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Effect of Copper Substitution on the Structural, Magnetic, and Electrical Properties of NiZn Ferrites

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Abstract: *NiCuZn ferrites were synthesized using the auto-combustion method, a self-propagating process involving metal nitrates and glycine. This study investigates the structural, magnetic, and electrical properties of the resulting ferrites. X-ray diffraction analysis revealed that the crystallite size of the NiCuZn ferrites was below 120 nm. The ferrite exhibited a single-phase cubic spinel structure after calcination at 600°C and sintering at 900°C, demonstrating typical ferrimagnetic behavior. Both NiZn and NiCuZn ferrite compositions displayed a polycrystalline structure with a space group number of 227. The saturation magnetization of the NiCuZn ferrites was approximately 70 emu/g, consistent with published values. Additionally, the ferrite compositions showed high electrical resistivity, reaching 10^6 Ω-cm. These findings suggest that the synthesized ferrites, with their high electrical resistivity and substantial saturation magnetization, are well-suited for use in the fabrication of multilayer chip inductors (MLCI)..*

Keywords: NiCuZn ferrite, Auto-combustion method, magnetization, XRD

I. INTRODUCTION

The miniaturization and integration of magnetic components are critical for advancing electronic device technology. Recently, there has been a growing trend toward more compact designs in electronics, leading to the development of surface-mount devices (SMDs). These devices, including multilayer chip inductors, LC filters, chip capacitors, and beads, are characterized by their small, lightweight, and compact nature, making them ideal for high-density packaging [1]. The miniaturization of SMDs not only enables more efficient use of space but also enhances the performance of electronic devices. With this shift toward smaller, denser, and lighter components, the demand for high-density packaging has grown.

Multilayer chip inductors (MLCIs), known for their magnetic shielding properties, play a vital role in suppressing high-frequency noise while allowing low frequency signals to pass through. Spinel ferrites, particularly nickel and nickel-zinc ferrites, are commonly used in multilayer applications. Nickel-zinc ferrite is a soft magnetic material with low coercivity and high electrical resistivity, making it highly suitable for inductors and transformers in power and telecommunication electronics, especially in the megahertz frequency range [2-3]. In addition to serving as core materials for these components, nickel-zinc ferrites are widely used for fabrication of MLCIs at lower megahertz frequencies. MLCIs, being laminated and equipped with magnetic shielding characteristics, effectively prevent electromagnetic interference while maintaining high performance.

For the fabrication of MLCIs, NiZn, and NiCuZn ferrite co-fired with pure silver below 900°C is a key factor in the sintering process. The sintering of silver (which has a melting point of approximately 960°C) is crucial for achieving high-quality inductors [4]. The substitution of copper ions in the NiZn ferrite composition reduces the densification temperature to around 900°C, compared to the typical 1100-1200°C for conventional ferrites, thus enabling use of silver conductors for component fabrication [5-7]. To ensure the reliable performance of these devices, it is essential to investigate ferrite compositions co-fired with silver electrodes at temperatures below the melting point of silver.

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This study aims to explore the impact of copper substitution on the electrical and magnetic properties of NiZn ferrites, sintered at lower temperatures with varying compositions. The NiZn and NiCuZn ferrite materials are synthesized using the auto-combustion chemical technique, which produces fine powders at the nanoscale. This method effectively promotes low-temperature sintering, enabling ferrite synthesis at approximately 900°C, making it suitable for multilayer chip inductor (MLCI) applications. Additionally, the study seeks to characterize the materials through various techniques, including X-ray diffraction (XRD) and Vibrating Sample Magnetometry (VSM).

