



Article

Role of Electrochemical Precipitation Parameters in Developing Mixed-Phase Battery-Grade Nickel Hydroxide

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Abstract: There is a high demand for nickel hydroxide as an engineering material used in the positive electrode of nickel metal hydride (Ni-MH) rechargeable batteries. These batteries are extensively used in various small instruments, disposable batteries, and electric vehicles. The structure of nickel hydroxide significantly influences the discharge capacity and energy density, key properties of Ni-MH batteries, and this structure is primarily determined by the synthesis method used. In this study, nickel hydroxide was synthesized using an electrochemical precipitation method, with current density acting as a parameter to control the desired phase of the product, whether α -nickel hydroxide, β -nickel hydroxide, or a combination of both. At a current density of 50 A/m², the synthesized nickel hydroxide demonstrated a smaller particle size and a superior discharge electrochemical property in comparison to that generated at 500 A/m². The effect of agitation in catholyte was also investigated to examine the change in discharge property of the precipitated material. The product synthesized at 500 A/m² from an agitated catholyte exhibited a tap density of 1.24 g/cc and an improved discharge capacity of 254 mAh per gram of Ni(OH)₂.

Keywords: nickel hydroxide; electrochemical precipitation; current density; agitation; discharge capacity



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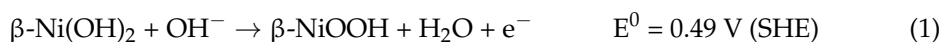
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1. Introduction

Nickel hydroxide is regarded a crucial energy material due to its application in nickel metal hydride (Ni-MH) batteries and other types of batteries [1,2]. It has a hexagonal close-packed arrangement where nickel atoms are located in the (001) plane and surrounded by six hydroxyl groups, which lie alternately above and below the (001) plane. Nickel hydroxide has two polymorphic forms: (i) β -Ni(OH)₂, which features a well-ordered brucite-type structure with a c-axis spacing of 4.8 Å, and (ii) α -Ni(OH)₂, which is characterized by a randomly stacked turbostratic structure in its hydrated state, exhibiting a c-axis spacing of 7.5 Å because of the presence of intercalated molecules of water between the (001) planes [3]. The charge–discharge cycle for β -Ni(OH)₂ can be outlined as follows:



The transition of β -Ni(OH)₂ to β -NiOOH requires protons to diffuse from the brucite lattice. During charging, the change from β -Ni(OH)₂ to β -NiOOH involves the transfer of one electron, which causes Ni²⁺ to be oxidized to Ni³⁺. As a result, β -Ni(OH)₂ is recognized

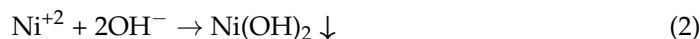
for its theoretical discharge capacity of 289 mAh/g. Nonetheless, during overcharging, γ -NiOOH is generated and can reversibly convert back to α -Ni(OH)₂ through electrochemical processes. The shift from α -Ni(OH)₂ to γ -NiOOH involves a considerable transfer of electrons, yielding a maximum discharge capacity greater than that of β -Ni(OH)₂ [4,5].

In contemporary nickel metal hydride rechargeable batteries, the theoretical discharge capacity is almost achieved with β -nickel hydroxide electrode material. However, it has been observed that the discharge capacity of the α -phase of nickel hydroxide surpasses that of the β -phase. Furthermore, the α -phase produced in the current context has a lower tap density compared to the commercially available β -phase nickel hydroxide [6]. This presents an opportunity to investigate the combination of both α - and β -phases to develop a superior battery electrode material with an enhanced discharge capacity and optimal tap density. For nickel hydroxide used in battery applications, the significance of tap density and discharge capacity are regarded as extremely important considering the targeted capacity density and power capacity of the material. Various structural features, such as crystal size, lattice disorders, crystal growth, degree of crystallinity, and the inclusion of anions or intercalated H₂O in the crystals influence the discharge capacity of nickel hydroxide. These factors are largely determined by the preparation technique used. Thus, the objective of this work was to regulate the process parameters in order to engineer the nickel hydroxide product.

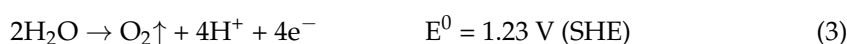
Furthermore, several researchers have explored the use of β -nickel hydroxide for other applications such as supercapacitors, nanofluids, and electrocatalysts in water splitting [7–11]. Lu and Hong [7] studied the incorporation of graphene as a layer between a 3D-foamed nickel substrate and β -nickel hydroxide by the chemical vapour deposition method. They reported that the resulting electrode material demonstrated excellent cycle life and specific capacitance, confirming its potential as a suitable material for supercapacitors and other energy storage applications. They examined the creation of a supercapacitor electrode that involved one-step hydrothermal method for depositing β -nickel hydroxide nanoparticles onto a substrate of nickel foam. As reported by the same authors, at a current density of 3 A/g, the maximum super capacitance achieved was 539 F/g [8]. Sen et al. developed nanofluids containing nickel hydroxide nanoparticles, which exhibited high flowability, excellent electrochemical performance, and significant loading capacity [9]. The formulation of dispersions with more than 60 wt% solids could be achieved by particle surface modification using small organic molecules. Krishnamurthy et al. explored how doping β -nickel hydroxide with Fe affects its performance as an electrocatalyst for water splitting. The study revealed that the developed iron-doped nickel hydroxide can possess enhanced electrocatalytic property for water splitting operation at a lower overpotential for both cathodic and anodic reactions [10].

Currently, battery-grade nickel hydroxide is prepared by employing the chemical precipitation route, and their characteristics are well understood. The factors affecting the charge–discharge characteristics of nickel hydroxide include the (i) particle size and morphology, (ii) surface area, (iii) intercalated water content, (iv) crystallite size, (v) phase, and (vi) structural characteristics such as stacking faults and cation vacancies [11]. These properties can be tailored through adjustment of the different precipitation conditions, such as the pH of the precipitation process, type of alkali reagent, temperature of the reaction, source of nickel ions, rate of mixing of the reactants, ageing of the product, hydrothermal treatment of the product, and additives [12–15]. The electrochemical precipitation process is one of the options for the production of battery-grade nickel hydroxide where the current density can be successfully utilized as a regulator for the supply of hydroxyl ions, which in turn controls hydroxyl/nickel ion ratio in the solution.

