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DEVELOPMENT OF LOW-LOSS NiZn-BASED FERRITE MATERIALS FOR HIGH-FREQUENCY APPLICATIONS

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DEVELOPMENT OF LOW-LOSS NiZn-BASED FERRITE MATERIALS FOR HIGH-FREQUENCY APPLICATIONS

By

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LIST OF SYMBOLS AND ABBREVIATIONS

AE	acoustic emmision
H	applied field
μ_B	Bohr magneton
H_c	coercive force
A	cross sectional area
T_c	curie temperature
ρ^*	density
f	frequency
γ	gyromagnetic ratio
μ''	imaginary part of permeability or magnetic loss parameter
B	induction
L	inductance
μ_i	initial permeability
D_i	inner diameter
σ	internal stress
l	length
$\tan\delta$	loss tangent
N	number of wire turns
D_o	outer diameter
μ_o	permeability of free space

PVA	polyvinyl alcohol
Q	quality factor
μ'	real part of permeability
RLF	relative loss factor
B_r	remanent induction
R	resistance
ρ	resistivity
B_s	saturated induction
M_s	saturation magnetisation
T	temperature
$\alpha_{\mu r}$	temperature coefficient of relative permeability
t	thickness
K_1	first anisotropy constant
R_T	total loss
$\gamma*$	wall energy
W	weight
XRD	x-ray diffraction

Abstract of the thesis presented to the Senate of Universiti Putra Malaysia in fulfilment of requirements for the degree of Master of Science

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February,1998

Chairman : Dr. Mansor Hashim
Faculty : Science and Environmental Studies

This work is an initial response to the demand for miniaturisation of electronic circuits and the shift to higher operating frequencies which has led to the development of high-density low-loss ferrites. In order to meet this demand, a number of necessities, such as fine-grained and homogeneous microstructure, low power loss, weak temperature dependencies of losses and of course reasonable cost of production are required. The first part of the work was to develop the required material by manipulation of composition. NiZn-based ferrites with various additives such as MgO, CuO, TiO_2, CoO and CaO were chosen and the low loss property was attained. Subsequently, a systematic crucial approach was started to further minimise the losses and to extend the operating frequency range. Green compacts with a particle size average (PSA) of $\approx 1.2\mu m$, a sintering temperature of about

1140^{0}C and only air atmosphere were needed to reach the research targets. In conclusion, a composition-microstructure design technique has been successfully developed, which is capable of producing ferrite materials with the desired magnetic properties.

Abstrak tesis yang dikemukakan kepada Senat Universiti Putra Malaysia bagi
memenuhi keperluan Ijazah Master Sains.

PENGHASILAN BAHAN-BAHAN FERIT NiZn DENGAN KEHILANGAN TENAGA RENDAH UNTUK KEGUNAAN FREKUENSI TINGGI

Oleh

NOORHANA YAHYA

Disember, 1997

Pengerusi Dr Mansor Hashim
Fakulti Sains dan Pengajian Alam Sekitar

Penyelidikan ini adalah suatu respons awal terhadap desakan ke arah pengecilan litar elektronik dan anjakan ke frekuensi operasi yang lebih tinggi yang telah menghalatujukan penghasilan ferit dengan ketumpatan tinggi dan kehilangan tenaga rendah Untuk memenuhi desakan ini, beberapa keperluan seperti mikrostruktur butir-seni dan homogen , kehilangan tenaga yang rendah, pergantungan lemah kehilangan tenaga terhadap suhu dan semestinya kos pengeluaran yang berpatutan, harus dipenuhi. Penyelidikan untuk menghasilkan bahan sedemikian dimulakan dengan melakukan manipulasi terhadap komposisi Ferit berasaskan NiZn dengan pelbagai bahan tambah seperti MgO, CuO, TiO_2 , CoO, dan CaO telah dipilih dan ciri-ciri kehilangan tenaga rendah telah dicapai.

