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Studies of Some Magnetic Properties on a Non-Stoichiometric Iron-Deficit NiZn Ferrite System

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Abstract: The effect of iron deficiency of $Ni_{0.30+2x}Zn_{0.70}Fe_{2-2x}O_{4-4x}$ with X = 0.00, 0.01, 0.02, 0.03 and 0.04 system on their magnetic properties was carefully studied. X-Ray Diffraction analysis identified single-phase ferrite. It was found that sample with 0.47 mole fraction of iron oxide in the series of iron-deficit NiZn-ferrite was the most effective value for the formation of densed samples. Grain size of 31.8µm and highest theoretical density, 0.78%, were obtained for the sample. On the other hand, sample with 0.48 mole fraction of iron oxide gave the highest experimental density (5.0905 gcm^{-3}) and saturation induction (2757Gauss). It is speculated that cationic and anionic vacancies were sufficiently formed for the diffusive transportation of metal ions to occur with relative ease during sintering thus profoundly affecting the magnetic properties. The magnetic properties of some of the samples obtained are suitable as core materials for inductor.

Key Words: Non-stoichiometry, Iron Deficit, Saturation Induction

Introduction

Nickel zinc ferrite cores are commonly used for high frequency applications. As such, obtaining high-density cores, which requires careful optimization of both intrinsic and extrinsic factors is vital. We know that the departure from stoichiometry could affect the materials transport during solid-state reaction, which depends greatly on the vacancies concentration (Okomoto, 1984). As such, we tried to obtain an effective value of iron-oxide mole-fraction in samples with iron deficit composition to optimize the magnetic properties, particularly those relating to quality factors, saturation induction and operating frequencies. The effect of microstructure due to the non-stoichiometry composition was also discussed briefly.

Experiment Procedure: All the samples were prepared via conventional method. XRD measurements were carried out in Siemen D5000 machine using Cuα radiation, with λ = 1.5418Å. The scanning speed of the counter is 2θ permin. Microstructure evolution of the sintered body was systematically done by using Scanning Electron Microscopy (SEM) , JOEL-MSZ 6400 machine. The magnetic measurements were carried out after each sample was wound with 5 turns of 0.3 mm diameter insulating copper wire. The wire was scraped by using a sand paper and was then coated with tin to ensure good contact during measurements. The sample was then connected to a Hewlett Packard 4284A Precision LCR meter. A series of inductance, Ls and Q factor was recorded from the lowest to resonance frequencies. The initial permeability values were calculated by introducing Ls to the equation below:

$$\mu_i = \frac{2\pi L_s}{N^2 \mu_0 t \ln(D_o/D_i)} \qquad (1)$$

Where Ls is the series inductance, N is the number of turns, μ_0 is the permeability of free space ($4\pi \times 10^{-7}$ H/m), t the thickness, D_0 the outer diameter, and D_i is the inner diameter and t is the height of the samples. Saturation induction graphs were obtained from Walker Scientific Hysteresisgraph.

Results and Discussion

XRD patterns of all the compositions indicate complete spinel formation.

Of individual oxides were seen. All the compounds of the system crystallized out with cubic structure.

The grain size, porosity and grain boundary phase of ferrites have great influence on the domain wall width, domain structure, and domain wall mobility (Okomoto, 1984). A strong influence of vacancies on the final density and microstructure were observed.

Table1: Theoretical Density/Vacancies and Experimental Properties for Samples N1F0D, N1F1D, N1F2D, N1F3D and N1F4D

Sample	N1F0D	N1F1D	N1F2D	N1F3D	N1F4D
Composition	X=0.00	X=0.01	X=0.02	X=0.03	X=0.04
Vacancies (Theoretical)	0.00	0.16	0.32	0.48	0.64
Density (Theoretical) g/cm^3	5.2818	5.2442	5.2066	5.1691	5.1315
Density (Experimental) g/cm^3	5.2198	5.0889	5.0450	5.0899	5.0905
% Porosity (Theoretical)	1.17	2.96	3.10	1.53	0.78
Average Grain Size (µm)	75.2	41.8	48.3	31.2	28.2

Referring to Table 1, it is obvious from the theoretical calculations that the density drops with the increasing vacancies assuming the vacancies occur only on site B of the spinel structure (Fig. 2). However, there is an increase in density for samples N1F3D and N1F4D.