II. EXPERIMENTAL

Auto-combustion synthesis was initiated by weighing and mixing the stoichiometric amounts of nitrate salts and glycine. Analytical-grade nickel nitrate [$Ni(NO_3)_2$. $6H_2O$], copper nitrate [$Cu(NO_3)_2.3H_2O$], zinc nitrate [$Zn(NO_3)_2.6H_2O$], ferric nitrate [$Fe(NO_3)_3.9H_2O$], and glycine [NH_2CH_2COOH], all obtained from Merck India Chemicals, were used as chemical reagents. Separate aqueous solutions of these reagents were prepared and then thoroughly combined. The solutions of nitrates and glycine were mixed with double-distilled water and stirred continuously for 1 hour, maintaining a molar ratio of nitrates to glycine at 1:1. The resulting homogeneous solution was gradually evaporated on a hot plate to form a viscous gel, which was then heated at 350°C for 30 minutes. During this process, the gel expanded into a fluffy mass, which eventually broke into brittle flakes. The decomposed powder was ground in a pestle and mortar, and then calcined at 600°C for 2 hours. The phase purity and crystallinity of the calcined powders were examined using powder X-ray diffraction (Bruker X8) with CuKα radiation. Following calcination, the ferrite specimens were sintered at 900°C for 1 hour in a muffle furnace, with a heating rate of 10°C/min. The sintered specimens were used to study its magnetic properties by measuring the M-*H* curve using a Vibrating Sample Magnetometer (Applied Research Laboratory, Princeton, USA). The magnetic moment per unit gram was measured across a magnetic field range of -10,000 to 10,000 Gauss at room temperature. Bulk resistivity measurements of the heat-treated samples were carried out by performing alternating polarity tests on samples mounted on a custom-made test fixture (with ring and guarded electrode) in combination with a high-impedance electrometer (Keithley 6517A) [8]. The calcined powder samples were compacted in circular shape (ϕ = 10 mm) with 1-2 drops of 2% polyvinyl alcohol (PVA) as a binder to prepare green pellets suitable for resistivity measurements. The pellets were sintered at 900°C for 1 hour in a muffle furnace.

III. RESULT AND DISCUSSION

The X-ray diffraction patterns of the as-synthesized ferrite composition i.e. $Ni_xCu_{0.5-x}Zn_{0.5}Fe_2O_4$ (x = 0.2 & 0.5) calcined at 600°C are shown in **Figure 1**. The X-ray diffraction pattern confirms the formation of good crystallization with a single phase of cubic spinel structure. The XRD patterns do not show impurity peaks, including those of residual reactants, implying the formation of single-phase NiZn and NiCuZn ferrite. The diffraction peaks of $Ni_{0.5}Zn_{0.5}Fe_2O_4$ could be identified with spinel structure of NiZn ferrite in space group Fd-3m (no. 227), corresponding to JCPDS reference no. 08-0234 [9]. The lattice constant for $Ni_{0.5}Zn_{0.5}$ ferrite was found to be 8.39 Å. The lattice constant decreased from 8.39 Å to 8.37 Å as the nickel content in the ferrite composition decreased. This reduction in lattice constant was attributed to the substitution of Ni^{2+} ion (0.74Å) with Cu^{2+} ion (0.70Å) at the *B*-site. [10]. It is well known that the Cu^{2+} and Ni^{2+} ion will prefer the octahedral *B*-site and the Zn ion will only prefer the tetrahedral *A*-site, which will eventually share with the remaining Fe^{3+} ions. It is well understood that the Zn^{2+} ions have a strong tendency to form covalent bonds directed towards the corner of a strong regular tetrahedron, which is the reason why the Zn ion will sit at the tetrahedral *A*-site with Fe^{3+} ion [11]. The lattice constant for $Ni_{0.2}Cu_{0.3}Zn_{0.5}Fe_2O_4$ was close to 8.37Å.

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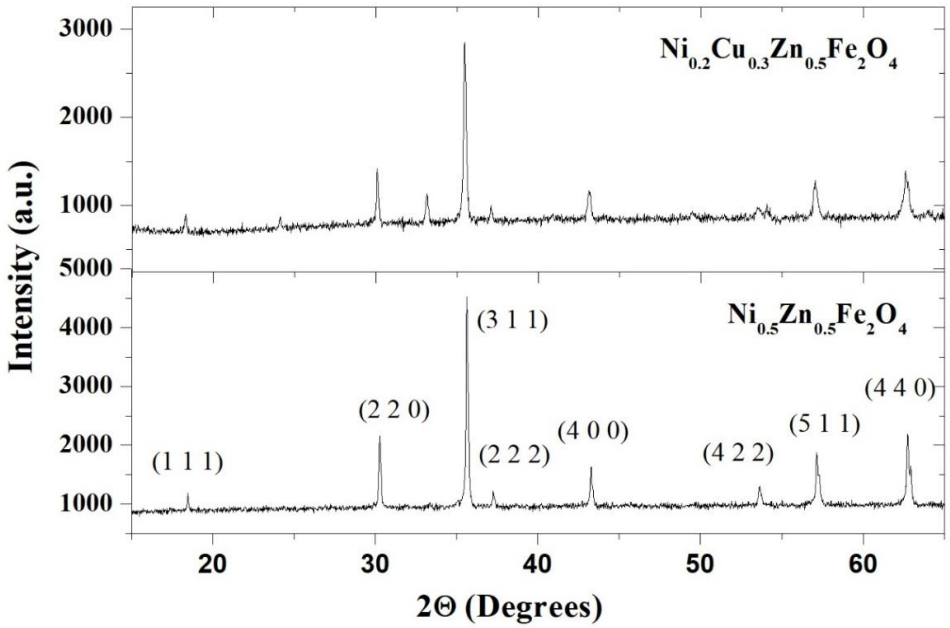

