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DETERMINATION OF THE POSSIBILITY OF OBTAINING BALLASTLESS NI(II)-NI(III) LAYERED DOUBLE HYDROXIDES AS PROMISING ACTIVE SUBSTANCES FOR SUPERCAPACITORS AND ELECTROCATALYSIS

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This study is aimed to investigate the synthesis of Ni^{2+}-Ni^{3+} LDH (layered double hydroxide) in the presence of sodium hypochlorite. Ni^{2+}-Ni^{3+} LDH is a promising active material for use in supercapacitors and electrocatalysis. The syntheses were carried out at a temperature of 60°C by coprecipitation under conditions of high supersaturation and at constant pH values (8, 10, and 12). To obtain the guest metal cation Ni^{3+} from the initial Ni^{2+} during synthesis, sodium hypochlorite was introduced as an oxidizing agent into the alkaline sodium carbonate solution. As control samples, Ni-Al-carbonate LDH were synthesized using the same methods and conditions. The formation of Ni^{3+} hydroxo compounds during synthesis was visually confirmed by a color change. The crystal structure of the samples was investigated by X-ray diffraction analysis, and the total Ni and Ni^{3+} contents were determined by trilonometric and iodometric titration. The samples synthesized in the presence of hypochlorite exhibited a black color, confirming the successful formation of Ni^{3+} hydroxo compounds. All control samples corresponded to Ni-Al LDH. The nickel hydroxide sample synthesized by coprecipitation at high supersaturation was identified as β-$Ni(OH)_2$, with a total Ni content of 59.5% and a Ni^{3+} fraction of 12.2%. The transformation of $Ni^{2+} \rightarrow Ni^{3+}$ occurred in the surface layer of the formed β-$Ni(OH)_2$ particles because the oxidation rate was lower than the hydroxide formation rate. The samples synthesized by coprecipitation at constant pH exhibited a layered structure consisting of β-$Ni(OH)_2$ and Ni^{2+}–Ni^{3+} LDH with an α-type lattice, suggesting that the oxidation rate exceeded the hydroxide formation rate. Thus, the possibility of synthesizing Ni^{2+}-Ni^{3+}-carbonate LDH was experimentally demonstrated. Under the optimal conditions (pH 8), the proportion of the Ni-Ni LDH phase reached 55.9%. The synthesized Ni^{2+}-Ni^{3+} LDH shows potential for application in supercapacitors and electrocatalysis, provided that its specific electrochemical characteristics are determined in further studies

Keywords: coprecipitation at high supersaturation, coprecipitation at constant pH, Ni^{2+}-Ni^{3+} LDH, sodium hypochlorite

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1. Introduction

Nickel hydroxides exhibit high electrochemical activity and are therefore widely used in various electrochemical devices.

Both single and double nickel hydroxides serve as active materials in nickel oxide electrodes of alkaline batteries [1] and as faradaic electrode materials in hybrid supercapacitors [2, 3]. Due to reversible transparency changes during redox processes, nickel hydroxide is also employed as an electrochromic material [4]. In addition, nickel hydroxides demonstrate high electrocatalytic [5, 6] and photocatalytic activity [7]. Ni-containing layered double hydroxides (LDHs), as well as the layered double oxides derived from them [8, 9], are applied in chemical catalysis and sensor technologies [10].

Nickel hydroxide exists in two forms [11]: β-hydroxide with the chemical formula $Ni(OH)_2$ and a brucite-type structure, and α-hydroxide commonly represented by the formula $3Ni(OH)_2 \cdot 2H_2O$ with a hydrotalcite-like structure. Previous studies [12] reported the formation of nickel hydroxide with a structure intermediate between α-$Ni(OH)_2$ and β-$Ni(OH)_2$, likely due to variations in the amount of crystallization water compared to the classical α-structure. [13] demonstrated the formation of nickel hydroxide with a layered $(\alpha + \beta)$-structure.

β-$Ni(OH)_2$ exhibits high stability during storage and cycling, which accounts for its widespread use in alkaline batteries and hybrid supercapacitors. However, α-$Ni(OH)_2$ possesses higher electrochemical activity. The pure α-modification of nickel hydroxide is metastable and, in concentrated alkaline media at elevated temperatures, transforms into the less active β-modification [14], resulting in a significant decrease in specific capacity and electrochemical characteristics. To stabilize the α-form, stabilizing additives are introduced into the nickel hydroxide structure, leading to the formation of layered double hydroxides (LDHs) [15]. LDHs consist of a host hydroxide lattice in which some of the "host" cations (Ni^{2+}) are replaced by "guest" cations, such as Al^{3+} [16]. To compensate for the excess charge in the crystal lattice, various anions are incorporated; these may stabilize the structure [17] or exhibit electrochemical activity [18].

The development and characterization of new types of layered nickel double hydroxides with enhanced activity and stability, particularly Ni-Ni LDHs, represent an important research direction for improving hybrid supercapacitors and electrocatalytic devices.

2. Literature review and problem statement

An analysis of numerous publications indicates that the most promising direction for further development is the improvement of LDHs with Ni^{2+} as the host metal cation. To stabilize the α-structure of such LDHs, Al^{3+} is most commonly introduced as the guest metal cation [19, 20]. The aluminum cation is electrochemically inert and, therefore, acts as a ballast component. However, its relatively low atomic mass is advantageous, since a lower atomic mass of the inert activator (Al) and the content of the electrochemically active component (Ni) decreases only slightly.

In addition to stabilizing the α-structure, Al^{3+} has been reported to enhance electrochemical activity, particularly by increasing the specific capacity of nickel LDHs [21, 22]. The introduction of the aluminum cation activates nickel hydroxide, and this effect is observed even for β-$Ni(OH)_2$ [23, 24]. This enhancement is most likely associated with increased polarization of the oxygen evolution reaction, which occurs as a side process during charging of nickel hydroxide. As a result, Ni-Al LDHs are used not only in supercapacitors [25], but also in electrocatalysis [26].

