THE INFLUENCE OF PARTICLE SIZE AND SUBSTITUENT CONTENTS ON THE MAGNETIC PROPERTIES OF Be OR Cu SUBSTITUTED NiZn FERRITES

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Summary Polycrystalline NiZn ferrites with the chemical formula $(Ni_{0.3}Zn_{0.7})_{1-x}Me_xFe_2O_4$ where Me being Be or Cu and x=0.05, 0.1 and 0.25 have been prepared by a ceramic method. Certain magnetic properties such as the coercivity H_c, initial permeability μ_i, real part of the complex permeability μ' and saturation magnetization M_s of the ferrites have been measured and discussed from the point of view of the substituting element type and contents as well as the average particle size.

1. INTRODUCTION

NiZn ferrites are a well-known class of technologically important ferrites. These materials are of special significance at high frequencies, because of high resistivity and thus low eddy current losses. The change of magneto-crystalline and anisotropy can provide the basis for tailoring soft magnetic properties at higher magnetic flux densities. These properties, i.e. the hysteresis loop shape, resistivity, etc. are known to be dependent upon the purity of starting compounds and the technological process nuances, such as the sintering temperature and atmosphere which influence the microstructure and final sample composition. The grain size, grain boundaries, porosity and stoichiometry are the important factors in regard to the technology. The substitution of selected diamagnetic ions offers an opportunity for tailoring the soft magnetic properties and/or the resistivity of NiZn ferrites [1-4].

The paper reviews the relationship between the chemical composition (associated with the microstructure and grain size variety) and magnetism in Cu or Be substituted NiZn ferrites. The substitution of ions offers one possibility for tailoring the magnetic properties in a wide frequency range.

2. EXPERIMENTS

NiZn ferrites with the chemical composition $(Ni_{0.3}Zn_{0.7})_{1-x}Me_xFe_2O_4$ with Me being Be or Cu substituents and x=0.05, 0.1 and 0.25 have been prepared by a ceramic method and the samples were sintered at 1200°C for 6 hours in air and then slowly cooled in a furnace. The saturation magnetization M_s was measured by means of a vibration sample magnetometer. The complex permeability of prepared ferrite samples were obtained by measuring the input complex impedance of a standard coaxial airline filled with a sample of toroidal shape using two types of impedance analysers in the frequency range from 100 kHz to 100 MHz.

3. RESULTS AND DISCUSSION

The reduction of particle size is a well-known method to subdivide the ferrite into single-domain particles, which increases the coercivity H_c towards the maximum value controlled by the sample anisotropies. Our experimental results are shown in Fig. 1, where the coercivity of Cu or Be doped NiZn ferrites is plotted against the average grain size D. The dependence of initial permeability μ_i for both Cu or Be substituted $(Ni_{0.3}Zn_{0.7})_{1-x}Me_xFe_2O_4$ ferrites where $x \in \langle 0, 0.25 \rangle$ on the grain size is shown in Fig. 1, too. The presented experimental data of H_c and μ_i fit well into the predictions following from the classical grain size theory, [5]. The coercivity and initial permeability are closely correlated to the effective anisotropy energy per unit volume A_e and the saturation magnetization M_s as follows:

$$H_c = g_c \frac{A_e}{M_s} \approx \frac{c_1}{M_s} \cdot \frac{1}{D} \qquad (1)$$

$$\mu_i = g_\mu \frac{M_s^2}{A_e} \approx \frac{M_s^2}{c_2} \cdot D \qquad (2)$$

where g_c and g_μ are the dimensionless factors close to unity; c_1 and c_2 are material constants. The relation of H_c proportional to $1/D$ for relatively large grain sizes (D>100 nm) reflects the classical rule that good soft magnetic properties require very large grains. The experimental data are in a good agreement with this rule. The grain size was determined by means of SEM.
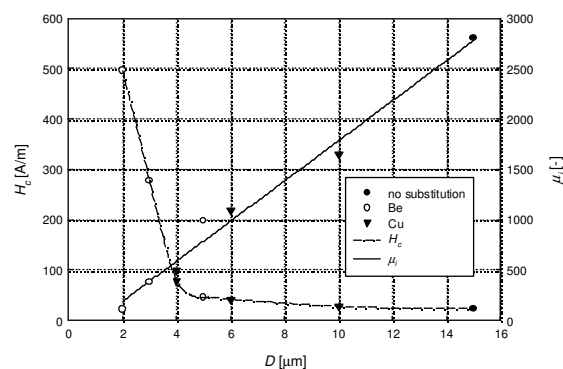

