Effect of Firing Temperature on the Electromagnetic Properties of Electronic Transformer Cores Developed by Using Nanosized Mn–Zn Ferrite Powders

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Abstract Current investigation deals with the effect of grain growth process as a function of sintering temperature on the electromagnetic properties (initial permeability, impedance, and gain) of Mn–Zn ferrite (MZF) cores (toroids). By employing auto combustion process, nanosized $[(20 \pm 5) \text{ nm}]$ MZF powders were synthesized and toroid shape cores were obtained after subsequent calcination and firing (sintering) process. It was observed that the submicron structure ($0.5 \mu\text{m}$) achieved in the ferrite core sintered at $1,000^\circ\text{C}$ was gradually transformed into micron size grains by increasing the firing temperature i.e., $1,100$, $1,200$, and $1,350^\circ\text{C}$. The results reveal that MZF core sintered at low temperature ($1,000^\circ\text{C}$) showed high impedance, low initial permeability, and narrow working frequency range i.e., $3\text{--}15 \text{ MHz}$. However, the improvement in initial permeability, sintered density, and operational frequency range ($100 \text{ kHz--}17 \text{ MHz}$) was observed at high temperature ($1,350^\circ\text{C}$) firing in the inert environment. This synthesized MZF core is more suitable for miniaturized switch mode power supply applications.

KEY WORDS: Mn–Zn ferrites; Nanosized powders; Impedance; Sintering

1 Introduction

Mn–Zn ferrites (MZF) are technologically very important class of soft ferrites due to possessing magnificent magnetic properties such as high initial permeability, moderate impedance, low power losses, and appreciable Curie temperature [1]. In this modern era, the demand of MZF is elevating due to their multidisciplinary applications. On the one hand, these are used in medical applications such as contrast agent in bio-imaging, hyperthermia treatment under magnetic field, drug delivery carriers, etc. [2–5]. While on the other hand, scientists are exploiting their water splitting property to produce hydrogen gas as an alternative

source of fuel [6]. These materials are also massively utilized by electronic industry in various forms such as toroids (rings), plates, rods, cylinders, beads, films, etc., for transmitting and receiving electronic data signals. Moreover, polymeric composites of soft ferrites have also drawn an important attention to get shielding against the electromagnetic pollution and microwave applications [7–9].

The utilization of MZF as a core material in electronic transformers is a more prominent and demanding application. Modern switch mode power supplies dealing with AC to DC or DC to DC conversion normally utilize these cores to modify the input frequency signals and demand the size of ferrite cores to be smaller and cost effective. However, it is challenging to produce compact size transformers cores (light weight and smaller dimensions) by conventional processing routes. Serious efforts have been made to meet this demand by using nanosized ferrite powders having low density and high surface area characteristics. Various synthesis routes have been employed to produce nanosized MZF powders [10, 11]. Furthermore, the intrinsic properties

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of MZF have also been tailored by adding foreign oxides in base compositions [12–14].

Sintering is an important step of ferrite core synthesis. It improves the strength of green cores and also initiates the grain growth process which directly influences the electromagnetic properties of MZF cores [15–18]. During sintering, the firing temperature provides sufficient thermal energy to mobilize the nano-crystallites and to enhance their coalescence which significantly affects the microstructure of ferrite cores. The developed microstructure composed of nano- or micro-grains remarkably effects the properties of MZF cores. Due to the significant role of microstructure, sintering of nanosized MZF powders is still an active area of research and demanded to explore more in detail.

Keeping in view the aforementioned facts, the current work is related with the grain growth study of nanosized MZF powders as function of sintering temperature. The effect of grain growth process on microstructure, sinter density, initial permeability and impedance of light weight MZF cores (toroids) was studied. Moreover, the gain response of fabricated MZF cores as a function of sintering temperature has also been discussed in this paper.

2 Experimental

Nanosized MZF ($\text{Mn}_{0.65}\text{Zn}_{0.35}\text{Fe}_2\text{O}_4$) powder added with 0.2 wt% ZrO_2 and 0.04 wt% CaO was synthesized by auto-combustion process. Metal nitrates, aqueous ammonia, citric acid, and demineralized water (DMW) were the main ingredients of this method; the detailed procedure of synthesized compositions by auto-combustion process has been described by authors elsewhere [19, 20]. Nanosized Mn-Zn ferrite powder [(20 ± 5) nm] obtained by this process was calcined at 450 °C in air to eliminate unwanted species. After that, it was uniaxial pressed in form of rings (toroids) and sintered at different temperatures for an appropriate time i.e., 1,000 °C (5 h), 1,100 °C (5 h), 1,200 °C (2 h), and 1,350 °C (2 h) under controlled inert (N_2) atmosphere.