Nevertheless, Ni-Al LDHs possess two intrinsic drawbacks related to the presence of Al^{3+} that cannot be eliminated within this material system. First, despite its activating properties, Al^{3+} cation has a ballast nature due to the lack of electrochemical activity. Most likely, the activation is associated not with the aluminum cation itself, but with the α-structure of the LDH. It should be noted that Ni-Al LDHs, which are single-phase systems in which the aluminum cation acts as a "guest" in the crystal lattice of the host hydroxide $Ni(OH)_2$, possess high electrochemical properties. Thus, the aluminum cation acts as a structural activator. However, during inevitable LDH degradation, partial dissolution and subsequent adsorption of aluminum species onto the nickel hydroxide surface may occur. In this case, aluminum acts as a poison for $Ni(OH)_2$, leading to a sharp decrease in electrochemical activity. Such poisoning of nickel hydroxide has been experimentally demonstrated for both the monohydroxide [23] and Al-containing LDHs [27]. This toxic surface effect represents a second inherent drawback of Ni-Al LDHs that cannot be eliminated within this material system, thereby limiting further development of this approach.

One possible way to overcome the disadvantages of Ni-Al LDHs is the use of electrochemically active Ti^{4+} as a guest metal cation, forming Ni-Ti LDHs [6, 7]. Although promising, a significant drawback is the low tonnage of titanium salts and the difficulty of LDH synthesis due to the strong hydrolysis of Ti^{4+} salts.

Another drawback of nickel hydroxides relates to their intrinsic electrical properties. The electrochemical process occurring in nickel hydroxide electrodes in batteries, supercapacitors, and electrocatalytic devices is solid-state in nature and is limited by H^+ mobility and electrical conductivity. Despite their electrochemical activity, Ni^{2+} hydroxide compounds exhibit very high electrical resistance. Partial oxidation of Ni^{2+} to Ni^{3+} increases electrical conductivity of the hydroxide particles by several orders of magnitude. Consequently, nickel hydroxide-based devices typically require conditioning charge–discharge cycles before operation. To overcome this drawback, it is necessary to develop nickel hydroxides containing some amounts of Ni^{3+}. This approach is particularly promising for LDHs with Ni^{2+} as the host metal cation. In principle, Ni^{3+} can act as a guest metal cation. However, water-soluble Ni^{3+} salts do not exist, and Ni^{3+} exists as NiOOH or other solid compounds. Therefore, synthesizing Ni^{3+}-containing LDHs is only possible by oxidizing Ni^{2+} during or immediately after synthesis.

Active chlorine compounds, especially the hypochlorite anion, are promising oxidizers for Ni^{2+}. Sodium hypochlorite is used to purify nickel salt solutions from cobalt salts by oxidizing Co^{2+} to Co^{3+} (CoOOH). It has also been used to prepare LDHs [28], including those with Co^{3+} as the guest metal cation [29]. However, the Ni^{2+} oxidation by hypochlorite is a slower and more complex process. Despite this, sodium hypochlorite remains the most promising oxidizing agent for synthesizing Ni^{3+}-containing LDHs.

In [30], Ni-Al/Ni^{3+} LDHs were synthesized by treating freshly precipitated Ni-Al LDHs with sodium hypochlorite. This resulted in partial oxidation of Ni^{2+} to Ni^{3+} and exfoliation of the LDH crystal structure. However, Ni^{3+} was localized only in the surface layer of the particles, which limits the effectiveness of this approach. This approach addresses the issue of increasing the electrical conductivity of Ni-Al LDHs, resulting in a slight increase in electrochemical activity. However, the Al^{3+} ballast effect remains with this approach. Therefore, such materials are mainly promising for electrocatalysis, where reactions occur at the surface of the nickel LDH, and the ballast effect of the aluminum cation is less critical.

A series of studies has also been devoted to the synthesis of LDHs with Ni^{3+} as the guest metal cation. The oxidation of Ni^{2+} to Ni^{3+} was carried out during synthesis with sodium hypochlorite. In [31], Mg-Al/Ni^{3+} LDHs were synthesized under various conditions. In [32], Mg-Al/Ni^{3+} LDHs were obtained using a 1.5-fold excess of sodium hypochlorite, indicating the difficulty of Ni^{2+} oxidation. The same authors synthesized several LDHs with the general formula $M_6^{2+}M_{1.5}^{3+}Ni_{0.5}^{3+}$, where Mg, Cu, or Ca were used as M^{2+} (the host metal), and Al, Fe, or Co were used as M^{3+} (the guest metal) [33]. These studies are promising; however, during the synthesis, the entire amount of Ni^{2+} is oxidized to Ni^{3+}. The Ni^{3+} cation is actually a guest metal cation, and the host metal cations have no electrochemical activity. Consequently, such LDHs generally exhibit limited electrochemical performance and are unsuitable for supercapacitor and electrocatalytic applications.

It should be noted that the most promising type of LDH for overcoming the limitations of nickel LDHs related to the Al^{3+} presence of is Ni^{2+}-Ni^{3+} LDH. This type of LDH is essentially free of ballast components. However, an analysis of numerous publications has shown that there are no studies focused on the synthesis of Ni-Ni LDHs. Therefore, it can be concluded that the challenge of synthesizing Ni^{2+}-Ni^{3+} LDHs still remains unresolved.

3. The aim and objectives of the study

The study aims to determine the feasibility of synthesizing Ni-Ni LDHs from Ni^{2+} salt solutions by modifying coprecipitation methods and conducting sample syntheses to establish the conditions required for obtaining ballastless Ni^{2+}-Ni^{3+} LDHs. This will make it possible to develop a new type of electrochemically highly active LDH and subsequently establish a technology for its production.

