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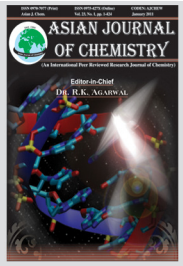

NiCuZn Nano Ferrite Prepared by Sol Gel Method for Multilayer Chip Inductor Applications†

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$Ni_{0.50}Zn_{0.45}Cu_{0.05}Fe_2O_4$ nanoparticles prepared through sol-gel process have been investigated by infrared spectra, X-ray diffraction analysis on the sintered samples. X-Ray diffraction patterns show single phase spinel structure. The far infrared spectra showed two absorption bands. The high frequency band ν_1 and low frequency band ν_2 are assigned to the tetrahedral and octahedral metal-oxygen complexes, respectively. The room temperature saturation magnetization (M_s) obtained from vibration sample magnetometer (VSM) shows M_s increases with increasing (M_s = 76.76 emu/g) sintering temperature up to 950 ºC. Initial permeability (μ_i) of the sintered samples increases with sintering temperature and for each sample μ_i is constant up to MHz frequency range showing high operating frequency of the material.

Key Words: Nanoparticles, Magnetization, Permeability.

INTRODUCTION

Recently, surface mounting devices (SMD) have been rapidly developed using multilayer chip inductors (MLCIs)[1]. They are the important components for the latest electronic products such as cellular phones, video cameras, notebook computers, hard and floppy drives *etc*[2]. Multilayer chip inductors are produced by coating and co-firing of ferrite with silver electrode material alternatively. Thus co firing of ferrite with Ag need low temperature sintering (because of melting point of Ag is 961 ºC) to avoid diffusion of Ag into ferrite which will deteriorate material properties[1]. In order to reduce sintering temperature of ferrite there are several techniques exist some of them are (1) preparation of powders with small particles and high surface area or (2) addition of sintering aids[3]. These multilayer chip inductors requires high permeability (μ) at higher frequencies, high resistivity (ρ) and high quality factor (Q). Permeability mainly depends on saturation magnetization (M_s), magnetic anisotropy (K) and grain size (D)[4]. In literature we found the composition of $Ni_{0.50}Cu_{0.05}Zn_{0.45}Fe_2O_4$ prepared through oxalate precursors method having high saturation magnetization (M_s) and density (d)[5]. Thus to increase magnetization further and for optimum sintering temperature to use the material as multilayer chip inductors we re-examined the same composition but prepared through sol gel poly(vinyl alcohol) method since the material properties are dependent on synthesis techniques. In the present paper structural and magnetic properties on samples sintered at three different temperatures were carried out.

EXPERIMENTAL

Synthesis: In order to prepare $Ni_{0.50}Cu_{0.05}Zn_{0.45}Fe_2O_4$ ferrite, analytical grade nickel nitrate [$Ni(NO_3)_2{\cdot}6H_2O$], zinc nitrate [$Zn(NO_3)_2{\cdot}6H_2O$], copper nitrate [$Cu(NO_3)_2{\cdot}6H_2O$] and ferric nitrate [$Fe(NO_3)_3{\cdot}9H_2O$] were weighed according to the required stoichiometric proportion and dissolved each separately in minimum amount of de ionized water. The formed cationic solutions were mixed and stirred for 1 h to improve homogeneity, the resulting solution is known as precursor. The molar ratio of total metal ions to poly(vinyl alcohol) was maintained at 1:3. The poly(vinyl alcohol) in measured quantities was added to the precursor and dehydrated around 100 ºC under continuous stirring. The gelation continued step by step till a slightly red gel type product was formed with the release of reddish brown gas around 100 ºC[6], where the gel further got converted into a fluffy ferrite mass which was annealed at 500 ºC for 3 h to remove poly(vinyl alcohol). The annealed powder was ground in agate mortar and mixed with 15 % of poly(vinyl alcohol) as binder in order to make pellets and toroids. These pellets and toroids were sintered at 900, 950 and 1030 ºC for 1 h with a heating rate of 5 ºC/min.

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The phase identification of the sintered samples were carried out using Inel XRG 3000 X-ray diffract meter (XRD) with Co-K_α radiation (λ = 1.78897 Å). The Infrared spectra of the annealed and sintered samples were recorded on Shimadzu 820/PC Fourier transform infrared in the wave number region from 4000 to 400 cm^{-1} by KBr pellet method. The vibration sample magnetometer (VSM, EV-7 VSM) was used to measure room temperature saturation magnetization (M_s) and intrinsic coercive force (H_c) on the sintered samples in an applied field of 20 kOe. Permeability studies in the frequency range of 1 MHz-1.8 GHz were carried out using 4291 B Agilent network analyzer.