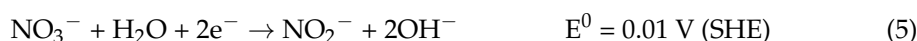
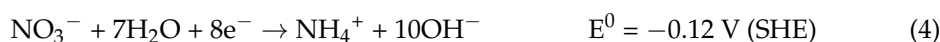
The fundamental phenomenon underlying the formation of nickel hydroxide, as illustrated by Equation (2), is consistent for both the chemical and electrochemical precipitation methods of synthesis.



Nonetheless, the origin of hydroxyl ions (OH^{-}) differs from one method to another. In the chemical method, the hydroxyl ions are provided through chemical reagents such as NaOH, KOH, $\text{NH}_4(\text{OH})$, or urea, whereas in the electrochemical precipitation method, the hydroxyl ions are generated in situ through electrolytic reactions. If nickel nitrate is used for preparing the electrolytic solution, the cathodic reaction involving the reduction in nitrate ions will generate OH^{-} ions leading to an increase in pH of the solution nearby the cathode surface. Nickel hydroxide precipitates from the solution when the solubility limit for its ionic product is surpassed. The use of an inert anode results in an anodic reaction according to Equation (3).



The major possible reactions of nitrate ion reduction at the cathode, reported by several researchers [16–19], can be illustrated as follows:



The main advantages of using the electrochemical precipitation technique are the following: (i) it provides a well-regulated OH^{-} ion supply by controlling the current density; (ii) the production of highly pure nickel hydroxide because of the use of only water and nickel nitrate in the electrolyte unlike chemical reagent-assisted nickel hydroxide production by the chemical precipitation method; (iii) all avenues are available for regulating the shape, size, and structure of the precipitated nickel hydroxide; and (iv) the process is environment friendly. The synthesis of thin films has been carried out by this method, and subsequently, the electrode is prepared by foaming with nickel hydroxide or the loading of a sintered nickel plaque [20]. In the electrochemical impregnation method, the hydroxyl ions, generated at the cathode due to reduction in nitrate ions in the nickel plaque pores, combine with nickel ions to produce nickel hydroxide. By controlling the parameters such as (i) lowering the current density gradually, (ii) raising the temperature of the electrolytic bath, and (iii) keeping the bath concentrations under the controlled addition of nickel nitrate and sodium nitrite, uniform coating of nickel hydroxide can be achieved. The electrolyte composition of 4M nickel nitrate was found to be the most appropriate formulation. An investigation carried out by Portemer et al. [21] shows that β -nickel hydroxide is formed at a temperature of 60 °C or higher, whereas α -nickel hydroxide tends to form at a comparatively lower temperature. However, the production of nickel hydroxide in bulk through the electro-precipitation route has not been studied extensively. Krynitz et al. [22] have patented a process wherein the nickel metal is oxidized in a medium containing mixture of chloride and sulphate solutions to produce nickel hydroxide as an intermediate product which eventually transforms to pure nickel hydroxide by alkaline treatment. Coates et al. [23] precipitated nickel hydroxide under operating conditions of 50–200 A/m² CD and 80–95 °C from an acidic solution of pH 1.8–2.2 containing nickel nitrate. Kong et al. [24] examined the process of electrochemically precipitating nickel hydroxide films onto a platinum electrode from a nickel nitrate solution, achieving densities between 1.25 and 1.5 g/cc. Subbaiah et al. [25] outlined the electrolytic formation of β -Ni(OH)₂ from a nickel sulphate solution, using nitric acid to supply nitrate ions, and

reported the resulting material exhibits 200 mAh/g of discharge capacity. The incorporation of different metallic additives in the nickel hydroxide lattice was found to bring an improvement by an enhancement in charge efficiency as well as utilization efficiency of the electrode material [26–28]. In recent years, much emphasis has been given on the preparation of such active materials possessing adequate discharge capacity and energy density, which needs the development of a structure constituting both the α - and β -phase and a higher tap density. Mixed-phase nickel hydroxide precipitation through the chemical method has been investigated widely and deeply [29–31]. Further, there have been several reports in the literature on nickel hydroxide precipitation from different aqueous systems [30]. Thus, considering the advantages of the electrochemical precipitation method mentioned above, there is a need to explore the possibility of improving the discharge properties of nickel hydroxide electrochemically precipitated from an aqueous system like nickel nitrate solution. Moreover, there is research highlighting that the current NiMH rechargeable batteries employ β -phase nickel hydroxide, owing to its impressive volumetric electrochemical capacity. Despite this, α -phase nickel hydroxide is increasingly preferred as the active electrode material because of its higher stability in structure, minimal volume variations, and improved electrochemical efficiency in charge–discharge cycles. The insignificant change in volume may be attributed to reduced absorption of water resulting in improved stability and cell design. However, β -phase nickel hydroxide offers the advantage of higher volumetric energy density compared to the α -phase. Thus, a mixed formulation of β - and α -Ni(OH)₂ could be a promising option to achieve the desired power density and discharge capacity in batteries.

Taking this into account, the electrochemical processing windows for the synthesis of mixed-phase nickel hydroxide were established. Efforts were made to incorporate the α -phase into the crystal structure of β -nickel hydroxide during synthesis via electrochemical precipitation, by controlling variables such as bath composition, current density, and agitation in the catholyte.

2. Experimental

2.1. Materials

In this work, all the chemicals used for conducting experiments were procured from Merck, Mumbai, India. An electrolytic cell of 1 L volume having two compartments for containing catholyte and anolyte, physically separated by means of a diaphragm made of polypropylene, was used for the electrochemical production of nickel hydroxide. In a two-electrode configuration system used for the production of nickel hydroxide, a stainless steel (SS) 304 sheet was connected to the cathode terminal while an IrO₂-coated titanium sheet worked as the anode because of its inertness and non-consumable property in an acidic medium.

2.2. Method

Nickel nitrate was dissolved in the distilled water to prepare both electrolytic solutions, i.e., catholyte and anolyte. Nickel hydroxide was produced through the process of electrochemical precipitation, employing a controlled power supply unit (APLAB, Mumbai, India) to deliver direct current (DC). All the experiments were conducted at ambient temperature (35 ± 2 °C) and atmospheric pressure. At the end of each experiment, the precipitates formed in the catholyte were separated physically by filtration and then washed using distilled water until free from nickel nitrate in the filtrate and subsequently dried for 24 h at a temperature of 100 ± 5 °C before storing inside a desiccator for further characterization.

