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OPTIMIZATION OF THE ELECTROCHEMICAL DEPOSITION METHOD FOR PREPARATION MAGNETIC/NON-MAGNETIC MULTILAYERED STRUCTURES

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Abstract. The new approach for electrochemical deposition of the magnetic/non-magnetic metal multilayered film structures was proposed. The joint complex electrolyte based on Ni^{2+}; Fe^{2+} and Cu^{2+} salts was developed. This complex electrolyte that contains compounds for magnetic (NiFe alloy) and non-magnetic (Cu) layers can be successfully used for permalloy/copper multilayered structures preparation. NiFe alloy and Cu electrodeposition kinetics was studied and optimal potential and current ranges for NiFe and Cu were estimated. The possibility of synthesizing single-layer ferromagnetic NiFe films, single-layer diamagnetic Cu films, as well as multilayer NiFe/Cu/NiFe films from a single complex electrolyte has been demonstrated. In the potential range from -0.25 to -0.4 V, a copper films without nickel and iron impurities were deposited. The Ni/Fe ratio in the alloy increases from 2.51 to 4.76 as the potential decreases from -0.92 to -0.95 V. The extra low Cu impurity in NiFe films was achieved (down to 3.2 at.%). It opens broad perspectives for application this approach for preparation of the layered film structures from one joint technological cycle. The critically low concentration of the non-magnetic impurities in magnetic layers as well as sharp interface boundaries between layers are key parameters for practical application.

Keywords: Combined electrolyte, NiFe alloy, potentiostatic electrodeposition, multilayer film.
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1 Introduction

Functional materials form the foundation of modern electronics. Therefore, obtaining these materials and studying their fundamental physical properties are crucial. Therefore, comprehensive studies of the physicochemical properties of functional materials are being conducted using modern research methods (Nazarov et al., 2024; Abiyev et al., 2026; Mamedova et al., 2025; Madatov et al., 2025; Ibrahimova et al., 2024). Studying the properties of materials allows us to identify their areas of application. Research into the effects of pressure, temperature, radiation, and electromagnetic radiation enables the development of converters suitable for use in aeronautical and space technology, as well as at nuclear centers (Azimova et al., 2020; Dashdemirov et al., 2024; Papailiou et al., 2010; Madatov et al., 2025; Abiyev et al., 2024).

Among functional materials, materials with magnetic properties occupy a special place. The electrochemical synthesis of multilayer ferromagnetic/diamagnetic structures is performed by alternate deposition of layers of an electrolyte containing ferromagnetic particles and an electrolyte containing diamagnetic particles in most studies (Seet et al., 2008). This complicates and lengthens the synthesis process, and also affects the adhesion of the layers. An alternative to this is deposition from a combined electrolyte containing both ferromagnetic and diamagnetic particles (Tench & White, 1990). For alternate deposition of ferromagnetic and diamagnetic layers, deposition is carried out at different potentials or current densities. For example, to obtain a multilayer Ni/Cu system from a combined electrolyte, copper layers are deposited at a higher potential -0.3 - -0.6 V (or at a low current density of 5-10 mA/cm^2), and nickel layers are deposited at a lower potential - 0.9 – 1.1 V (or at a high current density of 25-40 mA/cm^2) (Esmaili & Bahrololoom, 2012). This approach has the disadvantage of co-precipitating a less electronegative component (copper in the example above) with a more electronegative component (nickel in the example above). In this connection, the copper content in nickel layers can vary from 1 to 30% depending on the deposition conditions (Esmaili et al., 2011). The introduction of copper particles into the nickel lattice affects both the crystalline structure and grain structure of the nickel layers and significantly changes their magnetic properties (often worsening them, and therefore, by varying the synthesis parameters, they strive to reduce the copper content to a minimum).

For our study, we chose a multilayer NiFe/Cu film system based on alternating layers of the ferromagnetic permalloy alloy NiFe and diamagnetic layers of Cu. NiFe alloys have found application in the electronics industry, for recording and storing information in computers and other fields of technology due to their valuable magnetic properties (Kotelnikova et al., 2022). They are used as functional magnetic materials in magnetic recording devices, magnetic field sensors, devices based on the magnetic proximity effect, devices based on the spin Hall effect, electromagnetic screens, etc. (Fedkin et al., 2023; Sam et al., 2007; Horkans, 1981; Panasyuk et al., 2023).

2 Materials and methods

To study the kinetics of the deposition processes of the NiFe alloy and Cu from the combined electrolyte, three electrolytes were prepared: the combined electrolyte E-NiFeCu itself, and two additional simple electrolytes for comparison, E-Cu and E-NiFe.

