and pressure, slowly releasing hydroxide ions into the solution, which led to the formation of Ni-Co-LDH nanosheets on the nickel foam [2].

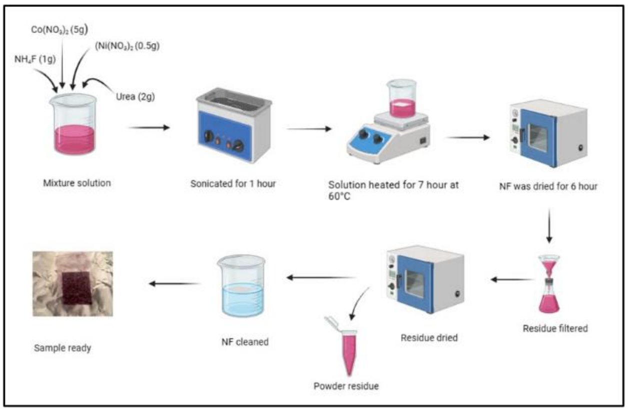

Fig. 1: Schematic diagram of synthesis of Ni-Co-LDH electrocatalyst.

NH_4F was used to refine the material's morphology by increasing the metal ion binding energy, aiding in the development of the LDH structure on the nickel foam. Additionally, NH_2CONH_2 and NH_4F created an alkaline environment that guided the material's morphological development. Residual carbon and nitrogen did not affect the chemical transformation. After synthesis, any remaining impurities were washed away with distilled water and ethanol multiple times, resulting in the pure Ni-Co-LDH. This study aims to improve our understanding of producing more effective electrocatalysts for water splitting. **Fig. 1** shows the synthesis methods for Ni-doped Co-LDH, with Ni-Co-LDH nanosheets uniformly grown on nickel foam, leading to a color change from gray to dark pink due to the hydrolysis of Ni and Co ions.

Crystallography and morphological analyses

X-ray Diffraction (XRD) is a useful technique for studying the structure of materials at the atomic level [25]. When used with Ni-Co Layered Double Hydroxide (LDH) electrocatalysts, which are important for hydrogen production, XRD helps us see what types of crystals are present and confirm that nickel and cobalt are part of the LDH structure. The powder X-ray diffraction (XRD) study was performed to confirm the successful synthesis of the designed electrocatalyst [4].

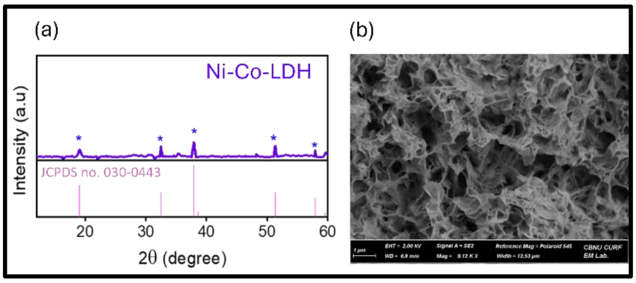

Fig. 2 (a): X-ray diffraction pattern of Ni-Co-LDH **(b)** SEM image of Ni-Co-LDH.

