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RESEARCH ARTICLE

MAGNETIC PROPERTIES OF PERMALLOY FILMS DEPOSITED ELECTROCHEMICALLY BY THE TIKHONOV METHOD

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Abstract

In this paper, the magnetic properties of permalloy films prepared by the local electrochemical deposition of a chloride electrolyte, with a specified ratio of Ni/Fe = 4.26, were investigated. The dependence of magnetization on the deposition mode and composition and geometry of local deposition was analyzed during electrolyte heating or at room temperature. The dependence of magnetization was identified as being caused by the main components and additives of hydrochloric acid and ammonia. The magnetization of multielement concentrators extended the range of the magnetic field before achieving saturation. Permalloy films used to fabricate magnetic field concentrators improved the sensitivity of the magnetic semiconductor microsystems.

Keywords: permalloy, electrochemical deposition of thin films, magnetic saturation, coercive force.

Introduction

Modern magnetic semiconductor microsystems (MSMSs) comprise passive elements for linearizing the magnetic field, which considerably increases the magnetic induction in the region of a magnetosensitive transducer. These elements are called magnetic field concentrators (MFCs). As part of MSMSs, they greatly increasethe magnetic field sensitivity, which broadens the application of MSMSs.

The NiFe alloy (i.e., permalloy) is the most popular material for MFCs due to its high soft magnetic properties. An 81%-nickel (Ni) permalloy has a high maximum specific permeability of μ ~ 100,000, which assumes a high coefficient of magnetic field straightening for a low coercive force (below 1 Oe); however, these properties manifest themselves within a narrow range of change in the composition. A permalloy with a lower Ni concentration (45–60%) has lower magnetic permeability (up to 10,000), but it can be used in the case of stronger saturation fields.

Electrochemical deposition is an effective method for coating silicon substrates with permalloy. MFCs are fabricated by depositing permalloys onto local regions of the MSMSs, which are bound by a photoresist mask. At the bottoms of these regions, metal areas are preliminarily formed.

The magnetization of permalloy films has been investigated to analyze the physical properties of nanostructures in thin films or nanowires. Relatively thick (10 μm) permalloy films have been investigated [1]; specifically, the magnetic properties of permalloy arrays with different geometric sizes and shapes, which were electrically deposited from sulfate-chloride electrolytes, were analyzed, and the $Ni_{80}Fe_{20}$ alloys were shown to possess better properties than the $Ni_{50}Fe_{50}$ and $Ni_{30}Fe_{70}$ alloys.

Applied electrolytes are characterized by a wide range of molar relationships (e.g., the relationship between Ni and Fe ranges from 50 to 0.1). The one-to-one relationship between electrolyte composition and beleaguered film has not been established. The anomalous effect of the preferential deposition of Fe was utilized to eliminate the introduction of electrolytes (which have notoriously large concentrations of Ni), various additives, and complex solutions based on ionic liquids instead of water. Deposition was performed using a rotating cathode on alternating or pulsating currents in a magnetic field.

The alloy $Ni_{81}Fe_{19}$ has the best magnetic properties and a molar ratio of Ni/Fe = 4.26. Studies on the deposition process of chloride electrolytes, with a molar fraction ratio maintained at 4.26 [2-6], and received alloy protectors with component ratios have been performed. Compared to the increase in growth rate for sulfate-fluoride acid and the increase in film thickness by 2-3 times, adding muriatic acid cleans electrolytes from sludge and makes the electrolytes stable.

This paper presents the results of a study on the magnetization and coercive force produced by permalloy films via chloride electrolytes from electrochemical deposition, with a mole fraction ratio of Ni/Fe equal to 4.26 at temperatures of 70°C and 22°C, which enables us to understand the causes of abnormal deposition and ensure normal and congruent deposition.

1. Technique for producing permalloy films by electrochemical deposition

Permalloys precipitate [2] from a specific molar ratio of Ni and Fe (4.26) for the electrolyte content in 2-liter electrolytic volume containing a graphite anode. Permalloy areas were obtained with a photoresist mask on metallized layers using the Ni surface of a silicon wafer coated with a layer of SiO_2. The electrolytes were heated with a submersible heater and mixed by a magnetic stirrer. Continuous amperage was maintained in areas with a Ni cathode.