2.3. Characterization

The concentrations of nickel in the as-prepared hydroxide precipitates were determined by a method involving the acid digestion of the sample followed by an analysis of the elements using Atomic Absorption Spectroscopy (Perkin Elmer Analyst 200, PerkinElmer, Waltham, MA, USA). Using a particle size analyzer (MALVERN Mastersizer 2000MU, MALVERN, Malvern, UK), the size of the particles was determined. The tap density of the particles was assessed using a Hall Flow Meter. The particles flowed through an opening under gravity, and then the powder collector, which has a 25 cm³ capacity, was tapped 200 times. The tap density was calculated by dividing the weight of nickel hydroxide powder contained in the collector by volume of collector (25 cm³). An X-Ray Diffraction system (PANalytical PW 1830, Philips, Amsterdam, The Netherlands) that utilized Cu K α radiation ($\lambda = 1.5404 \text{ \AA}$) was used for assessing the phases of the samples. In addition, the synthesized nickel hydroxide was analyzed morphologically using a scanning electron microscope (ZEISS Supra SEM, ZEISS, Oberkochen, Germany).

The discharge capacity of the material was evaluated by using an automatic battery charge–discharge unit (BITRODE, Fenton, MO, USA). A mixture of hydroxide sample and graphite powder enclosed in a stainless steel 304 mesh was pressed to produce disc-shaped pellets of 25 mm diameter and 3 mm thickness. Prior to the compaction of the mixture, nickel hydroxide and graphite powder were blended at a ratio of 2:1. For the discharge capacity evaluation, the working electrode terminal is connected to the disc pellet, with the nickel metal serving as the counter-electrode in a two-electrode configuration. A solution of concentration 6 M KOH was used as the electrolyte. The working electrode was subjected to charging in galvanostatic mode at 0.2 C rate for 7.5 h followed by a same rate discharging until it reached a cut-off voltage of 0.8 V. In this context, C denotes the current required to fully discharge the battery material's total capacity in one hour.

3. Results and Discussion

3.1. Effect of Nickel Concentration in the Bath

This study investigated how the initial concentration of nickel ions affected the synthesis, physical properties, and electrochemical characteristics of nickel hydroxide precipitates. Nickel concentrations in both the catholyte and anolyte were adjusted between 30 and 70 g/L. The electrochemical precipitation process was conducted under galvanostatic conditions with a cathodic current density of 200 A/m² over duration of 3 h. The results concerning the initial nickel concentration in the electrolyte and its influence on the percentage of nickel in the product, cell voltage, current efficiency, energy consumption, tap density, particle size, and discharge capacity of the precipitates have been analyzed.

Figure 1 illustrates that both the current efficiency and energy consumption do not show significant improvement with higher nickel concentrations in the electrolyte. However, a slight reduction in particle size occurs as nickel concentration in the bath rises. Charge–discharge tests indicate a moderate increase in discharge capacity as nickel concentration increases up to 60 g/L, followed by a slight decline at 70 g/L (Table 1). Keeping in view all the properties of battery-grade nickel hydroxide, the nickel concentration in the electrolyte for further studies was taken to be 60 g/L.

Table 1. Impact of bath concentration on the product properties, while employing the electrochemical precipitation method.

Ni Concentration (g/L)	Tap Density (g/cc)	Particle Size (μm)		Discharge Capacity (mAh/g)
		d ₅₀	d ₉₀	
30	1.18	30.97	81.25	230 \pm 5
40	1.16	27.85	75.97	233 \pm 7

Table 1. Cont.

Ni Concentration (g/L)	Tap Density (g/cc)	Particle Size (μm)		Discharge Capacity (mAh/g)
		d_{50}	d_{90}	
50	1.19	24.78	63.65	242 ± 4
60	1.21	21.56	55.70	245 ± 2
70	1.24	19.43	56.78	240 ± 3

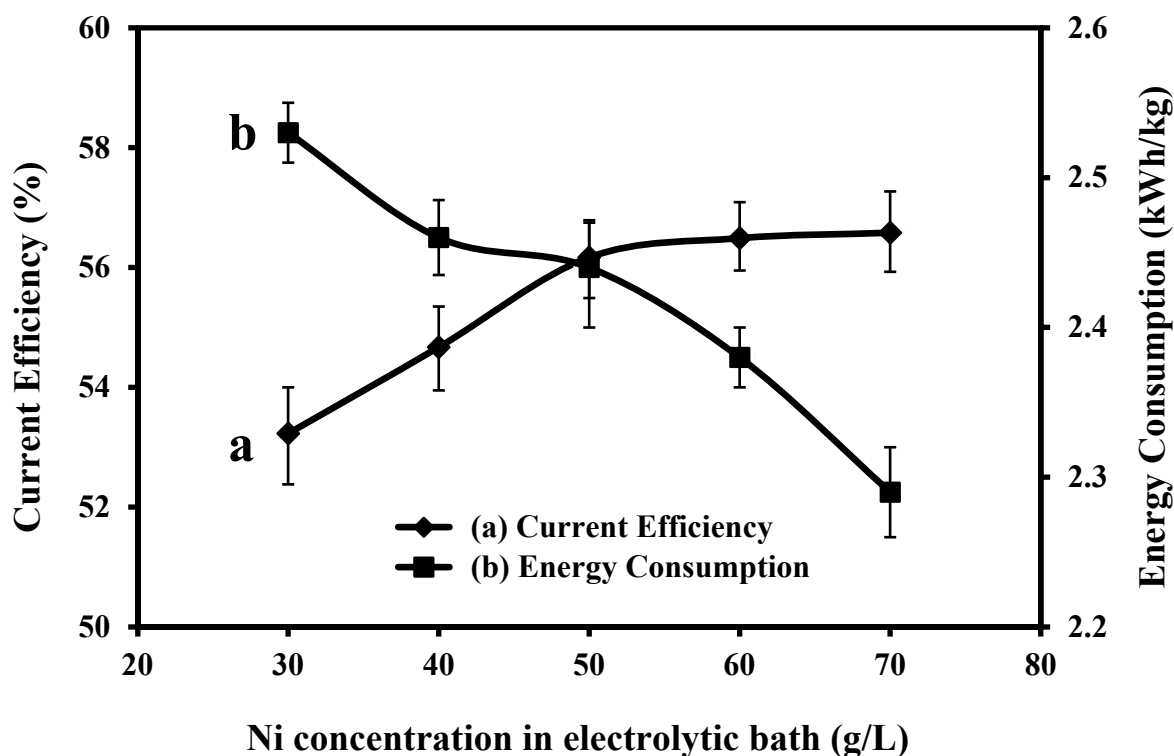


Figure 1. Changes in (a) current efficiency and (b) energy consumption while increasing nickel concentration in the electrolytic bath.