To achieve this aim, the following objectives were defined:

– to modify coprecipitation methods through the targeted selection and application of an oxidizing agent; and to synthesize nickel hydroxide materials using the modified procedures with visual monitoring of the oxidation process leading to the formation of Ni^{3+} hydroxo compounds;

– to investigate the crystal structure of the resulting samples, determine the Ni^{2+} and Ni^{3+} contents, and, based on comprehensive analysis, conclude the feasibility or impossibility of synthesizing Ni^{2+}-Ni^{3+} LDHs.

4. Materials and methods

4. 1. The object and hypothesis of the study

The object of the study is the synthesis of Ni^{2+}-Ni^{3+} hydroxides carried out by the coprecipitation in the presence of an oxidizing agent. The research hypothesis is that under a specific coprecipitation regime (either at high supersaturation or at constant pH) and appropriate synthesis conditions (pH), it is possible to obtain Ni^{2+}-Ni^{3+} LDHs.

For this study, it was assumed that during coprecipitation, the oxidation of Ni^{2+} to Ni^{3+} proceeds faster than the nucleation of nickel hydroxide, thereby enabling the formation of LDHs. It was further assumed, as a simplification, that other synthesis parameters, in particular temperature, do not significantly influence the formation processes of nickel hydroxide materials.

4. 2. Method for the preparation of nickel hydroxide samples

Sodium carbonate and nickel nitrate hexahydrate of analytical grade were used for the synthesis. Aluminum nitrate nonahydrate of analytical grade was used to synthesize control Ni-Al LDH samples. Sodium hydroxide was used in a granulated form of a higher purity grade (analytical grade, high purity). All reagents were used without additional purification. Sodium hypochlorite was used in the form of a commercial concentrated alkaline solution containing an equimolar amount of sodium chloride. The concentration of active chlorine was determined by iodometric titration.

The theoretical target was a Ni^{2+}/Ni^{3+} LDH with a molar ratio $Ni^{2+}:Ni^{3+} = 4:1$. Based on the planned yield of synthesized material, the required amounts of nickel nitrate, sodium hydroxide, and sodium carbonate were theoretically calculated according to stoichiometry.

Two synthesis methods were used to obtain nickel hydroxide samples. The first method was coprecipitation at high supersaturation [6, 27]. In this method, an alkali solution containing sodium carbonate was added to a nickel nitrate solution at 60°C under continuous stirring. The second method involved coprecipitation at a constant pH [18], with separate feeding of the reaction solutions and direct pH monitoring in the reaction mixture, followed by manual pH adjustment. A two-stream approach was employed. Solutions of 1) nickel nitrate and 2) alkali and carbonate were supplied at a constant rate using peristaltic pumps into a reaction beaker containing 100 ml of the initial solution adjusted to the required synthesis pH.

The synthesis was carried out at 60°C with continuous stirring. After completion of the precipitation, the resulting hydroxide suspension was aged in the mother liquor for 30 minutes at the same temperature and stirring conditions to allow crystallization. The precipitate was vacuum-filtered using a Bunsen flask and Buchner funnel, dried for 24 hours at 70°C, ground, and washed with distilled water to remove water-soluble compounds. The sample was then refiltered and dried under the same conditions. Control Ni-Al carbonate LDH samples were synthesized using the same methods and conditions. In this case, the nickel–aluminum nitrate solution (Ni:Al molar ratio = 4:1) was used instead of the nickel nitrate solution. When using coprecipitation at constant pH, the following values were employed: pH = 8, 10, and 12.

Designation of control samples:

– coprecipitation at high supersaturation **NiAl-HS-T60**, where Ni and Al denote the host and guest metals, respectively; HS indicates high supersaturation (synthesis method); T60 indicates synthesis temperature (°C), pH8 indicates synthesis pH;

– coprecipitation at constant pH **NiAl-T60pH8**, where Ni and Al are the host and guest metals, respectively; T60 indicates synthesis temperature (°C), pH8 indicates synthesis pH.

To obtain Ni^{2+}–Ni^{3+} hydroxide compounds from a solution containing Ni^{2+}, the coprecipitation methods must be modified to enable in situ oxidation reaction $Ni^{2+} \rightarrow Ni^{3+}$ during synthesis. This requires the introduction of an oxidizing agent into the reaction mixture during the synthesis. The targeted selection of oxidizing agent should be based on its high standard potential and, if possible, on the ability of its residual products to decompose during synthesis or post-treatment.

4. 3. Investigation of the characteristics of nickel hydroxide samples

Ni^{2+} to Ni^{3+} oxidation during synthesis. Since NiOOH and other Ni^{3+} compounds exhibit a brown-black color, the successful progression of Ni^{2+} oxidation during synthesis was assessed visually based on the color of the obtained sample.

Crystal structure analysis. The crystal structure of the samples was studied by X-ray diffraction (XRD) using a DRON-3 diffractometer. The measurements were carried out in a specialized X-ray diffraction laboratory according to a standard procedure under the following conditions: Co-Kα radiation, 2θ range of 10–90°, and a scanning rate of 0.1°/s.

