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Equivalent circuits for FeCoZr-Al_2O_3 nanocomposite films deposited in argon and argon-oxygen atmospheres

Abstract. *The investigation of the equivalent circuits for granular nanocomposite films was performed according to the method of the impedance spectroscopy. The films, consisted of the $Fe_{0.45}Co_{0.45}Zr_{0.10}$ ferromagnetic alloy nanoparticles embedded into amorphous dielectric alumina matrix, were deposited in pure argon or argon and oxygen mixture. The temperature dependences of active and reactive components in the equivalent circuits for the $(Fe_{0.45}Co_{0.45}Zr_{0.10})_x(Al_2O_3)_{(1-x)}$ nanocomposite films are compared and analyzed. The presence of the inductive part in the equivalent circuits for the samples deposited in Ar gas below and beyond percolation threshold is shown. It is revealed that the equivalent circuits of the $(Fe_{0.45}Co_{0.45}Zr_{0.10})_x(Al_2O_3)_{(1-x)}$ nanocomposites produced in argon + oxygen gas mixture show more strong inductive contribution than ones sputtered in pure argon.*

Streszczenie. *Przeprowadzono badania układów zastępczych ziarnistych nanokompozytowych warstw zgodnie z wymaganiami metody spektroskopii impedancyjnej. Warstwy, złożone z ferromagnetycznych stopów nanocząstek $Fe_{0,45}Co_{0,45}Zr_{0,10}$ osadzonych w amorficznym podłożu dielektrycznym, zostały wytworzone w atmosferze czystego argonu lub mieszaninie argonu i tlenu. Dokonano analizy porównawczej temperaturowych zależności składowej czynnej i biernej w układach zastępczych nanokompozytowych warstw $(Fe_{0,45}Co_{0,45}Zr_{0,10})_x(Al_2O_3)_{(1-x)}$. W przypadku próbek powyżej oraz poniżej progu perkolacji wytworzonych w atmosferze argonu wykazano obecność składowej indukcyjnej w układzie zastępczym. Zaobserwowano również, że układy zastępcze nanokompozytów $(Fe_{0,45}Co_{0,45}Zr_{0,10})_x(Al_2O_3)_{(1-x)}$ wytworzonych w atmosferze mieszaniny argonu i tlenu charakteryzują się znacznie silniejszym udziałem indukcyjności w porównaniu z warstwami naniesionymi w atmosferze czystego argonu. (**Układy zastępcze dla nanokompozytowych warstw FeCoZr-Al_2O_3 wytworzonych w atmosferze argonu oraz argonu i tlenu**).*

Keywords: equivalent circuits, impedance, ion-beam sputtering, nanocomposites.
Słowa kluczowe: układy zastępcze, impedancja, rozpylanie jonowe, nanokompozyty.

Introduction

Investigation of the granular nanocomposites containing dielectric matrix with granules of soft ferromagnetic metals and their alloys are of a great interest for the material science. The interest to such systems is mainly due to the possibilities for their application for engineering of shields protecting against the HF electromagnetic radiation, in highly-Ohmic resistors and for other purposes [1]. The fact that some nanocomposites possess the semiconductor properties but their manufacturing is usually cheaper, determines also the possibility to use them as active and reactive elements in electrical circuits [2]. As was noted in [3], some of the metal-dielectric nanocomposites can be recommended as coilless inductive elements.

It should be stressed that the particular contribution of each charge carriers transport mechanism that are realized in binary and more complicated nanocomposites is strongly dependent on the metal-to-dielectric phases volume ratio. As a result, when the metallic fraction x increases, the metal-dielectric composite in accordance with percolation theory [4] will pass from the weakly-conductive (dielectric) to the highly-conductive (metallic) state. This takes place when some critical concentration of metallic phase x_C is reached, while continuous conducting (or percolating) cluster is formed between electric probes from the contacting highly-conductive particles. In binary composites the mentioned x_C value is called the percolation threshold. When $x<x_C$ (below the percolation threshold) the composite will be at the dielectric side of the metal-insulator transition (MIT) whereas at $x>x_C$ (beyond the percolation threshold) composite is at the metallic side of MIT.