RESULTS AND DISCUSSION

X-Ray diffraction: Fig. 1 shows powder XRD patterns of the sintered samples. From the XRD patterns it is confirmed that there is a pure spinel phase and no other secondary phases exist. The annealed sample shows broad peaks whereas the sintered sample shows sharp peaks with high intensity indicating that crystallite size and crystallinity increasing with sintering temperature. The crystallite size is calculated from (311) peak using Debye Scherer's formula[7].

$$D = \frac{k * \lambda}{\beta \cos\theta}$$

where k is shape factor (k = 0.9); λ = is wavelength of X-rays (λ of CoK_α = 1.78897 Å); θ = peak centre; β = full width half maximum.

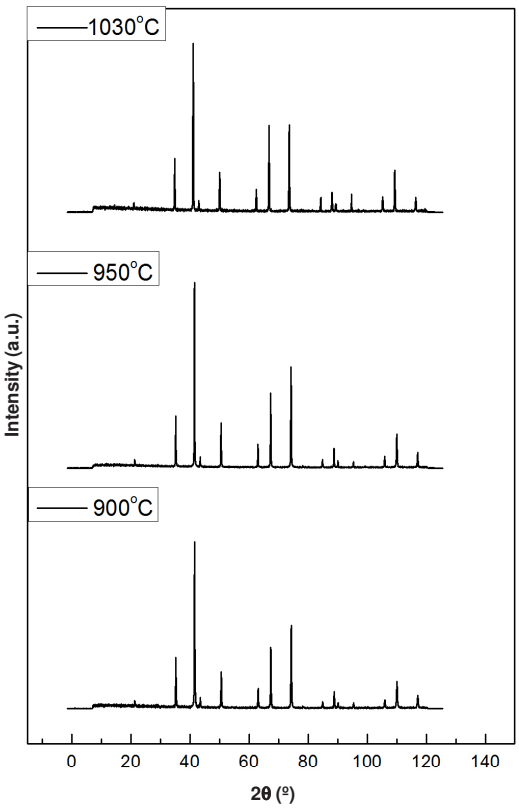

Fig. 1. XRD patterns of the sintered samples

Before estimating the crystallite size the correction to peak broadening due to the instrumental broadening is applied[8].

$$\beta = \sqrt{(\beta^2_{measured} - \beta^2_{instrumental})}$$

where β is the measured broadening of the peak at an angle 'θ', $\beta^2_{measured}$ is the measured full width at half maximum (FWHM) of experimental profile. $\beta^2_{instrumental}$ is the instrumental broadening. It is observed that the crystallite size is increasing with sintering temperature.

From XRD patterns the lattice parameter for each diffraction line is calculated using the eqns. 1 and 2:

$$2d \sin\theta = n\lambda \quad (1)$$

$$a = b * \sqrt{h^2 + k^2 + l^2} \quad (2)$$

The exact value of the lattice parameter is determined by drawing a graph between lattice parameter a *vs.* Nelson Riley function f (θ).

$$f(\theta) = \frac{\cos^2\theta}{2}\left(\frac{1}{\sin\theta} + \frac{1}{\theta}\right)$$

Table-1 shows change in lattice parameter, crystallite size with sintering temperature.

TABLE-1
CRYSTALLITE SIZE, LATTICE CONSTANT FOR THE SINTERED SAMPLES

S. No.	Sintering temperature (ºC)	Crystallite size (nm)	Lattice constant (Å)
1	900	52	8.3884
2	950	66	8.3857
3	1030	94	8.4131

FTIR: Infrared spectrum is used to correlate experimentally observed energy levels to the structure and force as a function of the molecules[9]. For spinel phase AB_2O_4, the cubic unit cell comprises 8 formula units and contains 64 tetrahedral and 32 octahedral sites, generally designated as A and B sites, respectively. Eight of the A sites and 16 of the B sites are occupied leaving all other sites vacant[10]. In the IR spectrum two main broad metal-oxygen bands are seen for all spinels and ferrites in particular. Waldron[11] attributed the band ν_1 around 600 cm^{-1} to the intrinsic vibration of tetrahedral metal oxygen complexes and band ν_2 at around 400 cm^{-1} to the intrinsic vibration of octahedral metal oxygen complexes[11]. The annealed sample shows characteristic bands at about 3427, 1622, 1384, 576 and 407 cm^{-1} corresponding to stretching mode of O-H group in the free and absorbed water and poly(vinyl alcohol), H-O-H bending vibration of the residual water, antisymmetric NO_2^- stretching vibration and characteristic bands for ferrites. The bands corresponding to O-H group and NO_2^- group disappeared at high sintering temperatures due to loss of water and decomposition of poly(vinyl alcohol) at high temperature heat treatment[12]. Fig. 2 shows variation of ν_1 and ν_2 with sintering temperature. For three samples sintered at different temperatures showed characteristic band ν_1 around 580 cm^{-1} confirming single phase ferrite formation.

