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Electromagnetic properties of Mn-doped NiCuZn-ferrites

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Abstract

In this paper, the effect of Mn^{3+} doping on the high- and low-frequency electromagnetic properties of NiCuZn-ferrites is investigated. There is an optimum Mn^{3+} concentration at which low-frequency properties such as initial permeability and power losses attain maximum and minimum values, respectively. Simultaneously, the high-frequency losses increase. This optimum Mn^{3+} concentration is found to depend on iron deficiency of material composition. The low- and high-frequency behaviors of the material are found to be very consistent with the characteristics of the microstructure. An explanation according to which manganese, when added at concentrations where the total trivalent cation amount of the material remains slightly below stoichiometry (i.e. $Fe^{3+}+Mn^{3+}\leqslant 2$), acts as grain growth promoter is proposed. At higher concentrations, the fill of the cation vacancies depresses grain growth and results to fine microstructures, which is negative for the low-frequency but positive for the high-frequency properties.
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Keywords: NiCuZn-ferrite; Manganese doping; Microstructure; Soft ferrite; Magnetic property

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1. Introduction

NiZn-ferrites constitute an important category of materials for manufacturing high-frequency bulk inductive components. Their superior high-frequency performance compared to their most important counterpart, MnZn-ferrites, is mainly due to their high specific resistivity (e.g. $\sim 10^2$–$10^3\,\Omega\,cm$ for MnZn-ferrites, $\sim 10^6$–$10^8\,\Omega\,cm$ for NiZn-ferrites [1]) that depresses the generation of ohmic eddy current losses. However, the magnetic permeability of NiZn-ferrites is generally less than that of MnZn-ferrites. Recently, NiZn-ferrites also found applications at low frequencies (e.g. ~50 kHz). Their higher losses when compared to MnZn-ferrites are counteracted by their ability to withstand breakthrough due to high voltages—a property that again resides in the high specific resistivity.

The development of surface mounting and function integration technology in the last decade brought about the necessity of manufacturing multilayer inductive components or multilayer semiconductor substrates that integrate inductive, dielectric or resistive functions. NiZn-ferrite materials with typical sintering temperatures higher than 1250 °C could only be compatible with other electronic materials and metal electrodes if their sintering temperatures were significantly reduced. Furthermore, the lower sintering temperature is of importance also to the bulk component industry since it is associated with reduction of zinc depletion during sintering and manufacturing costs related to energy consumption.

To this end, the effect of several additives on the sintering temperature of NiZn-ferrites has been investigated [2–4]. The most interesting in terms of magnetic performance has been the substitution of nickel with copper and these substitutions has given rise to the development of a new material family, NiCuZn-ferrites [5–7]. Although the mechanism by which copper enhances densification and lowers the sintering temperature is not fully clear, it is generally accepted that copper promotes densification through liquid-phase sintering.

The introduction of trivalent manganese, initially in NiZn-ferrites [8] and subsequently in NiCuZn-ferrites [9], has attracted considerable scientific attention, as it constitutes an alternative way of tailoring the magnetic

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properties. It is proposed that Mn^{3+}, when present at certain concentrations, induces magnetostriction compensation and subsequently affects magnetic permeability [10]. The effect of manganese doping on the microstructure and its implications to the low- and high-frequency magnetic behaviors have not been systematically investigated.

In this article, the effect of manganese doping on microstructure and on low- and high-frequency properties of NiCuZn-ferrites is investigated in relation to the iron deficiency. It appears that Mn^{3+}, besides being a magnetostriction compensation additive, acts also as a microstructure control additive that can be used to tailor the high- or low-frequency magnetic losses in NiCuZn-ferrites.

2. Materials and methods

NiCuZn-ferrites of the chemical formula $(\mathrm{Ni}_{0.3}\mathrm{Cu}_{0.07}\mathrm{Zn}_{0.63})\mathrm{Mn}_x\mathrm{Fe}z\mathrm{O}_{4-\delta}$ (z and x will be specified in the following paragraphs) are synthesized with the mixed oxide method. The oxide raw materials ($\mathrm{Fe_2O_3}$, ZnO, CuO, NiO, Merck, analytical grade) are initially weighed according to the proportions required by the chemical formula, up to a total 500 g synthesis batch. The raw materials are wet mixed for 3 h, dried and prefired to 750 °C for 2 h in air. The prefired powder is subsequently subjected to ball milling for 3 h. Prior to the wet mixing, $\mathrm{Mn_3O_4}$ is added to the powder at appropriate amounts. The dried and milled powder is then granulated in a roll granulator with the addition of 10 wt% binder consisting of 2 wt% aqueous solution of polyvinyl alcohol (Merck, Analytical grade, molecular weight 72,000). Ring-shaped specimens, with an internal diameter of 9 mm, an external diameter of 14 mm and a height of 5 mm, are formed by uniaxial pressing of the granulated powder at a compaction pressure of 10 MPa. The pressed specimens are finally sintered at 1125 °C in air.