3.2. Effect of Cathodic Current Density

The electrochemical precipitation of nickel hydroxide was performed at varying current densities ranging from 50 to 500 A/m², with both the catholyte and anolyte maintaining an initial nickel concentration of 60 g/L and a pH of 3. The output parameters and properties of the synthesized material are presented in Table 2. The cell voltage is significantly low at 50 A/m² in comparison to that at 500 A/m². As shown in Figure 2, the current efficiency and energy consumption have been determined on the basis of the weight of anhydrous and hydrated nickel hydroxide, respectively. It was noted that as the current density increases, both current efficiency and energy consumption also rise. The application of a lower current density results in the formation of smaller sized particles, and the difference between the particles formed at 50 and 500 A/m² is remarkable. The product, formed at 500 A/m², is well crystalline β -Ni(OH)₂, having a lower tap density as compared to that obtained at current density of 200 or 50 A/m².

Furthermore, Figure 3 illustrates the discharge capacity of the samples produced at varied current densities. It shows that the nickel hydroxide synthesized at 500 A/m² yields a discharge capacity of approximately 220 mAh per gram, while the products obtained at 200 A/m² and 50 A/m² demonstrate discharge capacities of around 246 mAh/g and 282 mAh/g, respectively.

Table 2. Electrochemical precipitation parameters and properties of nickel hydroxide synthesized under different conditions.

Output Parameters and Properties	50 A/m ² and Without Agitation	200 A/m ² and Without Agitation	500 A/m ² and Without Agitation	500 A/m ² and with Agitation
Current Efficiency, %	47.8 ± 1.3	52.4 ± 1.5	59.1 ± 2.0	66.7 ± 1.8
Cell voltage, V	2.2	2.9	4.0	3.6
Energy Consumption (kWh/kg)	1.69 ± 0.08	1.95 ± 0.06	2.87 ± 0.09	2.03 ± 0.06
FWHM β-(101)	0.4741	0.3216	0.2012	0.3097
Crystallite size, nm	30.91	45.57	72.85	47.32
Tap density, g/cm ³	1.28	1.26	1.19	1.24
Particle size (d ₅₀), μm	16.4	23.7	44.6	21.2
Discharge capacity, mAh/g	282 ± 6	246 ± 8	220 ± 7	254 ± 5

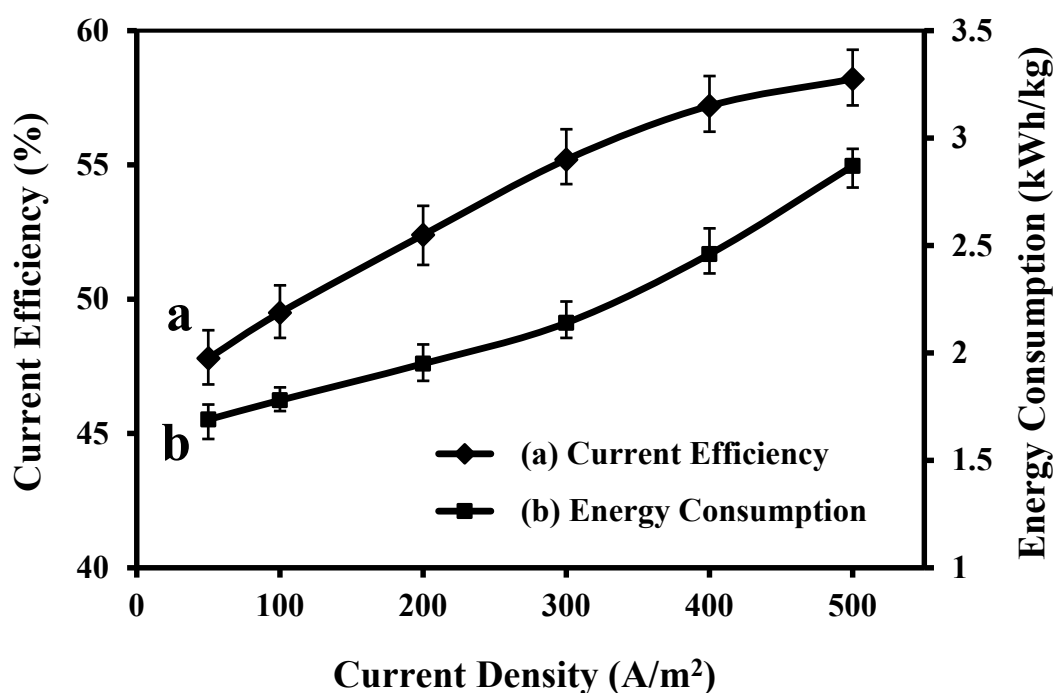
**Figure 2.** The relationship between increasing current density and its effects on (a) current efficiency and (b) energy consumption during the electrochemical formation of nickel hydroxide.

Figure 4 illustrates the XRD patterns of the material synthesized at varied current densities. The precipitates formed at 500 A/m² reveal peaks that represent β-nickel hydroxide with the sequence of planes (001), (100), (101), (102), (110), and (111). However, the product obtained at 50 A/m², shows an additional peak in excess of the diminished peaks similar to those obtained at 500 A/m², which corresponds to (001) plane of the α-phase. The relative peak heights suggest that the precipitates obtained at 50 A/m² and 200 A/m² are probably a mixture of β- and α-phase, with a lower proportion of the α-phase observed at high current densities. The rate of OH⁻ ion generation is quite low at a low current density, suggesting the prevalence of a low ratio of OH⁻/Ni²⁺ in the catholyte. As a result, at 50 A/m² during the precipitation of nickel hydroxide, water molecules may integrate into the lattice, promoting the development of the α-phase, which enhances the discharge capacity of the material. It suggests that a careful regulation of OH⁻/Ni²⁺ ratio through the control of the current density during electrochemical precipitation can lead to either β- or α-Ni(OH)₂ or a mixture of these phases for the improvement in discharge capacity. The relative peak heights also indicate that for β-Ni(OH)₂ synthesized at 50 A/m², preferential

growth occurs along the (101) plane, in contrast to the (001) plane observed at 500 A/m². Moreover, the XRD patterns show that the FWHM value of the β -(101) peak decreases as the current density is reduced from 500 to 50 A/m², indicating a reduction in the distortion of the lattice. According to Kohler et al., an increase in the degree of distortion of the lattice assists in improving the electrochemical property [32]. However, this may not explain the increased discharge capacity of Ni(OH)₂ produced at 50 A/m², as noted by Kohler et al. [32]. Thus, the enhancement in discharge capacity of the product formed at a lower current density can be primarily attributed to the existence of the α -phase.