The main goal of this paper is to investigate the equivalent circuits for granular nanocomposite films FeCoZr-Al_2O_3 using the method of impedance spectroscopy [5] and only AC impedance spectroscopy [6].

Experimental

Granular $(Fe_{0.45}Co_{0.45}Zr_{0.10})_x(Al_2O_3)_{(1-x)}$ nanocomposite films were prepared using ion-beam sputtering of compound target in vacuum chamber filled either with pure Ar gas under the pressure P_{Ar}=0,80 mPa (set 1) or with (Ar+O_2) gas mixture with partial pressure P_{O2}=(1,3−5,0) mPa (set 2) [7]. The samples under consideration were the films of 3-6 μm thicknesses deposited on glass ceramic substrates. The used deposition method (see details in [3, 7-9]) allowed to obtain in one technological cycle the whole set of samples with different metallic phase concentrations in the range of 30 at.%<x<65 at.%.

Temperature dependences of the AC resistance (impedance) were measured on the samples of 2 mm width and 10 mm length with indium ohmic contacts prepared by ultrasonic soldering. Impedance of the samples was measured by means of a 2-probe method in the temperature range of 100-340 K. Measurement setup included precise AC bridge HP-4284A and computer frequency and temperature control system. It allowed one to conduct measurements of current amplitude and phase in the frequency range of 10^2-10^6 Hz calculating then the real and imaginary parts of impedance in the studied samples.

The film thicknesses were measured with accuracy within 3-4 % on the samples chips by means of the scanning electron microscope. On the whole the relative error of admittance measurements was not more than 5 %.

The types of the equivalent circuits for granular nanocomposite films were designed according to the method of the impedance spectroscopy.

Results and Discussion

The impedance frequency dependences of the nanocomposites under study have given us two different types of the equivalent circuits for the samples of set 1 at dielectric (Fig. 1a) and metallic (Fig. 1b) sides of MIT.

As was noted in [9], the position of the percolation threshold x_C for the set 1 samples corresponds to the metallic phase concentration x~42 at.%. The change of type of equivalent circuits in Fig. 1 observed just at this concentration confirms this fact.

Let us study the temperature dependences of active and reactive contributions to equivalent circuits in Fig. 1. It should be stressed, that the parallel RCL scheme in equivalent circuits for the set 1 samples below the percolation threshold can be attributed mostly to the dielectric strata between metallic nanoparticles. This follows, in particular, from Fig. 2a where temperature dependence of the resistor $R(T)$ composing the active part of equivalent circuit in Fig. 1a.

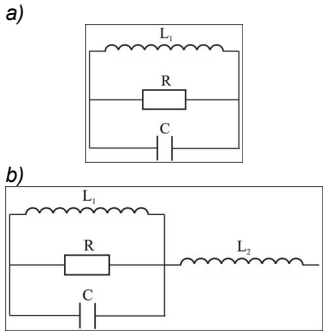


Fig.1. The equivalent circuits for set 1 nanocomposites below (30 at.%<x<42 at.%) (a) and beyond (48 at.%<x<65 at.%), (b) the percolation threshold.