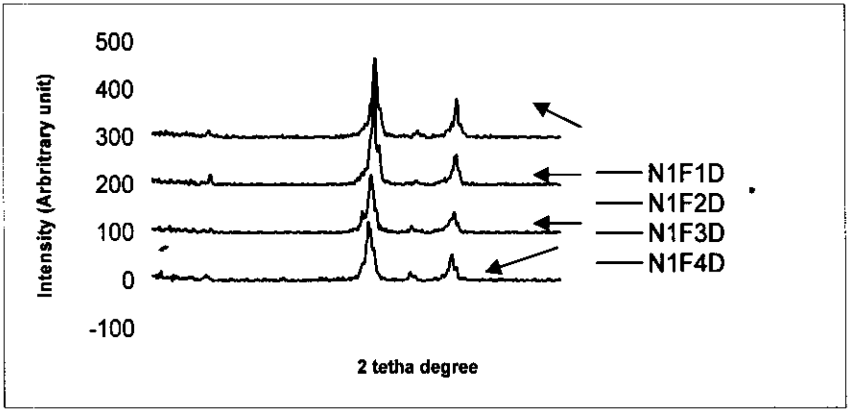


Fig. 1: X-Ray Diffraction Patterns for Samples N1F0D, N1F1D, N1F2D, N1F3D and N1F4D

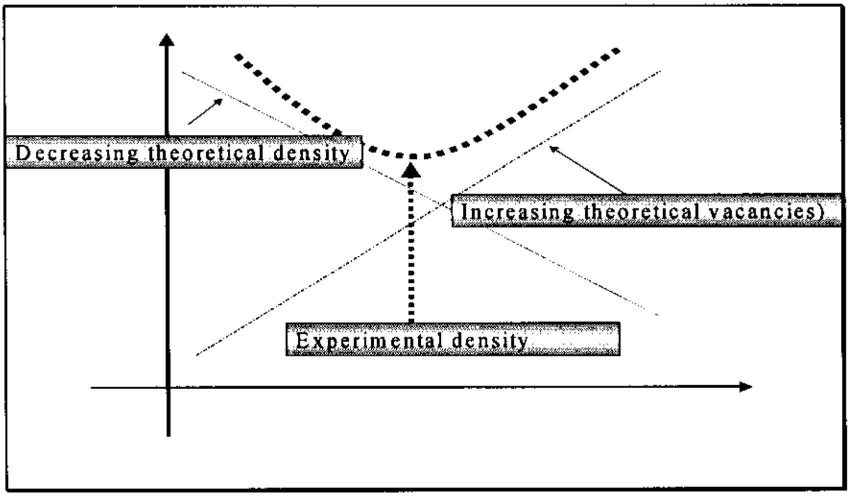
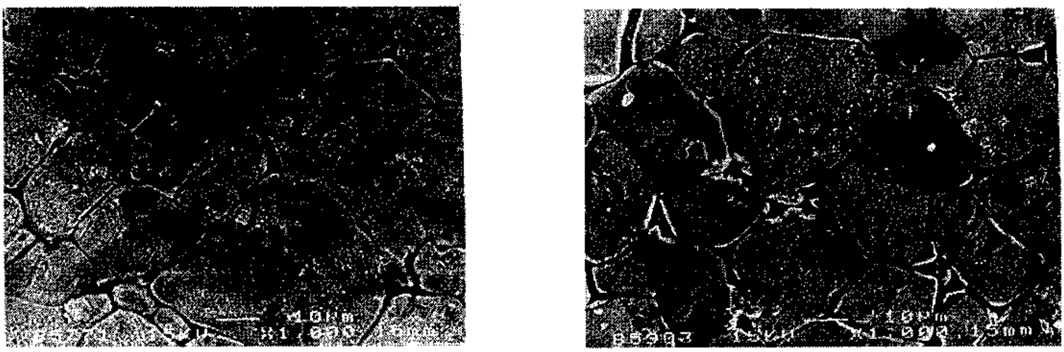


Fig. 2: A Scatch of Theoretical Density, Theoretical Vacancies and Experimental Density