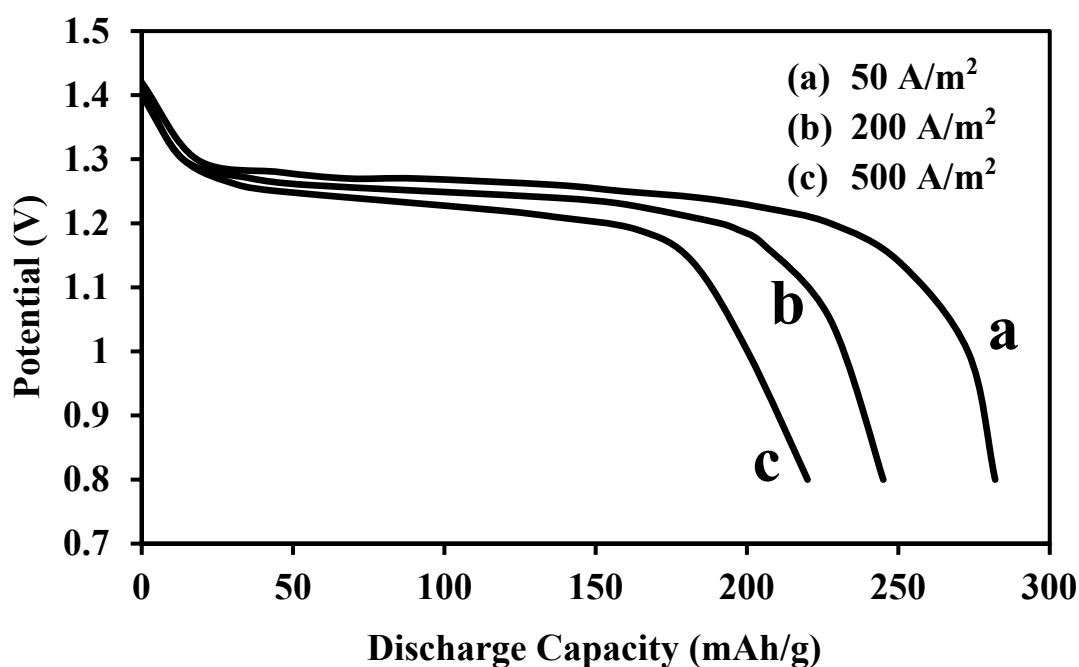


Figure 3. Discharge capacity of nickel hydroxide synthesized electrochemically at varied current densities (a) 50, (b) 200, and (c) 500 A/m² without agitation in the bath.

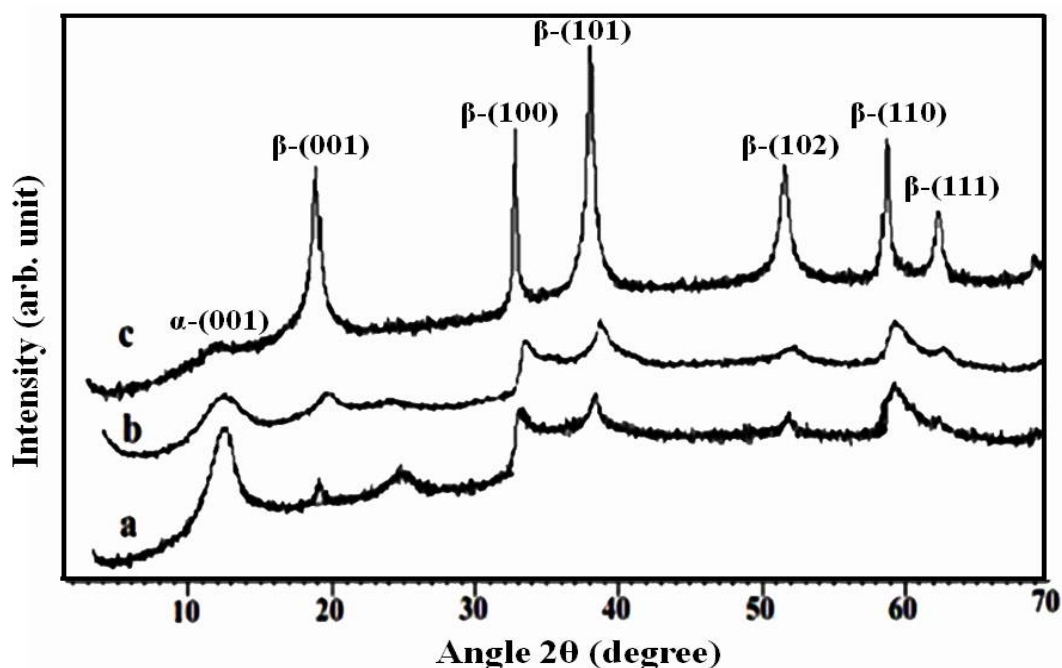


Figure 4. XRD patterns of nickel hydroxide synthesized electrochemically at various cathodic current densities: (a) 50 A/m², (b) 200 A/m², (c) 500 A/m².

The $[\text{OH}^-]/[\text{Ni}^{2+}]$ ratio in the bath during nickel hydroxide electrochemical precipitation is crucial in establishing whether the resulting product comprises a mix of the α - and β -phases (as displayed in Figure 4) or is purely β - $\text{Ni}(\text{OH})_2$. Figure 5 provides the XRD pattern for a standard β -nickel hydroxide product.