Nickel content in different oxidation states. The total nickel content was determined by trilonometric titration according to a standard method. For analysis, 50–100 mg of the sample was dissolved in 10% H_2SO_4, taken in slight excess. Since Ni^{3+} compounds are strong oxidizing agents in acidic media, they oxidize water molecules during dissolution, releasing Ni^{2+} cations into the solution; therefore, the total nickel content is determined. The resulting solution was titrated with a 0.1 N Trilon B in an ammoniacal medium in the presence of murexide until the color changed from yellow to blue. The total Ni content was calculated from the titration results. Since Ni^{3+} compounds are strong oxidizing agents, the Ni^{3+} content was determined by iodometric titration according to the standard procedure for oxidant determination. For titration, 0.1 g of the sample was dissolved in a 10% sulfuric acid. In contrast to the standard method, to prevent errors associated with the reduction of Ni^{3+} by water molecules, an excess of solid KI was added directly to the sample powder before dissolution in sulfuric acid. The mixture was kept in the dark in a closed conical flask for 5 minutes. The liberated I_2 was titrated with sodium thiosulfate solution until the solution became pale yellow. A starch solution was then added to the solution as an indicator, and the titration was continued until the violet coloration disappeared completely. The Ni^{3+} content in the sample was calculated from the titration data. The Ni^{3+}:Ni^{2+} molar ratio was calculated using the Ni^{2+} and Ni^{3+} contents. Based on the titration results, the Ni^{3+} content in the sample was calculated. Using the total nickel content and the theoretical molar masses of $Ni(OH)_2$ and Ni-Ni-CO_3 LDH, the fraction of Ni-Ni LDH in the experimental samples was estimated according to the additivity rule, assuming the nickel hydroxide sample to be a mixture of $Ni(OH)_2$ and Ni-Ni-CO_3 LDH. For correction of the calculated Ni–Ni–carbonate LDH content, the average molar mass of the NiNi-HS-T60 sample was used instead of the molar mass of $Ni(OH)_2$.

5. Results of sample preparation and characterization

5. 1. Results of modifying coprecipitation methods and synthesizing nickel hydroxide samples

To modify the coprecipitation methods at high supersaturation and constant pH, it was proposed to use a strong oxidizing agent. Sodium hypochlorite was selected for this purpose, as it is capable of oxidizing Ni^{2+} to Ni^{3+}, which is supported by literature data [31–33]. Furthermore, at the synthesis temperature of 60°C, any unreacted amount of NaOCl is expected to decompose during aging and drying stages. The volume of sodium hypochlorite solution required for oxidation was calculated stoichiometrically based on the theoretically proposed formula of Ni-Ni LDH. The calculated amount of NaOCl was added to the NaOH and Na_2CO_3 solution. Designation of experimental samples:

– coprecipitation at high supersaturation:

NiNi-NaClO-HS-T60, where Ni and Ni denote the host and guest metals, respectively; NaClO denotes the oxidizing agent; HS indicates high supersaturation (synthesis method); T60 indicates synthesis temperature (°C), and pH8 indicates synthesis pH;

– coprecipitation at constant:

pH **NiNi-NaClO-T60pH8**, where Ni and Ni denote the host and guest metals, respectively; NaClO denotes the oxidizing agent; T60 indicates synthesis temperature (°C), and pH8 indicates synthesis pH.

During the synthesis of control Ni-Al-CO_3 samples, successful formation of an emerald-colored precipitate was observed (Fig. 1, *a*). After drying, the powder retained the same color. Changes in the synthesis method and conditions had little effect on the color of the product.

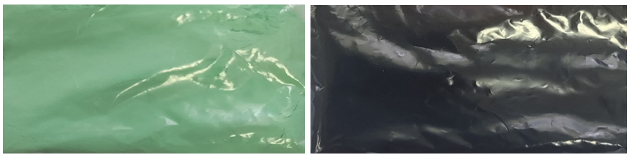


Fig. 1. Color of the resulting nickel hydroxide samples: *a* — **NiAl-HS-T60** (Ni-Al layered double hydroxide synthesized by coprecipitation at high supersaturation); *b* — **NiNi-NaClO-T60-pH8** (nickel-hydroxide sample synthesized by coprecipitation at constant pH = 8)

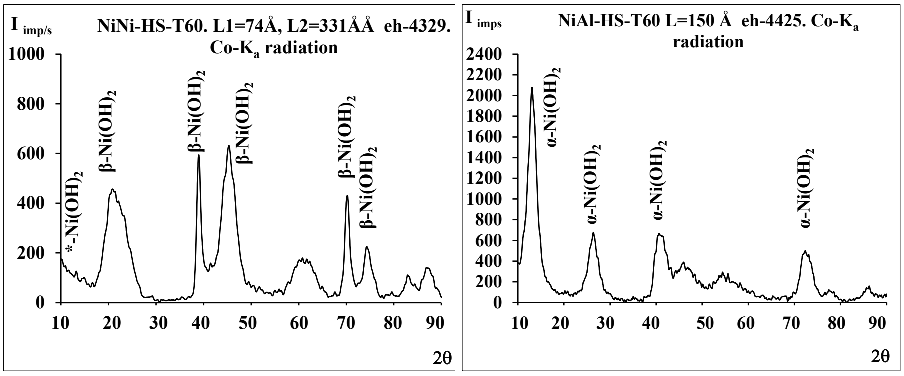
Nickel hydroxide samples synthesized from nickel sulfate in the presence of sodium hypochlorite exhibited a black color immediately after synthesis (Fig. 1, *b*), indicating successful oxidation of nickel to the trivalent state. After drying, washing, and repeated drying, the color of the product remained practically unchanged. It should be noted that the samples synthesized by coprecipitation, both at high supersaturation and at different constant pH values, exhibited nearly identical coloration.

5. 2. Results of the study of the phase structure and composition of the samples

The results of X-ray diffraction analysis of the experimental nickel hydroxide samples synthesized in the presence of NaClO and the control Ni-Al LDH samples synthesized without sodium hypochlorite are presented in Fig. 2, 3.

