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Obtaining Hexagonal Ferrites for Substrates Microstrip Microwave Devices of mm-Range of LTCC-technology

D.N. Chitanov, V.G. Kostishyn*, L.V. Kozhitov, A.Yu. Adamtsov

National University of Science and Technology "MISiS", 4, Leninsky prosp., 119049, Moscow, Russia

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In the work by the method low temperature co-fired ceramics (LTCC) obtained samples of isotropic and anisotropic polycrystalline hexaferrite $BaFe_{12}O_{19}$ and $SrFe_{12}O_{19}$. Using in the LTCC-technology the pressing operation for samples (tablets) in a magnetic field produces anisotropic hexaferrites, pressing without a magnetic field - isotropic hexaferrites. Application in the LTCC-technology molding process tape produces exclusively isotropic samples.

Keywords: Hexagonal ferrite, LTCC-technology, Microstructure, Reaction glass, Density

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1. INTRODUCTION

Perspective low temperature co-fired ceramics (LTCC) - hexaferrites, is of great interest for high frequency applications such as phase switches, circulators, antennas and wireless technologies for miniature electronic modules of the next generation. The rapid development of modern communication systems, such as mobile communications and satellite communications require miniaturization of microwave components. For reduction the size of microwave devices necessary to examine multilayer devices on the basis of the low temperature co-fired ceramics (LTCC), which consist of alternating ferrite and internal metallic electrodes. As a rule, as an internal electrode metal in multilayer LTCC devices using silver (Ag) due to its low losses and low electrical resistance at high frequencies [1, 2]. Since the melting point of Ag – 961 °C is necessary to develop LTCC materials which have a sintering temperature of about 900 °C, to prevent diffusion of Ag in the ferrites.

Hexaferrites usually get the classic ceramic technology [3, 4] at high sintering temperatures up to 1350 °C, which are not suitable for LTCC technology.

The development of LTCC magnets has trend to use small amounts of additives and ceramics glasses systems including softening glasses and magnetic ceramics with a high melting point. Typical additive for low temperature sintered hexaferrites prepared by low cost mixing oxide – Bi_2O_3. It is also possible to add lithium borosilicate glass or B_2O_3-Sb_2O_3. The main problem of the low-temperature burned polycrystalline magnetic phases such as $BaFe_{12}O_{19}$ – high porosity. Due to the high content of nonmagnetic phases (> 10 vol. %), such as glass or pores phase permeabilities sharply reduced. A new method that solves this problem and allows to simultaneously achieve sealing samples below 900 °C developed using sintering process hexaferrites with the addition of a small amount of reactive glasses, which are based on the Bi-B-Zn-Si-O (BBSZ).

2. OBTAINING RESEARCH OBJECTS

As starting materials for the M-type hexaferrites $BaFe_{12}O_{19}$ used $BaCO_3$, Fe_2O_3 grade HP (high purity) for $SrFe_{12}O_{19}$ - $SrCO_3$, Fe_2O_3 grade HP. As an additive to reduce the sintering temperature used borosilicate bismuth glass (BBSZ) based Bi_2O_3-B_2O_3-SiO_2-ZnO (3, 5, 7 vol. %).

Glass BBSZ with compositions $25Bi_2O_3$, $30B_2O_3$, $10SiO_2$ and 35ZnO prepared by a melting process. Starting powders of BBSZ glass weighed and mixed in accordance with their ratio in the chemical formula, and then the mixed powders are melted at 1200 °C for 2 hours in a platinum crucible, and quenched in water with formation amorphous glass. Then the glass dried and milled to powder. For studying the glass transition temperature and softening point glass part of the glass melt poured into a mold to make a glass column. The measured glass transition temperature and softening composed – 585 and 650 °C, respectively.

