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**Research Article**

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# Synthesis and Magnetic Properties of Composites Based on $CoFe_2O_4$ and Polyvinylidene Fluoride in an Alternating Magnetic Field

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**Abstract** $CoFe_2O_4$ cobalt ferrite nanoparticles, synthesized by the method of combustion of malonic acid dihydrazides of iron and cobalt, were used as dispersed filler in the preparation of thin-film composites based on polyvinylidene fluoride (PVDF) composites. It was established that as the intensity of an alternating magnetic field increases, the magnetic permeability increases monotonically, reaching a maximum value at a strength of 18 kA/m. However, with an increase in the thickness of the composite, magnetization and permeability values decrease significantly, which is associated with a reorientation of the domain structure.

**Keywords** nanoparticles, composites,$CoFe_2O_4$, thin-film,composite, PVDF

## Introduction

Nanocomposites based on polymers containing magnetic nanoparticles have a number of interesting magnetic and electrophysical properties, both from fundamental and applied points of view. Composite nanomaterials, whose functional properties (electrical, magnetic and electromagnetic) can be easily controlled by changing the composition, size, and concentration of nanoparticles in a polymer matrix can find application in the development of new generation microwave equipment and modernization of a wide range of radioelectronic devices: attenuators, equivalent loads, mode filters and harmonics of the radio signal, to ensure effective protection of bioobjects from electromagnetic radiation, as well as solve the problems of noise immunity, electromagnetic compatibility and the formation of adaptive characteristics of radio electronic equipment [3,5]. Therefore, cobalt ferrite acquires close attention of researchers [1,4,6,8,9].

The aim of the present work is to study the influence of the size effect on the magnetic characteristics of a composite based on a $CoFe_2O_4$ nanoparticle polymer.

## Experimental part

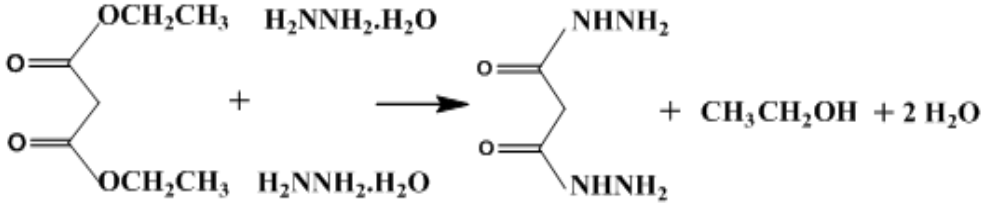
Synthesis of nanosized crystals on $CoFe_2O_4$ bases was carried out by burning cobalt and iron malonatedihydrazide according to the procedure described in [1].

Initial reagents for the synthesis were as follows:

- ferricnitratehexahydrate (III) - $Fe(NO_3)_3 \cdot 6H_2O$;
- cobaltnitratehexahydrate (II) - $Co(NO_3)_2 \cdot 6H_2O$;
- diethylmalonate- $C_7H_{12}O_4$;

- hydrazine hydrate - $NH_2NH_2 \cdot H_2O$;
- absolute alcohol- $C_2H_5OH$;
- nitric acid ($HNO_3$) with a concentration of 65%.

Monohydric acid dihydrazide was prepared by intensive mixing of diethyl malonate with hydrazine hydrate in molar ratios of 1: 2. The process proceeds according to the following chemical reaction:




To carry out the process, 25.8 ml (25.04 g) of hydrazine monohydrate were poured into a 1000 ml round-bottomed flask, then 42.3 ml (40.05 g) of diethyl malonate was added dropwise and the contents were diluted with 350 ml of absolute alcohol. The mixture was boiled in a water bath with stirring for 5 hours in a flask under reflux. White crystals that precipitated, have been filtered and dried. The yield is 23.5 g, that is 70% of the theoretical value.

Metal complexes are obtained by the interaction of malonic acid dihydrazide with the soluble salts of the corresponding metals. The best results for iron and cobalt dihydrazides are obtained using nitrate salts of these elements. In the next step, aqueous solutions of nitric acid salts of cobalt and iron with a concentration of 0.1 M were prepared. Then, precursor solutions were prepared by mixing the calculated amounts of solutions of nitric acid salts of cobalt and iron with an appropriate amount of malonic acid dihydrazide. Then they were slowly heated in heat-resistant glasses in one case to 60° C, in another to 300° C. Holding the solution at 60° C for 1 hour produces a solid substrate, and when heated to 250-300°C, already solid precursor burns. The resulting combustion products were further washed and, after pressing, annealed at high temperatures. Samples that were treated thermally at 700° C and above had crystalline structures of spinel type.