Boric acid serves as a complexion agent and increases the cathode current output. Saccharin provides the fine structure of the films. An electrolyte from a hydrochloric acid cleans up sludge and stabilizes the electrolyte. Electrolyte purification from Fe hydroxide and the stabilization of hydrochloric acid exclude abnormal codeposition and allow for the appropriate composition of the electrolyte film to be obtained, which congruent electrochemical deposition.

The proposed method for congruent deposition revealed that for the ratio of the components in the ion electrolyte $(FeCl)^+$, Fe^{2+} varied with temperature [6]. The complete ionization $(FeCl)^+$ and deposition of the electrolyte with Fe^{2+} corresponded to normal deposition, where the generally accepted notion of deposition used the NiFe alloy. The concept of the anomalous deposition of Ni in a suppressed Fe hydroxide was replaced with the concept of changing the deposition rate of Fe by one and two charged ions.

The dissociation of ferric chloride increases in weak electrolytes. The electrolyte receiving the permalloy films at room temperature, with the addition of ammonia, affirms the principle of charged Fe ions, which influences the composition of permalloy films.

The composition of films was studied using an X-ray (Philips XL 40 microanalyzer). The magnetization of the films on the plates was determined by a microsystems analyzer MESA-200. The film thickness and morphology were measured using an MSA-500.

2. Morphology of permalloy films

Figure 1 represents an image of the surface of the silicon wafer, with concentrators that are analyzed by the MSA-500 to measure the thicknesses of the transparencies. The films have a flat surface and a small number of defects, as well as a uniform thickness and good adhesion to Ni under low mechanical stresses.

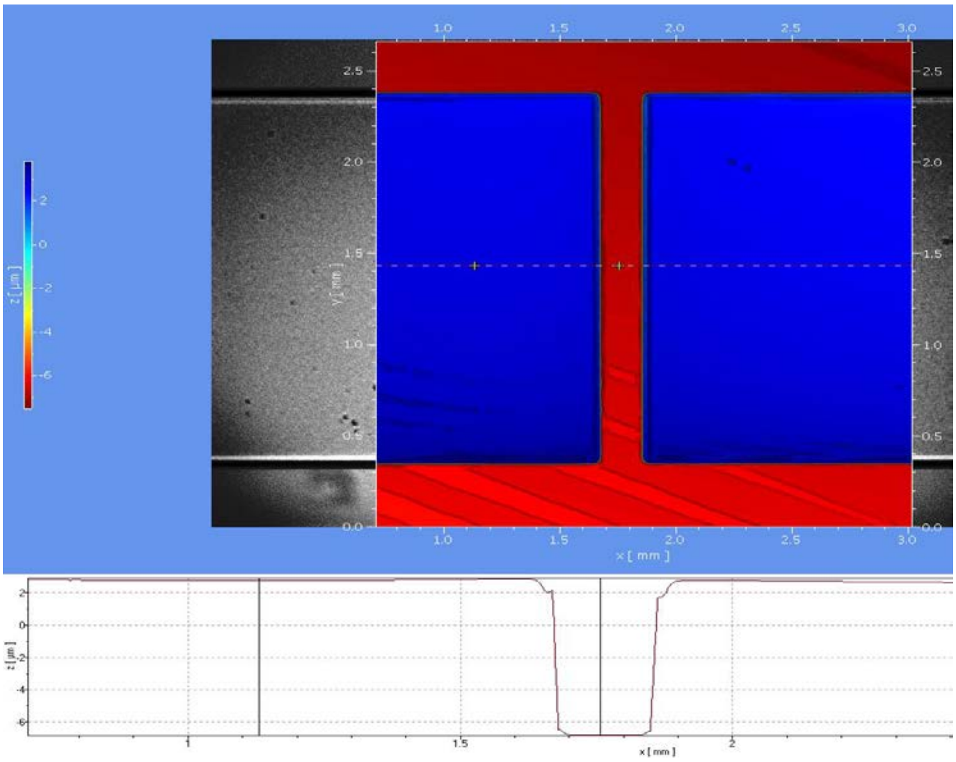

Figure 1. Micrographs of local regions of permalloy films with a thickness of 9.5 µm.

