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Magnetic Properties of Nickel-Cobalt Alloys: A Systematic Review of Alloys Obtained by Electrodeposition

Propriedades Magnéticas de Ligas de Níquel-Cobalto: Uma Revisão Sistemática de Ligas Obtidas por Eletrodeposição

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The development of Nickel-Cobalt alloys has been gaining interest in recent years as they present mechanical and magnetic properties suitable for applications in electronic and magnetic devices. One of the most used methods for manufacturing these alloys is electrodeposition, as it is a cheap and easy-to-execute process, as well as the possibility of controlling operational parameters. Thus, aiming to deepen knowledge about these alloys obtained by the electrodeposition process, this work was developed through a systematic review of the literature that made it possible to evaluate the parameters applied in Ni-Co alloy electrolytic baths and the influence of these parameters on the magnetic properties. The methodology was carried out in different stages: the first, with the establishment of keywords and search strings; the second, searches in the Scopus Preview and Science Direct databases, adopting primary research articles published from 2010 to 2023. After the search, inclusion and exclusion criteria were applied, limiting the study to 10 articles. From reading the selected articles, it was possible to identify that the magnetic properties of the Ni-Co alloy depend entirely on the operational parameters, the bath's composition, and the cobalt content in the coating. Studies also indicate that the crystalline structure of the alloy, pH variations, and heat treatment can influence these properties.

Keywords: Coercivity; saturation magnetization; thin films.

1. Introduction

The magnetic properties of metal alloys play a crucial role in several industry sectors, providing a wide range of fundamental applications. In sectors such as electronics and the automotive industry, for example, these alloys are essential to produce magnetic components, such as electric motors and sensors.[1] Furthermore, in the area of information technology, hard drives use magnetic alloys to store data efficiently.[2-3] However, the development of new alloys with specific magnetic characteristics faces significant challenges. The search for more efficient, sustainable, and economically viable materials is one of the main obstacles, as it requires advances in manufacturing processes and synthesis methods. Understanding magnetic properties and the ability to control these properties at the microscopic level are challenging but crucial aspects for the successful development of new metal alloys with innovative applications.

In this sense, (Ni-Co)-based alloys emerge as prominent materials due to their remarkable magnetic characteristics, such as high magnetic permeability, low coercivity and excellent thermal stability.[4-8] These properties make Ni-Co alloys ideal candidates for applications that require high-performance magnetic materials, such as transformers, sensors, electric motors and information storage devices.[9-11]

In the production scenario of Ni-Co alloys, several techniques are used, but electrodeposition stands out as a versatile and effective method.[12-21] Electrodeposition offers significant advantages compared to traditional deposition methods such as physical vapor deposition (PVD) and chemical vapor deposition (CVD). The ability to control the thickness of the deposited layer, the uniformity of composition distribution and the ability to produce coatings on complex substrates are distinctive features of electrodeposition. Furthermore, electrodeposition is a more economical and ecologically sustainable process, reducing waste and energy consumption compared to other methods.[22-25]

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It is essential to highlight that during the manufacturing process of these alloys by electrodeposition, the phenomenon known as "Anomalous Codeposition" occurs, characterized by the preferential deposition of the less noble metal rather than the more noble metal and generally occurs in metals that are in the Iron group.[26-28] Thus, in Ni-Co alloys, cobalt (less noble metal) tends to be preferably deposited in relation to nickel metal (more noble metal), and the study of this phenomenon becomes fundamental when the objective is to control the chemical composition of the coating.

Nickel-based alloys are well known due to their properties, mainly due to their excellent corrosion resistance. Adding other elements to these coatings can generate changes in their structure and morphology, providing interesting characteristics for various applications.[19] For instance, coatings based on Nickel bonded to Cobalt have magnetic and catalytic properties, resistance to oxidation at high temperatures, high hardness, adhesion, and stability around 55 °C.[29] Specifically, materials that exhibit magnetic activity and good wear resistance, such as Ni-Co alloys, are very prominent because they are used in computer components, including disks, memory cards, drums and microelectromechanical systems.[30]