The combined electrolyte E-NiFeCu designed by us has the following composition: $NiSO_4{\cdot}7H_2O$ (nickel sulfate heptahydrate) – 280 g/l, $FeSO_4{\cdot}7H_2O$ (ferrous sulfate heptahydrate) – 13 g/l, $CuSO_4{\cdot}5H_2O$ (copper sulfate pentahydrate) – 2 g/l, H_3BO_3 (boric acid) – 30 g/l, $MgSO_4{\cdot}7H_2O$ (magnesium sulfate heptahydrate) – 120 g/l, $C_7H_5NO_3S$ (saccharin) – 2 g/l.

E-Cu electrolyte has the composition: $CuSO_4{\cdot}5H_2O$ (copper sulfate pentahydrate) – 2 g/l, H_3BO_3 (boric acid) – 30 g/l, $MgSO_4{\cdot}7H_2O$ (magnesium sulfate heptahydrate) – 120 g/l, $C_7H_5NO_3S$ (saccharin) – 2 g/l.

E-NiFe electrolyte has the following composition: $NiSO_4{\cdot}7H_2O$ (nickel sulfate heptahydrate) – 280 g/l, $FeSO_4{\cdot}7H_2O$ (ferrous sulfate heptahydrate) – 13 g/l, H_3BO_3 (boric acid) – 30 g/l, $MgSO_4{\cdot}7H_2O$ (magnesium sulfate heptahydrate) – 120 g/l, $C_7H_5NO_3S$ (saccharin) – 2 g/l.

To study the kinetics of the deposition processes of the NiFe alloy and Cu from a combined electrolyte, measurements of polarization curves were performed using the electrochemical cell connected to the P-45X potentiostat-galvanostat. As a cathode, substrates made of copper foil

3×1 cm were used, treated with sandpaper (with fine grain). A silver electrode (silver-plated Pt wire) was used as an anode. A silver chloride AgCl/Ag electrode in a saturated KCl solution was used as a reference electrode.

The chemical composition of the films was determined using an AZtecLive Advanced with Ultim Max 40 energy dispersive X-ray spectrometer (EDX) (Oxford Instruments, Bognor Regis, UK). The thickness and surface microstructure of the films were studied using a Zeiss EVO 10 scanning electron microscope (SEM) (Zeiss, Oberkochen, Germany).

3 Results and discussion

The results of studying the deposition kinetics of the NiFe and Cu alloy in the linear scan mode are presented in Figure 1. According to the obtained polarization curves, during deposition from the combined electrolyte E-NiFeCu, as the potential decreases from 0 to -0.6 V, only copper ions are reduced. When the potential drops below -0.6 V, in addition to copper ions, nickel and iron ions begin to be reduced, which leads to the formation of the NiFe alloy. Then, as the potential decreases, the reduction of nickel and iron ions gradually increases up to a potential of -0.9 V, at which the reduction of nickel and iron ions increases sharply.

Based on the experimental cathode polarization curves in Figure 1 and literature data, the following optimal potential ranges for deposition of NiFe alloy layers were selected -0.915 – -0.95 V and current density -10 – -30 mA/cm^2, for Cu layers -0 .25 – -0.40 V and -0.6 – -0.8 mA/cm^2.

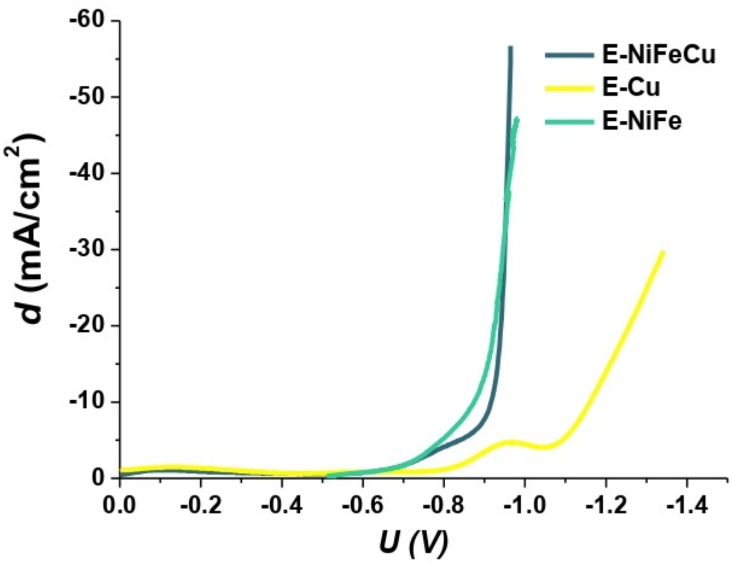

Figure 1: Polarization curves of E-NiFeCu, E-Cu and E-NiFe electrolytes.