The modified M-type hexaferrites phase composition $SrFe_{12}O_{19}$ or $BaFe_{12}O_{19}$ was prepared as follows. Reaction mixtures were prepared using the following powders: Fe_2O_3, $CaCO_3$ or Fe_2O_3, $SrCO_3$, respectively. Powders were weighed on an analytical balance with an accuracy of ± 0,1 mg in the required proportions. After weighing the components produced their joint grinding in a ball mill for 2 hours. Synthesis was performed ferrite charge (ferritization) in a furnace to a temperature of 1200 °C in air. This temperature range was maintained for 5 hours. Then made cooling to room temperature. For reducing the sintering temperature, the pulverized powder was mixed with a reaction borosilicate bismuth glass based on Bi_2O_3-B_2O_3-SiO_2-ZnO at 3, 5, 7 vol. %. Carried crushing, grinding in an attritor with high energy during 2 hours to particles size of 0.8-1.8 μm for compacting samples. Further carried out the preparation of press powder. To the synthesized charge was added binder (10% solution of polyvinyl alcohol in an amount of 15 wt.% of charge) and surfactants. The mixed powder was compressed into tablets, or poured into a tape. The press samples were made one-sided cold pressing. The most optimum compaction pressure of 200 MPa. The resulting products were sintered in the tunnel furnaces in air at T = 900-950 °C. Sintering duration was 5 hours. After time sintering furnace was

* drvgkostishyn@mail.ru

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turned off and was carried out natural cooling of the samples in air.

3. METHODS OF EXPERIMENTAL RESEARCH

X-ray diffraction and X-ray analysis of the objects of research carried out on the X-ray diffractometer DRON-3M (Russia).

During the X-ray diffraction was used CuKα-radiation, as well as a tube with an iron anode (operating current – 25 mA, voltage – 25 kV). Wavelength 0.193728 nm. When taking samples used the filter of Mn. Focusing was performed by the method of Bragg-Brentano with two slits Soller. The measurements were performed at room temperature.

Identification intense peaks in the diffraction pattern was carried out using PDWin 4.0 software. X-ray diffraction analysis of samples was limited to the determination of the series interplanar spacings, and compared them with the reference data base of powder diffraction data, which is based on the files of PDF2.

Investigation of microstructure and quantitative analysis of the samples was performed on a scanning electron microscope of the company «Carl Zeiss» brand SEM LEO-420 microprobe attachment INKA ENERGY-400.

In order to investigate the compatibility hexaferrites and Ag-electrode ferrite sheet with Ag electrode was prepared by tape casting and co-firing at 900 °C.

4. THE RESULTS OF EXPERIMENTAL RESEARCH

Figure 1 shows the complete process for producing polycrystalline hexagonal ferrites $BaFe_{12}O_{19}$ and $SrFe_{12}O_{19}$.