Figure 2 represents an image of the surface of the hub collected by a Philips XL 40 scanning electron microscope when measuring the compositions of the films. There is an even distribution of permalloy granules, with a size on the order of 5 nm.

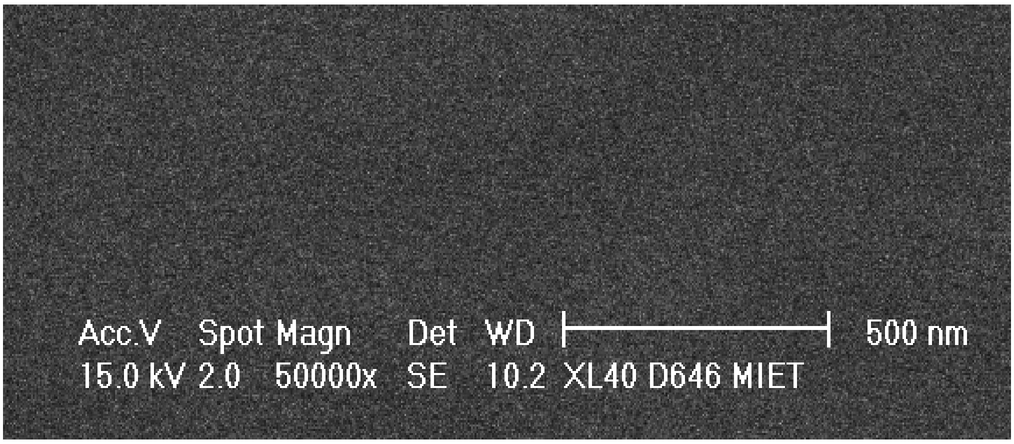

Figure 2. Scanning electron microscopeimage representing the morphology of permalloy films.

3. Dependence on the magnetic properties of permalloy film deposition

Compared with chloride in the sulfuric-chloride electrolyte, which has an atomic ratio of 4.26 in the alloy $Ni_{81}Fe_{19}$, there is an increase in the growth rate and film thickness by 2-3 times with the increase in saturation induction. The addition of the hydrochloric acid [2] electrolytes regulates pH, clears away the sludge, and stabilizes and affects magnetic properties, as shown in Figure 3. A small coercive force of less than 1 Oe is obtained within the pH range of 1.5-2.

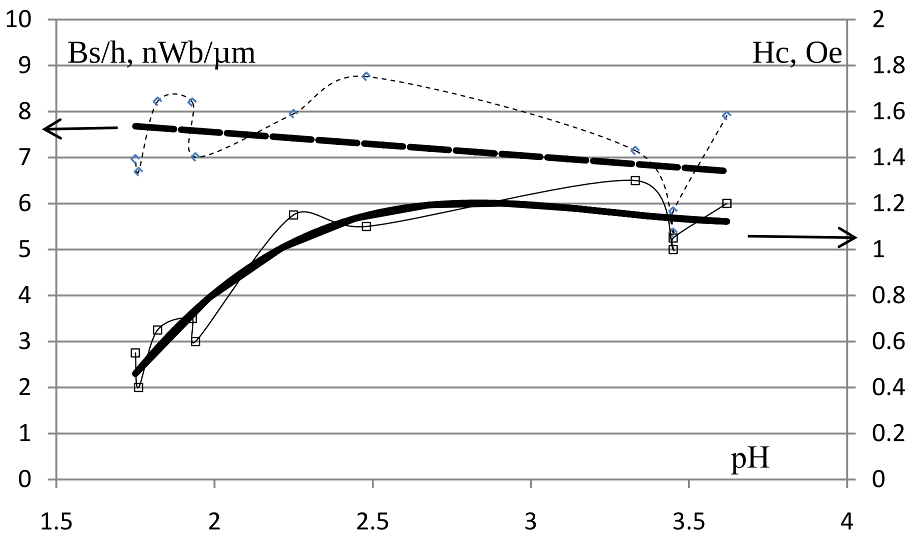

Figure 3. The dependence of the magnetization intensity and coercive force of permalloy films on the pH of electrolytes.