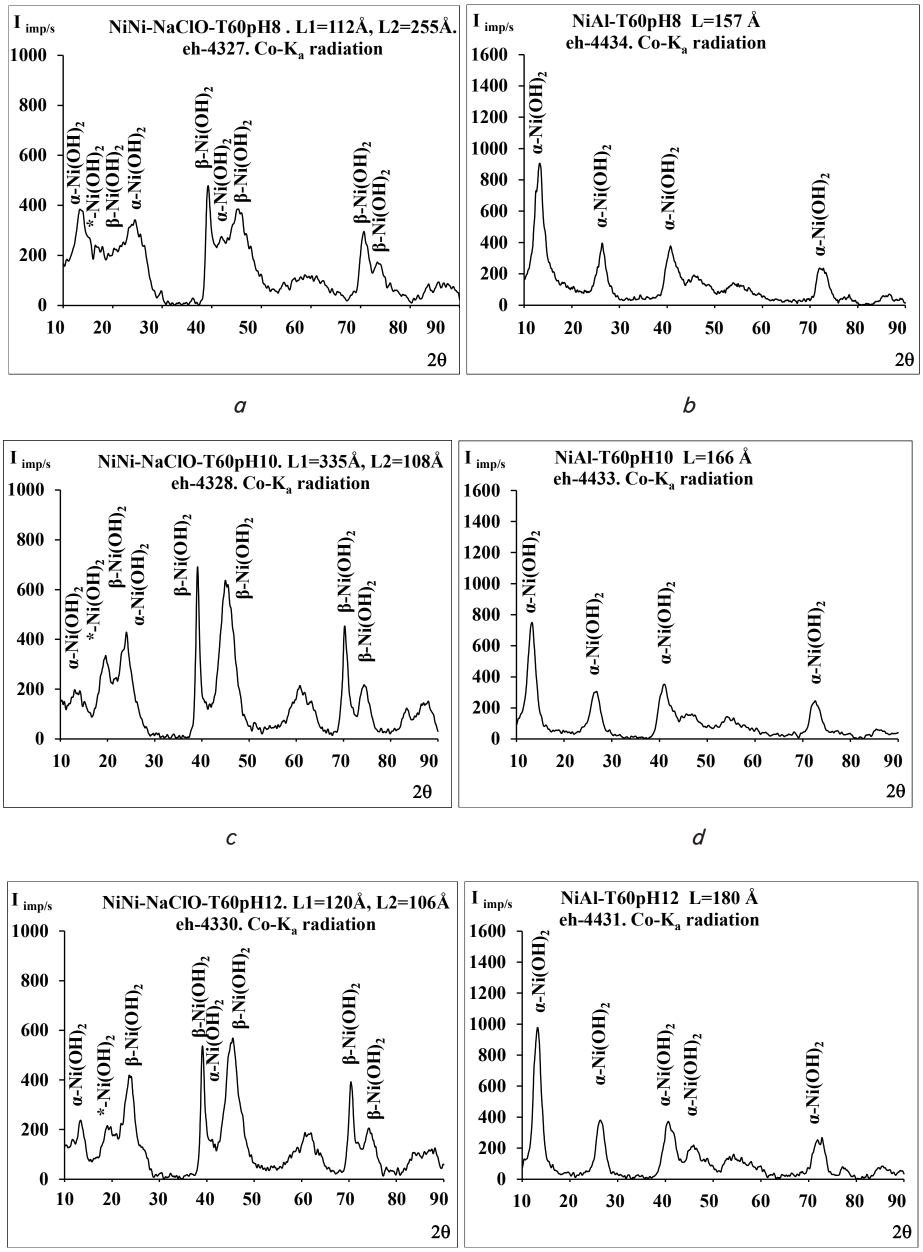
The X-ray diffraction patterns of the control samples synthesized from mixed nickel and aluminum nitrate solutions (Fig. 2, *b*, 3, *b*, *d*, *e*) exhibit only reflections corresponding to the α-structure, with a characteristic peak at $2\Theta = 13.5°$. These control samples are therefore identified as Ni-Al-carbonate LDHs with an α-$Ni(OH)_2$-type crystal lattice. Coprecipitation at high supersaturation produces Ni-Al-CO_3 LDHs with relatively high crystallinity. In this case, the characteristic peak is observed at $2\Theta \approx 13.1°$ (Fig. 2, *b*), indicating an increased interlayer spacing.

Samples synthesized by coprecipitation at constant pH values exhibit lower crystallinity. The lowest crystallinity is observed for the NiAl-T60pH10 sample synthesized at pH 10 (Fig. 3, *d*).



a *b*

Fig. 2. Diffraction patterns of samples synthesized by coprecipitation at high supersaturation: *a* — NiNi-HS-T60; *b* — NiAl-HS-T60



a *b*

c *d*

e *f*

Fig. 3. Diffraction patterns of samples synthesized by coprecipitation at a constant pH: *a* — NiNi-NaClO-T60pH8; *b* — NiAl-T60pH8; *d* — NiNi-NaClO-T60pH10; *d* — NiAl-T60pH10; *e* — NiNi-NaClO-T60pH12; *f* — NiAl-T60pH12

The diffraction patterns of the experimental nickel hydroxide samples synthesized in the presence of sodium hypochlorite are fundamentally different. The NiNi-HS-T60 sample synthesized by coprecipitation at high supersaturation showed only reflections of the β-structure with a characteristic peak at $2\Theta = 21.5°$. This sample is identified as poorly crystalline β-$Ni(OH)_2$. The diffraction patterns of the samples synthesized by coprecipitation at different constant pH values contain reflections corresponding to both α- and β-structures (Fig. 3, *a*, *c*, *d*). The detected α-structure corresponds to Ni-Ni-carbonate LDH. At the same time, the diffraction patterns contain peaks of the nickel hydroxide phase with a characteristic interplanar spacing intermediate between α-$Ni(OH)_2$ and β-$Ni(OH)_2$ (Fig. 3, *a*, *c*, *d*), this phase is designated as *-$Ni(OH)_2$. The ratio between the α- and β-structures depends strongly on the synthesis pH. The most pronounced α-$Ni(OH)_2$ peak is observed for the Ni-Ni-NaClO-T60pH8 sample (Fig. 3, *a*), where the α-phase predominates over the β-phase. The lowest amount of α-$Ni(OH)_2$ is found in the Ni-Ni-NaClO-T60pH10 sample (Fig. 3, *c*).