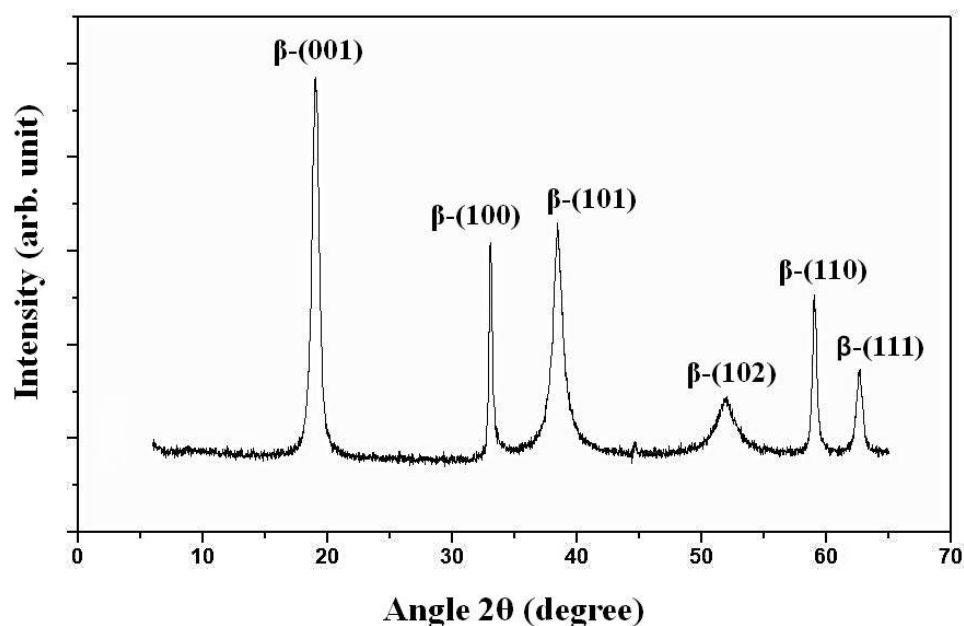


Figure 5. XRD pattern representing a standard β - $\text{Ni}(\text{OH})_2$ product.

It is important to highlight that the unit cell of the β - $\text{Ni}(\text{OH})_2$ crystal exhibits an octahedral configuration of Ni and OH species. The nickel atoms are all in the (001) plane and are surrounded by six hydroxyl groups, which lie alternately above and below the (001) plane. Therefore, six OH^- should be available per Ni^{+2} to obtain the β -nickel hydroxide crystal building up in the proper order. As a result, at a current density of 500 A/m^2 , when the $[\text{OH}^-]/[\text{Ni}^{2+}]$ ratio is above 6, the dominant product is β -nickel hydroxide.

3.3. Effect of Agitation in Catholyte

To examine the impact of agitation on nickel hydroxide electrochemical precipitation, the catholyte was stirred at a steady speed of 500 rpm using a motor-driven stirrer. The study was conducted at 500 A/m^2 , using a nickel concentration of 60 g/L in both the catholyte and anolyte, both with an initial pH of 3, to assess the independent effects of agitation on the characteristics of the nickel hydroxide precipitate. Table 2 displays the pertinent parameters of the electrochemical precipitation process along with the properties of the resulting product. It is observed that agitation has facilitated significant improvement in current efficiency from 59.1% to 66.7% and energy consumption from 2.87 to 2.03 kWh/kg. Since the agitation of the catholyte reduces the concentration gradient of Ni^{+2} ions across the cathode surface and bulk of the catholyte, the kinetics of production of nickel hydroxide improves. Consequently, the quantity of the product, formed in the stipulated period of time, increases ensuring higher current efficiency and lower energy consumption. Further, this has resulted in the production of finer particles with improved tap density. There is a substantial increase in the discharge capacity from 220 to 254 mAh/g, likely owing to the α -phase, resulting from water molecules intercalating into the lattice of β -nickel hydroxide. This improvement in capacity is further illustrated by the XRD data in Figure 6.

The XRD pattern displayed in Figure 6 demonstrates that agitation in the catholyte bath aids the intercalation of water molecules into $\text{Ni}(\text{OH})_2$ crystals, facilitating the generation of α - $\text{Ni}(\text{OH})_2$. This effect simultaneously diminishes the β -phase and removes some crystal

planes of β -Ni(OH)₂. This has resulted in a higher discharge capacity for nickel hydroxide prepared under the influence of the stirring action. If the peaks of α -(001) and β -(001) are compared, then under agitation, it shows that β -(001) is about 30% of that for α -(001), whereas without agitation, α -(001) was about 15 to 20% of that for β -(001). This indicates that agitation plays a crucial role in integrating α -Ni(OH)₂ into the crystal lattice during the precipitation of nickel hydroxide. On comparing the FWHM of β -(001), this indicates that under the stirring action, it increases, which results in the distortion of the lattice. This is also a reason for improving the discharge capacity of the material.