The magnetic activities observed in Ni-Co alloys are characterized as a phenomenon that occurs in materials capable of imposing a force of attraction or repulsion on other materials. The magnetic materials are characterized by parameters such as magnetic anisotropy, which is the magnetic dependence on crystallographic orientation.[31] The magnetic anisotropy of a material can be determined from the magnetization curves and it is related to the orientation of the spin magnetic moment, which depends on the crystallographic orientation of the material.[32] In addition to anisotropy, another parameter evaluated is the magnetic flux density, which represents the magnitude of the internal field inside a substance. This magnetic flux is entirely subjected to the intensity of the magnetic field.[31] Saturation magnetization (maximum possible magnetization) occurs when a ferromagnetic material is subjected to an external magnetic field, and all the magnetic moments of the material are aligned with this field. It is also possible to measure coercivity, which is defined as the intensity of the external magnetic field necessary to nullify ferromagnetic materials. In addition, there are other properties such as permeability and magnetic remanence. All these parameters depend on the material's chemical composition and the electrodeposition process variables, which can modify the structure of the deposit, grain size, internal tensions, and defects in the crystals.[30,33]

In Ni-Co alloys, the magnetic parameters are affected, mainly by the cobalt content; modification in amount of Co also causes changes in the structure and morphology of the alloy.[30] However, the composition of the bath and the deposition parameters can be controlled by electrodeposition. Consequently, high levels of adjustment of chemical composition and mechanical and magnetic properties can be achieved.[34-36] Saturation magnetization, for example, is highly dependent on the chemical composition of the alloy. Therefore, it can be adjusted and controlled based on electrodeposition parameters such as the current density applied to the deposit and the pH used in the solution.[35]

Therefore, one can notice the need to organize a systematic literature review to evaluate recent works focusing on the magnetic characterization of Ni-Co alloys obtained by electrodeposition. This review is necessary because several electrodeposition parameters can modify the alloy's magnetic properties, but no work elucidates this relationship. From this review, it will be possible to evaluate the types of electrolytic baths, substrates, temperature, pH, and current density used in the electrodeposition process and their relationship with the magnetic properties of the Ni-Co alloy.

2. Methodology

The study was carried out using a literature review in a systematic approach, based on the identification of a given subject or question. Data were collected from March to April 2023. The search was carried out on the Scopus Preview and Science Direct.

Initially, the following keywords were used: Ni-Co, Magnetic and electrodeposition, corresponding to the focus of the search, which is the magnetic characterization of Ni-Co alloys obtained by electrodeposition. Then, synonymous words and Boolean operators were inserted, thus forming the following search string: ("ni-co" OR "co-ni" OR "nico" OR "coni" OR "nickel-cobalt" OR "cobalt-nickel") AND ("electrodeposition" OR "electroplating" OR "electrochemical deposition" OR "electrodeposited" OR "electrochemically fabricated") AND ("magnetic" OR "ferromagnetic"). It is important to highlight that the Science Direct platform allows the use of up to 8 terms in each search field, so it was necessary to divide the search string using the following terms in the general search field: ("electrodeposition" OR "electroplating" OR "electrochemical deposition" OR "electrodeposited" OR "electrochemically fabricated") AND ("magnetic" OR "ferromagnetic") and the following terms in the fields of title, summary, and keywords: ("ni-co" OR "co-ni" OR "nico" OR "coni" OR "nickel-cobalt" OR "cobalt-nickel").

Furthermore, the following search filters were chosen: the period from 2010 to 2023 and research articles. Therefore, the search was limited to scientific research articles published in the last 13 years, and therefore, the following were excluded: review articles, short research, theses, books, and book chapters. After applying the search string and search filters, the data presented in Table 1 was obtained.

After carrying out the research with the search string and applying the filters, separating the articles to be studied began. From a prior reading of the titles and abstracts, works

Table 1. Search results carried out in September 2023

Database	Search String without filters	Search String with filters
Science Direct	766	508
Scopus Preview	511	213

that presented the focus of the present review were selected, and those that were not part of the theme were excluded. Next, an analytical reading of the texts to be studied was carried out, and ten articles were chosen presenting the subjects relevant to the study.