The crystalline (lattice) phase of the Ni-Co-LDH was achieved via XRD patterns. As mentioned in the experimental section, pure Ni-Co-LDH was obtained via the simple ageing method at room temperature. Peaks of the XRD pattern were consistent with different crystal phases crystal planes (001), (100), (101), (102), and (110), of Ni-Co-LDH at $2\theta = 19.0^0$, 32.4^0, 37.9^0, $51.4\ ^0$, and $57.9\ ^0$, respectively as shown in the **Fig. 2 (a)** (JCPDS No. 030-0443, this reference refers to the crystallographic data from the Joint Committee on Powder Diffraction Standards (JCPDS) database). The result confirms the formation of the expected crystal structure of Ni-Co-LDH. Furthermore, the absence of significant changes in the XRD pattern of Co-LDH including Ni dopant can be attributed to the similar ionic sizes of Ni^{2+} and Co^{2+} and the lower concentration of Ni. These results confirm the successful formation of the expected Ni-Co-LDH crystal structure [26, 29]. A Field Emission Scanning Electron Microscopy (SEM) image is a high-resolution photograph produced by a field emission scanning electron microscope [26]. This technique employs a concentrated electron beam to scan the surface of a specimen. As the electrons interact with the sample's atoms, they generate various signals that are captured and transformed into a detailed image. SEM is particularly known for delivering high-resolution images with remarkable depth of field, enabling the observation of intricate surface details and textures at the nanoscale level. In the SEM image of Nickel (Ni) doped Cobalt Layered Double Hydroxides (Co-LDH) **Fig. 2(b)** designed for the HER, the material exhibits a characteristic layered nanosheet morphology. The SEM image of Ni-Doped Co-LDH for HER reveals a highly porous and layered structure, indicative of the potential for high surface area and active site availability [8,14]. The SEM image shows a network of interconnected nanosheets and voids, which are crucial for hydrogen evolution reaction (HER) [8,12,14]. Porosity and layered morphology enhanced electron transfer and gas diffusion pathways, which are beneficial for catalytic performance [27]. The doping with Ni appears slight changes in the morphology, with increase in potential in surface roughness, which enhance the materials active sites for HER [12,28,29]. The presence of Ni further enhanced the electronic properties of Co-LDH, potentially improving their catalytic efficiency [12]. Compared to undoped Co-LDH, doped shows a more uniform distribution of flakes, which contributes to more consistent HER performance.

Electrocatalytic characterization for HER

Linear sweep voltammetry and Electrochemical impedance spectroscopy analyses

In the synthesis and analysis of Ni-Co-LDH electrocatalysts, Linear Sweep Voltammetry (LSV) and Electrochemical Impedance Spectroscopy (EIS) are crucial for understanding their electrochemical properties. LSV measures the current response (current density) as a function of the applied potential, which helps evaluate the Ni-Co-LDH effectiveness in catalyzing reactions such as the Hydrogen Evolution Reaction (HER). By examining the shape and position of peaks in the LSV curve, we can determine the catalyst's onset potential and efficiency.

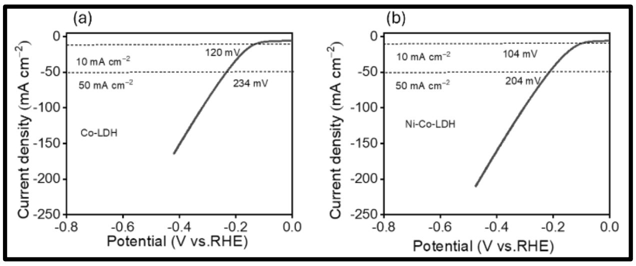

Fig. 3. **(a)** LSV curve of Co-LDH **(b)** LSV curve of Ni-Co-LDH.

