A comparative analysis of magnetic properties and microstructure of high coercivity Sm(CoCuFe)$_5$ quasi-binary alloys in the framework of fractal geometry

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Abstract. The results of the micro- and nanostructures investigation of Sm(CoCuFe)$_5$ alloys by means of scanning electron and atomic force microscopy are presented. It was shown that sequential high- and low-temperature heat treatments lead to the formation of a homogeneous microstructure with nanoscale compositional heterogeneities. Such a structure provides a coercive filed of up to 32 kOe. The coercivity and remanent magnetization of the samples in the temperature range from 300 to 700 K linearly decrease. The predominant coercivity mechanism in these materials is pinning on nanoscale inhomogeneities with an increased copper concentration. The fractal dimension of the surface of the Sm(CoCuFe)$_5$ alloy metallographic specimen was determined at different stages of heat treatments. The comparative analysis of magnetic properties and microstructure in the framework of fractal geometry was carried out.

1. Introduction
Despite the significant results achieved in the formation of a highly coercive state of Sm – Co – Fe – Cu – Zr alloys, its reversal mechanism may be explained in many ways in publications of various authors [1–4]. These materials are characterized by a regular microstructure with nanoscale heterogeneous, which is formed during heat treatments. At the same time, the main structural components have a hexagonal crystal structure and stoichiometric ratios of 1:5 and 2:17. According to [5], these alloys with a prevalent content of the 1: 5 structural component have increased temperature stability.

In this work, the Sm(CoCuFe)$_5$ alloy, considered as a model of one of the structural components, uses as objectives. Moreover, these type of alloys attracts attention due to the results obtained by authors in [6, 7]. It was found that prolonged low-temperature annealing allows to form a regular structure, which is formed by alternating layers of 3d-metal, copper and rare-earth metal atoms. This structure is characterized by the appearance of the exchange interaction between the layers. Besides, it provides a high-energy barrier to the motion of the domain wall.

It is well known that the parameters of microstructure determine the coercivity of the materials. In turn, for a description of the complex geometry of the microstructure fractal geometry can be used. Moreover, the relationship between the fractal dimension of the soft magnetic materials samples microstructure and their magnetic properties was experimentally investigated in [8]. Thus, fractal characteristics can be used for the evaluation of the processes responsible for the formation of...
structurally sensitive magnetic properties. In [8], using various technological schemes for modification of the soft magnetic materials samples surface, the value of the fractal dimension of the profile was estimated in the range of \(1.106 \leq D_L \leq 1.424\), which characterizes a sufficiently undeveloped fractal relief.

The aim of this paper is a comprehensive study of the micro- and nanostructures and magnetic properties of Sm(Co,Cu,Fe)\(_5\) alloys after heat treatments and their comparative analysis in the framework of fractal geometry.

2. Experimental

The Sm(Co\(_{0.45}\)Cu\(_{0.4}\)Fe\(_{0.15}\))\(_5\) alloy were prepared by arc melting and annealed using following two-step regime: 1) homogenization at 1100°C during 6 hours and 2) subsequent low-temperature treatment at 400°C during 55 hours.

The microstructure was studied using a JEOL JSM-6610LV scanning electron microscope (SEM). The nanostructure was revealed on a scanning probe microscope (SPM) in the contact mode. Magnetic measurements were performed using a commercial Quantum Design VSM vibrating sample magnetometer (VSM) in fields up to 100 kOe, measurements in the temperature range from 300 to 700 K were made on LakeShore VSM 7410. Two types of samples were used for measurements: 1) separate grains of less than 10 mg; 2) cylinder shape samples from the textured powder of 80 \(\mu\)m in diameter embedded in a nonmagnetic epoxy.

3. Experimental results

A) Microstructure

Figure 1 depicts BSE images of the typical microstructures of the as-cast (a), homogenized (b) and annealed (c) alloys. One can see the evolution of the alloy structure during heat treatments from multiphase to microscopically homogeneous.