The evaluation of these articles was carried out based on 4 Research Questions (presented in Fig. 1). This procedure organizes the presentation of results so that the discussion is carried out clearly and concisely, following what was defined as the central focus of the search.

3. Results and Discussion

Based on the search strategy established in the methodology, it was possible to select 10 articles relevant to the focus of this work. They were each organized by an ID, the title of the article, the source where it was found, and the year of publication, as shown in Table 2, and from now on, they will be referenced using their ID.

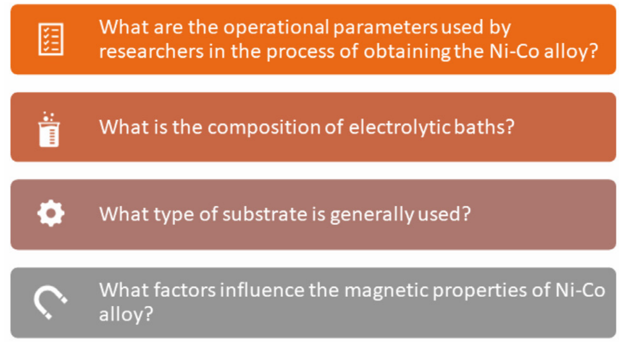

Figure 1. Research Questions

After thoroughly reading the articles presented in Table 2, the information necessary to answer the Research Questions QP1, QP2, and QP3 (listed in Figure 1) were extracted and are organized in Table 3.

The data presented in Table 3 indicate that the compositions of the electrolytic baths have similarities. Nickel sulfate and cobalt sulfate are the primary precursors. In addition, the presence of boric acid is observed in all works. In some studies, when electrolytes such as nickel chloride and cobalt chloride are used, support reagents are

Table 2. Set of articles selected for review

ID	Title	Source	Reference
A01	The influence of saccharin on the electrodeposition and properties of Co–Ni alloy thin films	Transactions of the IMF	37
A02	Role of substrate on film thickness, structural, compositional and magnetic properties of CoNi alloy thin films by low temperature electrodeposition technique	Journal of Saudi Chemical Society	29
A03	Electrodeposited Ni–Co films from electrolytes with different Co contents	Applied Surface Science	38
A04	Investigation of Structural and Magnetic Properties of Co, Ni and CoNi Alloy Thin Films by Fabricated with Electrodeposition	El-Cezeri Journal of Science and Engineering	40
A05	Morphology, structure and magnetic properties of cobalt–nickel films obtained from acidic electrolytes containing glycine	Electrochimica Acta	35
A06	Study on Electrochemical and Magnetic Properties of CoNi Alloy Coating Electrodeposited on Semiconductor Silicon	International Journal of Electrochemical Science	41
A07	The influence of pH and bath composition on the properties of Ni–Co coatings synthesized by electrodeposition	Vacuum	42
A08	The Influence of Applied Current Density on Microstructural, Magnetic, and Morphological Properties of Electrodeposited Nanocrystalline Ni–Co Thin Films	Journal of Superconductivity and Novel Magnetism	43
A09	The electrodeposition behaviors and magnetic properties of Ni–Co film	Applied Surface Science	44
A10	Correlation between crystallographic texture, microstructure and magnetic properties of pulse electrodeposited nanocrystalline Nickel–Cobalt alloys	Journal of Magnetism and Magnetic Materials	36