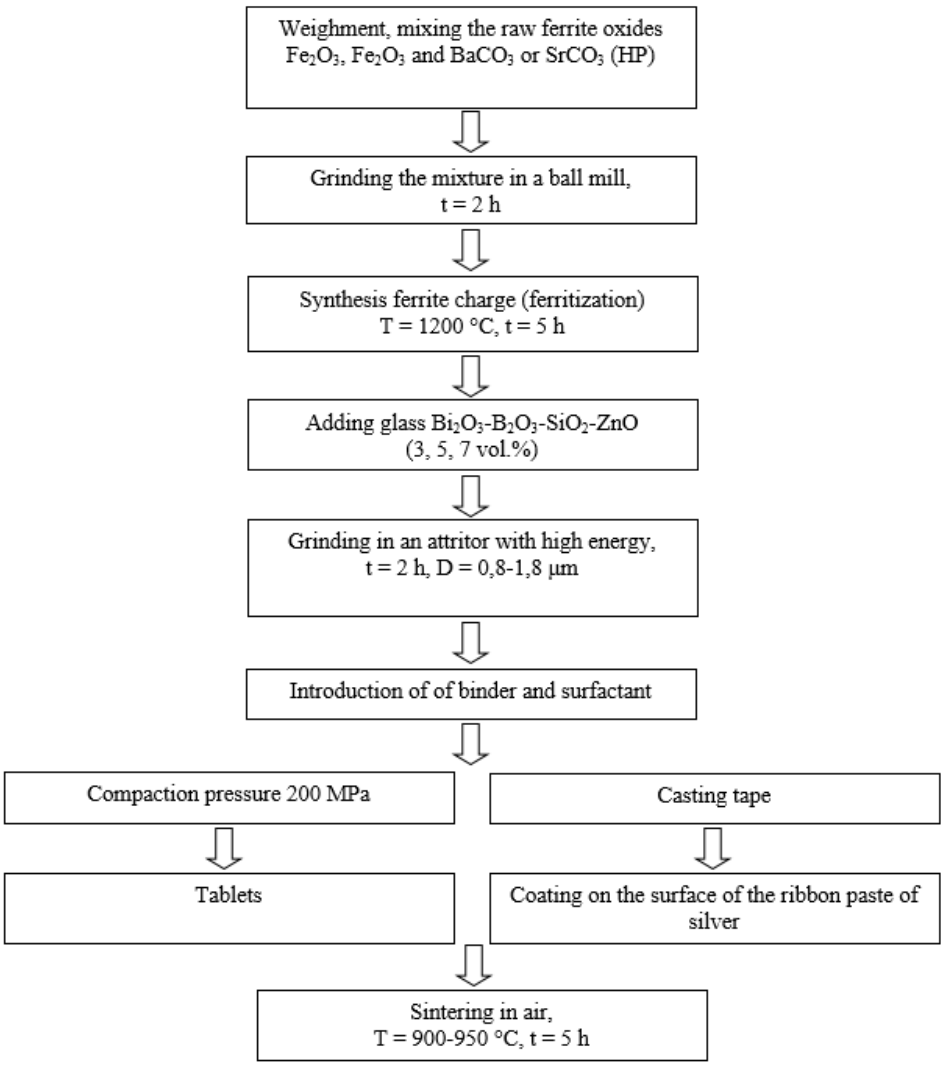

Fig. 1 – Technological scheme of the hexaferrites $BaFe_{12}O_{19}$ and $SrFe_{12}O_{19}$

For the manufacture anisotropic hexaferrites workpieces pressing occurs in a magnetic field applied along the direction of compression. For this purpose used a special press equipped with two coils (electromagnet), which create a magnetic field. In the upper coil includes the plunger press with reinforced tip on it, the form of which contributes to the concentration of the magnetic field. In the lower coil is a base for the mold with a hole to drain water ending with fitting for fastening a hose connected through a trap with a mechanical vacuum pump. Source of feeding electromagnet provides the DC up to 10 A at voltages up to 20 V.

X-ray studies have confirmed that as a result of the used technology obtained as a polycrystalline isotropic and anisotropic hexagonal ferrites $BaFe_{12}O_{19}$ and $SrFe_{12}O_{19}$. The characteristic X-ray diffractions are shown in Figures 2 and 3.

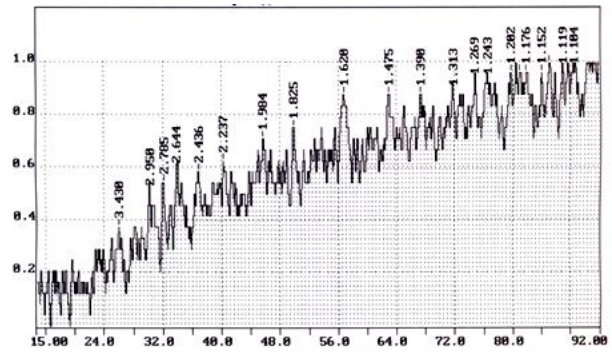

Fig. 2 – The characteristic X-ray diffraction pattern of the sample of polycrystalline isotropic hexaferrite $BaFe_{12}O_{19}$

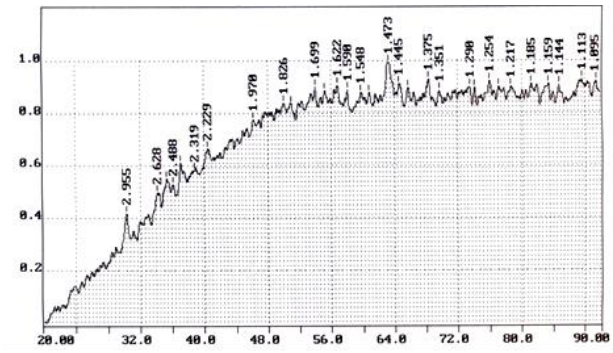

Fig. 3 – The characteristic X-ray diffraction pattern of the sample of polycrystalline anisotropic hexaferrite $SrFe_{12}O_{19}$

Dilatometric studies thermal treatment of samples with different amounts of glass BBSZ were conducted in order to establish a shrink effect on the density of the obtained samples. The results are shown in Fig. 4. The shrinkage increases with the amount of glass BBSZ. The porosity estimated using the method of Archimedes, decreases with increasing amount of glass BBSZ.