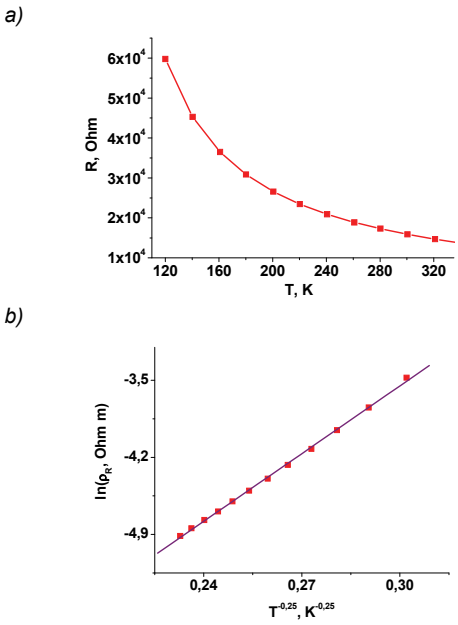


Fig.2. The temperature dependences of the active part of impedance $R(T)$ for frequencies 100 Hz<f< 200 kHz in direct scale (*a*) and resistivity $\rho_R(T)$ in Mott scale (*b*) for equivalent circuit in Fig. 1a

Linearization of this dependence in Mott scale $\ln(\rho_R)$–$(1/T)^{-0.25}$ shown in Fig. 2b corresponds to the known Mott law [10]:

$$\rho_R \propto \exp\left(-\frac{T_0}{T}\right)^{-0.25}, \quad (1)$$

that confirms the predominance of hopping conductance mechanism of carrier transport at dielectric side of MIT (at x_C<42 at.%) for the nanocomposites deposited in pure Ar atmosphere earlier noted in [8] from DC measurements.

Linearization in direct scale of temperature dependence of R in equivalent circuit in Fig. 1b for the nanocomposites of the set 1 beyond the percolation threshold (see, Fig. 3) confirms the predominance of the metallic conductance when x_C>42 at.% as was observed earlier in [8] for DC measurements.

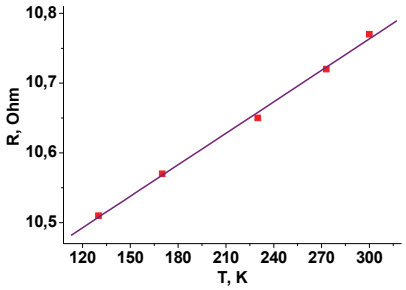


Fig.3. The temperature dependence of the active part of impedance $R(T)$ for frequencies 100 Hz<f<200 kHz in direct scale for equivalent circuit in Fig. 1b

The presence of small inductive contribution in the equivalent circuits for the samples of set 1 below and beyond percolation threshold (see, Figs. 1a and 1b) can be explained by the coil-like percolating routes of the charge carrier as was noted earlier in [11].

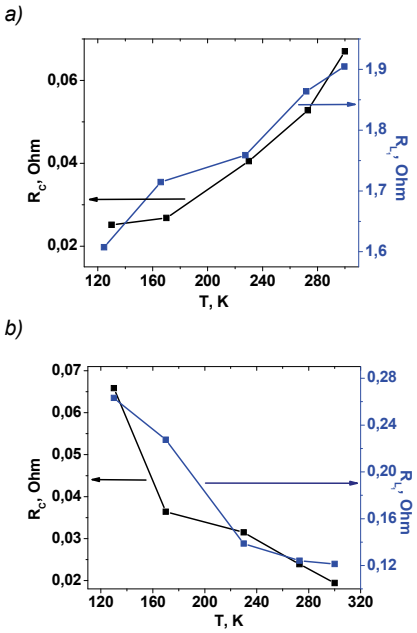


Fig.4. The temperature dependences of reactive parts RC and RL_1 for frequencies 100 Hz<f<200 kHz in equivalent circuit for the nanocomposites of the set 1 below (*a*) and beyond (*b*) the percolation threshold.

As is seen from Fig. 4, the character of temperature dependences of reactive parts R_C and R_{L1} in equivalent circuits for the nanocomposites of the set 1 is changed when crossing the percolation threshold. This is probably caused by the formation of metallic net and, as a consequence, change of sign of derivatives dRC/dT and dR_{L1}/dT from negative below the percolation threshold to positive beyond it.