Fig. 3 shows the results of chemical analysis for total nickel content and Ni^{3+} content in the experimental samples.

It should be noted that the histogram (Fig. 4), in addition to the experimental and calculated values, also includes data obtained by theoretical calculation for β-$Ni(OH)_2$ (formula $Ni(OH)_2$) and Ni-Al-CO_3 LDH (formula $Ni_{0.8}Ni_{0.2}(CO_3)_{0.1}(OH)_2{\cdot}0.66H_2O$). Based on the total nickel content, the samples can be divided into two groups. The NiNi-NaClO-HS-T60 and NiNi-NaClO-T60-pH10 samples exhibit high total nickel contents (59.5 % and 58.6 %, respectively), close to the theoretical nickel content in β-$Ni(OH)_2$ (63.3 %). It should be noted that these samples are characterized by a reduced Ni^{3+} content (12.2 % for NiNi-NaClO-HS-T60 and 11.5 % for NiNi-NaClO-T60pH10). The second group of samples (NiNi-NaClO-T60pH8 and NiNi-NaClO-T60pH12) is characterized by a reduced total nickel content (55.7 % and 56.7 %, respectively) and an increased Ni^{3+} content (14.2 % and 15.5 %, respectively).

Based on the total nickel content and the molar masses of $Ni(OH)_2$ and $Ni_{0.8}Ni_{0.2}(CO_3)_{0.1}(OH)_2{\cdot}0.66H_2O$, the molar fraction of LDH in the experimental samples was calculated according to the additivity rule. The calculation results are shown in Fig. 5, 6.

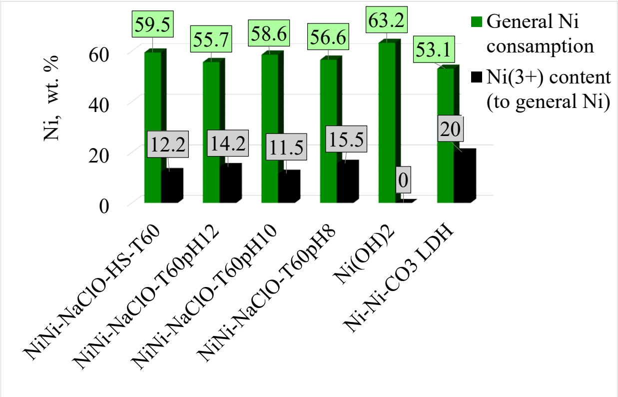


Fig. 4. Results calculated from the chemical analysis of the experimental samples for total nickel and trivalent nickel content

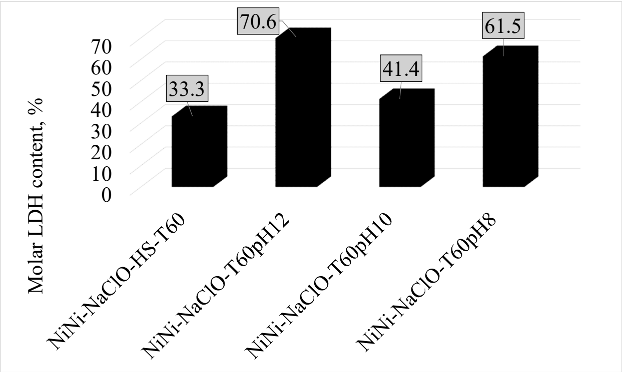


Fig. 5. Mole fraction of Ni-Ni carbonate layered double hydroxide in the experimental samples

A low fraction of Ni-Ni LDH was found for the NiNi-NaClO-HS-T60 (33.3 %) and NiNi-NaClO-T60pH10 (41.4 %) samples. The NiNi-NaClO-T60pH8 and NiNi-NaClO-T60pH12 samples contained a significantly higher LDH fraction (61.5 % and 70.6 %), which correlated with more pronounced α-structure peaks (Fig. 3, *a*, *e*).

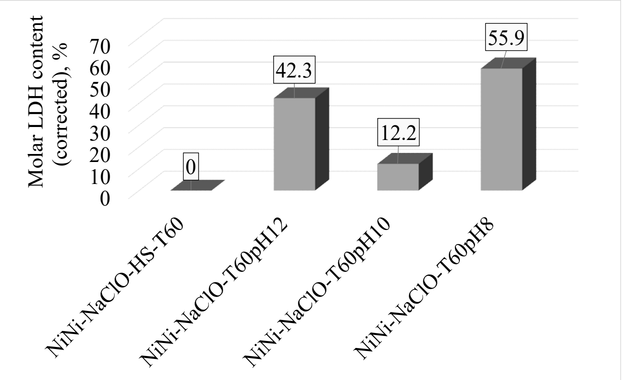


Fig. 6. Corrected molar fraction of Ni-Ni-carbonate layered double hydroxide in the experimental samples

After correction (Fig. 6), the LDH fraction in the NiNi-NaClO-T60pH8 and NiNi-NaClO-T60pH12 samples was found to be 42.3 % and 55.9 %, respectively.

6. Discussion of the results of modifying the synthesis methods, their implementation, and characterization of the properties of nickel hydroxide samples.