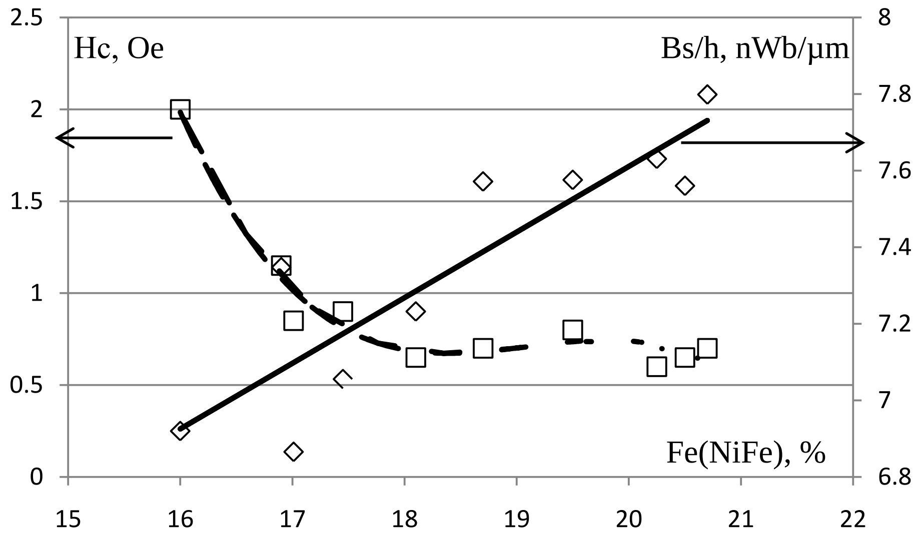

Figure 4. The dependence of the magnetization of magnetic density (Bs/h) and coercive force (Hc) on the Fe content in the film of the NiFe permalloy when the Tikhonov method is used for local electrochemical deposition.

In [4], the effect of the magnetization of permalloy films on the Fe content was studied. Figure 4 shows that a small coercive force of less than 1 Oe was obtained in the NiFe alloy when the Fe concentration ranged from 17% to 21%. Meanwhile, the specific magnetization of the films increased slightly from 7.1 to 7.8 nWb/µm.

In [2], the electrochemical composition of permalloys was shown to be strongly dependent on the temperature of the electrolyte.

The electrochemically deposited films of NiFe alloys containing 19% Fe from different electrolyte concentrations in atomic Fe are presented for the selected concentrations of Fe in the solution (Figure 5). At low concentrations of Ni chlorides, both the Fe content and deposition rate at room temperature are significantly lower than those at a higher temperature of 70°C.

