

Effect of Nb₂O₅ additions on the power loss of NiZn ferrites

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Abstract Nickel–zinc ferrite system, Ni_{0.65}Zn_{0.35}Fe₂O₄ + x Nb₂O₅ where x varies from 0.0 wt% to 1.5 wt% in steps of 0.3 wt%, has been prepared by conventional ceramic technique. The samples were sintered at 1250 °C for 4 h in air atmosphere followed by natural cooling. The power loss and microstructures of these materials are examined. Microstructures reveal that niobium oxide additions promoted grain growth with an increase in grain size from 4 μm to 13.2 μm with the increase in niobium concentration. The measured power loss at frequencies from 100 kHz to 10 MHz under different exciting flux densities from 5 mT to 30 mT was found to be low up to 3 MHz, thus making the materials suitable for power applications up to this frequency. In the total power loss, hysteresis loss is predominant below 500 kHz and eddy current loss component is much higher at higher frequencies.

Introduction

Continuous growth in power electronics industry demands processing of high performance ferrite cores, capable of operating at increasingly high frequencies, for switch mode power supplies. Many efforts have been made so far to

develop low power loss ferrite cores for work at frequencies around 1 MHz [1]. Theoretically, power loss arises mainly from hysteresis and eddy current loss sources leading to heat rise in the core and dismal performance beyond a certain frequency. There may be some residual losses also from dielectric and wall related sources, but this contribution is assumed to be negligibly small under large amplitudes of exciting magnetic flux [2], which is a requirement for these applications. Mn–Zn–Fe ferrites with high saturation magnetizations are largely used as core materials in the switching power supplies. However, in spite of several efforts to enhance the grain boundary resistivity contributions in these materials, the rather high n-type electrical grain conductivity in them restricts their usage in the said applications beyond 1 MHz due to increased eddy current losses [3]. Alternately, Ni–Zn ferrites with high resistivity are considered for improvements in switching frequencies [4]. But, the low saturation magnetizations of these ferrites and almost free ferrous content under normal preparation conditions are the factors that work against the low power loss objectives. It has been reported as quite mandatory that certain quantity of ferrous content is essential as well in the final product to realize soft saturation and low hysteresis loss, particularly, when the materials are aimed at high frequency switching power supplies [5]. Therefore, careful design of compositions and optimized synthesis in their development is required to obtain balance between the low power loss and high switching frequency.

High saturation magnetization Ni_{0.65}Zn_{0.35}Fe₂O₄ composition in the Ni_{1-x}Zn_xFe₂O₄ system is considered as basic composition for the present study. Since Nb₂O₅ is known to improve core losses in Mn–Zn ferrites [6], it is aimed at studying effect of niobium oxide additions in the above Ni–Zn ferrite composition. This paper reports and discusses the

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influence of Nb₂O₅ additions on the microstructure, permeability and power loss of Ni–Zn ferrites at high frequencies.

Experimental details

Using high purity chemicals in targeted proportions, a series of Ni–Zn ferrites with niobium oxide additions under the general formula, Ni_{0.65}Zn_{0.35}Fe₂O₄ + *x* Nb₂O₅ where *x* values ranging from 0.0 wt% to 1.5 wt% in steps of 0.3 wt%, have been prepared by conventional ceramic technique. Sintering of the samples was done at 1250 °C for 4 h in air atmosphere. The adopted sintering schedule was described elsewhere [7], but is done in such a way to retain certain quantity of divalent iron in the final product. X-ray diffraction patterns of the samples confirm single phase spinel structures, and the sample densities were approximately 90–93% of their theoretical limits [7]. The estimated lattice constant, Curie temperature and saturation magnetization of the basic composition were compared well with the respective parameters of the same composition reported earlier [8]. Micrographs are taken on fractured samples using Philips XL-50 microscope. Permeability and power loss measurements are made on toroidal samples from 100 kHz to 10 MHz using Iwatsu B-H Analyzer. Apart from the relative permeability (μ_a) values, the DC resistivity (ρ) values measured using standard two-probe method are also listed in Table 1 along with power loss and grain size data for help in analyzing the results.

Results and discussion

The micrographs of the fractured samples of Ni_{0.65}Zn_{0.35}Fe₂O₄ + *x* Nb₂O₅ (*x* = 0.0 and 1.5 wt%) are shown in Fig. 1. It is clearly evident that the additions of Nb₂O₅ have brought about marked changes in the microstructures as the multi-faceted grains in the micrographs have become increasingly sharper and larger as the niobium concentration increases. Initially, there appears a discontinuous grain

growth, as seen in the micrograph of N2, with inter-granular bridges indicating that the densification process is yet to be completed. However, subsequent niobium oxide concentrations promoted grain growth as evidenced by the increase in grain size from 5.7 μ m at 0.6 wt% concentration to 13.2 μ m at 1.5 wt% concentration, as listed in Table 1. Though there is an increase in the grain size, the grains are observed to be neither round nor uniform. The advantage of contributions to the magnetic performance by the increase in grain size may be largely offset by their irregular shapes due to demagnetizing effects. Further, there also appears more inter-granular pores as the niobium concentration increases, which may be another factor working adversely to the magnetic properties.