Magnetization (M_s): Saturation magnetization (M_s) is influenced by intrinsic factors such as preferential site occupancy of the ions, composition and is also influenced by wextrinsic factors like microstructure and bulk density of the ferrites[13]. Room temperature hysteresis loops for samples

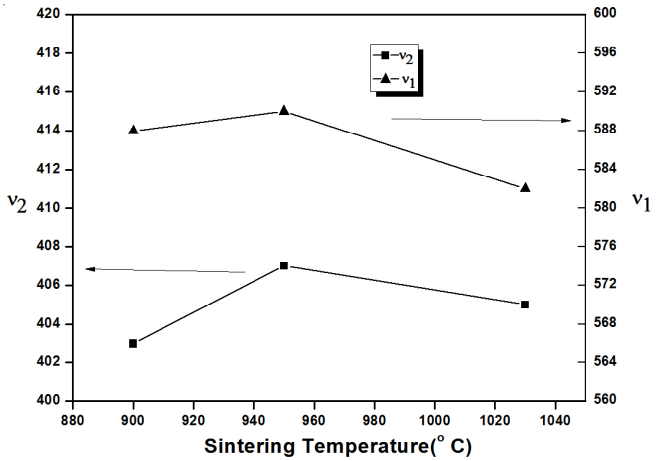

Fig. 2. Variation of ν_1, ν_2 with sintering temperature

sintered at different temperatures (Fig. 3) with the increasing sintering temperature the magnetization increases up to 950 ºC and maximum value of mass magnetization σ_s is 76.76 emu/g with bulk density of 4.99 g/cm^3. The increase in the magnetization with temperature may be because of increase in the crystallite size[4].

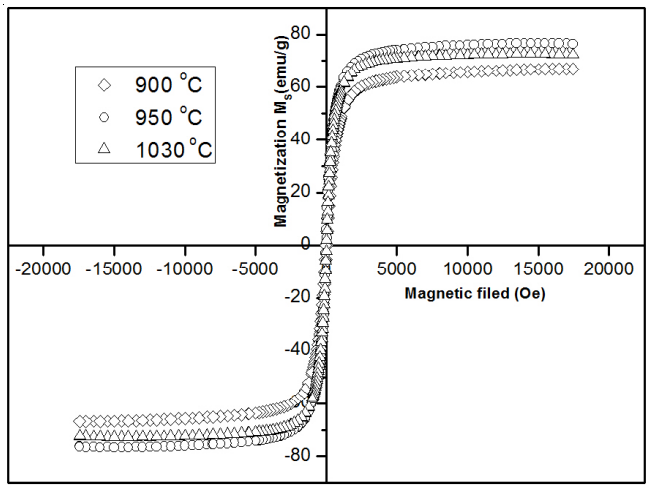

Fig. 3. Room temperature hysteresis loops for sintered samples

Initial permeability (μ_i): Fig. 4 shows real part of permeability with frequency. In general the permeability is related to two different magnetizing mechanisms, the spin rotational magnetization and domain wall motion. Spin component is dominated by chemical composition while the domain component is affected by both composition and the microstructure[1]. In the present case with increasing sintering temperature permeability increases but the critical field for resonance decreases

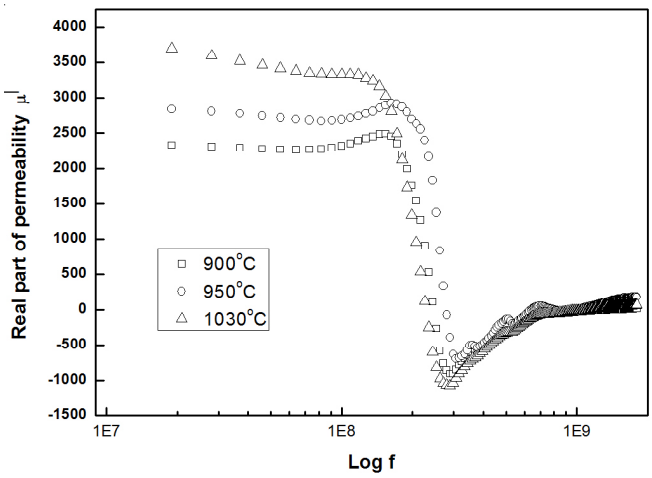

Fig. 4. Real part of permeability with frequency for sintered samples

according to Snoek's law[4]. The increase in μ_i with sintering temperature could be attributed to the increase of crystallite size causing easy domain wall motion[14].

Conclusion

NiCuZn nano ferrite prepared by sol gel poly(vinyl alcohol) method having single phase spinel structure with no secondary phase was obtained. High saturation magnetization, (σ_s = 76.76 emu/g), density obtained at a sintering temperature of < 961 ºC (melting point of Ag) which is one of the requirements to use the material as a multilayer chip inductor. High permeability with operating frequency in the range of MHz was obtained.

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