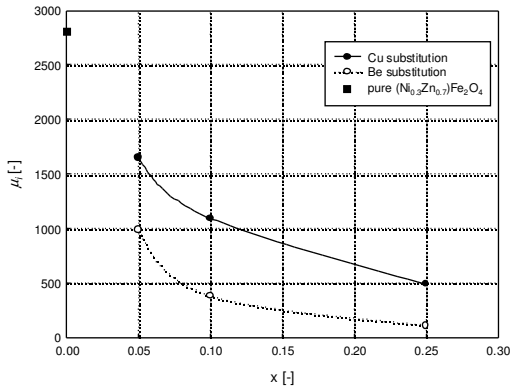*Fig. 1. The coercivity H_c and initial permeability μ_i as a function of grain size.*

Fig. 2. The initial permeability μ_i as a function of substitution coefficient x.

The measured data of initial permeability show an analogous behaviour being essentially inversely proportional to the coercivity, i.e. proportional to *D*. Obviously, the "non-magnetic" parts fill the gaps between the ferrite grains (particles) in the sample. Thus, the variation of grain size is due to Cu, Be substitution of NiZn composites and/or the sintering conditions. Probably, the increase of Be (Cu) contents reduces simultaneously the size of grains in the shown case. This fact again fits well within the assumptions of grain size theory.

On the other hand, the dependencies of initial permeability against the substitution coefficient x show the variation of μ_i values for Be or Cu substituted NiZn ferrite, see Fig. 2. The incorporation of both doping ions causes a rapid decrease of μ_i with increasing x. One can conclude that the increase of Cu (or Be) ions contents causes the decrease of Zn contents in NiZn ferrite, which is the main reason of μ_i drop with x in both NiZnCu and NiZnBe ferrite systems. From the results shown in Fig. 2 a question arises − why NiZnCu systems have in general higher values of initial permeability in comparison with NiZnBe systems. Probably, the answer is associated with the saturation magnetisation related to substitution coefficient x, see Eq. (2). To support this hypothesis, the variation of saturation magnetisation as a function of x can be seen in Fig. 3.
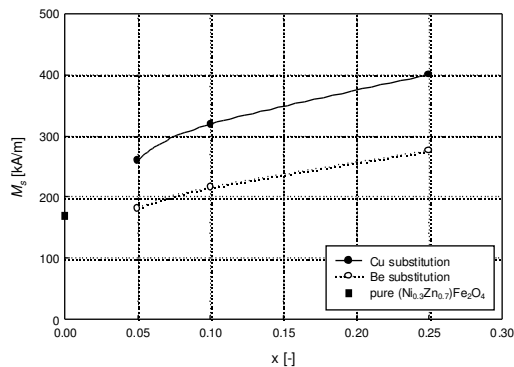

Fig. 3. The saturation magnetisation M_s as a function of substitution coefficient x.
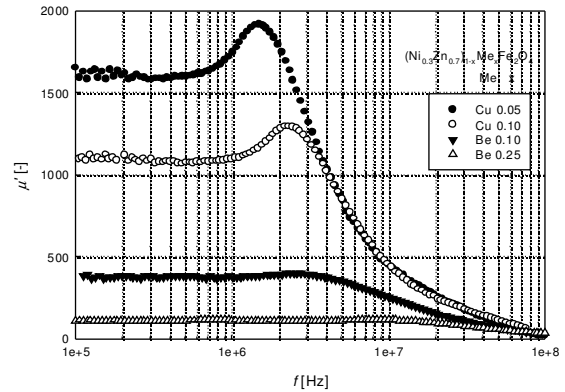

Fig. 4. The real component of complex permeability for Be or Cu substituted NiZn ferrites.