The typical frequency response of complex permeability for the basic Ni–Zn ferrite composition is shown in Fig. 2. The real permeability, the behavior being the same in respect of all the samples, is quite stable up to a few megahertz, beyond which it takes a small peak and then decreases rapidly which is coincided by a sharp peak exhibited by the imaginary part. Interestingly, the frequency at which the dispersion takes place has been observed to be higher for higher permeability (listed in Table 1) samples in these samples. Further, the imaginary part of the permeability in any of the samples has not been marked by broad peaks as was normally the case with relaxation. Phase difference between the applied field and magnetization of the ferrite occurs normally due to the damping of either spin motions or domain walls. If there is no damping, the imaginary part is zero for all frequencies except at relaxation frequency. But, magnetic relaxations appear as μ' decreases with increasing frequency and μ'' has a maximum near the relaxation frequency. The μ'' in the present study is not marked with maximum values good enough for relaxation. Then, if the permeability relaxation phenomena in ferrites cannot be solely explained as a superposition of domain wall motion and spin rotation, it may well be related to the behavior of the electromagnetic waves within the core. This had been well demonstrated in earlier case [9] as a resonance due to sample dimensions in Mn–Zn ferrites. Thus, the observed behavior of permeability spectra in the present study could be attributed to

Table 1 Grain size, saturation magnetization and power loss of Ni_{0.65}Zn_{0.35}Fe₂O₄ + *x* Nb₂O₅

Sample, wt%	Grain size, μ m	ρ , Ω cm	μ_a $H_m = 0.1$ Oe at 1 MHz	Power loss, kW/m ³	
				at 500 kHz	at 1 MHz
N1/0.0	~6	215	168.9	30.62	89.44
N2/0.3	~4	162	176.1	35.09	97.93
N3/0.6	5.7	162	169.0	35.08	106.24
N4/0.9	7.3	192	169.4	38.98	119.17
N5/1.2	9.7	134	160.3	37.29	113.83
N6/1.5	13.2	163	165.8	42.13	130.1

Fig. 1 Microstructure of $\text{Ni}_{0.65}\text{Zn}_{0.35}\text{Fe}_2\text{O}_4 + x \text{Nb}_2\text{O}_5$

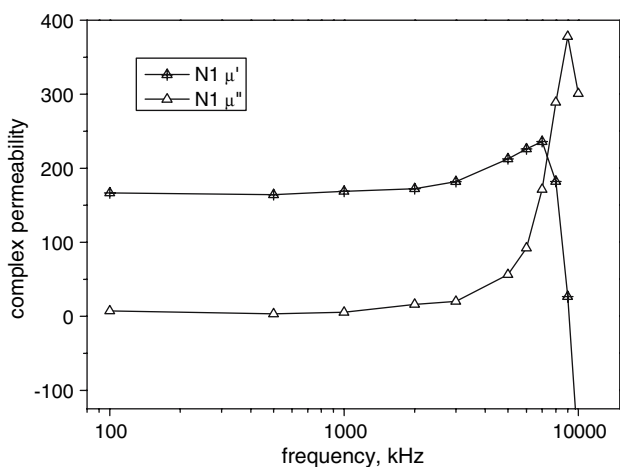
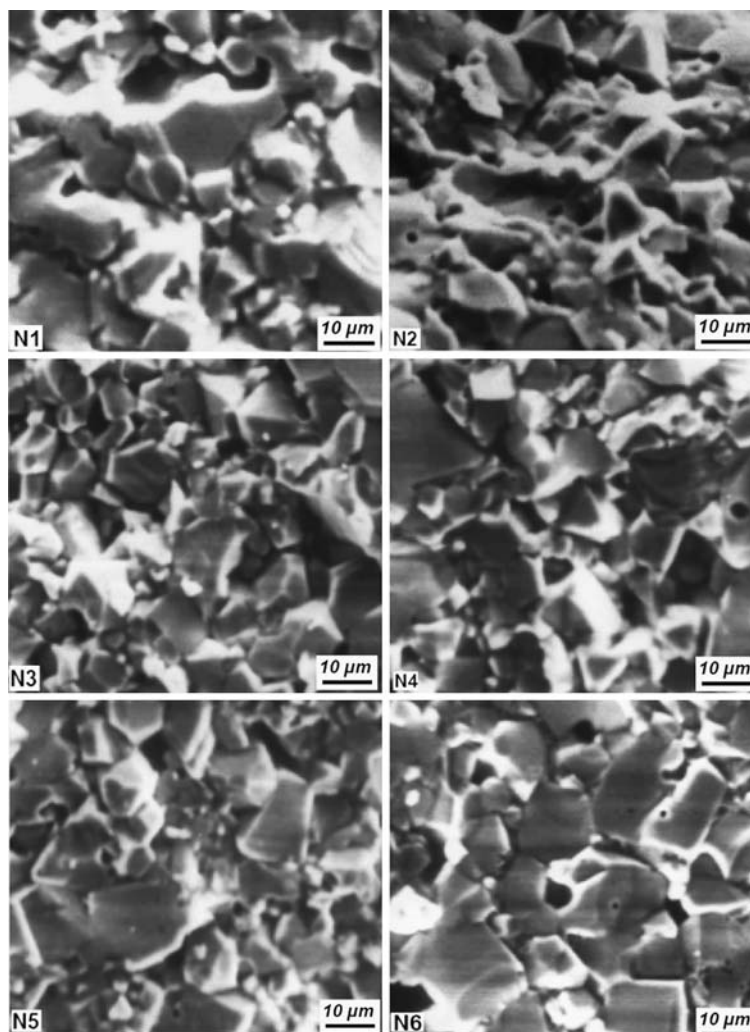