The magnetic properties of the NiZn ferrite and NiCuZn ferrite have been determined at room temperature using a Vibrating Sample Magnetometer. The M-*H* curve of the $Ni_xCu_{0.5-x}Zn_{0.5}Fe_2O_4$ (x = 0.2 and 0.5) calcined at 600°C is shown in **Figure 2**. Further, Table 1 shows the magnetization values of the $Ni_xCu_{0.5-x}Zn_{0.5}Fe_2O_4$ (x = 0.2 and 0.5) calcined at 600°C and subsequently sintered at 900°C. As shown by the VSM data, NiZn and NiCuZn ferrite particles exhibit a ferrimagnetic nature. The saturation magnetization, σ_s values decrease with the addition of copper content as seen from the results tabulated above. The saturation magnetization for NiZn ferrite composition varies from ~ 70 emu/gm and decreases to ~40 emu/gm with the addition of the copper content, regardless of the processing temperature. This reduction is attributed to the smaller magnetic moment of the Cu^{2+} ion (1.3 μ_B) compared to that of Ni^{2+} (2.3 μ_B). Consequently, substituting Ni^{2+} with Cu^{2+} reduces the overall magnetic moment and weakens the exchange interaction between the A and B sublattices. Additionally, the remanent magnetization (σr) of the samples shows a similar trend to that of the saturation magnetization. The saturation magnetization (σ_s) tends to be higher after sintering at 900°C compared to calcination at 600°C for both compositions. The increase in saturation magnetization with sintering suggests that higher temperatures lead to increased particle size and crystallinity, which enhances the saturation magnetization in the sintered specimens. The coercivity (H_c) of the sintered sample is lower than that of the specimen calcined at 600°C. This decrease in coercivity with increasing sintering temperature may be attributed to the promotion of grain growth at higher temperatures, which results in larger magnetic domains. The enlargement of these domains makes it more challenging to reverse the magnetization direction, thereby leading to a reduction in the coercive field.

Composition	**Calcination/ Sintering**	σ_s **(emu/gm)**	σ_r **(emu/gm)**	H_c **(Gauss)**
$Ni_{0.5}Zn_{0.5}Fe_2O_4$	Calcination at 600°C	70	12	109
	Sintering at 900°C	73	8	83
$Ni_{0.2}Cu_{0.3}Zn_{0.5}Fe_2O_4$	Calcination at 600°C	41	7	136
	Sintering at 900°C	44	5	87

Table 1: Magnetization values of NiZn and NiCuZn ferrite composition calcined at 600°C and sintered at 900°C

The room-temperature bulk resistivity values for the NiZn and NiCuZn ferrite are of the order of 10^6 Ω-cm. The bulk resistivity in excess of 10^6 Ω-cm together with the optimum magnetic properties, make these materials suitable for MLCI applications.

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IV. CONCLUSION

In this study, NiCuZn ferrites were successfully synthesized using the auto-combustion method, and their structural, magnetic, and electrical properties were thoroughly investigated. The X-ray diffraction analysis confirmed the formation of a single-phase cubic spinel structure for both NiZn and NiCuZn ferrites, with crystallite sizes below 120 nm. The lattice constant was found to decrease from 8.39 Å to 8.37 Å with the substitution of Ni^{2+} ion by Cu^{2+} ion. Vibrating Sample Magnetometry (VSM) revealed that the ferrites exhibited ferrimagnetic behavior, with saturation magnetization values for NiZn ferrite around 70 emu/g, which decreased to approximately 40 emu/g with the incorporation of copper. The sintering process at 900°C led to an increase in saturation magnetization, likely due to enhanced particle size and crystallinity. Additionally, the coercivity decreased upon sintering, which is attributed to grain growth at higher sintering temperatures.

The synthesized ferrites also demonstrated high electrical resistivity, exceeding 10^6 Ω-cm at room temperature, making them suitable for applications in multilayer chip inductors (MLCIs). The combination of high resistivity, low coercivity and moderate to high saturation magnetization, indicates that these materials are well-suited for use in high-frequency electronic devices. In conclusion, the synthesized NiCuZn ferrites exhibit a distinct combination of structural and magnetic characteristics, positioning them as promising candidates for multilayer chip inductor (MLCI) applications and other advanced electronic devices that require low-temperature sintering.

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