N1F3D

N1F4D

Fig.3: Scanning Electron Micrographs (SEM) for Samples N1F3D and N1F4D

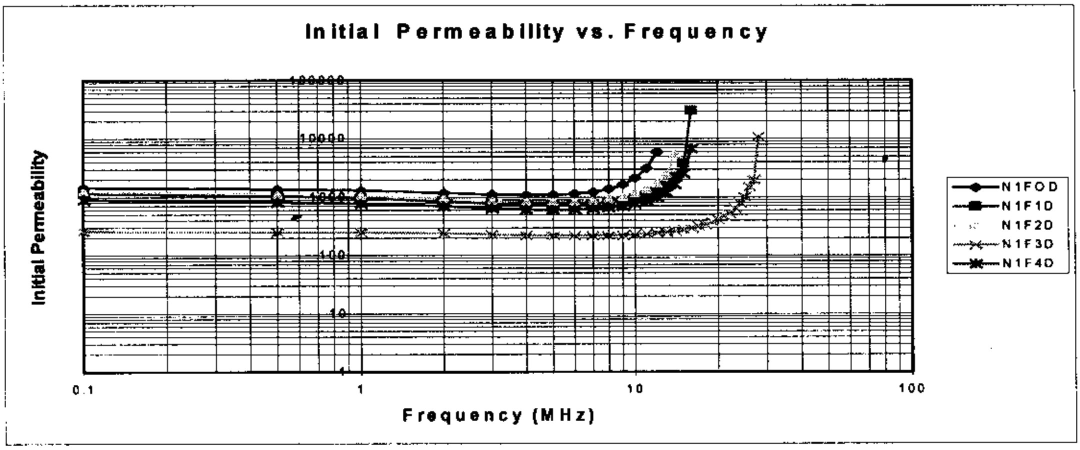


Fig.4: Initial Permeability (μ_i) vs. Frequency (MHz) for Samples N1F0D, N1F1D, N1F2D, N1F3D and N1F4D Measured at Room Temperature

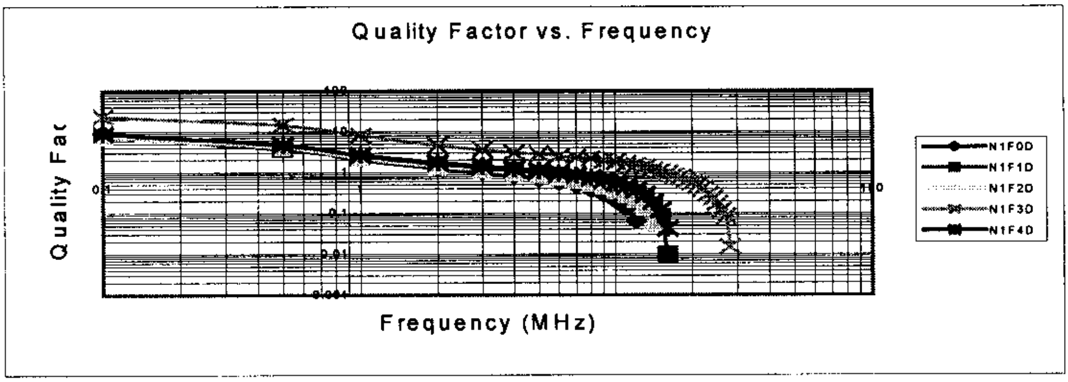
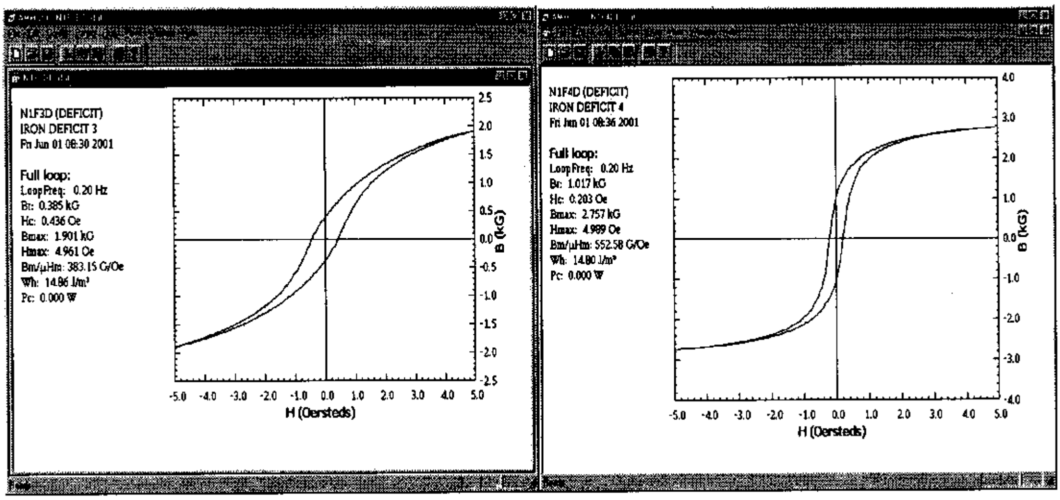


Fig.5: Variation of Quality Factors (Q-factor) vs. Frequency (MHz) for Samples N1F0D, N1F1D N1F2D, N1F3D and N1F4D Measured at Room Temperature


N1F3D (DEFICIT)
IRON DEFICIT 3
Fri Jun 01 08:30 2001

Full loop:
LoopFreq: 0.20 Hz
Br: 0.385 kG
Hc: 0.436 Oe
Bmax: 1.901 kG
Hmax: 4.961 Oe
Bm/μHm: 383.15 G/Oe
Wh: 14.86 J/m³
Pc: 0.000 W

