Electron beam sintering of Mn-Zn ferrites using a forevacuum plasma electron source

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Abstract. The article presents the results of electron beam sintering without applying pressure of Mn-Zn ferrites in an oxygen environment. Samples for sintering were made from fine powders and pressed at various pressures into compacts in the form of disks. Measurements of the elemental composition and structure of the sample after sintering are presented. It is shown that the result of sintering depends on the pressing pressure of compacts, time and temperature of sintering.

1. Introduction
Ferrites are semiconductor materials of great technological importance. The high resistivity of ferrites allows them to be used in microelectronics and microwave technology [1]. The rapid growth of electro-ceramic production of ferrites and the need for dielectric resonators are associated with the rapid development of communication technologies of the fifth and sixth generations - 5G and 6G. Currently, much attention is paid to the development of materials with improved microwave properties. Such materials are widely used in wireless communications technologies such as mobile phones, intelligent transport systems (ITS), ultrafast wireless LAN, and satellite broadband. The electrophysical properties of ferrites depend on several factors, such as the chemical composition, structure or grain size, and the method of production [2]. Manganese-zinc ferrites are soft magnetic materials that are used as a magnetic circuit for products operating in weak sinusoidal magnetic fields in the frequency range up to 0.45 MHz. Such ferrites are intended for the manufacture of chokes, inductors operating in the elements of electronic equipment and communication equipment.

The properties of ferrites are influenced by the methods of their synthesis, and mainly researchers obtain manganese-zinc ferrites by the ceramic method [3], the solid-phase reaction method [4], hydrothermal technology [5], the sol-gel method [6, 7], the coprecipitation method [8], the combustion method [9] and others. A variety of methods for producing ferrites makes it possible to obtain ferrites with slightly different properties. One of the new methods for synthesizing materials using ceramic technology is electron beam sintering in the forevacuum pressure range. For operation in this pressure range, relatively simple forevacuum electron sources have been developed, which make it possible to obtain beams with an electron energy of up to 30 keV and a current of up to 200 mA. The peculiarity of such sources is their ability to operate at pressures of 1-100 Pa. The use of a narrowly focused electron beam propagating at relatively high pressures makes it possible to sinter ceramic and cermet samples in a much shorter time compared to traditional methods [10]. The aim of this work is to study
the possibility of electron-beam sintering of Mn-Zn ferrites in an oxygen gas atmosphere at a pressure of tens of pascal.

2. Experimental setup
As a starting material for sintering, a ferrite powder was used, consisting of grains with a size of no more than 20 μm, figure 1. Samples in the form of disks 10 mm in diameter and 3 mm thick were made from the powder by the method of isostatic uniaxial pressing. The pressing pressure for all samples was 200 kgf / cm². At lower pressure, the resulting compacts were destroyed in the process of extracting from the mold.

![Figure 1. SEM image of Mn-Zn ferrite powder used for sintering.](image)

The composition of the starting powder is shown in table 1.

| Element      | [norm. wt.%] | [norm. at.%] |
|--------------|--------------|--------------|
| Carbon       | 1.64         | 4.67         |
| Oxygen       | 23.53        | 50.16        |
| Manganese    | 14.30        | 8.88         |
| Iron         | 52.86        | 32.29        |
| Zinc         | 7.67         | 4.00         |

Electron beam sintering of preliminarily pressed samples was carried out in a vacuum chamber in an oxygen atmosphere at a pressure of 10 Pa. For sintering, an electron-beam vacuum setup equipped with a forevacuum plasma electron source was used. The sintered sample was placed in the vacuum chamber on a holder made of a refractory material. The holder was installed at a distance of 30 cm from the electronic source on its axis. For uniform heating, the electron beam was scanned over a square with side dimensions larger than the sample diameter. The power of the electron beam was increased for 20 minutes for a more uniform heating of the entire volume of ferrite. Then, when the temperature of the ferrite surface reached 1000 or 1250 degrees, the heating was stopped and the sample was kept at this temperature for 10 or 60 minutes. Thus, it was possible to determine the influence of both the temperature and the duration of the electron-beam exposure on the possibility of ferrite sintering. The surface temperature was monitored with a Raytek 1NM infrared pyrometer with a measurement range of 550-3000°C. To study the microstructure of the initial powders, as well as the elemental composition, a Hitachi-3400Sn scanning electron microscope equipped with an energy dispersive analysis system was used.
3. Experimental results and discussion