The discussion of the results of the attempt to obtain ballastless Ni-Ni-carbonate layered double hydroxide should be based on an understanding of the LDH formation mechanism and an assessment of the ratio between the rates of LDH formation and the oxidation of Ni^{2+} to Ni^{3+}. The chemical mechanism of LDH formation consists of two stages. In the first stage, a guest metal hydroxide is formed. In the second stage, the resulting guest metal hydroxide reacts with host metal cations, hydroxide ions, and intercalated anions to form LDH. This mechanism is quite clearly implemented for control samples, since the solution contains guest metal cations (Al^{3+}), for which the hydrate formation pH is significantly lower than that of the host metal cation (Ni^{2+}). In the case of possible formation of Ni^{2+}-Ni^{3+} LDHs, the initial solution does not contain a guest metal cation (Ni^{3+}). This cation must be formed directly in solution via oxidation by hypochlorite anions. In this case, the ratio between the oxidation rate and the hydroxide formation rate plays a significant role. If the oxidation rate is lower than the rate of nickel hydroxide formation, nickel hydroxide will form first, and the oxidizing agent will oxidize Ni^{2+} on the nickel hydroxide surface. Ni-Ni LDH can form only when the oxidation rate exceeds the hydroxide formation rate.

Visual examination of the obtained samples and evaluation of sodium hypochlorite as an oxidizing agent. As a result of synthesizing control samples of Ni-Al carbonate LDH, turquoise powders were obtained (Fig. 1, *a*), corresponding to the color of Ni-Al-carbonate LDHs synthesized in previous studies [18, 27]. It should be noted that β-$Ni(OH)_2$ is light green [13] and differs significantly in color from Ni-Al LDH. Experimental samples obtained from a nickel nitrate solution in the presence of sodium hypochlorite (as an oxidizing agent) are black. This clearly indicates the successful oxidation of nickel to the trivalent state. NiOOH (black-brown), being an oxidizing agent, is known to be unstable, especially in neutral or slightly acidic solutions. The fact that the samples retained their black color after drying, rinsing with distilled water for 24 hours, and repeated drying indicates stabilization of the Ni^{3+} hydroxo compounds and their presence within the particle structure of the resulting material.

Composition and structure of the samples. Analysis of the X-ray diffraction patterns of the control samples (Fig. 2, *b*, 3, *b*, *d*, *e*) revealed that all samples possess an α-$Ni(OH)_2$ crystal structure with a distinct peak at $2\Theta = 13.1–13.5°$, characteristic of the (100) plane. Thus, it can be concluded that Ni-Al carbonate LDHs are synthesized both by coprecipitation at high supersaturation (sample NiAl-HS-T60) and by coprecipitation at different constant pH values. The highest crystallinity is characteristic of the NiAl-HS-T60 sample under high supersaturation conditions. The X-ray diffraction patterns of the experimental samples differ significantly. The experimental NiNi-HS-T60 sample, obtained by coprecipitation at high supersaturation, shows only β-$Ni(OH)_2$ reflections in the diffraction pattern, with a characteristic (001) peak at $2\Theta = 21.7°$. It can be concluded that the NiNi-HS-T60 sample is β-$Ni(OH)_2$ and that Ni-Ni LDH was not formed under high supersaturation conditions. The observed black color of this sample indicates that, under these synthesis conditions, the oxidation rate of the nickel cation is lower than the hydroxide formation rate, and the oxidation of the nickel cation occurs on the surface of the already formed β-$Ni(OH)_2$. As a result, NiOOH is formed in the surface layers. The additional charge of Ni^{3+} is compensated by the removal of H^+ from the brucite-like layers. The relatively low crystallinity is probably due to the incorporation of carbonate anions into the β-$Ni(OH)_2$ crystal lattice, which may enhance the chemical and electrochemical properties of nickel hydroxide materials [34].

The diffraction patterns of experimental nickel hydroxide samples obtained by coprecipitation at constant pH (NiNi-T60pH8, NiNi-T60pH10, NiNi-T60pH12) contain reflections of both α-$Ni(OH)_2$ and β-$Ni(OH)_2$ (Fig. 3, *a*, *c*, *e*). The component with the α-$Ni(OH)_2$ crystal structure corresponds to Ni-Ni LDH intercalated with a carbonate anion. Under these synthesis conditions, the oxidation rate of the nickel cation is higher than the rate of LDH formation, which leads to the formation of $Ni^{2+-}Ni^{3+}$ LDH. exceeds the LDH formation rate, leading to the formation of Ni^{2+}-Ni^{3+} LDH. Most likely, these samples are single-phase materials with a layered structure consisting of alternating sets of β-$Ni(OH)_2$ layers and Ni-Ni-carbonate LDH layers with an α-type lattice, similar to the layered structures reported in [13]. The influence of synthesis pH on the phase composition of the samples should be emphasized. Synthesis at pH = 10 yields the NiNi-T60pH10 sample with the lowest content of the α-structure component (Ni-Ni LDH). Increasing the synthesis pH to 12 and decreasing it to 8 leads to an increase in the Ni-Ni LDH content (α-component), while in the NiNi-NaClO-T60pH8 sample, synthesized at pH = 8, the Ni-Ni LDH content is comparable to the β-$Ni(OH)_2$ content or even exceeds it. The observed pH effect might be attributed to experimental error. However, analysis of the influence of pH on the crystal structure of the control Ni-Al carbonate LDH samples revealed a similar trend: synthesis at pH 8 produced a sample with the lowest crystallinity. This confirms the presence of some effect that currently remains unexplained and requires further investigation.