To study the effect of synthesis conditions on the composition and growth rate of ferromagnetic NiFe and diamagnetic Cu layers, three series of samples were synthesized: a series of single-layer Cu films, a series of single-layer NiFe films and a series of multilayer NiFe/Cu/NiFe films (Table 1).

Table 1: Deposition parameters of experimental samples.

Sample	U_1, V	t_1, s	U_2, V	t_2, s	U_3, V	t_3, s
Cu-1	-	-	-0.25	300	-	-
Cu-2	-	-	-0.3	300	-	-
Cu-3	-	-	-0.35	300	-	-
Cu-4	-	-	-0.4	300	-	-
NiFe-1	-0.92	300	-	-	-	-
NiFe-2	-0.93	200	-	-	-	-
NiFe-3	-0.94	150	-	-	-	-
NiFe-4	-0.95	100	-	-	-	-
NiFe/Cu/NiFe	-0.94	150	-0.3	300	-0.94	150

For the deposition of experimental samples, a combined electrolyte E-NiFeCu with a volume of 2.5 liters was prepared. Si (100) silicon wafers with a sputtered layer of Au gold (100 nm) 1×2 cm in size were used as substrates. The sample deposition parameters are given in Table 2. The electrolyte temperature was maintained at 35 °C and pH at 2.4.

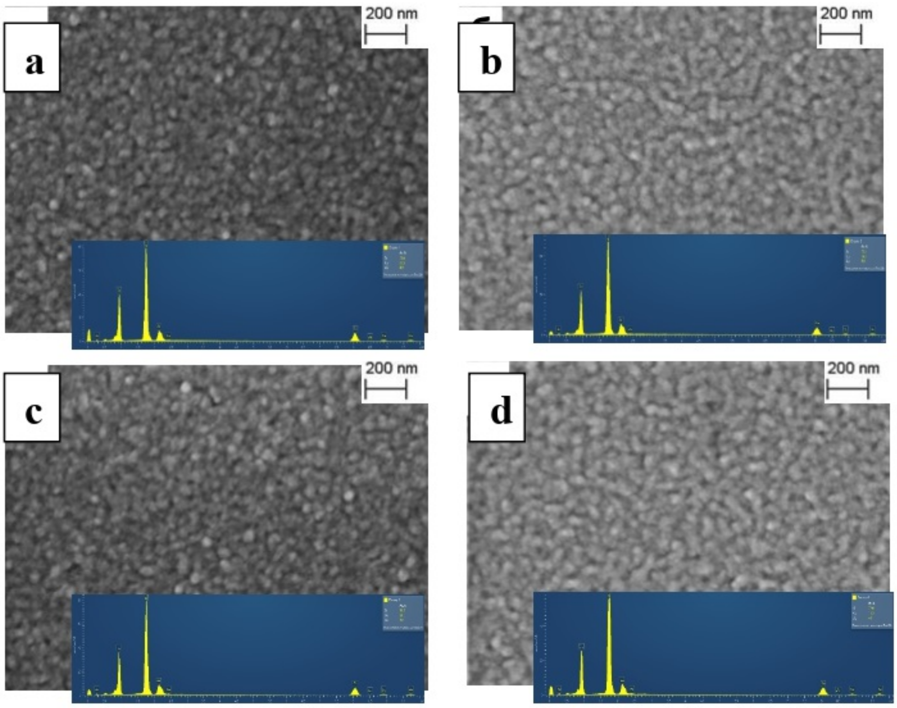

Figure 2: Microstructure of the surface and X-ray energy dispersive spectra of the Cu films: (a) Cu-1; (b) Cu-2; (c) Cu-3; (d) Cu-4.

Table 2: Characteristics of thicknesses and growth rates of samples Cu-1 – Cu-4.

Sample	U_2, V	d_{av}-, nm	d_{max}-d_{min}, nm	v, nm/min
Cu-1	-0.25	326	31	65.2
Cu-2	-0.3	332	27	66.4
Cu-3	-0.35	329	35	65.8
Cu-4	-0.4	341	34	68.2

Analysis of the chemical composition of the surface of samples Cu-1 – Cu-4 using X-ray energy dispersive spectroscopy (EDX) showed that in all cases Cu films were deposited. Due to the small thickness of copper films, the EDX spectra presented in Figure 2 also contain peaks from the silicon substrate and the conductive gold sublayer.