In the as-cast alloy (figure 1a), there are two main phases (light and dark grey regions) corresponding to 1:4 and 1:5 stoichiometric ratios, respectively. Dark elongated impurity phase, occupying less than 8% of the volume, have copper-rich composition. Homogenization at 1100°C for 6 hours (figure 1b) does not change the number of phases, however, each phase acquires clear boundaries. An analysis of the chemical composition indicates a change in the stoichiometry of the phases to 1:5 and 1:6, correspondingly. In addition, according to EDX analysis, the redistribution of the copper among the phases was revealed.

![Figure 1](image1.png)

**Figure 1.** The microstructure of Sm(Co\(_{0.45}\)Cu\(_{0.4}\)Fe\(_{0.15}\))\(_5\) alloy detected by scanning electron microscopy: the state after melting (a); after homogenization at 1100 °C (b); after low temperature treatment at 400 °C (c).

The last stage of heat treatment is low-temperature annealing at 400°C. It allows one to achieve a microscopically homogeneous state in the alloy (figure 1c). The results of EDX analysis have shown
that the chemical composition of the alloy is very close (± 2 at. %) to the nominal one Sm(Co$_{0.45}$Cu$_{0.43}$Fe$_{0.12}$)$_{5.9}$.

The study of the surface of the metallographic specimens using contact atomic force high-resolution microscopy revealed the presence of a nanoscale structure. AFM images of a microscopically homogeneous alloy after electrochemical etching are presented in figure 2.

![AFM images of surface of the Sm(Co$_{0.45}$Cu$_{0.4}$Fe$_{0.15}$)$_5$ alloy after a complete heat treatment cycle. Scans size: 3x3 μm (a), 1.5x1.5 μm (b).](image_url)

**Figure 2.** AFM images of surface of the Sm(Co$_{0.45}$Cu$_{0.4}$Fe$_{0.15}$)$_5$ alloy after a complete heat treatment cycle. Scans size: 3x3 μm (a), 1.5x1.5 μm (b).

Electrochemical etching visualizes the micro- and nanostructure of the metallographic specimen, creating a topography which reflects the phase distribution. As shown, close-packed round shaped areas 100–200 nm in diameter are observed on the surface. The presence of the depicted features of the surface is due to regular nanoscale composition inhomogeneities in the alloy volume. It is most likely that observed periodic concentration inhomogeneities in copper similar to liquation.

Thus, the variation of the heat treatments regimes leads to formation of the regular micro- and nanostructure in the samples.

**B) Magnetic properties**

The results of magnetic measurements performed on textured powders indicate a significant coercive filed of $H_{c1} = 7$ kOe and 32 kOe for a homogenized sample and the alloy after low-temperature heat treatment, respectively. This result is in good agreement with the previous data [7].

To study the reversal process, the major and minor hysteresis loops of the samples after homogenization and a full heat treatment cycle were measured (figure 3). The type of the obtained dependences for both samples corresponds to the pinning mechanism of coercivity. An increase in the magnetizing field does not change the type of the hysteresis loops. As shown, the main hysteresis characteristics linearly increase with rising of magnetizing field and reach a constant value in magnetic fields of more than 60 kOe. This result does not confirm the assumption that in this type material there are periodic energy barriers, formed by exchange bonds between atomic layers since in this case, the hysteresis loops should have at least a high degree of rectangularity. Indeed, the demagnetization curves in the entire range of magnetizing fields monotonically decrease.

The temperature dependences of the coercive field and the remanent magnetization, presented in Figure 4, linearly decrease. Such behaviour of the main hysteresis characteristics with increasing temperature also proves the pinning mechanism.
Figure 3. Full and partial hysteresis loops of powder samples Sm(Co_{0.45}Cu_{0.4}Fe_{0.15})_5 after homogenization (a) and a complete heat treatment cycle (b).

