Preparation of low–power loss MgCuZn ferrites using the microwave sintering method

A. BHASKAR, B. RAJINI KANTH, S. R. MURTHY
Department of Physics, Osmania University, Hyderabad-500 007, India

A reduction in size and weight of power supplies can be achieved by using switched mode or resonant concepts. For the voltage conversion several circuit designs are in use with ferrites as transformer core materials. Transformer ferrites must show low energy losses at high induction levels at higher and higher frequencies [1–3]. This requires development of new ferrite materials with constantly improving loss characteristics.

To reduce the power loss, both hysteresis loss and eddy current loss should be suppressed by the available preparation technique. In this connection, it was found that the microwave sintering technique markedly enhanced the densification rate at lower sintering temperature of the material. In this method of preparation, the material can absorb the microwaves and self-generate heat [4–6].

Recently, we have developed low temperature sintered MgCuZn ferrites used for multilayer chip inductors [7]. These ferrites were conventionally sintered at 910 °C/12 hr. In continuation of this work, microwave sintered MgCuZn ferrites were selected for this investigation. The total power loss was measured.

Pure (99.99%) MgO, ZnO, CuO, and Fe_2O_3 powders with different particle size were used for the preparation. The α-Fe_2O_3 coarse particles (average diameter 0.13 μm) were synthesized by the oxidation of Fe_3O_4 particles at 800 °C. Industrial grade particles of magnesium oxide, zinc oxide, and copper oxide were used as the coarse particles with the average diameters of 0.8, 0.15, and 0.75 μm, respectively. Fine particles of magnesium oxide, zinc oxide, and copper oxide were synthesized by the thermal decomposition of the oxalates of MgO, ZnO, and CuO at 400 °C in air. These powders were mixed together with wet attrition milling. Their molar ratio was adjusted to obtain the composition $Mg_{0.58}Cu_{0.12}Zn_{0.30}Fe_2O_4$, and then the powders were made into two batches. One batch (Batch-I) of the powders was calcined at 800 °C/4 hr using the conventional ceramic method. The second batch (Batch-II) of the powders was calcined at 800 °C/30 min using a microwave furnace. Then the two batches were reground by wet ball milling for 10 hr to form slurry. To these powders, 2 wt% poly-vinyl-alcohol was added as a binder. The granules were compacted at a pressure of 190 MPa for 10 min, into pelletes (12-mm diameter, 3-mm thickness) and rods (60-mm length, 12-mm diameter) and toroids (12-mm outer diameter, 6-mm inner diameter, 4-mm thickness). The specimens were heated up to 400 °C to remove the binder and the lubricant.

The first batch of the samples was conventionally sintered (CS) at 870 °C/6 hr, 890 °C/6 hr, 910 °C/6 hr, and 930 °C/6 hr in air at atmospheric pressure. The second batch (Batch-II) of the samples was sintered using the microwave sintering (MS) method at 870 °C/20 min, 890 °C/20 min, 910 °C/20 min, and 930 °C/20 min in air at atmospheric pressure.

The microwave sintering process was carried out using a specially designed applicator which consists of a domestic microwave oven having an output power level tunable up to a maximum of 800 W and an operating frequency of 2.45 GHz.

The structure and microstructure of the sintered materials were examined using X-ray diffractometry and scanning electron microscopy (SEM), respectively. The bulk density was measured by the Archimedes method. The room temperature magnetic properties were obtained by recording hysteresis loops with the help of a vibrating sample magnetometer (VSM). The initial permeability (μ_i) of the toroidal sample was measured using the HP 4191A impedance analyzer in the frequency range of 1–100 MHz. The power loss, P_t, was measured at 100 kHz–1 MHz with the flux density of 50 mT with the B-H/Z analyzer (HP- E5060 A).

Fig. 1a and b, c and d shows the X-ray diffraction patterns for the few typical samples prepared by conventional and microwave sintering methods, respectively. It can be seen from the figure that the samples contain only single phase, irrespective of whether the materials were densified by the conventional or microwave sintering techniques. The average size of the ferrite grain, geometrically estimated from SEM photographs, and the results are presented in Table I. The grain size for the present samples varies from 3 to 16 μm irrespective of sintering methods. The grain size of MS samples is smaller than that of CS ferrites.

