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Co-firing process using conventional and microwave sintering technologies for MnZn- and NiZn-ferrites

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Abstract

Effect of co-firing process on the characteristics of MnZn-ferrite materials was examined. The crystal structure and microstructure are not markedly modified, whereas the initial permeability-temperature ($\mu_i - T$) and power loss (P_L) properties are pronouncedly altered due to co-firing process, no matter whether the materials are densified by conventional sintering (cs) or microwave sintering (ms) process. The degradation of the magnetic properties is attributed to the strain induced in the MnZn-ferrites. The decreases in initials permeability (μ_i) and the increases in power loss (P_L) is less extent for the materials densified by the microwave sintering process. Such a phenomenon is attributed to the smaller grain size of MnZn-ferrite when the materials are microwave sintered.

Keywords: Co-firing; Ferrites; Microwave processing

1. Introduction

MnZn-ferrite materials possess highest initial permeability and magnetic inductance among the ferrite materials.[1–4] However, applicability of those materials for multilayer chip inductors is limited due to intrinsically low electrical resistivity of the materials, since the directly screen printed conductor winding on top of the ferrite layers will be shorted out. Coating an insulation layer outside the MnZn-ferrite materials is a plausible approach to circumvent such a difficulty.[5–9] NiZn-ferrite materials possess high magnetic inductance and electrically insulating characteristics. Moreover, they have crystal structure and lattice parameters similar with those of MnZn-ferrite. However, the co-firing behavior and the interfacial characteristics, which are necessary informations in order to apply the NiZn-ferrite as insulating coating outside the MnZn-ferrite,[10,11] are still not well understood.

Therefore, in this paper, we systematically investigate the influence of co-firing process on the magnetic properties of MnZn-ferrite and NiZn-ferrites materials is examined and the possible mechanism is discussed.

2. Experimental

The samples were prepared from commercial calcined $(Mn_{0.73}Zn_{0.21}Fe_{0.06})Fe_2O_4$ (Saikei, Co. Japan) and $(Ni_{0.48}Zn_{0.52.})Fe_2O_4$ powders. Either pure MnZn-ferrites toroids or MnZn/NiZn-composite ferrite toroids were prepared. In the latter the MnZn-ferrite layer is sandwiched in between the NiZn-ferrite with thickness ratio 1:5:1. In the conventional sintering (cs) process, the toroids were sintered at 1250–1330°C for 3 h (in air) and cooled in N_2-atmosphere to suppress the oxidation of the MnZn-ferrite materials. In microwave sintering (ms) process, a 2.45 GHz microwave generated from a 3 kW magnetron (Gerling, GL 107) was fed into an applicator made of WR284 waveguide. The samples were heated at a rate of 30°C/min to 1250–1330°C, soaking for 30 min (in air), and then cooled at a rate of 10°C/min down to 800°C. Both the cs- and ms-processed samples were post-annealed at 1200°C (2 h) under 1.5°C Po_2 to adjust the oxidation state (Fe^{2+}/Fe^{3+} ratio).

The crystal structure and the microstructure of the samples were examined using x-ray diffractometry (XRD, Rigaku D/max-IIX) and scanning electron microscopy (SEM, Jeol JSM-840A). The initial permeability- temperature (μ_i-T) characteristics, were estimated from the

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inductance of a coil wound on the toroids, which were measured using HP 4284A LCR-meter. The power loss of the toroids was evaluated at 500 kHz operation frequency regime (50 mT), in the range of 25–105°C, by using Iawatz 8623 B-H tracer.

3. Results and discussion

3.1. Sintering on pure MnZn-ferrites

Fig. 1(a) (open squares) indicates that when sintered by the cs-process, the density of the materials increases with sintering temperature, achieving 4.76 g/cm^3 (95.6% T.D.) when sintered at 1330°C. By contrast, it requires only 15 min to achieve the same density [solid squares, Fig. 1(a)] when firing the MnZn-ferrites by using the microwave sintering (ms) process. Extending the soaking time to 30 min further increases the density of the materials [solid diamond, Fig. 1(a)]. The grain size distributes uniformly for both cs- and ms-processed MnZn-ferrites. The grains are about 10 μm in size when conventional-sintered and is about 6.5 μm in size when microwave-sintered, as shown in Fig. 2. These results demonstrate the beneficial effect of microwave sintering (ms) processes on enhancing the densification of MnZn-ferrites, as compared with the cs-processes.

The ms-processed MnZn-ferrites possess similar initial permeability-temperature (μ_i-T) characteristics to the cs-processed samples [Fig. 3(a)], but exhibit markedly better power loss characteristics than the conventional-sintered ones [Fig. 3(b)]. The room temperature (25°C) power loss for ms-materials [$(P_L)=220$ kW/m^3] is more than 25% smaller than that for the cs-materials [$(P_L)=280$ kW/m^3], which is presumably due to the small grain size of the ms-materials, as compared to that of the cs-samples.