Fig. 2 Typical frequency dependence of complex permeability of $\text{Ni}_{0.65}\text{Zn}_{0.35}\text{Fe}_2\text{O}_4$ (N1)

the combining effects of both relaxation due to damping of spins and domain walls and resonance due to sample dimensions.

Power loss was measured under different exciting conditions from 5 mT to 30 mT. The variation of power loss of Ni–Zn ferrites with niobium oxide concentration at different exciting flux densities is shown in Fig. 3. The power loss has been quite low ($<1000 \text{ kW/m}^3$) for all the samples under all the exciting conditions up to 25 mT; thus making them highly suitable under these conditions as core materials for power applications. But, interestingly the Nb_2O_5 additions have not really shown improvements in the power loss in these materials. As mentioned above, despite the increase in grain size the microstructures of these samples due to their irregular grain shapes could do little to improve the magnetic loss. Also, for the same reason, the rich niobium oxide grain-boundary resistivity component in the bulk resistivity (listed in Table 1) was apparently missing, thus leading to enhanced eddy current loss in the power loss. However, the control of power loss beyond 0.6 wt% Nb_2O_5 concentration from going out of proportion is attributed to the increased grain size.

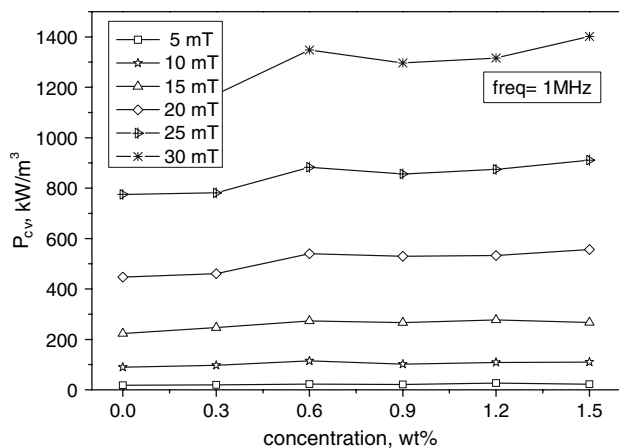


Fig. 3 Variation of power loss with niobium oxide concentration under different exciting flux densities at 1 MHz

The variation of power loss of Ni–Zn ferrites with niobium oxide concentration under the exciting flux density of 10 mT and at different frequencies up to 3 MHz is shown in Fig. 4. Once again, the power loss has been quite low (<1000 kW/m³) for all the samples up to this frequency.

In order to reveal the power loss trend over the whole frequency range studied, the frequency dependence of power loss for all the samples in the system between 100 kHz and 10 MHz at the exciting condition of $B_m = 10$ mT is shown in Fig. 5. As mentioned earlier, the power loss has remained low for all the samples up to 3 MHz beyond which it has been observed to increase rapidly. The samples with zero and small quantity of niobium oxide have recorded relatively low losses. The contributions of hysteresis and eddy current losses to the power loss as a function of frequency for the low Nb₂O₅ concentrated samples are shown as inset in Fig. 5. The hysteresis loss is predominant loss mechanism in the lower