Table 3. Bath composition, substrate, and operational parameters

ID	Bath composition	pH	Bath temperature (°C)	Current density	Substrate
A01	Cobalt chloride, nickel chloride, potassium chloride, boric acid and saccharin	3.8	20	Defined by Cyclic Voltammetry	Si(100)/Cr/Ru
A02	Cobalt sulfate, nickel sulfate, boric acid, and ammonium chloride	5-7	40-60	5-20 mA/cm²	Stainless steel, Aluminum, Tin Oxide, Copper
A03	Nickel sulfamate, cobalt sulfate, and boric acid	2.8	24	Defined by Cyclic Voltammetry	Titanium
A04	Cobalt sulfate, nickel sulfate, nickel chloride, and boric acid	4.5	50	50 mA/cm²	Aluminum
A05	Cobalt sulfate, nickel sulfate, nickel chloride, cobalt chloride, boric acid, saccharin, glycine and, sodium lauryl sulfate	2-5	55 e 80	5 - 40 mA/cm²	Copper
A06	Cobalt sulfate, nickel sulfate, ammonium citrate, boric acid, and sodium sulfate	6	50	1.5 A/dm²	semiconductor silicon
A07	Cobalt sulfate, nickel sulfate, boric acid, and saccharin	2 – 5.4	25	2 A/dm²	Copper
A08	Nickel sulfate, cobalt sulfate, and boric acid	4.4	25	10-20 mA/cm²	Glass coated with indium tin oxide (ITO)
A09	Cobalt sulfate, nickel sulfate, boric acid, and saccharin	2.5	25	2 A/dm²	glass coated with indium tin oxide (ITO)
A10	Cobalt sulfate, nickel sulfate, nickel chloride, boric acid, and saccharin	2	65	0.2 A/cm²	Titanium

used concomitantly to improve the efficiency of the cathodic current and complexing agents. Baths that contain chlorides in their composition generally tend to produce structures with high internal tension. Baths that contain sulfates (nickel/or cobalt) generate coatings with lower internal tension and are stable at acidic pH.[30]

Electrolytic baths can also be combined with complexing agents and additives. According to Table 3, the primary additive used in electrolytic baths is boric acid, as it is a buffering agent, with the function of controlling the pH of the solution. In addition to boric acid, saccharin, glycine, sodium dodecyl sulfate, lauryl, and others can make up the bath, having the function of surfactants, and are added to improve the coating surface and relieve possible tensions.[30] Studies indicate that saccharin when introduced into baths, reduces the growth rate of crystals on the surface and can also modify their size, which occurs because the addition of saccharin promotes a more negative potential to the bath, which results in obtaining coatings with finer crystals.[19]

Article **A01** of Table 3 refers to the work of Tebbakh *et al.*,[37] which deals with the effect of adding saccharin to the Ni-Co alloy electrodeposition mechanism. The authors found that adding saccharin to the electrolytic bath modifies the deposition process and favors anomalous deposition. Consequently, the preferential deposition of cobalt was favored, generating coatings rich in cobalt. The results also indicated greater corrosion resistance, grain refinement, and low coercivity values for the coatings obtained in a saccharin bath, indicating they are suitable for magnetic applications.

The data in Table 3 also shows that different substrates and varied operational parameters were used. The temperatures were studied in the range of 20 to 80 °C, and the pH of all the baths analyzed are in the acidic pH range (2-7). It is known that the variation of these parameters has a direct influence on the chemical composition and the structure and morphology of coatings obtained by electrodeposition and, consequently, on the magnetic properties.

Article **A02** reveals a study on the role of the substrate on film thickness structural, compositional, and magnetic properties of the Co-Ni alloy obtained by electrodeposition. The results presented by Thanikaikarasan *et al.*,[29] indicated that changing the substrate modifies some properties of the alloy. All coatings obtained presented a face-centered cubic structure and smooth surface. However, the change in substrate led to deposits with varied compositions. The Co content (at.%) was between 79.42 and 85.28, while the Ni content (at.%) was between 14.72 and 20.62. Changes in magnetic properties were also observed, and the authors concluded that the copper substrate generated coatings with properties suitable for magnetic applications.

To answer question PQ4, a descriptive analysis was carried out on the articles between **A03** and **A10**, thus observing the influence of electrodeposition parameters on the magnetic properties of alloys and their structures. See below the description of the extracted information.

In article **A03**, Karpuz *et al.*,[38] show that electrodeposited coatings presented high values for saturation magnetization. The authors also observed that the increase in saturation magnetization values was due to increased cobalt content in the alloy. This behavior corroborates with the literature that indicates that magnetization is highly associated with the chemical composition of the material. Therefore, when the cobalt content is greater than the nickel content in the coating, the magnetization values will be higher, considering that the magnetic dipole moment of cobalt is greater than that of nickel. Consequently, if the coating has high levels of nickel in its composition, the saturation magnetization will be lower. According to Bakhit and Akbari[39] Ni-Co alloys with high cobalt content are used for magnetic applications, while Ni-Co alloys with high nickel content are used for applications requiring more corrosion-resistant materials. The authors also evaluated coercivity and verified that microstructural parameters, such as average grain size and crystalline structure, influenced this property. It is essential to highlight that the electrolytic bath used was composed of sulfamate-sulfate, and there are few records of this combination in the literature.