Analysis of the total nickel content in the various experimental samples (Fig. 4) revealed that the NiNi-HS-T60 sample synthesized by coprecipitation at high supersaturation contained the highest amount – 59.5 %. This value is very close to the nickel content in β-$Ni(OH)_2$ (63.3 %). It should be noted that industrially produced nickel hydroxide used for alkaline batteries production must contain 60% nickel or more. Together with the X-ray phase analysis data, this clearly confirms that the NiNi-HS-T60 sample is β-$Ni(OH)_2$, in which 12.2 % of nickel is present as Ni^{3+}. The low Ni^{3+} content confirms the heterogeneous mechanism of nickel cation oxidation. The NiNi-NaClO-T60pH10 sample, obtained at pH = 10, also exhibits a relatively high total nickel content (58.6%) and a low percentage of trivalent nickel (11.5 %). However, its diffraction pattern (Fig. 2, *d*) contains peaks of α-$Ni(OH)_2$ corresponding to Ni-Ni LDH. This indicates that during coprecipitation at pH = 10, the oxidation rate of nickel cations slightly exceeds the hydroxide formation rate. Nevertheless, the oxidation rate of the nickel cation is insufficient to produce a significant amount of Ni-Ni LDH. Under coprecipitation conditions at pH = 12 and especially at pH = 8, the oxidation rate of the nickel cation increases, leading to the formation of a sufficient amount of Ni-Ni LDH. As a result, the total nickel content decreases, while the fraction of Ni^{3+} increases.

Based on the total nickel content and theoretical molar masses of $Ni(OH)_2$ and Ni-Ni-CO_3 LDHs, the proportion of Ni-Ni LDHs in the experimental samples was calculated (Fig. 5). The LDH content correlates with the proportion of Ni^{3+}. However, Fig. 4 reveals an obvious contradiction: for the NiNi-HS-T60 sample, the calculated fraction of Ni–Ni LDH is 33.3 %, yet such an amount of LDH should have produced α-structure peaks in the diffraction pattern; however, these peaks are completely absent (Fig. 2, *a*). Therefore, for recalculating the LDH content, the average molar mass of the NiNi-HS-T60 sample was taken as the molar mass of the β-structure. The corrected data on the content of Ni-Ni-carbonate LDH are shown in Fig. 6.

Analysis of the mole fraction of Ni-Ni-carbonate LDH (Fig. 6) shows that the maximum LDH proportion was found for the NiNi-NaClO-T60pH8 sample (55.9 %), obtained by coprecipitation at a constant pH = 8.

In summary, the possibility of synthesizing Ni^{2+}-Ni^{3+}carbonate LDH has been experimentally confirmed. This fact represents scientific novelty and has scientific significance for the synthesis and application of this new type of LDH, for which no literature data has been found. This new type of LDH, namely Ni^{2+}-Ni^{3+} carbonate, can be successfully used in power sources, particularly supercapacitors. This may lead, firstly, to the elimination of the primary processing stage in supercapacitor production and, secondly, to an increase in the capacity of these power sources. In addition, Ni^{2+}-Ni^{3+} carbonate LDH can be used for electrocatalysis, increasing the rate of wastewater purification from organic contaminants. However, a prerequisite for practical application of the new LDHs is experimental confirmation of their high electrochemical activity.

However, the resulting materials have a layered β-$Ni(OH)_2$+α-Ni-Ni LDH structure. The incomplete conversion of the required amount of nickel cations is explained by the fact that a stoichiometric amount of sodium hypochlorite was used to synthesize Ni-Ni-carbonate LDH with a Ni^{2+}:Ni^{3+} ratio of 4:1. Most likely, part of the sodium hypochlorite decomposes during the synthesis process at pH = 8, and an excess of NaOCl is required to obtain predominantly Ni–Ni LDH.

The limitation of this study lies in the use of only two synthesis methods and a limited number of pH values for coprecipitation at a constant pH. Another disadvantage is that an excess of sodium hypochlorite was not used to directly and conclusively confirm the synthesis of Ni–Ni carbonate LDH. Further development of this study should include determining the electrochemical and electrocatalytic characteristics of the resulting materials.

7. Conclusions

1. A modification of the high-supersaturation coprecipitation and constant pH coprecipitation methods (pH 8, 10, and 12) has been proposed by introducing sodium hypochlorite into an alkaline solution as an oxidizing agent. Previously unsynthesized nickel hydroxide materials have been obtained using the modified methods. Visual observation shows that the synthesized samples have a black color, confirming oxidation of the nickel cation with the formation of Ni^{3+} hydroxo compounds.

2. X-ray diffraction analysis and chemical determination of total Ni and Ni^{3+} content show that the sample synthesized by coprecipitation at high supersaturation is β-$Ni(OH)_2$ with a total Ni content of 59.5 % and a Ni^{3+} content of 12.2 %. A reasonable hypothesis has been proposed that hypochlorite oxidized Ni^{2+} via a heterogeneous reaction in the surface layer of forming β-$Ni(OH)_2$ particles due to the nickel cation oxidation rate being lower than the hydroxide formation rate.

It has been found that samples synthesized by coprecipitation at a constant pH had a layered structure consisting of β-$Ni(OH)_2$ and an α-component corresponding to Ni^{2+}-Ni^{3+} carbonate LDH. Under the most favorable conditions (at pH = 8), the nickel cation oxidation rate exceeds the hydroxide formation rate. Under these conditions, the Ni-Ni LDH fraction reaches 55.9 %. The incomplete oxidation is likely associated with partial decomposition of hypochlorite during synthesis. Therefore, the possibility of synthesizing Ni^{2+}-Ni^{3+}carbonate LDH has been experimentally demonstrated. To maximize the LDH fraction, it is recommended to use an excess of sodium hypochlorite during synthesis.

Conflict of interest

The authors declare no conflict of interest regarding this article or the published research results, including financial aspects of conducting the study, obtaining and using its results, and any non-financial personal relationships.

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Data availability

The manuscript has no associated datasets. Any additional explanations and materials can be obtained from the corresponding author upon request.

Use of artificial intelligence

The authors confirm that they did not use artificial intelligence technologies in the preparation of this work.

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