These data are vital for comparing the Ni-Co-LDH electrocatalyst's performance with other Co-LDH and for confirming its catalytic activity. The electrochemical performance of the Ni-Co-LDH electrode was tested in a 1.0 M KOH alkaline solution. **Figs. 3 (a)** and **(b)** displayed LSV curves representing the HER on a Co-LDH and Ni-Co-LDH electrode under alkaline conditions, respectively, with the electrode having dimensions of 1 cm × 1 cm. The polarization curves from the LSV measurements indicated that the Co-LDH electrode (**Fig. 3(a)**) required an overpotential (η) of 120 mV to achieve a current density of 10 mA cm^{-2}. The Co-LDH demonstrated relatively poor electrocatalytic performance, with overpotentials of 120 mV and 234 mV for current densities of 10 mA cm^{-2} and 50 mA cm^{-2}, respectively. This suggests that the Co-LDH has limited efficiency in the HER process due to its higher overpotential requirements. In comparison, the Ni-Co-LDH electrocatalyst required a significantly lower overpotential of 104 mV to achieve a current density of 10 mA cm^{-2}. For a higher current density of 50 mA cm^{-2}, the Ni-Co-LDH showed an overpotential of 204 mV. Under alkaline conditions, Ni-Co-LDH demonstrated excellent HER performance, with overpotentials of 104 mV and 204 mV at current densities of 10 mA cm^{-2} and 50 mA cm^{-2}, respectively (**Fig. 3 (b)**). The enhanced electrocatalytic activity of the Ni-Co-LDH is attributed to the combined effects of Ni and Co dopants, which were incorporated into the crystal lattice with specific precursor concentrations [28,29]. The similar atomic radii of Ni and Co allowed them to replace lattice atoms, creating defects that likely contributed to the improved HER performance. Electrochemical Impedance Spectroscopy (EIS) is a powerful method used to measure the electrical properties of materials, particularly in energy storage and catalysis. By applying an alternating current signal, EIS measures key parameters like charge transfer resistance (R_{ct}) and capacitance [30]. A lower R_{ct} indicates improved electron movement between the electrode and electrolyte, enhancing catalyst efficiency, while higher capacitance reflects better charge storage and release, crucial for effective catalysis [31]. EIS also assesses capacitance, reflecting the material's ability to store and release charge quickly, which is crucial for effective catalysis [32]. In Nyquist plot, the real part of impedance (Z') is shown on the horizontal axis, and imaginary part (Z") on the vertical axis, forming a semicircular pattern that reflects the material's electrochemical behavior. In **Figs. 4 (a) and (b)**, Ni-Co-LDH exhibited smaller semicircle (**Fig. 4 (b)**) in the Nyquist plot compared to Co-LDH (**Fig. 4 (a)**). This pattern reflects significant changes in charge transfer resistance and capacitance, which are critical for assessing Co-LDH and Ni-Co-LDH performance in energy storage and catalysis. The Ni-Co-LDH in Fig 4(b), EIS results show that incorporating Ni reduces R_{ct}, increases capacitance, and enhances the active surface area, enabling more efficient charge transfer and superior catalytic performances [12]. These findings, as shown in the Nyquist plots, underline the material's effectiveness and durability for processes like the HER.

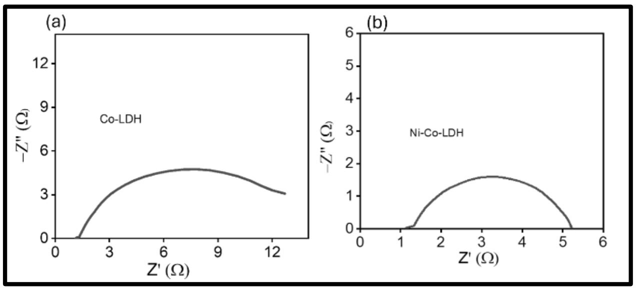

Fig. 4. (a) EIS curve of Co-LDH **(b)** EIS curve of Ni-Co-LDH.

Conclusions

The study demonstrates that Ni-Co-LDH can be synthesized effectively using a straightforward room-temperature aging method. This process allows for the uniform integration of Ni into the cobalt-LDH structure, enhancing its catalytic activity for the HER. The Ni-Co-LDH catalysts exhibit excellent performance in alkaline environments, achieving low overpotentials of 104 mV at 10 mA cm^{-2} and 204 mV at 50 mA cm^{-2}. These improvements are attributed to the synergistic effects of Ni doping and the cobalt-LDH framework, which together optimize the catalytic surface and facilitate efficient charge transfer. The findings suggest that Ni-Co-LDH represents a promising and cost-effective electrocatalyst for HER, offering significant potential for large-scale hydrogen production and supporting the shift towards renewable energy solutions.

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Author's Contribution Statement

Ashim Chaudhari: Methodology, Investigation, Formal analysis, Data curation, Writing-original draft preparation, **Mani Ram Kandel**: Conceptualization, Resources, Funding acquisition, Writing-original draft, Writing review and Editing, supervision, **Abid Ali**: Formal analysis and Writing-review **Dasu Ram Paudel**, **Bipeen Dahal, Subhangi Subedi:** Formal analysis

Conflict of Interest

The authors do not have any conflict of interest throughout this research work.

Data Availability Statement

The data supporting this study's findings are available from the corresponding authors upon reasonable request.

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