Development of magnetic phase (spinel) in sintered samples was confirmed by XRD analysis using Bruker diffractometer (D-5005) with CuK_α radiation source ($\lambda = 0.154$ nm). Sintered density was measured by employing Archimedes principle. Scanning electron microscope (SEM, Philip, XL-30) was used to observe the morphology of grains. Electromagnetic properties such as initial permeability and impedance were measured using precision impedance analyzer (Agilent-4294A) having magnetic test fixture (Model-16454A). Gain response of sintered cores (toroids) was measured by laboratory built electronic circuit [20].

Fig. 1 XRD patterns of MZF composition sintered at 1,000, 1,100, 1,200, and 1,350 °C

3 Results and Discussion

3.1 X-ray Diffraction Analysis

X-ray diffraction patterns of MZF cores sintered at 1,000, 1,100, 1,200, and 1,350 °C are shown in Fig. 1. Single-spinel (magnetic) phase can be observed in each pattern. This was achieved by supplying the nitrogen at some appropriate time during the sintering cycle. Nitrogen prevented the oxidation of MZF during sintering and also tends to revert the partially developed haematite (non-magnetic phase) to spinel phase. The low temperature sintering study of nanosized MZF powders has been already discussed by authors somewhere else [18, 19].

3.2 Microstructure Analysis

Micrographs of MZF samples (cores) sintered at 1,000, 1,100, 1,200, and 1,350 °C are shown in Fig. 2. Sharp edge grain morphology can be observed in these samples. Such type of morphology tends to suppress the grain growth process. Choi *et al.* [21] have also pointed out that the grains having sharp corners required greater energy than the circular grains for their coalescence with neighboring ones as a result grain growth process decreased during sintering. In the present case, sharp edges grain morphology was achieved by adding zirconia in MZF. It is well known that zirconia acts as grain growth inhibitor and tends to control the grain size of base composition by increasing the interfacial energy of adjacent grains [20–22]. Therefore, in present research smaller grain sizes were observed even at high sintering temperature (1,350 °C). An average grain size of 3 μm having cubic-type morphology is clearly observed in Fig. 2d. The presence of cubic morphology confirms the complete development of spinel phase in MZF samples. The effect of sintering

Fig. 2 Microstructures development in MZF cores at different sintering temperatures: **a** 1,000 °C; **b** 1,100 °C; **c** 1,200 °C; **d** 1,350 °C

Fig. 3 Grain size and sintered density of MZF cores as a function of sintering temperature

Inner diameter=13.2 mm
Outer diameter=18.44 mm
Height=1.2 mm
Weight=0.70 g

Fig. 4 Dimensions of slim size MZF cores obtained after sintering at 1,350 °C

temperature on density and grain size is graphically shown in Fig. 3. The density of samples (toroids) increases from 91 to 95% as a result of grain growth process (0.5–3 μm). The increasing trends of both variables (density and grain size) is due to the rise in sintering temperature which acts as driving force to enhance the grain size and density of MZF samples.

The picture of core (toroid) obtained after sintering the nanosized MZF powder at 1,350 °C is shown in Fig. 4, having 1.20 mm in thickness and around 0.70 g in weight. These dimensional features indicate that by choosing appropriate synthesis and sintering conditions, MZF cores (toroids) can be fabricated in compact sizes to facilitate the modern miniaturized electronic industry.

Fig. 5 Initial permeability curves of MZF cores sintered at different temperatures

3.3 Initial Permeability and Impedance

Figure 5 shows the initial permeability (μ_i) curves of MZF cores sintered at different temperatures. The increase in the initial permeability with rise in sintering temperature (1,000–1,350 °C) can be observed in this figure. It can also be noticed that initial permeability lines remain flat up to 30 MHz (maximum test frequency of instrument) at each temperature. The stability of initial permeability with increase in frequency is an encouraging sign regarding the utilization of cores for high-frequency applications. The value of initial permeability depends upon the amount of magnetic induction produced in soft ferrites under an applied field and also on the fraction of magnetic phase present in the MZF cores. As discussed previously, MZF cores sintered at different temperatures have fully developed magnetic phase (spinel) which significantly contributed in the initial permeability value. Furthermore, the increase in average grain size as a function of sintering temperature (Fig. 3) further improved the permeability of ferrite cores. In other words, grain growth process significantly influences the initial permeability values. Under an applied field, larger grains permit more magnetic lines of forces to pass through them and tend to enhance the magnetic induction which ultimately improves the permeability of ferrite cores. The similar concept has been exploited in this research and is schematically explained in Fig. 6.