It can be seen from the table that the densification rate has been significantly increased in the microwave sintering process. In both preparation techniques, the spinel ferrite formation is completed at a low temperature of 870 °C. Conventional sintering process requires at least 16 hr to reach the sintering temperature of 870 °C and soaking time of 6 hr to obtain a sample with 91% of theoretical density (TD). Similarly, the density of the samples increased to 94% of TD with an increase of the sintering temperature from 870 to 930 °C, and time taken was 24 hr. In contrast, the microwave sintering process needs only 30 min to reach 870 °C, and a soaking period of 20 min to obtain a sample with the density as high as 94% of TD. The

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TABLE I Preparation and magnetic properties data for MgCuZn ferrites at room temperature

Sample no.	Sintering temp./time.	% theoretical density (TD)	Grain size (μm)	M_S (mT)	μ_i (at 1 MHz)	dc reistivity (Ω-Cm)	P_t (kW/m^3)
CS 1	870 °C/6 hr	91.0	16	345	1120	5.5×10^5	840
CS 2	890 °C/6 hr	92.0	15	380	1200	4.3×10^5	790
CS 3	910 °C/6 hr	93.0	12	396	1800	5.6×10^6	760
CS 4	930 °C/6 hr	93.5	18	410	2450	4.0×10^6	710
MS 1	870 °C/20 min	93.5	5.0	450	150	5.5×10^9	400
MS 2	890 °C/20 min	94.2	6.5	475	200	4.5×10^{10}	340
MS 3	910 °C/20 min	95.6	5.4	525	250	6.8×10^{10}	320
MS 4	930 °C/20 min	96.0	4.0	550	285	3.5×10^{10}	250

