PRELIMINARY STUDIES OF AC CONDUCTIVITY OF $(FeCoZr)_x(SiO_2)_{(100-x)}$ NANOCOMPOSITES PRODUCED IN THE ATMOSPHERE OF PURE ARGON

[1]TOMASZ N. KOŁTUNOWICZ, [2]KAROLINA CZARNACKA, [3]ALEKSANDER K. FEDOTOV, [4]JULIA A. FEDOTOVA

[1,2]Department of Electrical Devices and High Voltage Technology, Lublin University of Technology, Lublin, Poland
[3]Department of Energophysics, Belarusian State University, Minsk, Belarus
[4]Research Institute for Nuclear Problems of Belarusian State University, Minsk, Belarus
E-mail: [1]t.koltunowicz@pollub.pl

Abstract: The paper presents preliminary studies of AC conductivity of $(FeCoZr)_x(SiO_2)_{(100-x)}$ nanocomposites. Samples by ion-sputtering in the atmosphere of pure argon were produced. Samples of various metallic phase content were prepared. With the use of AC apparatus, the basic electrical parameters were determined. From measured values the dependence of conductivity on frequency and temperature the was determined.

Keywords: Nanocomposites, Electrical Properties, Conductivity, Frequency-Temperature Relationship.

I. INTRODUCTION

Among the wide range of nanostructures, nanogranular layers with metallic nanoparticles of a ferromagnetic alloy or semiconductor located inside a silicon oxide dielectric matrix occupy a special place [1-4]. Numerous experiments have shown that such nanomaterials, in a number of cases, are characterized by unique combinations of electrical [5], magneto-transport [6, 7], magnetic [8, 9], optical [10-12] and other properties. The basic factors which are determining the electrophysical properties of granular nanocomposites are: chemical composition of nanoparticles and matrix, size and morphology of metallic nanoparticles and also their degree of order. Such nanocomposites, from the point of view of charge transport, are model structures for the study of percolation phenomena [13]. Charge transfer on the dielectric side should take place by hopping exchange of electrons (tunneling) between neighboring nanoparticles [14, 15]. It is easy to understand taking into account, that nanoparticles of metallic or semiconductor phases are separated by a stratum of dielectric with very high resistivity, and during the current flow the carriers must overcome the insulation barriers which are separating nanoparticles of metallic or semiconductor phase.

The aim of this work is preliminary analysis of changes in conductivity of nanocomposites with metal-dielectric structure (where: metal is FeCoZr and dielectric is SiO_2) and the influence of temperature on the properties of these nanocomposites

II. EXPERIMENTAL DETAILS

Nanocomposites with a metal dielectric structure where the metallic phase is formed by FeCoZr ferromagnetic alloy particles and the dielectric matrix is silicon dioxide SiO_2 were selected for the study. For the production of nanocomposites with a metal-dielectric structure the method of two-source ion-sputtering was used. Sputtered target consisted of a $Fe_{45}Co_{45}Zr_{10}$ metal alloy plate and SiO_2 dielectric stripes attached to this plate. This plate configuration enables the performance of thin film with thickness of 1 to 6 μm with a different metallic to dielectric ratio in one technological process.

Sputtered with pure argon ions, the shield atoms settle on the rotating substrate. A component with a higher surface energy value (metal alloys) creates elements in the dielectric matrix, which are similar to spherical shapes. In this way, structures consisting of metallic nanograins randomly arranged in the dielectric matrix are formed.

By varying deposition parameters such as argon gas pressure, rotational speed of the substrate and other conditions, structures differing in the metallic phase content can be obtained. A description of the production device and its operation is presented in the work [16-19].

Using the ion sputtering method in argon atmosphere, nanocomposites $(FeCoZr)_x(SiO_2)_{(100-x)}$ with different ration of metallic phase content x to the dielectric phase (100-x) were produced. Samples with those metallic phase content were selected for measurements: x_1= 58.2 at.%, x_2= 67.0 at.% and x_3= 76.3 at.%.