Fig. 6 The grain growth development (nano to micro size) in MZF cores as a function of sintering temperature and its effect on magnetic induction

Figure 7 shows the effect of sintering temperature on the impedance (Z) response of MZF cores. The trend of curves shows that impedance initially increases and then starts to decrease after reaching its maximum. For each sintering temperature, the maximum impedance ($Z_{\max}$) value and the resonance frequency (f_r) are noted and tabulated in Table 1. These values show that the impedance and the resonance frequency are inverse function of sintering temperature. Both of the variables ($Z_{\max}$ and f_r) reached their peak values when MZF was sintered at 1,000 °C. Impedance loosed 36% of its maximum value and resonance frequency (f_r) descended 17% when sintering temperature was increased from 1,000 to 1,350 °C. This behavior indicates that although high temperature firing helps in improving the sintered density and initial permeability; on the other hand, it deteriorates the electrical properties by enlarging the grain sizes and reducing the grain boundary population in ferrite core microstructure [23]. Moreover, large grains induce polarization effect in MZF, as a result, hopping or polarization conduction (ionic conduction) may start within or across the grains and then reduces the AC resistance of ferrites [24].

From these findings, it can be deduced that submicron grains structure obtained in ferrite cores under low temperature sintering (1,000 °C) produces high impedance and resonance effects. Such type of sintering condition is more

Fig. 7 Impedance trend of MZF cores sintered at different temperatures

Table 1 The impedance and the resonance frequency of MZF cores sintered at different temperatures

Sintering temperature (°C)	Impedance	Resonance frequency (MHz)
1,000	395	64
1,100	350	62
1,200	303	62
1,350	251	54

suitable for making ferrite beads which may be used to suppress electromagnetic interference (EMI) in the electronic devices. While the MZF cores which were made from nanosized powder and sintered at high temperature (1,350 °C) have moderate impedance, good resonance frequency, and acceptable initial permeability values for modern switch mode power supply modules.

3.4 Effect of Sintering Temperature on Gain Response

The performance of soft ferrite cores in the electronic circuits is generally checked by gain response measurements as reported in Ref. [20]. Figure 8 shows the gain response curves of sintered cores (toroids) obtained at 1.0 V input signals for the frequency range of 100 kHz–20 MHz. From these curves, working frequency range can be noted as 3–10, 2–10 MHz, 200 kHz–15 MHz, and 100 kHz–17 MHz for MZF cores (toroids) sintered at 1,000, 1,100, 1,200, and 1,350 °C, respectively. These results reveal that cores sintered at 1,000 °C started to operate when the frequency is above kHz order. It is evident that magnetic induction (low hysteresis loss) is more important than electrical resistance of ferrite cores in kHz order frequency range [25]. Mn–Zn ferrite cores with large size grains work well in kHz frequency range due to their better magnetic induction and low hysteresis losses. In

Fig. 8 Frequency-gain curves of MZF cores sintered at different temperatures

present case, core sintered at 1,000 °C had smaller grains (submicron sizes) and was unable to operate at low frequency signals (kHz order). However, when the sintering temperature was increased i.e., 1,100, 1,200, and 1,350 °C, the grains gradually started to grow in micron sizes and magnetic induction began to increase (as mentioned in Fig. 6) as a result ferrite cores also started to operate in kHz order frequency range. The broader frequency range was covered by the MZF core sintered at 1,350 °C because of possessing medium size ($\sim 3 \mu\text{m}$) grains and low electrical losses due to the presence of adequate number of grain boundaries in ferrite microstructure. The peak in each gain curve represents the occurrence of resonance phenomenon in synthesized MZF cores. However, the broadening in resonance peaks may be due to the accumulative resonance effects coming from various circuitry components like connecting cables, primary and secondary winding wires, and MZF core material itself. Each of these components resonates at its own particular frequency and their collective effect tends to wider the resonance peak area as it was observed in the present research.

4 Conclusions

- (1) Nanosized MZF powders can be best utilized to make lighter and small size transformer cores for various electronic applications.
- (2) Temperature driven grain growth phenomenon in nanocrystalline MZF powders significantly affected the electromagnetic properties (initial permeability, impedance, and gain) of synthesized transformer cores.
- (3) High temperature firing (1,350 °C) of nanosized MZF powders gives optimum results regarding the

development of MZF cores for compact size switch mode power supplies working in kHz order to MHz order frequency range.

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