Seterusnya, suatu pendekatan yang lebih sistematik telah dijalankan untuk mengurangkan lagi kehilangan tenaga dan menambahkan lagi julat frekuensi operasi. Penyelidikan ini telah mencapai matlamatnya melalui "kompak hijau" yang purata saiz zarahnya (PSZ) $\approx 1.2\ \mu m$, suhu pensinteran sekitar 1140^0C dan dengan hanya atmosfera udara. Kesimpulannya, suatu teknik rekabentuk "komposisi-mikrostruktur" yang berupaya menelurkan bahan ferit dengan ciri-ciri magnet yang diperlukan telah berjaya dihasilkan.

CHAPTER I

GENERAL INTRODUCTION

Soft Magnetic Ferrites

A ferrite is a metal oxide which contains magnetic ions arranged in a manner which produces spontaneous magnetisation (Gerald,1975; Standley,1972; Crangle,1991). Soft magnetic materials become magnetised by relatively low-strength magnetic field. When the applied field is removed, they return to a state of relatively low residual magnetism. The converse, the need for high magnetising field and high remnant magnetism, is true for hard magnetic materials . Ferrites have three distinct crystal structures: The hexagonal magnetoplumbite, dodecahedral garnet and the spinel structure (Crangle, 1991; Standley,1972). The first structure is that of hard ferrites, the later two being those of soft ferrites.

The remarkable capacity of these materials to survive in the intense competition from the growing technologies and their ability to enter into newer areas of applications have promoted them to many disciplines. Consequently, the growing information-oriented society and the expanding roles of ferrites in electronic gadgets and other industrial pursuits have, in return, motivated this study.

The general concern and direction of this study is guided by the push for miniaturisation and the fact that many electronic devices are moving towards higher operating frequencies . In order to comply with these demands , ferrite materials with homogeneous and fine-grained microstructure , high density, narrow property tolerances, weak temperature dependencies of losses, high saturation induction and reasonable cost of production are essentially required (Suresh et al., 1989).

Magnetic Parameters

It is recognised that the response of magnetic moments to oscillating magnetic field excitations is the dominant phenomenological basis on which most science-driven and application-driven soft ferrite research rests. Hence, the following very early ,though brief, introduction to the main response parameters is thought to be helpful and appropriate. The centrally important properties in non-microwave non-rectangular ferrite applications are the permeability and it's frequency spectrum (Wolfarth, 1980). The initial permeability, μ_i, simply called the permeability of a material is defined as the limit of change of the induction B with respect to the applied field H in the demagnetised state as H approaches zero and is represented as

$$\mu_i = (1/\mu_o) \lim_{H \to 0} (B/H) \qquad [1]$$

where μ_o = magnetic constant ≈ permeability of free space

$= 4\pi \times 10^{-7}$ Henries/meter

B and H are in Weber/m^2 [10^4G] and A/m [0.0126 Oe] respectively.

Strictly speaking, if part of the energy carried by H is lost in producing B, the μ_i in equation [1] should be written μ_i^*, a complex number. The permeability concept can be extended to include the losses. For time harmonic fields,

$$H = H_o \exp(j\omega t) \qquad [2]$$

where ω is the angular frequency and t is the time; the dissipation can be described by the phase difference, δ, between H and B. In the complex notation, the frequency dependency of permeability becomes

$$\mu(\omega) = \frac{B \exp j(\omega t + \delta)}{H \exp(j\omega t)} \qquad [3]$$

$$= \mu' - j\mu'' = \mu_0(\mu_r' - j\mu_r'') \qquad [4]$$

where r signifies a relative, dimensionless quantity.

For vanishing H fields, as used exclusively in this work,

$$\mu(\omega) = \mu_0\mu_i^* = \mu_0(\mu_i' - j\mu_i'') \text{ giving } \mu_i' \equiv \mu_r' \quad \text{and} \quad \mu_i'' \equiv \mu_r''$$

The real part , μ', describes the stored energy expressing the component of B in phase with H and the imaginary part , μ'', describes the energy dissipated (lost) expressing the component of B out of phase with H. μ' or μ_i' relates to the inductance L (see Appendix A). μ'' can be obtained from the relation (for series L-R circuit)

$$\frac{\mu''}{\mu'}\left[= \frac{\mu_r''}{\mu_r'} = \frac{\mu_i''}{\mu_i'}\right] = \tan\delta \qquad [5]$$

In normal practice, μ_r', μ_r'', μ_i' and μ_i'' are written as μ_r, μ'', μ_i or μ' or μ, and μ'' respectively; however they remain the same dimensionless and relative quantities. It is carefully noted that the μ' and μ'' of equation [5] are proportionality parameters with a dimension or unit.