The main elements of the test station are the helium cryocooler and the LCR bridge. The cryocooler range of measurement temperatures T_p is 15 K – 400 K. HIOKI 3522-50 LCR HiTESTER low-frequency meter is designed for measuring many AC parameters, including: phase angle θ, resistance R_p, capacity C_p and loss tangent tgδ and their frequency

dependencies. The frequency measurement is in the range of 10^{-3} Hz to 10^5 Hz. The methodology of research has been presented in detail in papers [19, 20].

III. RESULTS AND DISCUSSION

From measured values the dependence of conductivity σ on frequency and temperature the was determined. Due to the large number of obtained measuring curves corresponding to different measuring temperatures T_p (90 curves), only 6 examples from the range 20 K-375 K are presented on the graphs.

Figure 1 shows the frequency dependence of the conductivity for a nanocomposite $(FeCoZr)_x(SiO_2)_{(100-x)}$ with a metallic phase content x_1= 58.2 at.%.

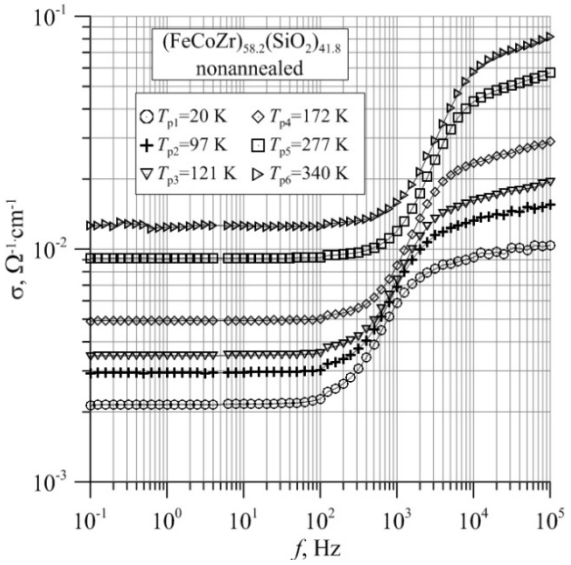


Fig. 1. Dependence of conductivity versus frequency for nanocomposite $(FeCoZr)_x(SiO_2)_{(100-x)}$ with metallic phase content x_1 = 58.2 at%, produced in pure argon atmosphere.

It can be observed that as the measured temperature T_p increases, the conductivity values also increase. S-shaped curves with clearly defined conductivity values σ_L appear in the low frequency range (10^{-1} Hz – about 10^2 Hz) and conductivity at the highest frequencies σ_H (about 4×10^4 Hz for T_p = 20 K). Determined values at the highest measurement frequencies, along with the increase of the measuring temperature , move towards to higher frequencies (for non-measured values beyond the measuring range). In the frequency range between the determined regions, the conductivity change can be described as [21]:

$$\sigma_M \sim f^{\alpha}$$

where: σ_M – conductivity for the middle range of frequency; f – frequency, α – frequency coefficient.

For nanocomposites with a higher content of metallic phase x_2= 67.0 at.% (Fig. 2) the conductivity constant value with frequency increase and its increase with temperature in the range of 10^{-1} Hz to approx. 10^4 Hz, are observed. The area of the change in conductivity σ_M is observed from the frequency of 10^4 Hz to the end of the measuring range. There is no visible range of stable conductivity σ_H at higher frequencies. It should be presumed that in the higher measuring range for the frequency it will be possible to determine the slope in the area of σ_M.