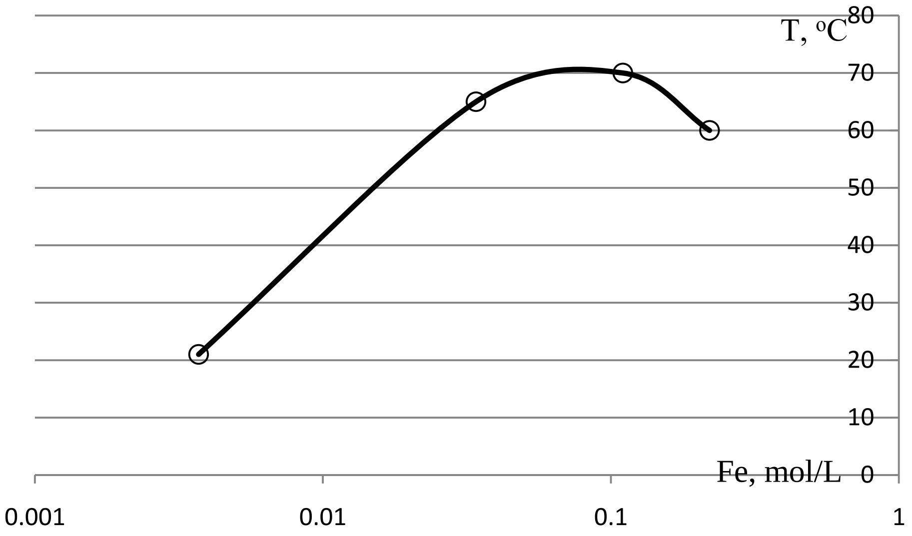

Figure 5. Electrochemically deposited films on the NiFe alloy containing 19% Fe from different electrolyte concentrations of atomic Fe: 1) 0.0037 mol/L at room temperature, 2) 0.034 mol/L at a temperature of 65°C, 3) 0.11 mol/L at a temperature of 70°C, and 4) 0.22 mol/L at a temperature of 60°C.

Making the NiFe alloy electrochemically deposit from the charges of Fe ions during the change in electrolyte temperature allows for a new approach for the collection of electrolytes. The charge of the ions in the electrolyte depends not only on temperature but also on the concentration of Fe. The reduction in concentration is well known to exacerbate the electrolytic dissociation of salts. To ascertain the nature of the phenomena occurring in the electrolyte, we study chloride electrolytes and the deposition of NiFe alloy films at room temperature.

The results of the electrochemical deposition of permalloys on the chloride electrolyte, with an Ni/Fe molar ratio of 4.26, enabled us to understand the causes of abnormal deposition to ensure normal and congruent deposition [5].

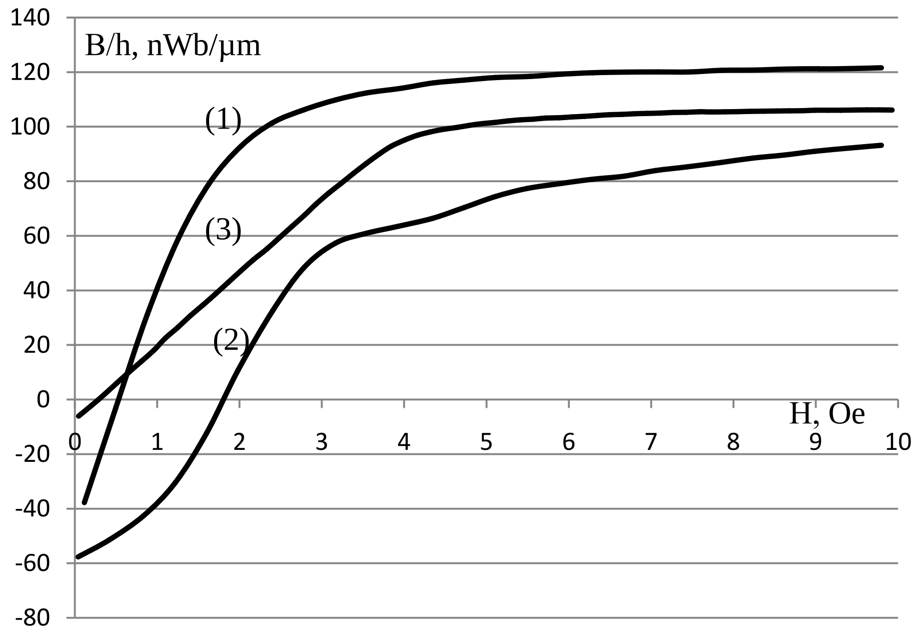

Figure 6. The dependence of the magnetization of permalloy films on plates, as shown in Table 1.

The study of the deposition of NiFe alloy films at room temperature and a comparison of their magnetic properties (Figure 6 and Table 1) among films were obtained at a temperature of 70°C. Obtaining weak solutions of permalloy films increases the electrolytic depletion of ferric chloride and affirms the principle of the influence of charged Fe ions on the composition of permalloy films. Chloride electrolytes laced with ammonia are deposited with permalloy films at room temperature and have good magnetic properties. These results reveal the possibility of obtaining nanofilms in permalloys by electrochemical deposition.