Figure 4. Temperature dependences of the coercive force and the remanent magnetization of the Sm(Co_{0.45}Cu_{0.4}Fe_{0.15})_5 sample after a complete heat treatment cycle.

C) Fractal geometry analysis

Below, a comparative analysis of the magnetic properties of the samples and the fractal characteristics of their surface was carried out.

Measurement of the fractal dimension of nanostructures on metallographic specimen surface of the alloys at various stages of the technological process will allow us to establish a correlation between the former and the surface morphological factors of the as-cast, homogenized and annealed samples. This will allow one to determine the optimal parameters of the formation of the most developed hard magnetic material surface.

As a rule to characterize the basic properties of fractal cluster aggregates – the self-similarity of their internal structures – the fractal dimension \( D_c \) is determined using the equation [11, 12]:

\[
N = \left( \frac{d}{a} \right)^{D_c},
\]

where \( N \) is the number of particles in the cluster (the number of monomers), \( d \) is the linear size, i.e. cluster (aggregate) diameter, \( a \) is the size of the particles that the cluster consist of (average monomer...
size). As already mentioned above, the fractal dimension of the profile $D_L$ was determined in [8]. The fractal dimension $D_f$ of the surface can be calculated as follows:

\[ D_L = D_f - 1, \]  

(2)

i.e. it would be expected that the values of the fractal dimension of the surface in the range of $D_f = 2.106 \pm 2.424$.

In this paper, the estimation of the fractal dimension and processing of AFM images of the surface was performed in the Image Analysis software package (version 3.5.30.19856). The proposed method for determining the fractal dimension is described in [13]. Below a brief overview of its provisions is presented. The following relation is used to determine the fractal dimension

\[ D_c = 3 - \alpha, \]  

(3)

where $\alpha$ is the scaling coefficient called the roughness index (Hurst index $H$). The Hurst exponent is determined through the slope of the initial section $tg\beta$ of the height-height correlation function for the selected direction, constructed in logarithmic coordinates according to the equation (4):

\[ H = \tan \beta / 2. \]  

(4)

Moreover, as was shown in [14], the fractal dimension of the surface $D_f$ can be identified with the corresponding cluster dimension of the three-dimensional aggregates. Analysis of Figure 2 shows that the fractal dimension of the surface of the sample is $2.21 \leq D_f < 2.35$. This range of values corresponds to the undeveloped fractal relief and the values, obtained for soft magnetic materials. The average fractal dimension of $2.25 \pm 0.02$ for the scales of $1.5 \times 1.5 \mu m$ and $3 \times 3 \mu m$ is practically the same, that confirms the stability and adequacy of the proposed in [12, 13, 15, 16] method for determining the fractal dimension. During the analysis of larger scales (in particular $5x5 \mu m$) of the Sm(Co$_{0.4}$Cu$_{0.4}$Fe$_{0.15}$)$_5$ sample surface agglomerates with a fractal dimension of 2.46 were observed. This value corresponds to a moderately developed fractal relief. Perhaps, these fractal agglomerates in the region of the sample did not degrade in the process of a heat treatment cycle.

4. Conclusion

Data on the microstructure, nanostructure and chemical composition of the phases of Sm(Co,Cu,Fe)$_5$ alloys were obtained. It was shown that microheterogeneity is not observed in the samples after low-temperature heat treatment. At the same time, the nanostructure is regular and represents rounded precipitations of 100-200 nm in diameter.

Based on a comprehensive analysis of micro- and nanostructure together with magnetic properties, it was determined that the pinning mechanism governs the magnetic behavior of the explored alloys.

For the first time, for the Sm(Co,Cu,Fe)$_5$ alloy a possible correspondence between the magnetic properties and the values of the fractal dimension of the surface is shown. The adjustment of the regime and the number of heat treatment cycles make it possible to produce samples with the required value of the fractal dimension.

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