Polymer composites were prepared by simple mixing of $CoFe_2O_4$ nanoparticles (with dimensions 35-45 nm) with powders of polyvinylidene fluoride (PDVF) in a weight ratio of 30:70 and 50:50, followed by hot pressing. On the opposite surfaces, the resulting films are printed with silver paste. The measurement of the magnetic parameters is carried out according to the work methodology [7].

**Discussion of Experimental Results**

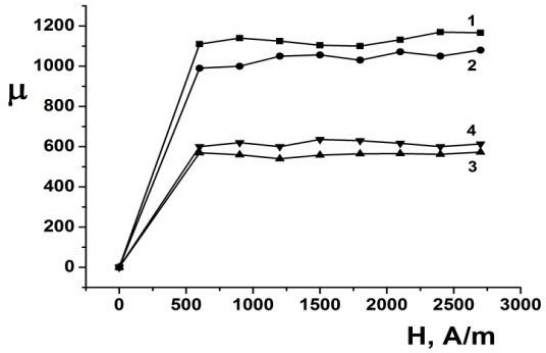



*Figure 1: The dependence of the magnetic permeability on the strength of an alternating magnetic field: 1-30% (s) + 70% (PVDF), 2-50% (s) + 50% (PVDF), (h=180 μm, 3- 30% (s) + 70% (PVDF), 4- 50% (s.) + 50% (PVDF), (h=360 μm)*

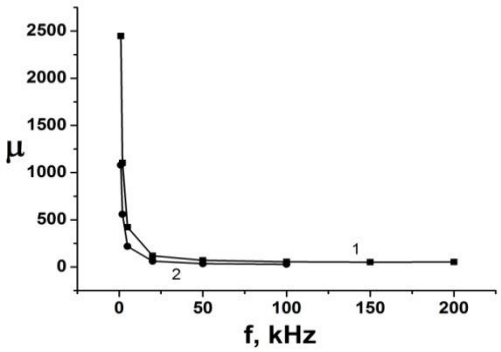



*Figure 2: The dependence of the magnetic permeability on the frequency of the alternating field: 1- 30% (s) + 70% (PVDF), (h=180 μm); 2- 30% (s) + 70% (PVDF), (h=360 μm); H=300, А/м*

All the composites studied according to the magnetic characteristics, namely, the magnetic permeability, the magnetization as a function of the intensity and frequency of an alternating field at room temperature.

The results of the studies are shown in Fig. 1-4.

From the analysis of the results obtained, the following conclusions can be drawn:

- Regardless of the percentage content of components with increasing alternating-field strength (at f = 2 kHz), the magnetic permeability increases, reaches a maximum value, and then tends to saturation with increasing field. In this case, the maximum value of the magnetic permeability on samples with a thickness of 180 μm is almost 1.6 times larger than with a thickness of 360 μm;
- The frequency dependence of the magnetic permeability, regardless of the percentage content of the components, as well as, the thickness of the samples, sharply decreases to a minimum, and then changes monotonically with increasing purity;

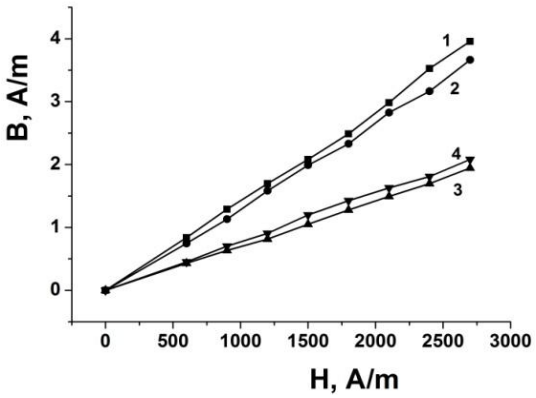



*Figure 3: The dependence of magnetic induction on the strength of an alternating magnetic field: 1-30% (s) + 70% (PVDF); 2-50% (s) + 50% (PVDF), (h=180 μm); 3- 30% (s) + 70% (PVDF); 4-50% (s + 50% (PVDF), (h=360 μm)*