The thicknesses of the copper film layers in samples Cu-1 – Cu-4 are given in Table 2. According to the results obtained, the average thickness of the films changes nonlinearly when the current potential decreases from -0.25 V to -0.4 V and is 329-341 nm. In this case, the most uniform in thickness is the copper film in sample Cu-2, for which the difference between the maximum and minimum thickness is 27 nm. The film growth rate with decreasing potential also changes nonlinearly and amounts to 65.2-68.2 nm/min. Based on the results obtained, the mode in which the Cu-2 copper film was synthesized was chosen as optimal for further experiment and synthesis of multilayer structures. The choice of this mode is due to the greatest uniformity of the film thickness.

Analysis of the chemical composition of the surface of samples NiFe-1 – NiFe-4 using X-ray energy dispersive spectroscopy showed that in all cases films of the NiFe alloy with a Cu impurity were deposited (Figure 3).

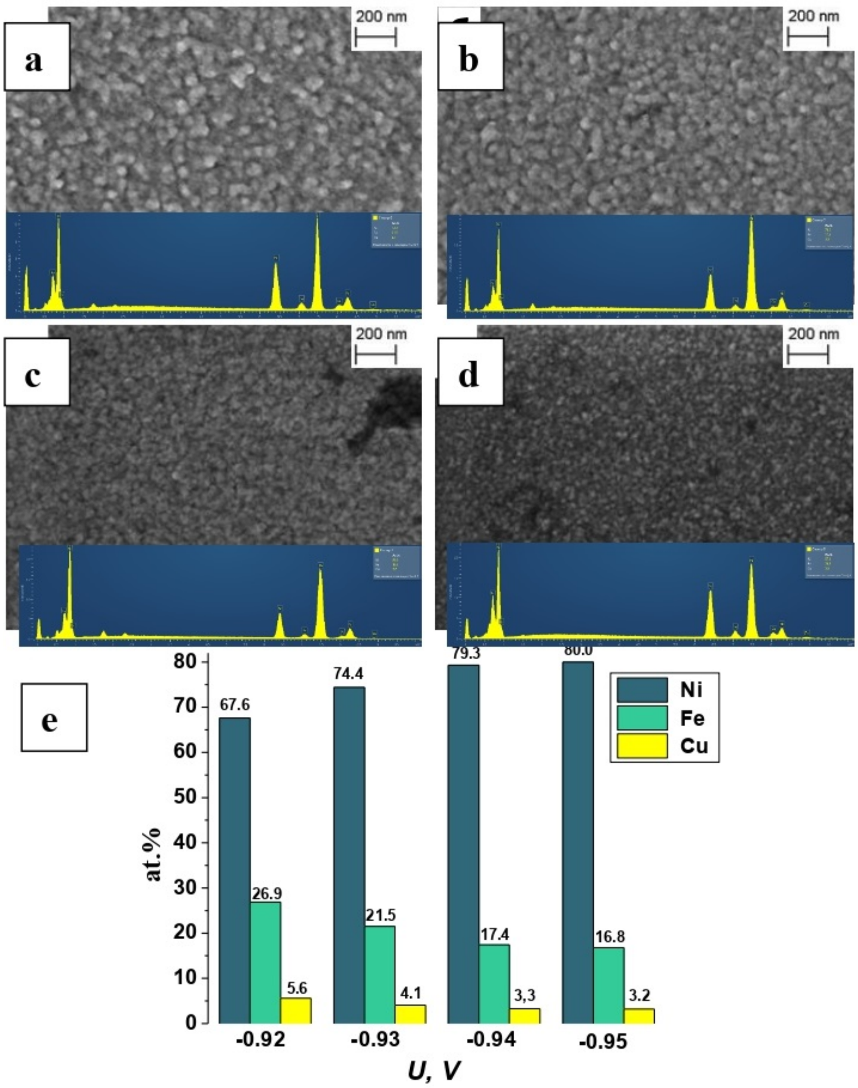

Figure 3: Microstructure of the surface and X-ray energy dispersive spectra of the NiFe films: (a) NiFe -1; (b) NiFe -2; (c) NiFe -3; (d) NiFe -4; (e) chemical composition of the NiFe films.