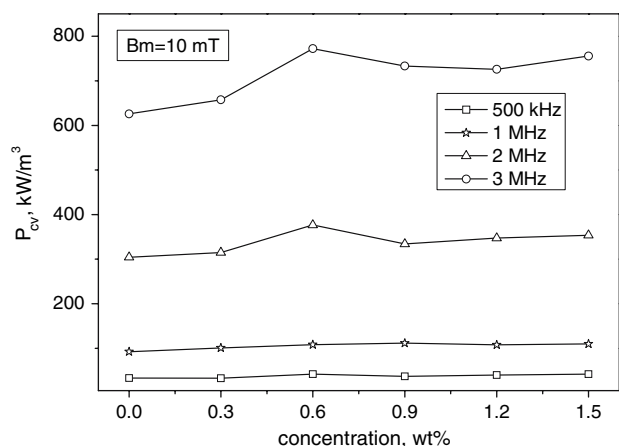


Fig. 4 Variation of power loss with niobium oxide concentration at different frequencies for the 10 mT exciting flux density

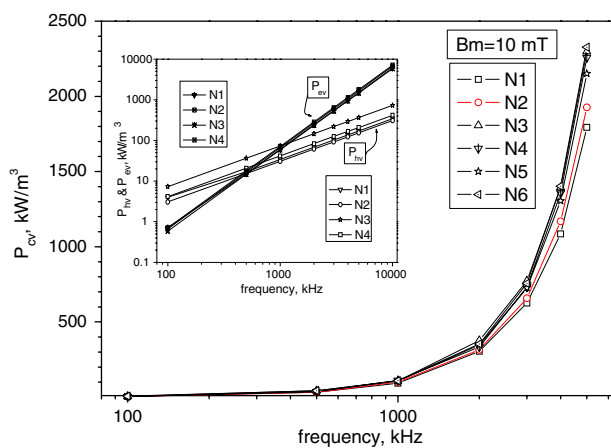


Fig. 5 Frequency dependence of power loss (P_{cv}) of Ni_{0.65}Zn_{0.35}-Fe₂O₄ + x Nb₂O₅. Inset in the figure shows contributions made by hysteresis (P_{hv}) and eddy current loss (P_{cv}) components for different samples

frequencies approximately below 500 kHz, and beyond this frequency the eddy current loss increases gradually with increasing frequency and becomes predominant in the power loss at higher frequencies.

The variation of power loss can be explained as follows: Since the basic composition in the present study is a high room temperature saturation magnetization ferrite among the entire Ni–Zn series, additions of Nb⁵⁺ ions to this composition, without a direct change in the density of the magnetic cations, are not likely to bring out major changes in the magnetic environment and the corresponding net magnetization. However, due to their incorporation particularly in small quantities, they are likely to enter the lattice and convert some Fe³⁺ ions into Fe²⁺ ions to maintain the charge balance, and thereby indirectly alter the magnetic environment slightly. In the present system, though the addition of niobium ions has not degraded the magnetization with increase in niobium oxide concentration [7] the relative permeability has been observed to degrade marginally, as listed in the Table 1. This is contrary to expectations because production of a small quantity of divalent iron both during synthesis and in the addition of Nb⁵⁺ ions is more likely, and in such case the resulting Fe²⁺ ions could be expected to make indirect anisotropy compensations and thereby to enhance the permeability to some extent. Notwithstanding to these expectations and also to the slight increase in grain size, the resulting slight degradation of permeability could be understood in terms of the defect microstructures of these samples. The microstructures with insoluble secondary phase imperfections and porosities usually pin the domain walls and decrease their contributions to the permeability. As a result of this, the hysteresis loss component has become substantial even after the niobium oxide additions.

Also, as discussed earlier, the irregular shaped grains with more inter-granular pores could have nullified the advantage of the insulating grain-boundary component of the resistivity out of niobium oxide additions. As can be seen from Table 1, all the niobium oxide doped samples have displayed smaller resistivities compared to the basic Ni–Zn ferrite sample. This is in support of the arguments made above, and explains not only the predominance of the eddy current loss component, particularly, at high frequencies but also the frequency dependence of the total power loss. However, optimum sintering schedule in combination with minor compositional modifications in these ferrites with very small quantities of niobium oxide additions would help to obtain better microstructures and higher resistivities [10], which could probably decrease power loss considerably and push resonances further up by a few megahertz.

Conclusions

Niobium oxide additions in nickel–zinc ferrites seem to have promoted densification and grain growth. Over addition of Nb₂O₅ brings, perhaps due to acceleration in grain growth, a number of open pores trapped in between the grains in microstructures and affects the high frequency magnetic behavior. The undoped and the low niobium

oxide containing materials have, particularly, displayed low power losses up to 3 MHz; thus making them suitable for power applications up to this frequency. Optimum sintering schedule with additions of small quantities of niobium oxide in high magnetization basic ferrite composition would be favorable for further studies.

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