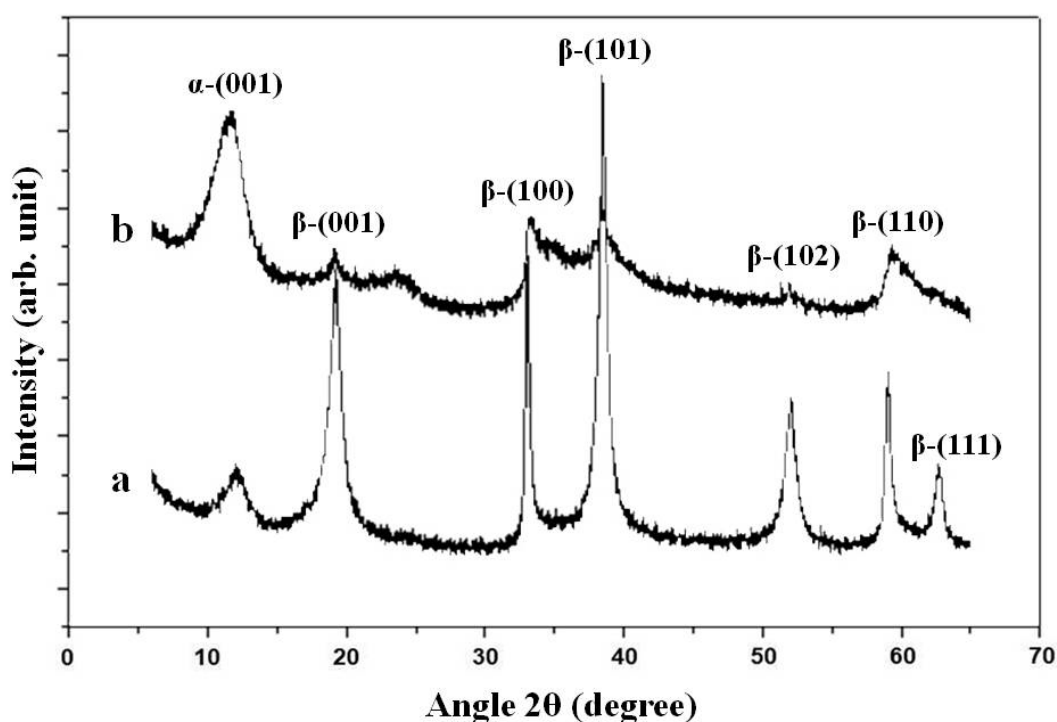


Figure 6. XRD pattern of nickel hydroxide synthesized at current density of 500 A/m² (a) without agitation and (b) with agitation in the catholyte bath.

The SEM image shown in Figure 7 indicates that the nickel hydroxide precipitated from agitation-free catholyte displays a compact and dense structure. However, the product obtained from the agitated catholyte constitutes of quasi-spherical shape particles agglomerated together (Figure 7b).

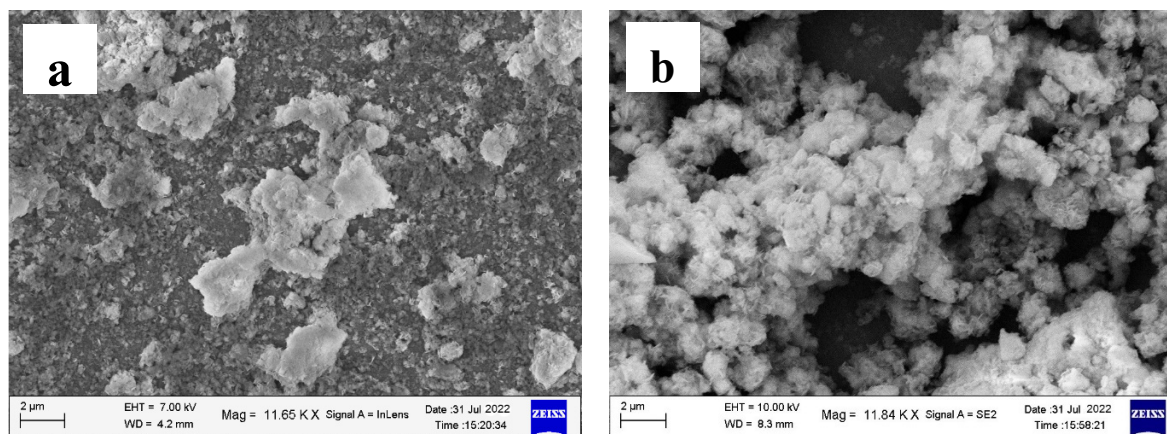


Figure 7. SEM images of nickel hydroxide generated under various conditions: (a) without agitation and (b) with agitation.

4. Conclusions

This study investigated the effects of electrolyte composition with respect to nickel concentration, applied current density, and agitation in the catholyte to determine how these factors contribute to the improvement of both the physical and electrochemical properties of nickel hydroxide intended for battery applications during its electrochemical precipitation from nitrate solutions. Current density plays a pivotal role in modulating the generation of hydroxyl ions, thereby impacting the nickel hydroxide crystal structure, which can manifest as the β -phase, α -phase, or a mixture of both. Additionally, this parameter can be used to regulate the shape and size of the particles, contributing to improved tap density and consequently, higher volumetric capacity. Increasing the discharge capacity of the electrochemically synthesized material is also facilitated by agitation of the catholyte.

Author Contributions: Conceptualization, C.K.S.; methodology, C.K.S. and R.K.P.; validation, G.L.A.; formal analysis, G.L.A.; investigation, C.K.S.; resources, T.S.; writing—original draft preparation, C.K.S.; writing—review and editing, G.L.A., R.K.P. and S.K.R.; visualization, S.K.R.; supervision, R.K.P. and S.K.R.; project administration, T.S.; funding acquisition, T.S. All authors have read and agreed to the published version of the manuscript.

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References

1. Ying, T.K.; Gao, X.P.; Hu, W.K.; Wu, F.; Noreus, D. Studies on rechargeable NiMH batteries. *Int. J. Hydrogen Energy* **2006**, *31*, 525–530. [[CrossRef](#)]
2. Geng, M.; Northwood, D.O. Development of advanced rechargeable NiMH and Ni/Zn batteries. *Int. J. Hydrogen Energy* **2003**, *28*, 633–636. [[CrossRef](#)]
3. McBreen, J. *Modern Aspects of Electrochemistry*; White, R.E., Bockris, J.O.M., Conway, B.E., Eds.; Plenum Publishing Corporation: New York, NY, USA, 1990; pp. 29–58.
4. Dong, L.H.; Chu, Y.; Sun, W.D. Controllable synthesis of nickel hydroxide and porous nickel hydroxide nanostructures with different morphologies. *Eur. J. Chem.* **2008**, *14*, 5064–5072. [[CrossRef](#)] [[PubMed](#)]
5. Dixit, M.; Jayashree, R.S.; Kamath, P.V.; Shukla, A.K.; Kumar, V.G.; Munichandraiah, N. Electrochemically impregnated aluminium stabilized α -nickel hydroxide electrodes. *Electrochem. Solid-State Lett.* **1999**, *2*, 170–171. [[CrossRef](#)]
6. Hu, W.K.; Gao, X.P.; Noreus, D.; Burchardt, T.; Nils, K.N. Evaluation of nano crystal sized α -nickel hydroxide as an electrode material for alkaline rechargeable cells. *J. Power Sources* **2006**, *160*, 704–710. [[CrossRef](#)]
7. Lu, Y.M.; Hong, S.H. Preparation of electrodes with β -nickel hydroxide/CVD-graphene/3D-nickel foam composite structures to enhance the capacitance characteristics of supercapacitors. *Materials* **2024**, *17*, 23. [[CrossRef](#)]
8. Lu, Y.M.; Hong, S.H. Controlling the cooling rate of hydrothermal synthesis to enhance the supercapacitive properties of β -nickel hydroxide electrode materials. *Materials* **2023**, *16*, 5576. [[CrossRef](#)]
9. Sen, S.; Moazzen, E.; Acuna, S.; Draxler, E.; Segre, C.U.; Timofeeva, E.V. Nickel hydroxide nanofluid cathodes with high solid loadings and low viscosity for energy storage applications. *Energies* **2022**, *15*, 4728. [[CrossRef](#)]
10. Krishnamurthy, P.; Maiyalagan, T.; Panomsuwan, G.; Jiang, Z.; Rahaman, M. Iron-doped nickel hydroxide nanosheets as efficient electrocatalysts in electrochemical water splitting. *Catalysts* **2023**, *13*, 1095. [[CrossRef](#)]
11. Reisner, D.E.; Salkind, A.J.; Strutt, P.R.; Xiao, T.D. Nanostructured active materials for advanced batteries. In Proceedings of the 14th International Primary and Secondary Batteries Technology and Application Conference, Fort Lauderdale, FL, USA, 10–13 March 1997.