Table 1. The parameters of permalloy films.

Electrolyte composition: the ratio of the concentrations of nickel (Ni) and iron (Fe) was 4.26, the concentration of Ni, and the addition of 2 mL/L of NH_3 (ammonia) or hydrochloric acid (HCl). The cathode current density (J).The timing of the deposition process (t). Film thickness of the permalloy (h) and deposition speed (V). Fe content in the film of the permalloy. Magnetization of the films (B).Specific magnetization (B/h).Coercive force (H).

№	Electrolyte composition Ni, g/l	pH	J, mA/cm^2	t, min	h, µm	V, nm/min	Fe, %	B, nWb	B/h, nWb/µm	H, Oe
1	6,6 NH3	5,6	4,3	60	1,9	32	20,05	242	127	0,5
2	6,4	4,3	4,3	60	2,3	38	18,7	212	92	1,8
3	56 HCl	1,75	900	80	24	300	21,6	2609	109	0,4

This study shows the magnetization of permalloy films on electrolytes with a low impurity content at room temperature. Figure 6 shows that a small coercive force of less than 1 Oe and a maximum specific magnetization of the permalloy films were obtained by deposition at room temperature, as well as the addition of the electrolyte to ammonia.

4. The results of measuring MFC magnetization

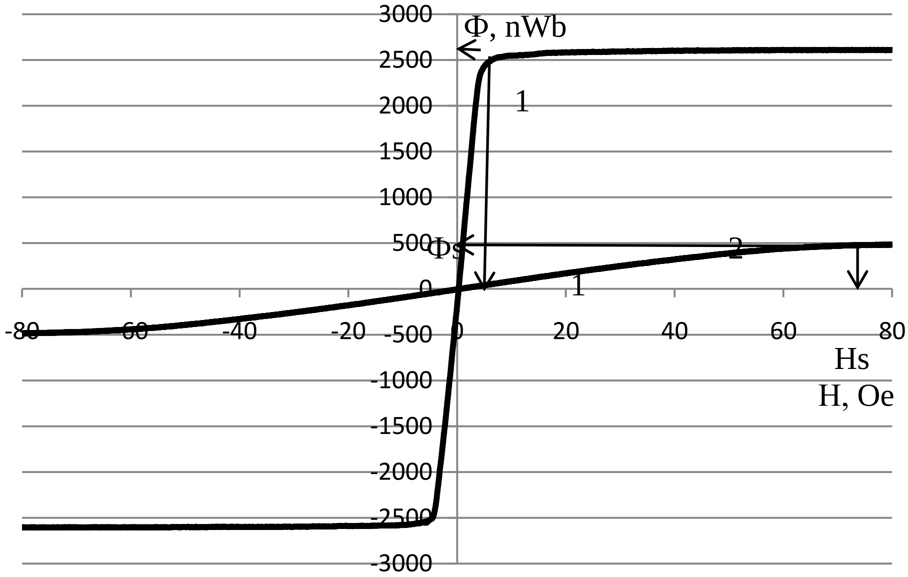

Figure 7. Magnetization flux (*Φ*) versus magnetic field strength (*H*) for the 21 μm thick permalloy films: (1) full coating of the substrate surface and (2) 43.4% coating of the substrate surface.

The experimental [7] magnetic-strength (H) dependencies of the magnetization flux Φ on the permalloy films (21 μm thick) are shown in Figure 7: (1) continuous film and (2) film with MFCs in the form of regions separated by a nonmagnetic gap.

The arrows show the saturation flux (Φs) and saturation induction (Hs). Magnetization line 1 (for the continuous permalloy film) has a small saturation field of Hs ≈ 5 Oe and a large saturation flux of Φs ≈ 2,600 nWb. The small value of Hs limits the range of the magnetic field where the magnetic flux varies, which is a disadvantage of the MFCs when applied to MSMSs.

The film regions separated by the nonmagnetic gap are characterized by magnetization line 2,with a saturation field up to Hs ≈ 75 Oe and saturation flux of $Φ_S$≈ 500 nWb. These multipart concentrators have a wider magnetic field (within which the magnetic flux varies) and a lower saturation flux.