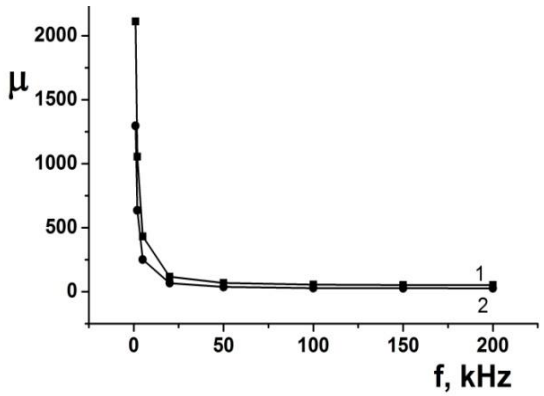



*Figure 4: The dependence of the magnetic magnetic permeability on the frequency of the alternating magnetic field: 1-50% (s) + 50% (PVDF), (h=180 μm), 2- 50% (s) + 50% (PVDF), (h=360 μm). H=300, А/м*

- As it can be seen from the figure, regardless of the percentage of components, the magnetization increases linearly with increasing alternating-field strength (at f = 2 kHz) and does not reach technical saturation. In this case, the magnetization on samples with a thickness of 180 μm is about 1.5 times larger than for samples of 360 μm. We note that such a linear dependence of magnetization is a characteristic of magnetodielectrics, which are widely used as cores in oscillatory circuits of electro-radio engineering. Proceeding from this, itcan be concluded that the composites obtained can also be used as cores in oscillatory circuits operating at different frequencies.
- From the analysis of the obtained results it follows that in the investigated composites a dimensional effect takes place.

The observed growth of the magnetization under the action of an external alternating field is due to two main processes: a) displacement of the domain walls, b) rotation of the magnetic moments of the domains. According to [2], the initial section of the magnetization curve corresponds to an elastic displacement of the domain walls. This increases the volume of those domains whose magnetic moments form the smallest angle with the direction of the external field, and conversely, the sizes of domains with an unfavorable orientation of the spontaneous magnetization vector decrease. After removing the weak field, the domain walls return to their previous positions; the residual magnetization in the sample does not arise.

With a further increase in the magnitude of the alternating field, the displacement of the domain walls has an irreversible spasmodic character. In this case, the magnetization curve has the greatest steepness in the region of irreversible displacement of domain walls. With further increase in the magnetic field strength, the role of the second magnetization mechanism, namely, the rotation mechanism, in which the magnetic moments of domains

from the direction of easy magnetization forming a small angle with the field, gradually turn in the direction of the field, i.e., in the direction of more difficult magnetization. The reason for the observed decrease in the magnetic permeability from the frequency, along with other factors, is the inertness of the domain walls at high frequencies, which is confirmed by the experimental results (Fig. 1-4).

In our opinion, the reason for the size effect is mainly the following factor: In the synthesis of composites, magnetic particles are most likely distributed chaotically, because of which free poles appear at their ends when magnetized. This creates an internal magnetic field directed against the external field. It should be emphasized that the presence of an internal field in the composite can also cause the observed linear dependence of the magnetization.

Note that the reason for the decrease in magnetization and magnetic permeability can also be magnetic losses. The main magnetic losses are loss of hysteresis and eddy current. The fact is that domains grow due to the displacement of boundaries, in turn, the substance of the sample is resisting this process. In addition, impurities, internal stresses, and also crystalline anisotropy, usually prevent the displacement of domain walls and to overcome these resistance forces it is necessary to spend additional energy.

## Conclusion

The influence of the size of magnetic particles on the magnetization, magnetic permeability and coercive force was experimentally determined. It is revealed that in the investigated composites, the magnetization and magnetic permeability values decrease with increasing sample thickness. It is established that with increasing alternating-field strength, regardless of the percentage ratio of the components, the magnetization increases, and the magnetic permeability reaches a maximum value and goes further to saturation with a further increase in the field. It is shown that the reason for the decrease in the values of the parameters studied is the internal magnetic field directed against the external field, which is created by the composite poles of randomly distributed magnetic particles.

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