The change of magnetisation vector is generally brought about by rotation of magnetisation or domain wall displacement (Ishino et al. 1987). These motions lag behind the change of magnetic field and cause the increase of μ''. Moreover , the resonance-relaxation phenomenon will be induced when the frequency increases and approaches the characteristic frequency of the rotation of the magnetisation or the domain wall displacement. The frequency at which μ'' maximises (loss resonance) is nearly inversely proportional to the low-frequency permeability according to the equation given by Snoek (Goldman, 1991 ;Wolfarth, 1980)

$$f_r (\mu - 1) = 4/3 \ \gamma \ M_s \qquad [6]$$

where f_r is the loss resonance frequency, γ is the gyromagnetic ratio and M_s is the saturation magnetisation.

Basis of Work

The advances in technology have undoubtedly led to studies in modern ferrites being undertaken extensively. Fortunately, in dealing with interactions involving intrinsic and structural features which affect the permeability, energy losses, coercivities, resistivities, etc., it becomes apparent that many of the contributing factors are common to ferrites generally. However, special attention should be drawn to the magnetic losses, which are the most important factors that govern the properties of any ferrite materials (Tebble and Craik, 1976). Ultimately, a work of this nature attempts to obtain a magnetic material with low loss properties. It is notable that applications at high frequencies frequently involve small-amplitude and weak signals. Hence the receiving antennae must not lose/dissipate much of the signal energy. Besides the low loss properties, this research work also hopes to produce a material that can be used over a wide operating frequency range. This would allow a single ferrite component to have a flexible use at various frequencies with the highest upper limit possible.

Hence, detailed investigations of the factors that cause energy losses are essential for a proper perspective understanding of the physics involved. This knowledge could then be tapped for the fabrication of high-quality low-loss ferrite materials. In particular, preparation techniques aimed at achieving desired loss properties are satisfactory only when their uses can be unambiguously related to meet the specific requirements of a certain application, for example, as antenna cores.

Thus this research work has been designed to have the following four main premises of effort concentration:

a) The first premise concerns the preparation of samples with a fine-grained, very dense and homogeneous microstructure which has been convincingly proven to confer low loss properties (Goldman, 1991). These properties are associated with high electrical resistivity (Standley, 1972) and restrained domain wall movement resulting from such a microstructure.
In single phase ferrites, the microstructure is characterised by the structure of pores, surfaces and the grain boundaries which are determined not only by the surface energy, but also by the initial state before sintering (Okamoto et al., 1984).

b) The second premise focuses on the exploitation of the large positive anisotropy value (Pyun et al., 1985) of the Co^{2+} ion in order to produce excellent frequency extension. However, the skin depth into which alternating magnetic flux can penetrate becomes thinner as the frequency rises. Beyond this depth, the eddy current effect becomes prominent. Consequently, this reduces the effectiveness of the material to support the frequency increase. As such, the contradiction between achieving the low loss properties and the high operating frequencies is acknowledged.

The importance of this effort derives from the keen interest in new wireless digital technology which has led to a kind of modern day gold rush for more portions of the radio spectrum. However there is a limiting amount of 'space' within the radio spectrum. So these wireless gadgets and services must compete with increasingly narrow bands and unused or reassigned 'space'.

c) The third premise focuses on the measurement of frequency-dependent complex impedance used to analyse the electrical response of the polycrystalline ferrite sample. An experimental equivalent circuit is to be proposed to explain the observed resistance-reactance (R-X) dispersion. To enable meaningful analysis of the samples, separation of the grain and the grain boundary effects on the sample's electrical properties must be carried out.

In general, a single phase ceramic material can be mathematically described as a material with complex conductivity .The crystal grain and grain boundary are the two main components that determine the microstructure. The bulk conductivity of the ferrite material is significantly higher than that at the grain boundary phase. Since the resistivity of the grain boundary phase is usually much higher than that of the crystallite material, most of the applied