In the studies by Özdemir and Korkmaz,[40] referring to article **A04**, the Ni, Co, and Ni-Co films presented the behavior of hard ferromagnetic materials. It was also observed that the magnetic moment of the alloy is proportional to the cobalt content, corroborating with the studies by Karpuz *et al.*[38] In XRD analyses, crystalline structures of the hcp (compact hexagonal) and fcc (face-centered cubic) types were observed, with differences in crystal size and anisotropy.

A05 shows that Ergeneman *et al.*,[35] used an electrolytic bath containing glycine as a complexing agent, which is intended to stabilize acid-base baths. The experiments were carried out with a pH variation between 2 and 5, and it was possible to observe the anomalous behavior characteristic of the Ni-Co alloy. The results indicated a decrease in coercivity values as a function of increasing the pH of the solution, but in the experiment with pH 5.0, the coercivity value increased. Another critical point of the work is X-ray diffraction (XRD) analysis, as depending on the conditions for obtaining the Ni-Co alloys, they may present crystalline structures of the fcc and/or hcp type. Alloys with a hcp-type structure contain a higher cobalt content and, consequently, higher coercivity values, while alloys with mixed phases (hcp/fcc) have lower coercivity values. The authors added glycine to the bath and concluded that there were no significant changes in the coercivity and magnetization of the deposits. However, when they replaced the chloride-based reagents with sulfates, they noticed radical changes in the material's magnetic properties.

The research by YI *et al.*,[41] **A06**, deals with the effect of varying the concentration of cobalt sulfate on the thickness, roughness, morphology, microstructure, and magnetic properties of the Co-Ni coating obtained by electrodeposition. The authors reported that increasing the concentration of cobalt sulfate in the electrolyte solution from 5 to 20 g L^{-1} generates thicker coatings with higher cobalt content. Furthermore, it was possible to observe grain refinement and a reduction in the surface roughness of the Co-Ni coatings, which influenced the improvement of magnetic properties (coercivity and saturation magnetization). The authors also indicated that when using a Cobalt Sulfate concentration of 40 g L^{-1}, the coatings present a black and rough surface, and the magnetic property values are reduced, indicating that increasing the Cobalt Sulfate concentration is beneficial, but should be carried out with caution up to a specific concentration value ranging from 20 to 40 g L^{-1}.

Article **A07**, referring to the work of Tian, Xu, and Xiao,[42] shows the influence of pH on the structure, composition, morphology, and magnetic properties of the Ni-Co alloy. The authors found that increasing the pH of the solution causes an increase in the saturation magnetization of the coatings obtained. On the other hand, it generates a decrease in coercivity. Regarding chemical composition, the authors observed that increasing pH led to coatings with higher cobalt content. This behavior corroborates other studies and does not deviate from expectations, as saturation magnetization and coercivity are highly dependent on the cobalt content in the coatings. Furthermore, it was possible to identify that when the pH of the solution was in the lower range, such as 2, the coating morphology showed more imperfections than the coatings obtained at a higher pH, which explains the higher coercivity values in this pH range because the greater the number of defects, the greater the coercivity of the material.

Sarac, Baykul and Uguz[43] – **A08** – applied different current densities (-10, -15, and -20 mA cm^{-2}) to evaluate the effect of this parameter on the microstructure, chemical composition, morphology, and magnetic properties of the Ni-Co alloy. From the results, the authors observed that the nickel content in the coatings increases due to the increase in current density and that this increase causes grain refinement. Regarding magnetic properties, it was observed that the coercivity and remanence ratio decrease as the current density increases. Thus, the authors concluded that the coatings obtained with the highest current density (-20 mA cm^{-2}) showed higher nickel content, grain refinement, lower saturation magnetization, and lower coercive field, attesting that current density affects the microstructure, composition, morphology, and magnetic properties of the Ni-Co alloy.