As a result of electron beam sintering, four batches of samples were obtained, differing in sintering temperature and holding time at this temperature. The temperature of isothermal holding for samples No. 1 and No. 2 was 1000°C, for samples No. 3 and No. 4 the temperature was higher and amounted to 1250°C. The holding time was also different - samples 1 and 3 were kept at a constant temperature for 10 minutes. For samples No. 2 and No. 4, the time was 60 minutes. The choice of such sintering modes is due, on the one hand, to the desire to preserve the elemental composition by lowering temperatures (samples No. 1 and No. 2), and on the other hand, to accelerate the sintering process due to a higher temperature (samples No. 3 and No. 4). After sintering, the internal structure of the samples was investigated. The results are presented in the form of photographs of a cross section, figure 2.

As can be seen from figure 2 only sample No. 4 has a uniform structure in depth. Samples 1 and 2 sintered at temperature contain an unsintered central region (figure 2, a and b). The size of this area decreases with increasing exposure time, however, it is not possible to completely eliminate the unsintered area even after exposure for 60 minutes. The possibly longer duration of the sintering process will lead to a more homogeneous sample, but this will also be associated with higher energy costs.

![Figure 2](image)

**Figure 2.** Micrographs of the cross-section of samples sintered at different temperatures $T_s$: a, b - $T_s = 1000^\circ$C; c, d - $T_s = 1250^\circ$C; and holding times $t_s$: a, c - $t_s = 10$ min; b, d - $t_s = 60$ min.

For sample No. 3 sintered at a temperature of 1250°C, delamination is also observed in the central part, however, an increase in the sintering duration to 60 minutes leads to the complete elimination of this defect. The internal structure of sample No. 4 turns out to be more uniform in depth, and such a sintering mode should be considered satisfactory.
Preservation of the elemental composition on the surface and along the depth of the samples during sintering is important. Studies of the content of Fe, Zn, Mn, and O in all four samples showed that a change in the content of elements is observed on the surface and depends on the sintering temperature. Thus, samples No. 1 and No. 2 sintered at 1000°C showed a slight change in the zinc content. The difference in the percentage of zinc between the irradiated and non-irradiated sides was about 1%. For samples No. 3 and No. 4 sintered at 1250°C, the difference in the zinc content on the surface is more significant. So, for samples No. 3 and No. 4 on the side exposed to irradiation, the zinc content turned out to be close to zero, at the same time, on the side not exposed to electron beam irradiation, zinc was present, at least in a smaller percentage, figure 3.

![Figure 3. Elemental composition on irradiated a and unirradiated b surfaces of sample No. 3.](image)

As shown by measurements of the elemental composition in depth, the distribution of zinc for all samples turned out to be uniform. The difference is observed only in its surface content for samples No. 3 and No. 4.

4. Conclusion
The results of electron-beam sintering of manganese-zinc ferrites in an oxygen-containing environment are presented. It is shown that the result of sintering depends on the sintering temperature and the holding time at this temperature. In the sintering mode with a holding time of 60 minutes at a temperature of 1250°C, it is possible to obtain a homogeneous sintered sample. A change in the content of zinc on the surface of a sintered specimen exposed to electron beam irradiation was found. In this case, zinc losses are observed only on the surface of the samples; the content practically does not change with depth. Thus, electron beam sintering in the forevacuum pressure range makes it possible to obtain homogeneous sintered samples of Mn-Zn ferrites from a fine-grained powder.

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