It was found that as the potential decreases from -0.92 to -0.95, the copper content in the alloy decreases from 5.6 to 3.2 at.%, and the iron content also decreases from 26.9 to 16.8 at.%. The nickel content, on the contrary, increases from 67.6 to 80 at.%. It is worth noting that the Ni/Fe ratio in the alloy increases from 2.51 to 4.76 as the potential decreases. These differences in the composition of the samples are mainly due to the influence of potential on the deposition of nickel. The reduction of nickel ions is an activation-limited process. A decrease in potential and an accompanying increase in current density lead to an increase in the cathodic overvoltage on the surface of the growing film, which promotes the reduction of nickel. At the same time, the reduction of iron and copper ions are diffusion-limited processes and a decrease in potential does not contribute to their acceleration.

Based on the composition results, the mode in which the NiFe-3 and NiFe-4 films were synthesized were chosen as optimal. The choice of these modes is due to low copper content in it and Ni/Fe composition ratio close to 80/20 (which is known to have best magnetic properties). However, NiFe-3 film has lover thickness nonuniformity, than NiFe-4 film. Thereby NiFe-3 film electrodeposition mode was chosen for further experiment and synthesis of multilayer structures.

Table 3: Characteristics of thicknesses and growth rates of samples NiFe -1 – NiFe -4.

Sample	U_2, V	d_{av}-, nm	d_{max}-d_{min}, nm	v, nm/min
NiFe-1	-0.92	1635	60	327
NiFe-2	-0.93	1659	61	498
NiFe-3	-0.94	1671	65	668
NiFe-4	-0.95	1753	73	1052

The deposited multilayer film NiFe/Cu/NiFe sample cross-section was examined by scanning electron microscopy, and elements distribution maps and elements distribution profiles were obtained by X-ray energy dispersive diffraction. During the cross-section preparation Au/NiFe/Cu/NiFe film was separated from the Si/Au substrate and thus free film was examined. The elements distribution maps and the elements distribution profiles shown in Figure 4 confirm the synthesis of multilayer films with alternating layers of NiFe and Cu alloy.

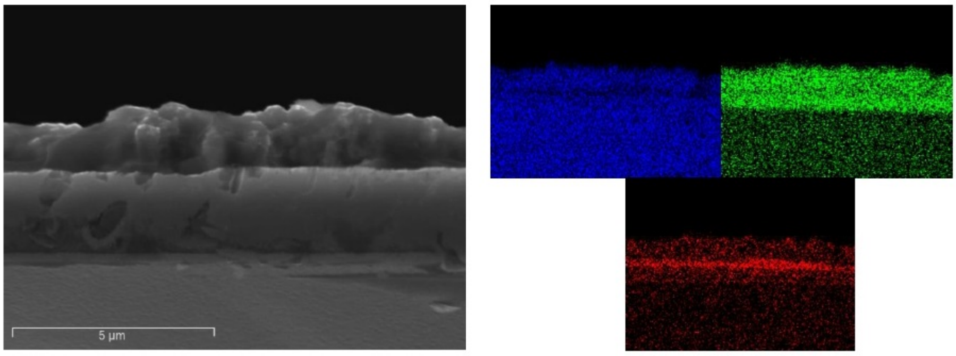

Figure 4: Maps of the elements distribution in Au/NiFe/Cu/NiFe free film (Ni – blue, Fe – green, Cu – red).

4 Conclusions

A combined electrolyte has been developed for the electrochemical deposition of multilayer ferromagnetic/diamagnetic (NiFe/Cu) systems. The kinetics of electrochemical deposition of an alloy of NiFe and Cu has been studied. It has been established that in the potential range from -0.25 to -0.4 V, a copper film without nickel and iron impurities is deposited. It is shown that as the potential decreases, the film growth rate increases from 65.2 to 68.2 nm/min, and the most uniform film thickness is formed at -0.3 V. It was found that in the potential range from -0.92 to -0.95 V, a NiFe alloy film is deposited. As the potential decreases, the copper content in the alloy decreases from 5.6 to 3.2 at.%, and the iron content also decreases from 26.9 to 16.8 at.%. The nickel content increases from 67.6 to 80 at.%. It is worth noting that the Ni/Fe ratio in the alloy increases from 2.51 to 4.76 as the potential decreases. The heterogeneity in film thickness with decreasing potential also increases from 60 to 73 nm. The most uniform film thickness is formed at a potential of -0.92 V. It opens broad perspectives for application this approach for preparation of the layered film structures from one joint technological cycle. The critically low concentration of the non-magnetic impurities in magnetic layers as well as sharp interface boundaries between layers are key parameters for practical application such technology for GMR devices.

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