N1F4D (DEFICIT)
IRON DEFICIT 4
Fri Jun 01 08:36 2001

Full loop:
LoopFreq: 0.20 Hz
Br: 1.017 kG
Hc: 0.203 Oe
Bmax: 2.757 kG
Hmax: 4.989 Oe
Bm/μHm: 552.58 G/Oe
Wh: 14.80 J/m³
Pc: 0.000 W

Fig. 6: Maximum Saturation Induction (Bmax) for Samples N1F3D and N1F4D Measured at Room Temperature

The increase of density for these samples could be due to the sufficient space or vacancies for an effective transportation of metal ions. The spinel lattice is primarily considered as a lattice of close-packed oxygen ions and thus metal transport depends strongly on the anionic vacancies (Tebble and Craik, 1976) concentration. However, due to the iron deficiency adopted in this work, more and more cationic vacancies are formed. Metal ions are speculated to be transported along both, the anionic and cationic vacancies, along the grain boundaries into the pores. Observing the microstructure of N1F3Dand N1F4D, it is clear that the grains of both the samples are clear, small and dense compared to the other samples.

Initial permeability (μ_i), one of the most important magnetic properties measured at room temperature, is shown in Fig. 3. There was a general decreased of μ_i for samples N1F0D, N1F1D,N1F2D and N1F3D. except for sample N1F4D. The decrease of μ_i is corresponding to the decrease in grain size. The equilibrium partial pressure, PO_2, of NiZn ferrites is generally known to increase with the temperature between 1000^{0}C to 1400^{0}C (Pyun and Baek, 1985). It was also reported that at 1140^{0}C the equilibrium oxygen partial pressure is 0.213 bar (air) for $Ni_{0.32}Zn_{0.68}Fe_2O_4$ (Pyun and Baek, 1985). Therefore, the equilibrium oxygen pressure at 1300^{0}C is expected to be higher than 0.213 bar (air), so that the equilibrium between the spinel ferrite phase and oxygen,

$$Fe^{3+} + 3/8\ V_e + \tfrac{1}{2}\ O^{2-} \leftrightarrow Fe^{2+} + \tfrac{1}{4}\ O_2 \qquad (2)$$

is shifted to the right . We speculate some formation of excess Fe^{2+} with the loss of oxygen in the ferrite for all the samples are due to the high sintering temperature. It is well established that the excess Fe^{2+} contributes to the positive magnetocrystalline constant (Wolfarth, 1980). This is speculated to be one of the reasons for the degradial of μ_i for all the iron-deficit samples. Sample N1F3D however exhibit a marked decrease of permeability from sample N1F2D to N1F3D. This could be due to the more densed microstructure of N1F3D resulting from a greater number of cationic vacancies. It should be noted that μ_i is very microstructure sensitive; grain size, pore size and pore distribution could alter the permeability value. The smaller grain size causes the domain walls to be pinned at the grain boundary. Observing Table 1, the theoretical porosity is 1.53%, slightly higher than that of sample N1F4D that gives 0.89%. Sample N1F4D, which has less pores should be the reason to the rise of the μ_i (Fig. 3) and the fall of resonance frequency.

Resonance frequency is an effective limit to the product of frequency and permeability, so that high permeability and high frequency is mutually incompatible.

The quality factor, which is defined as the reciprocal of tan δ, is the figure of merit of a material (Pyun and Baek, 1985). Fig.5 illustrates the Q-factor vs. frequency. According to Snoek's theory, the Q factor should remain high until the spin resonance is approached. Sample N1F3D exhibits the highest Q thus the lowest losses and the widest operating frequency compared with that of other samples.

Observing Fig. 6, the saturation magnetization value is higher for sample N1F4D (2.757 kG) compared with that of sample N1F3D (1.901 kG). It could be speculated that saturation is done easily without much constrain. The domain structure is regular, without much pinning centers. The crystal for sample N1F4D does not contain much compositional inhomogeneities, non-magnetic inclusions such as voids and second phase and thus is not subjected to random mechanical stress resulting in higher saturation induction as well as higher permeability.

Conclusion

It is concluded that vacancies concentration in an iron-deficit $Ni_{0.30+2x}Zn_{0.70}Fe_{2-2x}O_{4-4x}$ is be considered a good route to obtain high density ferrites. The 0.47 mole fraction of the iron-deficit NiZn ferrite series gives the optimum number of vacancies for the transportation of metal ions during solid-state reaction. However, 0.48 mole fraction of the series gives higher saturation induction and initial permeability.

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