12. Yang, C.C. Synthesis and characterization of active materials of Ni(OH)₂ powders. *Int. J. Hydrogen Energy* **2002**, *27*, 1071–1081. [[CrossRef](#)]
13. Acharya, R.; Subbaiah, T.; Anand, S.; Das, R.P. Effect of precipitating agents on the physicochemical and electrolytic characteristics of nickel hydroxide. *Mater. Lett.* **2003**, *57*, 3089–3095. [[CrossRef](#)]
14. Fierro, C.; Zallen, A.; Koch, J.; Fetcenko, M.A. The influence of nickel hydroxide composition and microstructure on the high temperature performance of nickel metal hydride batteries. *J. Electrochem. Soc.* **2006**, *153*, A492–A496. [[CrossRef](#)]
15. Zhang, W.G.; Jiang, W.Q.; Yu, M.; Fu, Z.Z.; Xia, W.W.; Yang, M.L. Effect of nickel hydroxide composition on the electrochemical performance of spherical Ni(OH)₂ positive materials for Ni-MH Batteries. *Int. J. Hydrogen Energy* **2009**, *34*, 473–480. [[CrossRef](#)]
16. Law, H.H.; Sapjeta, J. A Novel substrate for nickel-cadmium batteries. *J. Electrochem. Soc.* **1988**, *135*, 2418–2422. [[CrossRef](#)]
17. Jayashree, R.S.; Kamath, P.V. Nickel hydroxide electrodeposition from nickel nitrate solutions: Mechanistic studies. *J. Power Sources* **2001**, *93*, 273–278. [[CrossRef](#)]
18. Streinz, C.C.; Motupally, S.; Weidner, J.W. The effect of temperature and ethanol on the deposition of nickel hydroxide films. *J. Electrochem. Soc.* **1995**, *142*, 4051–4056. [[CrossRef](#)]
19. Murthy, M.; Nagarajan, G.S.; Weidner, J.W.; Van Zee, J.W. A model for the galvanostatic deposition of nickel hydroxide. *J. Electrochem. Soc.* **1996**, *143*, 2319–2327. [[CrossRef](#)]
20. Corrigan, D.A.; Bendert, R.M. Effect of co-precipitated metal ions on the electrochemistry of nickel hydroxide thin films: Cyclic voltammetry in 1M KOH. *J. Electrochem. Soc.* **1989**, *136*, 723–728. [[CrossRef](#)]
21. Portemer, F.; Delahaye-Vidal, A.; Figlarz, M. Characterization of active material deposited at the nickel hydroxide electrode by electrochemical impregnation. *J. Electrochem. Soc.* **1992**, *139*, 671–678. [[CrossRef](#)]
22. Krynitz, U.; Naumann, D.; Olbrich, A. Process for the Production of Pure Nickel Hydroxide and Its Use. U.S. Patent 5,391,265, 21 February 1995.
23. Coates, D.K.; Chiappetti, D. Process of Forming a Nickel Electrode. U.S. Patent 6,193,871, 27 February 2001.
24. Kong, F.; Kosteki, R.; McLarnon, F. In-situ ellipsometric study of the electroprecipitation of nickel hydroxide films. *J. Electrochem. Soc.* **1998**, *145*, 1174–1178. [[CrossRef](#)]
25. Subbaiah, T.; Mallick, S.C.; Mishra, K.G.; Sanjay, K.; Das, R.P. Electrochemical precipitation of nickel hydroxide. *J. Power Sources* **2002**, *112*, 562–569. [[CrossRef](#)]
26. Dixit, M.; Kamath, P.V.; Gopalkrishnan, J. Zinc substituted α - nickel hydroxide as an electrode material for alkaline secondary cells. *J. Electrochem. Soc.* **1999**, *146*, 79–82. [[CrossRef](#)]
27. Pralong, V.; Chabre, Y.; Delahaye, V.A.; Tarascon, J.M. Study of the contribution of cobalt additive to the behavior of the nickel oxy-hydroxide electrode by potentiodynamic techniques. *Solid State Ion.* **2002**, *147*, 73–84. [[CrossRef](#)]
28. Han, E.; Kang, H.; Wei, Z. Study on the preparation and performance of multiphase nickel hydroxide. *Chin. J. Power Sources* **2007**, *5*, 396–399.
29. Yuan, A.; Cheng, S.; Zhang, J.; Cao, C. The influence of calcium compounds on the behaviour of the nickel electrode. *J. Power Sources* **1998**, *76*, 36–40. [[CrossRef](#)]
30. Chen, J.; Bradhurst, D.H.; Dou, S.X.; Liu, H.K. Nickel hydroxide as an active material for the positive electrode in rechargeable alkaline batteries. *J. Electrochem. Soc.* **1999**, *146*, 3606–3612. [[CrossRef](#)]
31. Li, X.; Li, S.; Li, J.; Dong, H. Structure and electrochemical properties of nickel hydroxide electrodes with cobalt additives. *J. Appl. Electrochem.* **2009**, *39*, 377–381. [[CrossRef](#)]
32. Kohler, U.; Antonius, C.; Bauerlein, P. Advances in alkaline batteries. *J. Power Sources* **2004**, *127*, 45–52. [[CrossRef](#)]

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