M_s values were found to be increasing with the substituent concentration x for both Be and Cu ions. It was shown [1], that the increments of saturation magnetization ΔM_s(Cu) and ΔM_s(Be) are in the coincidence with decreasing Zn contents in $(Ni_{0.3+y}Zn_{0.7-y})Fe_2O_4$ if the coefficient of Zn contents y varies from 0 to 0.25. It was also described in [1] why the Cu substituted NiZn ferrite exhibits higher values of saturation magnetisation comparing to Be substitution. As long as the initial permeability is proportional to M_s^2, higher values of initial permeability in case of Cu substitution in Fig. 2 are probably associated with higher increments ΔM_s(Cu) in the given interval of x.

The frequency dependencies of the real part of the complex permeability μ' have been studied as a function of the composition. In Fig 4, four frequency spectra of the real component of the complex permeability $\mu'(f)$ are observed to decrease with increasing of Cu or Be contents. For both substituents at x=0.05 the value of μ' at low frequencies, referred to as the initial permeability μ_i, can be explained mainly by the reversible domain walls displacement due to higher grain size values (>4nm), [6-8]. The dependencies in Fig. 4 also show, that the value of μ' is practically independent on the frequency up to few hundreds of kHz. Beyond this limit it shows a resonance peak whose frequency increases with increasing of dopant contents. The resonance peaks are more evident in case of copper substitution. In this case the initial permeability is higher, too. The increase of Cu and Be contents probably enhances the influence of effective magnetic anisotropy and the resonance is observed at higher frequencies.

The hysteresis loops for all ferrite samples were obtained at certain values of exciting magnetic field strength (H_m=25 A/m and 100 A/m) and frequencies (*f*=1 kHz and 10 kHz) by exciting the toroidal samples with a sinusoidal waveform. As an example, Fig. 5 presents the minor hysteresis loops for all ferrite samples, measured at the magnetic field intensity of H_m=25 A/m and frequency *f*=10 kHz.
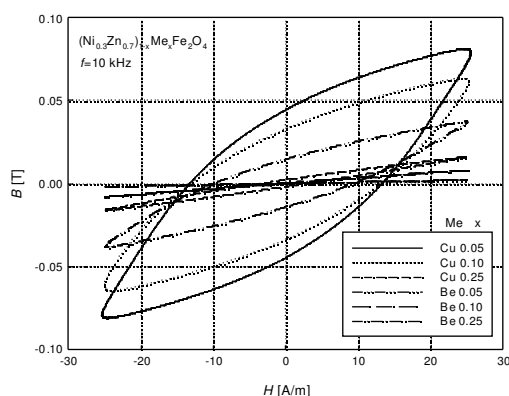
Fig. 5. The hysteresis loops for Be or Cu substituted NiZn ferrites.

Both the Be^{2+} and Cu^{2+} ions have shown predominant impact on measured hysteresis loops. Be substituted NiZn ferrites showed a little lower values of maximum magnetic flux density compared to Cu substituted NiZn ferrites. The study of the hysteresis loops also showed that with increasing the frequency the shape of the hysteresis loops at low exciting field amplitude (H_m=25 A/m) changes from straight line to elliptical one and the mean slope decreases.

4. CONCLUSION

The substitued NiZn ferrites offer an interesting system for study of the influence of divalent ions (Cu, Be, etc.) for the case in which the exchange correlation between tetrahedral and octahedral sublattices is controlled. The substitution ions offer probably one way for changing the grain size. In summary they offer a new possibility for tailoring superior soft magnetic properties comparable to those of NiZn and MnZn ferrites. The advantage is also a significantly better thermal stability of the soft magnetic properties.

The results confirmed, that the substitution of Cu or Be plays very important role in controlling the magnetic properties of $(Ni_{0.3}Zn_{0.7})_{1-x}Me_xFe_2O_4$ ferrites. Our experiments have shown a strong influence of Cu and Be contents on the initial permeability, saturation magnetisation, coercivity as well as hysteresis loop area associated with the energy loss. Thus, the substituent concentration along with proper technology becomes a strong tool for the preparation of the ferrites tailor-made for any particular application.

Acknowledgement

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