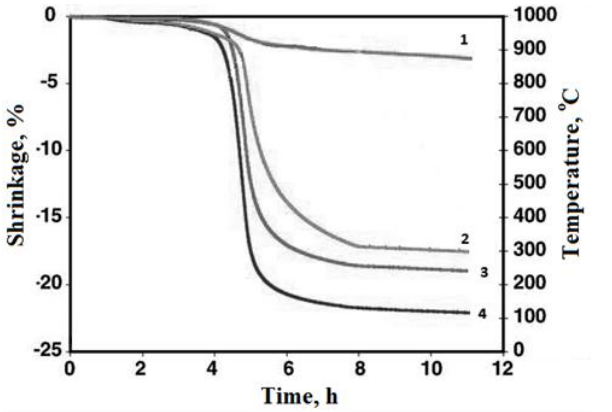

Fig. 4 – Impact BBSZ-glass content in the linear shrinkage $BaFe_{12}O_{19}$: 1 – 0 vol. % BBSZ; 2 – 3 vol. % BBSZ; 3 – 5 vol. % BBSZ; 4 – 7 vol. % BBSZ

Figure 5 shows the microstructure of the surface $BaFe_{12}O_{19}$ hexaferrite with different content BBSZ additives. By adding 3 vol. % BBSZ can be observed porous microstructure (Fig. 5 (b)). This suggests that the amount of liquid phase is not enough to seal the hexaferrite. In [5, 6], it was reported that increasing the glass content of the dielectric ceramics becomes denser microstructure. In our work also found that by increasing the content of additives in BBSZ geksaferrite microstructure is compacted. When the content of 5 vol. % BBSZ can be observed a homogeneous microstructure. The grain size of 0.2-0.5 μm (Fig. 3 (c)). This can be explained by the fact that the ferrite densification promoted BBSZ-additive, at the same time, grain growth was inhibited by the high surface energy [6].

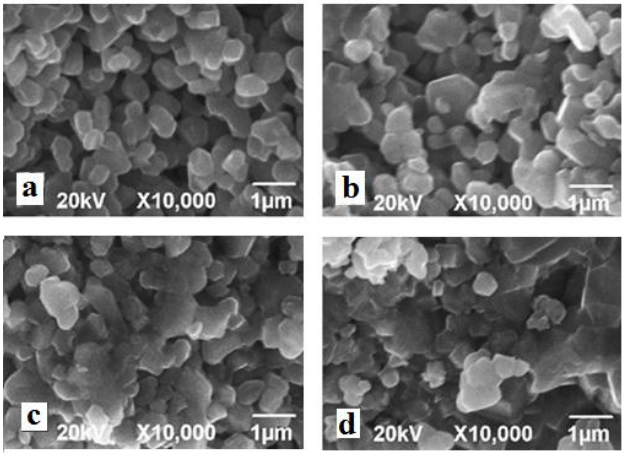

Fig. 5 – Microstructure hexaferrite $BaFe_{12}O_{19}$ sintered at 900 °C: (a) without adding BBSZ, (b) about 3 vol. %, (c) 5 vol. % and (d) about 7 % BBSZ, respectively

5. CONCLUSION

In the work obtain polycrystalline hexagonal ferrites substrate of subminiature microstrip FRP short-wave part of the centimeter and millimeter wavelength ranges for LTCC-technology.

The developed methods allow to achieve compacting the samples at 900 °C during sintering hexaferrites with the addition of a small amount of reaction glasses composition Bi_2O_3-B_2O_3-SiO_2-ZnO (BBSZ).

Using in LTCC-technology the pressing operation of samples (tablets) in a magnetic field produces anisotropic hexaferrites, pressing without a magnetic field - isotropic hexaferrites.

Application in LTCC-technology molding process tape produces exclusively isotropic samples.

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