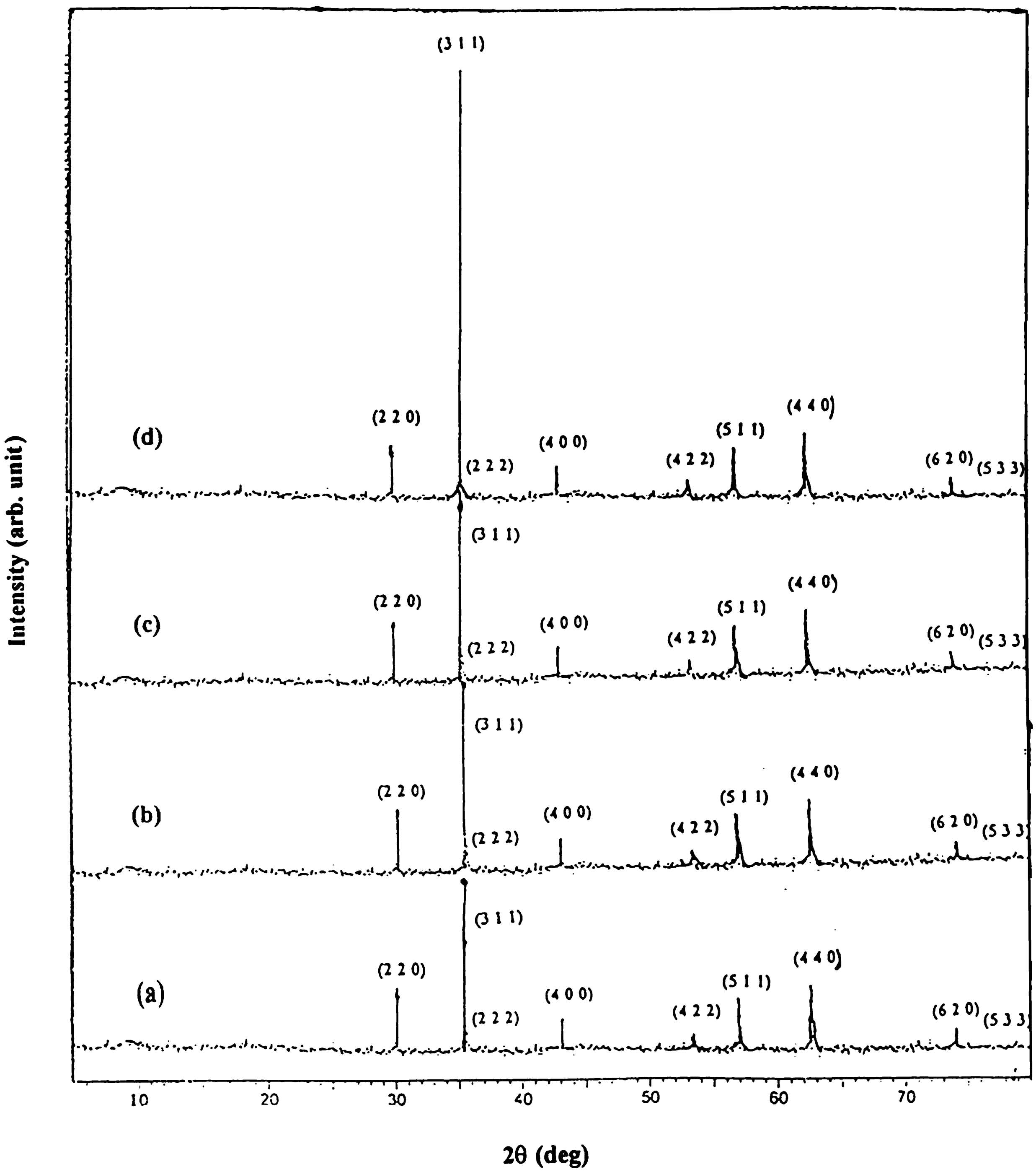

Figure 1 XRD patterns of conventional (a and b) and microwave (c and d) sintered MgCuZn ferrites.

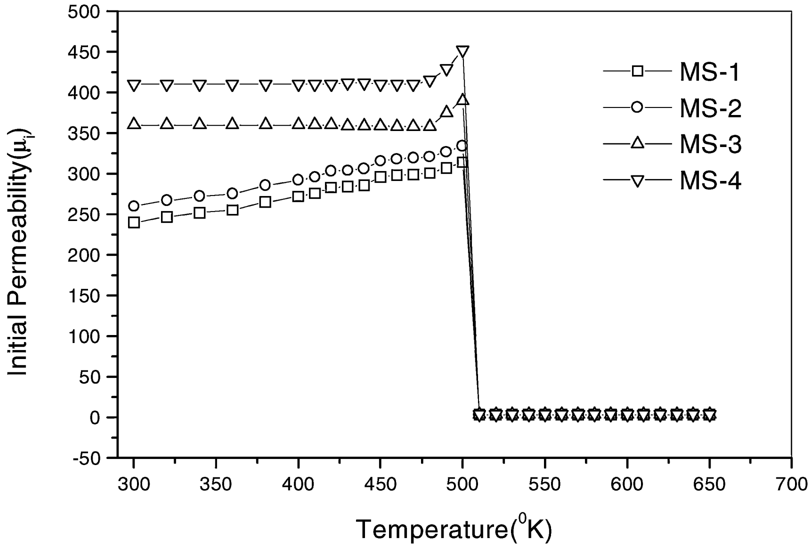

Figure 2 Temperature variation of initial permeability (μ_i) for microwave sintered (MS) MgCuZn ferrites.

density of MS ferrites has increased to 96% of TD with an increase of the sintering temperature from 870 to 930 °C and the time taken was 82 min only. Thus, higher densification can be achieved in shorter period by using the microwave sintering process.

Room temperature data of saturation magnetization (M_S) and initial permeability (μ_i) are also presented in Table I. It can be seen from the table that the values of M_S and μ_i are found to increase with increase in the sintering temperature. It can also be observed that the microwave-sintered samples possess a high value of saturation magnetization and permeability than that of the CS samples. The linearity between the sintering density and magnetization implies that the magnetization is conserved. The initial permeability of the MS samples varies with the sintering temperature in a similar way to that of the conventionally sintered samples, i.e., the higher the density, the larger the μ_i value.