In **A09** Tian, Xu, and Qiang[44] carried out studies separately to understand the Ni-Co alloy's behavior. Initially, the coatings were subjected to x-ray diffraction tests to observe the structure of the alloy, thus obtaining what was expected for a characteristic crystalline structure of the Ni-Co alloy without heat treatment. The samples were then subjected to heat treatment, with temperatures of 250, 350, and 450 °C, for 2 hours, hoping for increased crystallinity. After this test, the samples were characterized regarding saturation magnetization and coercivity.

The results indicated that as the temperature varies from 25 to 450 °C, the saturation magnetization increases, and the coercivity decreases. In a second step, the alloy was subjected to annealing for 2 hours at a temperature of 450 °C. It was observed that the saturation magnetization and coercivity increased after annealing the alloy and that this increase is also related to the cobalt content in the electrolyte, making it possible to reach a consensus that the saturation magnetization of the Ni-Co alloy is dependent on the cobalt content, corroborating the studies of other authors.

For Sharma *et al.*,[36] **A10**, when coatings were subjected to heat treatment, saturation magnetization increases and coercivity decreases. For the authors, the increase in magnetization was due to the reduction of grain boundaries and defects during grain growth. Based on other works in the literature, the authors explain that these defects act by fixing the movement of the material domains and that during heat treatment, heating caused a reduction in these defects, leading to a decrease in coercivity.

Sharma *et al.*,[36] also evaluated the influence of the addition of saccharin on magnetic properties. Saccharin, as already explained throughout the text, is used as an additive, and in this study, the solution was added with a variation of 1 and 2 g L^{-1}. The authors analyzed the effect of adding saccharin based on hysteresis curves. It was observed that the increase in saccharin concentration caused a reduction in saturation magnetization. The authors explained that this behavior could be related to the co-deposition of sulfur (present in saccharin). For coercivity, it was not possible to observe any significant changes. Unfortunately, the authors did not present graphs of the hysteresis curves of the coatings before adding saccharin to the bath, limiting themselves to just representing the concentration variation.

4. Conclusions

Given the study carried out based on articles published in the last 13 years on the operational parameters and composition of electrolytic baths used in the Ni-Co alloy electrodeposition process, it is possible to conclude that:

- Cobalt sulfate, nickel sulfate, and boric acid are the reagents most commonly used to obtain these coatings. It was observed that coatings obtained from sulfate baths have excellent magnetic properties. However, some authors use chloride-based baths and use additives such as glycine and saccharin to reduce internal tensions and improve the deposit's surface. Furthermore, it was found that changing the type of substrate leads to changes in the chemical composition and, consequently, in the magnetic properties of the alloy.
- The magnetic properties are entirely dependent on the chemical composition of the deposit. The saturation magnetization increases as a function of the increase in the cobalt content in the alloy.
- The variation in pH also influences the properties of coatings obtained by electrodeposition. Increasing the pH of the solution causes an increase in saturation magnetization. On the other hand, it generates a decrease in coercivity.
- Variations in current density generate changes in the chemical composition and, consequently, changes in the magnetic properties of the alloy. Changes such as higher nickel content, grain refinement, lower saturation magnetization, and lower coercive field.
- Thermal treatments modify the chemical composition and morphology of the coatings, leading to changes in the magnetic properties. Increasing heat treatment temperatures causes a reduction in defects in the crystal lattice, increasing saturation magnetization values and reducing coercivity values.

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Author contributions

Évany Silva dos Santos (Investigation; Methodology and Writing – Original Draft). Josiane Dantas Costa (Conceptualization; Methodology and Formal Analysis). Arthur Filgueira de Almeida (Investigation; Visualization and Writing – Review & Editing). Mikarla Baía de Sousa (Conceptualization; Visualization and Writing – Review & Editing). José Anderson Machado Oliveira (Visualization; Validation and Writing – Review & Editing). Heleno da Costa Neto (Investigation; Visualization and Validation). Aureliano Xavier dos Santos (Conceptualization; Methodology and Visualization). Renato Alexandre Costa de Santana (Supervision; Validation and Writing – Review & Editing). Ana Regina Nascimento Campos (Supervision; Validation and Writing – Review & Editing)

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