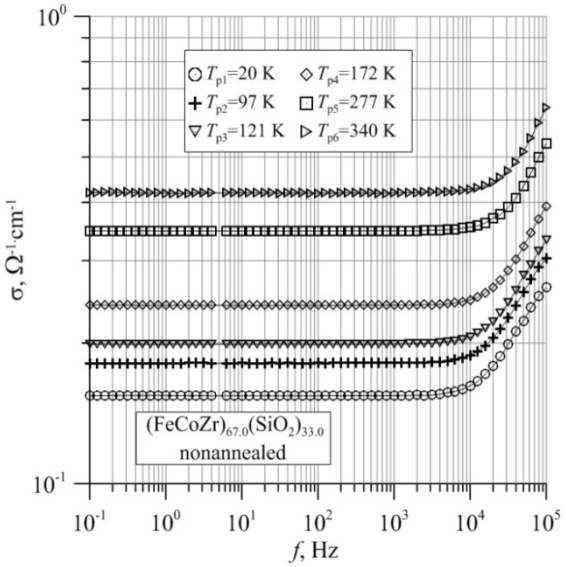


Fig. 2. Dependence of conductivity versus frequency for nanocomposite $(FeCoZr)_x(SiO_2)_{(100-x)}$ with metallic phase content x_2= 67.0 at.%, produced in pure argon atmosphere.

For the nanocomposite with the highest content of the metallic phase x_3= 76.3 at.% (Fig. 3), a similar situation is observed for the nanocomposite with the metallic phase content x_2= 67.0 at.% – movement of the regions for σ_M and σ_H towards to the higher frequencies (beyond the measuring range of the used bridges).

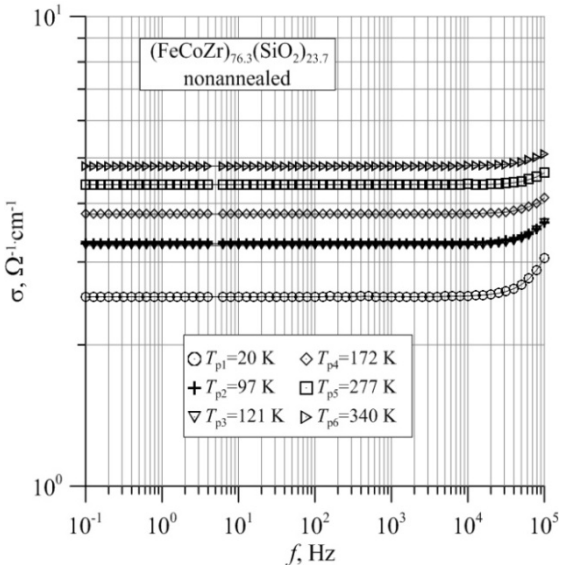


Fig. 3. Dependence of conductivity versus frequency for nanocomposite $(FeCoZr)_x(SiO_2)_{(100-x)}$ with metallic phase content x_3= 76.3 at.%, produced in pure argon atmosphere.

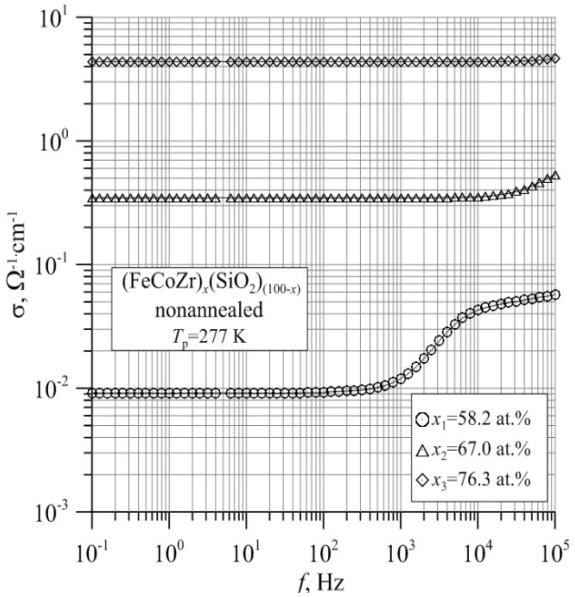


Fig. 4. Dependence of conductivity versus frequency for nanocomposites $(FeCoZr)_x(SiO_2)_{(100-x)}$ with metallic phase contents x_1= 58.2 at.%, x_2= 67.0 at.% and x_3= 76.3 at.% determined at measurement temperature T_p = 277 K.