The dc resistivity of the samples was measured using a two-probe method and the results are also presented in Table I. It can be seen from the table that the values of resistivity for both batches increase with an increase of the sintering temperature. This increase of 'ρ' may be attributed to increase in the bulk density and uniform grain growth occurred in the samples. Higher values of resistivity are observed for the microwave-sintered samples.

Figs 2 and 3 show the plots of initial permeability (μ_i) versus temperature for the samples sintered by both microwave and conventional sintering methods, respectively. It is evident from the figure that the μ_i remains constant over a wide temperature range for all the MS samples. It can also be seen from Fig. 2 that the MS ferrites show good thermal stability. The CS ferrites (Fig. 3) show a broad peak in the vicinity of Curie temperature than that of the MS samples. This shows that MS ferrites possess higher homogeneity. The Curie temperature (T_c) was measured from these plots and its value for the present samples varies from 500 to 510 K.

Losses in magnetic materials have two main contributions: one is due to hysteresis and originates from

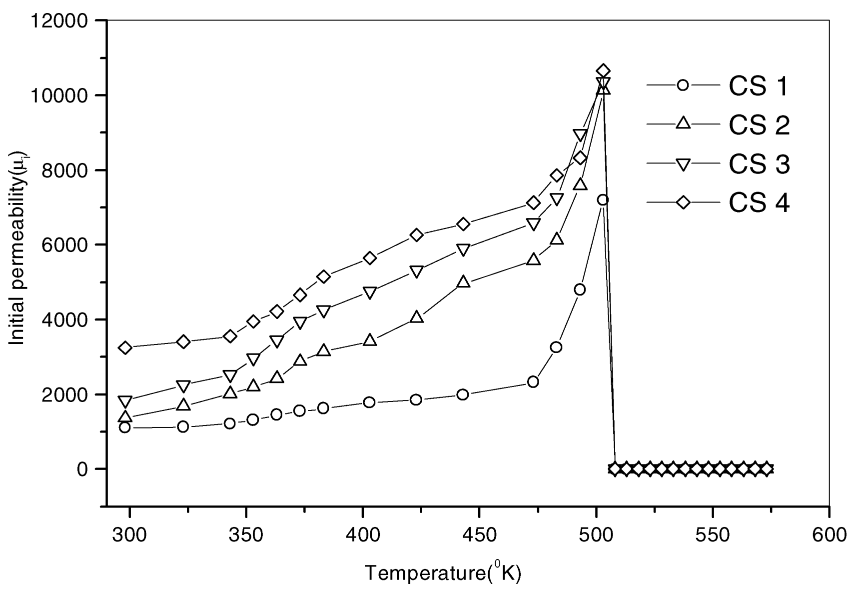

Figure 3 Temperature variation of initial permeability (μ_i) for conventionally sintered (CS) MgCuZn ferrites.

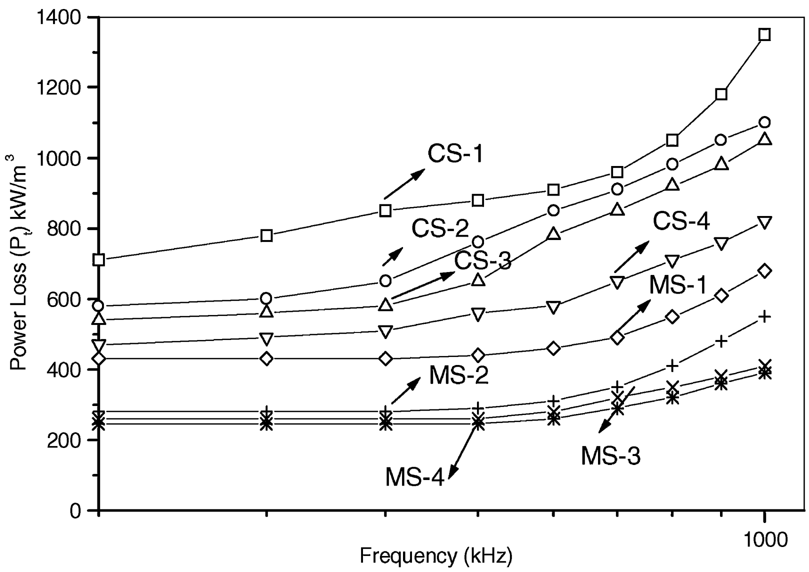

Figure 4 Frequency dependence of P_t for conventional (CS) and microwave (MS) sintered MgCuZn ferrites.