It was revealed from frequency dependences of impedance the existence of the only one type of equivalent circuit for nanocomposites of set 2, deposited in Ar+O_2 gas mixture, for the whole range of metallic phase concentrations 30 at.%<x<65 at.% (see, Fig. 5). This confirms the lack of the percolation threshold in such nanocomposites as was noted in [8] on the base of DC measurements.

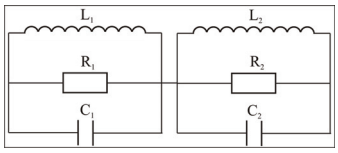
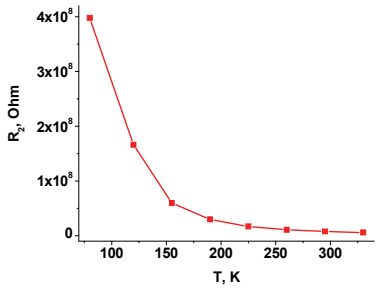


Fig.5. The equivalent circuits for set 2 nanocomposites for the whole set of samples with metallic phase concentrations in the range of 30 at.%<x<65 at.%

It should be noticed the presence of two RCL schemes with parallel connected R, L and C elements in equivalent circuit in Fig. 5 as compared to the equivalent circuit in Fig. 1a for set 1 samples. It is naturally that the first RCL scheme probably corresponds to the alumina strata between metallic nanoparticles whereas the second one characterizes the oxide „shells" [9].

a)



b)

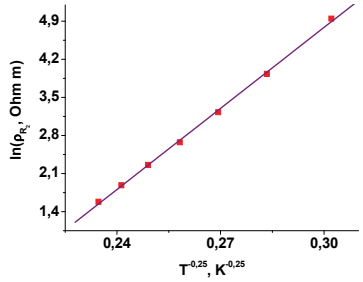


Fig.6. The temperature dependences of $R_2(T)$ contribution in the equivalent circuit in Fig.5 for set 2 nanocomposites in direct scale (*a*) and resistivity $\rho_R(T)$ in Mott scale (*b*) for frequencies 100 Hz<f<200 kHz

Note that, in accordance with Fig. 2b, R_1 contribution has Mott-like temperature dependence. This allowed us to estimate temperature dependence of R_2 contribution (see, Fig. 6a) in the equivalent circuit in Fig. 5. As is seen from Fig. 6b, $R_2(T)$ also obeys Mott law (1) that also denotes the predominance of hopping conductance mechanism in oxide "shells".

Data of [3] confirm that the impedance of granular $(Fe_{0.45}Co_{0.45}Zr_{0.10})_x(Al_2O_3)_{(1-x)}$ nanocomposites prepared in Ar+O_2 gas mixture reveals more strong inductive contribution in their equivalent circuit than it was observed in the samples sputtered in pure argon.

Resume

The carried out study of granular $(Fe_{0.45}Co_{0.45}Zr_{0.10})_x(Al_2O_3)_{(1-x)}$ nanocomposites prepared in pure Ar gas or in Ar+O_2 gas mixture have shown the presence of the inductive parts in the equivalent circuits which was dependent both on the metallic phase concentration and type of gas at deposition.

The presence of the inductive part in the equivalent circuits for the samples deposited in Ar gas below and beyond percolation threshold could be explained by the coil-like type of the charge carrier routes. The change of equivalent circuit type with x increase in set 1 nanocomposites is caused by the formation of the metallic net when crossing the percolation threshold. Linearization of temperature dependence of the equivalent circuit active part in Mott scale for the set 1 nanocomposites confirmed the predominance of hopping conductivity mechanism on dielectric side of MIT (x_C<42 at.%). Linearization of the temperature dependence of active part of equivalent circuit beyond the percolation threshold for set 1 samples displayed the predominance of the metallic conductivity when x_C>42 at.%.

The existence of the only one type of equivalent circuit for nanocomposites deposited in Ar+O_2 gas mixture for the whole set of samples with 30 at.% <x<65 at.% could be explained by the absence of the percolation threshold in this samples (due to their non-binarity).

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