Investigation of Microwave Characteristics of a Composite Based on Copper-Substituted Nickel-Zinc Ferrite

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Abstract. The results of ferrite synthesis and composite preparation and investigation are presented. The Cu substituted Ni-Zn ferrite was prepared in single phase form. The morphology and chemical composition were obtained. The composites on the base of experimental Ni₀.₂₈Zn₀.₆₂Cu₀.₁Fe₂O₄ and commercially available one were prepared. The microwave properties of all samples were investigated. It was checked that the used synthesis method (temperature and time range) can be used for ferrite producing for industry. The microwave properties of composites made of synthesized ferrites are very close to those of composites based on commercially available one. All samples exhibit broad magnetic loss peaks associated with a natural ferrimagnetic resonance.

1. Introduction

Ferrite materials are widely used today. Since 1950, nickel-zinc ferrites with a spinel structure have been actively used as a material for creating various components. The properties that they possess make it possible to use them, for example, in electronics, as choke coils, inductors, transformers [1-3], as well as a material for absorbing electromagnetic radiation [4-7], which must have high electrical resistance, low dielectric loss, high hardness, high Curie temperature and chemical stability.

The production of such materials is possible using various synthesis methods, from which the final properties of the material are very strong. The most popular methods are considered: solid-phase synthesis [8], coprecipitation [9], sol-gel [10-11], auto-combustion [12-13], etc. In this work, as the most promising from the point of view of ease of preparation, method of solid-phase synthesis.

The creation of new materials is one of the main tasks for the development of science and technology. Based on this principle, scientists are faced with the task of producing materials whose properties can be manipulated. By changing the composition of the initial mixture, the properties of the material change dramatically [14-17]. There are a huge number of publications describing certain results of obtaining Ni-Zn ferrites by various methods and with the addition of one or more additional elements. Spinel ferrites with varying chemical substitution and strong correlation between functional properties attract much attention due to fundamental and practical importance [18-21]. The authors of [22] showed that the introduction of Dy³⁺ cations into Ni-Cu-Zn-ferrite leads to an increase in the magnetic properties of the product. Sm³⁺ cations, according to [23], increase the magnetic permeability of Cu-Zn-ferrite, La³⁺ cations decrease the magnetization and coercive force of Zn-Cu-Ni-ferrite [24], and also increase the activation energy of conduction. An increase in the magnetic and electrical characteristics of spinel ferrites is associated with the magnetocrystalline anisotropy of rare earth elements and with the exchange interaction between Fe³⁺ and Ln³⁺ cations.

This work presents the results of obtaining and studying nickel-zinc ferrite doped with copper.
2. Experimental

The initial components for preparing the samples were powders of iron oxides (Fe₂O₃), nickel (NiO), zinc (ZnO), and copper (CuO). All the chemicals used were of analytical grade.

The starting materials were measured in the specified stoichiometric ratios, mixed and ground for 30 minutes in an agate mortar. Table 1 shows the calculation of the mass fraction of the starting materials required for the synthesis of samples in the total mass of the charge.

| Target composition | Charge composition (wt.%) |
|--------------------|---------------------------|
| Ni₀.₂₄ZnC₀.₆₅Cu₀.₁Fe₂O₄ | Ni  | Zn  | Cu  | Fe  |
|                     | 0.08 | 0.22 | 0.03 | 0.67 |

After grinding, the resulting mixture was compressed into a tablet. The pressing was carried out in a metal mold with a diameter of 20 mm using a laboratory hydraulic press. The pressing force was 3 t/cm².

The resulting tablets were placed on a platinum sheet in a high-temperature electric furnace and sintered at temperatures of 1150 °C for 5 hours. The platinum backing is required to prevent the sample from interacting with the furnace lining elements. The furnace heating rate was 400 °C/hour. The furnace cooling rate to 900 °C was 100 °C/hour, at lower temperatures the cooling rate was not controlled.

The phase composition and structure of the obtained samples were studied using a Rigaku diffractometer, model Optima IV (Cu radiation). The elemental composition was studied using a Jeol JSM7001F scanning electron microscope equipped with an INCA-max 80 X-ray energy dispersive spectrometer (Oxford Instruments). Figure 1 shows SEM image of obtained ferrite sample. The crystals are typically shaped for spinel structure. Figure 2 shows the X-ray diffraction spectra of the synthesized sample. Figure 1 shows that the obtained sample is monophasic and have a spinel structure. The dashes indicate the literature data [25]. Table 2 presents the chemical composition and average sample formula.

![Figure 1. Electronic image obtained from a sample cleavage Ni0.24ZnC0.65Cu0.1Fe2O4.](image-url)
Figure 2. X-ray diffraction pattern of experimental Ni0.24Zn0.65 Cu0.1Fe2O4 sample. The dashes indicate the literature data [25].

Table 2. The chemical composition and average sample formulas.

| Target composition | Charge composition (wt.%) | Sample formula |
|--------------------|----------------------------|----------------|
| Ni_{0.24}Zn_{0.65}Cu_{0.1}Fe_{2}O_{4} | O 31.11 Fe 31.81 Ni 4.26 Cu 1.49 Zn 9.34 | Ni_{0.25}Zn_{0.62}Cu_{0.1}Fe_{2}O_{4} |

The frequency dependences of complex permeability and permittivity are measured in the frequency range of 0.1 to 20 GHz by the transmission-reflection (Nicolson-Ross) technique [26] in a 7/3 mm coaxial line. The sintered ceramics and composite samples for microwave measurements are shaped to fit the coaxial line.

Composite samples for microwave study are prepared by manual mixing of the ferrite powder with molten paraffin wax. The volume fractions of the powder in composite samples are 10% and 30%. The thickness of the samples is about 2 mm. Figure 3 presents the measured frequency dependences of permeability and permittivity of composite samples.

The microwave properties of composites made of synthesized ferrites are very close to those of composites based on commercially available one. All samples exhibit broad magnetic loss peaks associated with a natural ferrimagnetic resonance. A small discrepancy in the permittivity may be due to the presence of inhomogeneities in the samples or to a gap in the line during measurement.

The magnetic loss peak shifts toward low frequencies, from about 1 GHz to about 0.6 GHz with volume fraction increase. This can be attributed to the interaction between ferrite particles in composites.

The measured frequency dependences of permeability and permittivity of sintered sample are shown in Figure 4.

The frequency of the magnetic peak loss is beyond the region if the measurement. The shift of the magnetic peak loss of composites toward high frequency compared to sintered samples may be due to the influence of a demagnetizing field on the ferrite particles [27].

3. Conclusion

In the current paper the results of ferrite synthesis and composite preparation and investigation are presented. The Cu substituted Ni-Zn ferrite was prepared in single phase form. The morphology and chemical composition were obtained. The composites on the base of experimental Ni_{0.25}Zn_{0.62}Cu_{0.1}Fe_{2}O_{4} and commercially available one were prepared. The microwave properties of all samples were investigated. It was checked that the used synthesis method (temperature and time range) can be used for ferrite producing for industry.
**Figure 3.** The frequency dependences of permeability and permittivity of composite samples.

**Figure 4.** The frequency dependences of permeability and permittivity of ferrite samples.
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Acknowledgments
This research was funded by RFBR, project no. 20-52-53020. In part of the ferrites preparation the work was supported by RFBR, project no. 20-08-00716.