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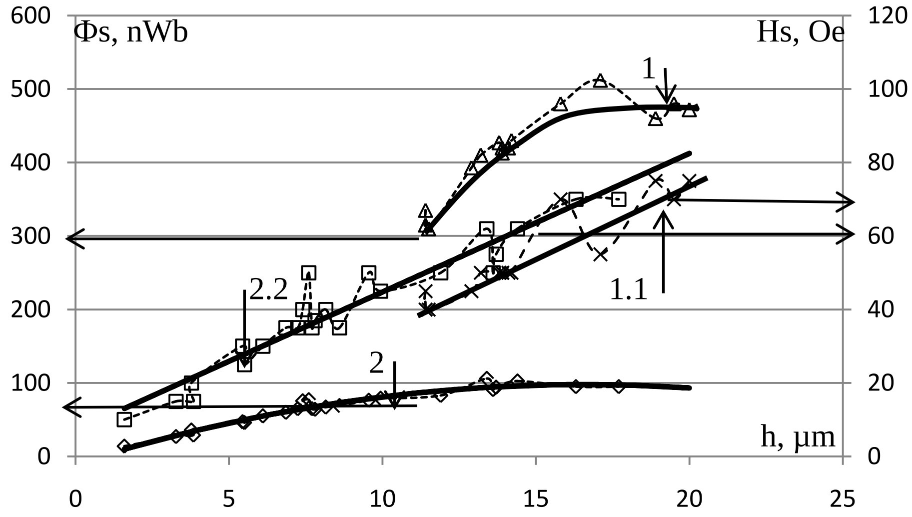

Figure 8.Saturation flux Φsand saturation induction *Hs*versus film thickness.

Figure 8 shows the experiment depending on the film thickness of the permalloy magnetic field (i.e., saturation flow, Φs, and saturation induction, Hs) with parts of the film size separated by a nonmagnetic gap, with two varying fill densities of 43.4% and 11%, respectively.

In these versions of the topology of the magnetic field, the saturation flux concentrators (Φs) (lines 1 and 2) differ 5 times, and the saturation induction (Hs) (lines 1.1 and 2.2) varies in the same way depending on the thickness of the permalloy films. Saturation induction of the magnetic field does not depend on the saturation flow, but it does depend on the thickness of the permalloy films in the h1 hub. To obtain a wide range of saturation changes in the induction magnetic field, the thickness of the transparencies in the hub should be increased. High saturation values in the magnetic field flow are obtained at the maximum filling of the area occupied by hubs with a minimal gap between them.

This specific behavior of magnetization in the case of multielement concentrators [8] allows one to extend the variation range for the magnetization of concentrators up to saturation.

Conclusion

The technological Tikhonov method for local, congruent electrochemical deposition from chloride electrolytes makes it possible to obtain uniformly thick permalloy ($Ni_{81}Fe_{19}$) films, with low mechanical stresses and high adhesion to Ni without high temperature annealing.

The film system for magnetic field generation is used to design various magnetic field sensors based on magnetoresistors, Hall sensors, and magnetotransistors. This system possesses a new quality for the increased sensitivity to magnetic induction directed parallel to the substrate surface and an extended range of sensitivity, which is due to the concentrators consisting of many elements in the permalloy films. To increase the strengthening coefficient, the MFC configuration must be selected according to the calculation results obtained from the configuration of concentrators suitable for the magnetosensitive element of a given design. During the process of electrochemical deposition, changing the composition of a permalloy film allows one to obtain an MFC magnetization curve that is more suitable for a particular MSMS [9].

A reviewed explanation of the anomalous deposition of the NiFe alloy based on the hydrolysis of ferric chloride not only explains why the phenomenon was discovered 60 years ago but also gives the solution to the problem of using films in the frequent reception of structured, normal deposition.

When selecting electrochemical concentration ratio Ni/Fe = 4.26 at chloride electrolyte and appropriate temperature provide permalloy films Ni81Fe19.

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