The morphological investigations of the microstructures are performed with a scanning electron microscope (SEM) on the polished surfaces of the cross sections of the specimens that have been subjected to thermal etching in order to reveal the grain boundaries. The two-dimensional grain size distributions are calculated by digital processing of the microstructure images. Chemical analysis for composition determination of sintered specimens is performed with X-ray fluorescence (XRF; Philips PW1480).

The low-frequency magnetic properties are measured by a programmable unit with an impedance-gain analyzer (Agilent 4284A) equipped with an oscilloscope (Tektronix TDS 714L), power amplifier (Crown micro-tech 600) and frequency generator (Agilent 33120A) on specimens wound with copper wires to form inductors, from the alternating voltage across the winding and the in-phase component of the current before and after the specimen. The high-frequency magnetic properties are measured by an impedance/materials analyzer (E4991A, 1 MHz to 3 GHz) equipped with a magnetic material test fixture (Agilent 16454A) for toroidal one-turn coils. Calibration follows the short-open-load method.

3. Results

In Fig. 1a, the effect of the weighed Mn additions on the coercivity field of $(\mathrm{Ni}_{0.3}\mathrm{Cu}_{0.07}\mathrm{Zn}_{0.63})\mathrm{Mn}_x\mathrm{Fe}_z\mathrm{O}_{4-\delta}$ is shown for different weights in iron contents z. As can be seen, the effect of Mn additions depends strongly on the iron deficiency of the ferrites. For $z = 1.93$ or 1.94, the coercivity field initially decreases, passes through a minimum and subsequently increases. For $z = 1.98$, no initial decrease is observed. Instead, the coercivity field increases with increasing Mn content. The Mn content that corresponds to the minimum coercivity field also seems to depend on the iron content. The lower the iron content

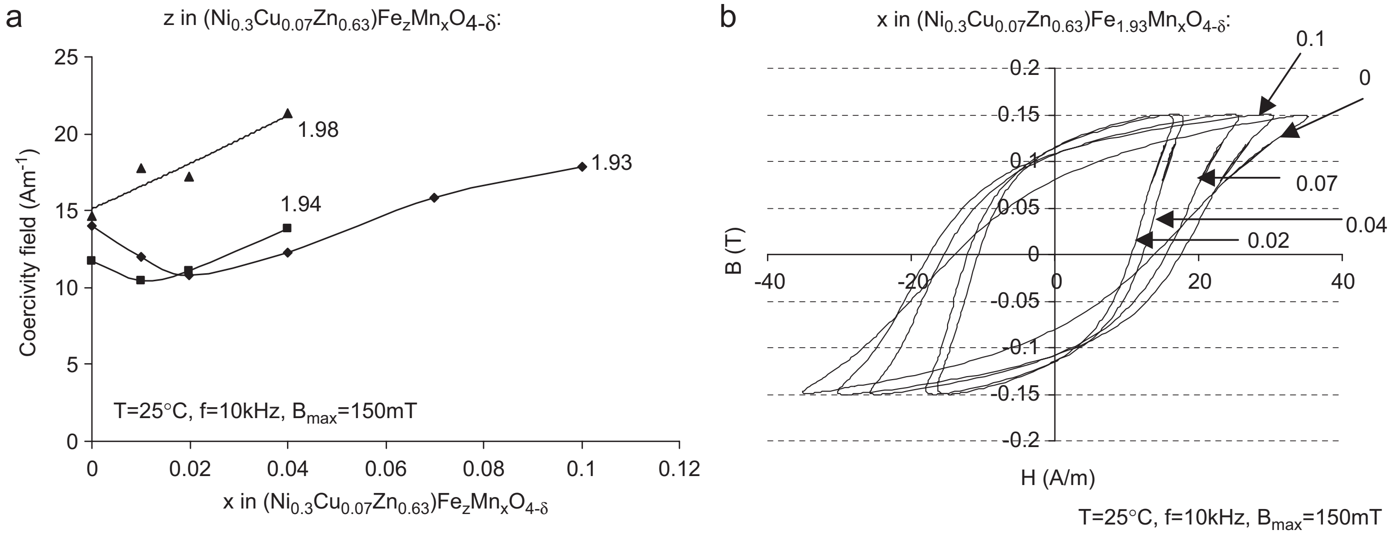

Fig. 1. (a) The effect of manganese additions x on the coercivity field of $(\mathrm{Ni}_{0.3}\mathrm{Cu}_{0.07}\mathrm{Zn}_{0.63})\mathrm{Mn}_x\mathrm{Fe}_z\mathrm{O}_{4-\delta}$ ferrites, for different iron contents z. (b) The hysteresis loops as a function of manganese content in $(\mathrm{Ni}_{0.3}\mathrm{Cu}_{0.07}\mathrm{Zn}_{0.63})\mathrm{Mn}_x\mathrm{Fe}_{1.93}\mathrm{O}_{4-\delta}$ ferrites.

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