Figure 4 presents dependence of conductivity versus frequency for nanocomposites $(FeCoZr)_x(SiO_2)_{(100-x)}$ with different metallic phase contents: x_1= 58.2 at.%, x_2= 67.0 at.% and x_3= 76.3 at.% which were determined at measurement temperature T_p = 277 K. There is an increase in the conductivity associated with the increase in the content of the metallic phase.

CONCLUSIONS

In order to determine the full-form (S-shaped) conductivity curves, measurements should be made in a wider frequency range, because with the increase of the metallic phase content x in the nanocomposites, the $\sigma_M \sim f^{\alpha}$ region moves towards the higher measurement frequencies.

ACKNOWLEDGMENTS

The work was partly supported by the statute tasks of the Lublin University of Technology, at the Faculty of Electrical Engineering and Computer Science, 8620/E-361/S/2017 (S-28/E/2017), entitled "Researches of electrical, magnetic, thermal and mechanical properties of modern electrotechnical and electronic materials, including nanomaterials and electrical devices and their components, in order to determination of suitability for use in electrical engineering and to increase the efficiency of energy management".

REFERENCES

[1] Williams G.V.M., Prakash T.; Kennedy J., The effect of fluence on the magnetic properties of superparamagnetic iron-nickel nanoparticles in SiO2 made by dual Ni and Fe low energy ion implantation, Nuclear Instruments & Methods in Physics Research Section B-Beam Interaction with Materials and Atoms, vol. 409, pp. 187-191, 2017

[2] Prakash T., Williams, G.V.M., Kennedy J. Rubanov S., Formation of magnetic nanoparticles by low energy dual implantation of Ni and Fe into SiO2, Journal of Alloys and Compounds vol. 667, pp. 255-261, 2016

[3] Chulapakorn T., Sychugov I., Suvanam S.S., Linnros J., Primetzhofer D., Hallen A., Influence of swift heavy ion irradiation on the photoluminescence of Si-nanoparticles and defects in SiO2, Nanotechnology, vol. 28 n. 37, Article Number: 375603, 2017

[4] Tyschenko I.E., Voelskow M., Cherkov A.G., Popov V.P., Ion-beam synthesis of InSb nanocrystals in the buried SiO2 layer of a silicon-on-insulator structure, Semiconductors, vol. 48 n. 9, pp. 1196-1201, 2014.

[5] Koltunowicz T.N., Zukowski P., Czarnacka K., Bondariev V., Boiko O., Svito I.A., Fedotov A.K., Dielectric properties of nanocomposite (Cu)x(SiO2)(100-x) produced by ion-beam sputtering, Journal of Alloys and Compounds, vol. 652, pp. 444-449, 2015.

[6] Balashev V.V., Vikulov V.A., Dimitriev A.A., Pisarenko T.A., Pustovalov E.V., Korobtsov V.V., Evolution of the structural and magnetotransport properties of magnetite films depending on the temperature of their synthesis on the SiO2/Si(001) surface, Physics of Metals and Metallography, vol. 118 n. 7, pp. 644-651, 2017.

[7] Kasiuk J.V., Fedotova J.A., Svito I.A., Kalinin Yu.E., Sitnikov A.V.: Influence of the short-range order in FeCoZr nanoparticles on the electrical and magnetotransport properties of FeCoZr-CaF2 nanocomposites Physics of the Solid State, vol. 54 n. 10, pp. 2091-2099, 2012.

[8] Fedotova J., Ivanou D., Ivanova Y., Fedotov A., Mazanik A., Svito I., Streltsov E., Saad A., Tyutyunnikov S., Koltunowicz T.N., Demyanov S., Fedotova V., Magnetoresistance in n-Si/SiO2/Ni nanostructures manufactured by Swift Heavy Ion-Induced Modification Technology, Acta Physica Polonica A, vol. 120 n. 1, pp. 133-135, 2011.

[9] Fedotova J., Saad A., Ivanou D., Ivanova Y., Fedotov A., Mazanik A., Svito I., Streltsov E., Koltunowicz T.N., Tyutyunnikov S., Gigantic magnetoresistive effect in n-Si/SiO2/Ni nanostructures fabricated by the template-assisted electrochemical deposition, Przegląd Elektrotechniczny, vol. 88 n. 7a, pp. 305-308, 2012.