the non-reversibility of the magnetization process; the other is dynamic. For time harmonic fields, losses in magnetic materials can be depicted by a difference in phase between the external applied magnetic field and the resulting magnetization. Whatever is its physical origin, this time lag is closely related to magnetization mechanisms, which occur in the material. These mechanisms are the domain wall displacements and the spin rotations. Their relative importance depends on basic magnetic parameters such as magnetocrystalline and magnetoelastic anisotropies and the magnetic structure of the Weiss domains. Thus, a detailed study of power losses in ferrites gives an insight into the understanding of magnetization mechanism and increases their increase in the high-frequency applications.

The total power loss (P_t) for all samples was measured in the frequency range of 100 kHz to 1 MHz with 100 mT and the obtained results are plotted in Fig. 4. It can be seen from the figure that the power loss is markedly lower for the MS samples than that of the conventionally sintered samples. The power loss for the conventionally sintered samples increased with increase in the frequency. But, the power loss for the microwave-sintered samples remains constant up to a frequency of 500 kHz and increases for higher frequencies. The decrease in power loss for MS ferrites compared to that of CS samples may be ascribed to the decrease of internal strain. The internal strain for MS ferrites is decreased due to smaller grain size.

Fig. 5 shows the plots of the temperature dependence of power loss for all the MgCuZn ferrites. The power loss for the CS and MS samples decreases with an increase in temperature and shows a minimum value at a temperature which is denoted by T_{min} [8, 9]. The temperature gradient of the power loss for the MS samples is smaller with high T_{min} temperature.

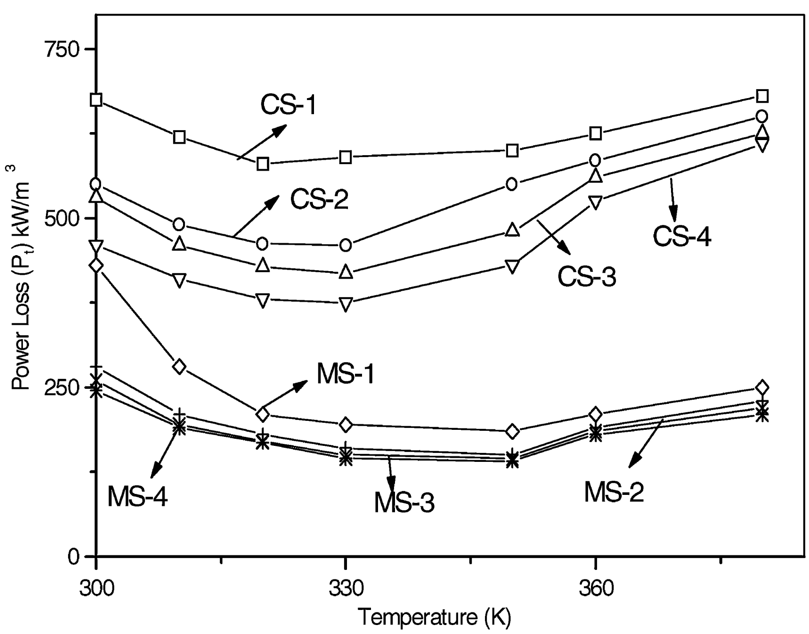

Figure 5 Temperature dependence of power loss for conventional (CS) and microwave sintered (MS) MgCuZn ferrites.

In conclusion, we have prepared the higher density, low-temperature microwave sintered MgCuZn ferrites with low power losses as compared with the conventional MgCuZn ferrite in the frequency range from 100 kHz to 1 MHz.

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