[10] Kermadi S., Agoudjil N., Sali S., Boumaour M., Bourgeois S., de Lucas M.C.M., Sol-gel synthesis of xTiO(2)(100-x)SiO2 nanocomposite thin films: Structure, optical and antireflection properties, Thin Solid Films, vol. 564, pp. 170-178, 2014.

[11] Girgis E., Wahsh M.M.S., Othman A.G.M., Bandhu L., Rao K.V., Synthesis, magnetic and optical properties of core/shell Co1-xZnxFe2O4/SiO2 nanoparticles, Nanoscale Research Letters, vol. 6, Article Number: 460, 2011.

[12] Fermento R., Gonzalez-Diaz J.B., Cebollada A., Armelles G., Diaz M., Martinez L., Roman E., Huttel Y., Ballesteros C., Optical and magneto-optical properties of Co-SiOx thin films, Journal of Nanoparticle Research vol. 13 n. 6, pp. 2653-2659, 2011.

[13] Koltunowicz T.N., Zukowski P., Czarnacka K., Svito I., Fedotov A.K., Percolation phenomena in nanofilms Cux(SiOy)100-x produced by ion beam-sputtering, Acta Physica Polonica A, vol. 128, n.5, pp. 908-911, 2015.

[14] Svito I., Fedotov A.K., Koltunowicz T.N., Zhukowski P., Kalinin Y., Sitnikov A., Czarnacka K., Saad A., Hopping of electron transport in granular Cux(SiO2)1-x nanocomposite films deposited by ion-beam sputtering, Journal of Alloys and Compounds, vol. 615 Supplement 1, pp. S371-S374, 2014.

[15] Fedotov A., Mazanik A., Svito I., Saad A., Fedotova V., Czarnacka K., Koltunowicz T.N., Mechanisms of carrier transport in Cux(SiO2)1-x nanocomposites manufactured by Ion-Beam Sputtering with Ar Ions, Acta Physica Polonica A, vol. 128, n.5, pp. 883-886, 2015.

[16] Kalinin Yu.E., Sitnikov A.V., Stognej O.V., Ponomarenko A.T., Granular metal-insulator nanocomposites with amorphous structure, Physics and Chemistry of Materials Treatment, vol. 5, pp. 14-20, 2001.

[17] Zolotukhin I.V., Kalinin Y.E., Ponomarenko A.T., Shevchenko V.G., Sitnikov A.V., Stognei O.V., Figovsky O., Metal-dielectric nanocomposites with amorphous structure, Journal of Nanostructured Polymers and Nanocomposites, vol. 2, pp. 23-34. 2006.

[18] Larkin A.V., Fedotov A.K., Fedotova J.A., Koltunowicz T.N., Zhukowski P., Temperature and frequency dependences of impedance real part in the FeCoZr-doped PZT nanogranular composites, Materials Science-Poland, vol. 30 n. 2, pp. 75-81, 2012.

[19] Koltunowicz T.N., Measurement station for frequency dielectric spectroscopy of nanocomposites and semiconductors, Journal of Applied Spectroscopy, vol. 82, n.4, pp. 653-658, 2015.

[20] Komarov F.F., Zhukowski P., Krivosheev R.M., Munoz E., Koltunowicz T.N., Rodionova V.N., Togambaeva A.K., Effects of surfactant and fabrication procedure on the electrical conductivity and electromagnetic shielding of single-walled carbon nanotube films, Physica Status Solidi A – Applications and Materials Science, vol. 212 n. 2, pp. 425-432, 2015.

[21] Kołtunowicz T.N., Żukowski P., Boiko O., Bondariev V., Czarnacka K., Fedotova J.A., Fedotov A.K., Svito I.A., Impedance model of metal-dielectric nanocomposites produced by ion-beam sputtering in vacuum and its experimental verification for thin films of (FeCoZr)x(PZT)(100-x), Vacuum, vol. 120 Part B, 2015, pp. 37-43.

★★★