3.2. Co-firing of MnZn/NiZn-composite ferrites

The sintering behavior of the MnZn/NiZn-composite ferrites is not markedly different from that of the pure MnZn-ferrites. Fig. 1(b) shows that the density of the co-fired samples varies with the sintering temperature and soaking time in exactly the same manner as those of the pure MnZn-ferrites, no matter whether they were microwave or conventional sintered. The microstructure of MnZn-ferrite of the co-fired materials is the same as that of the pure MnZn-ferrite (not shown). However, the μ_i-T characteristics of the materials were markedly modified due to the co-firing process. The secondary maximum peak of the μ_i-T curve shifted from 85 to around 50°C and the μ_i-value was reduced from 3200 to around 2450 for both ms- and cs-processed samples [Fig. 4(a)]. It is expected that the magnetic properties of the co-fired materials are still predominated by that of

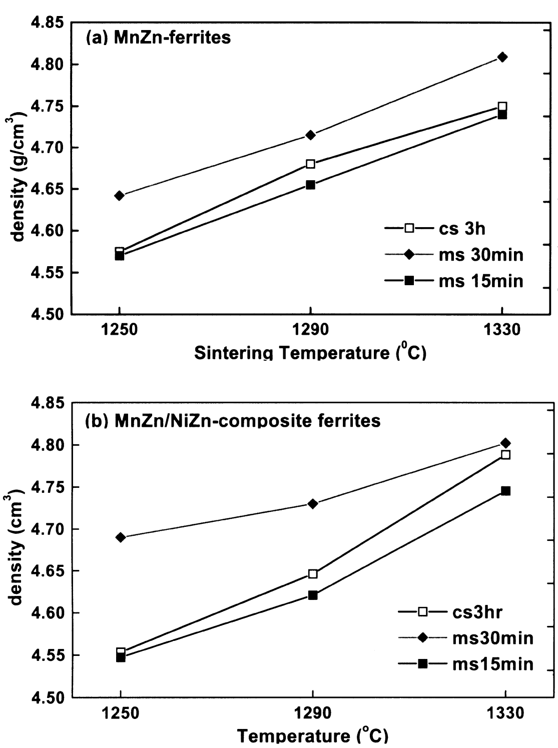

Fig. 1. Variation of density of (a) pure MnZn-ferrites and (b) MnZn/NiZn-composite ferrites with sintering temperature.

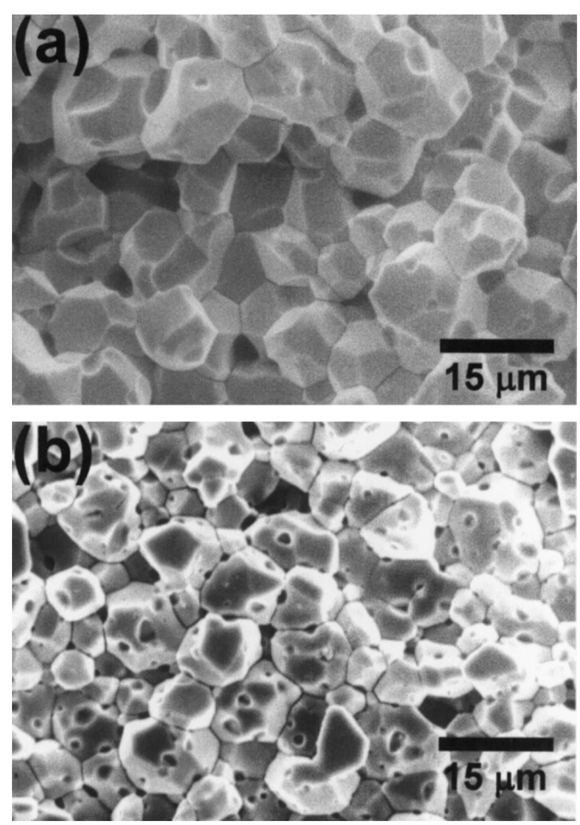

Fig. 2. Typical SEM microstructure of MnZn-ferrites densified by (a) convectical furnace sintering, cs, or (b) microwave sintering, ms, process.

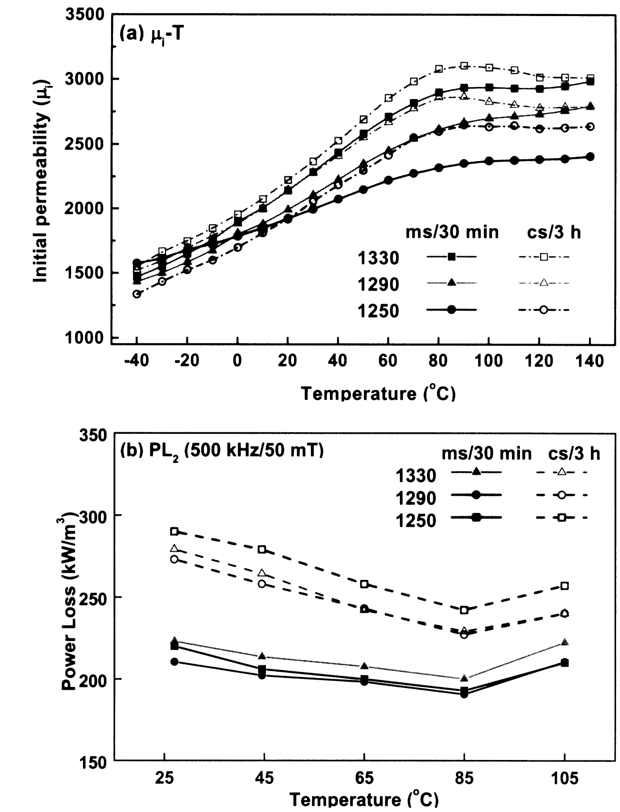

Fig. 3. Temperature dependence of (a) initial permeability, μ_i, and (b) power loss at 500 kHz/50 mT for pure MnZn-ferrites.

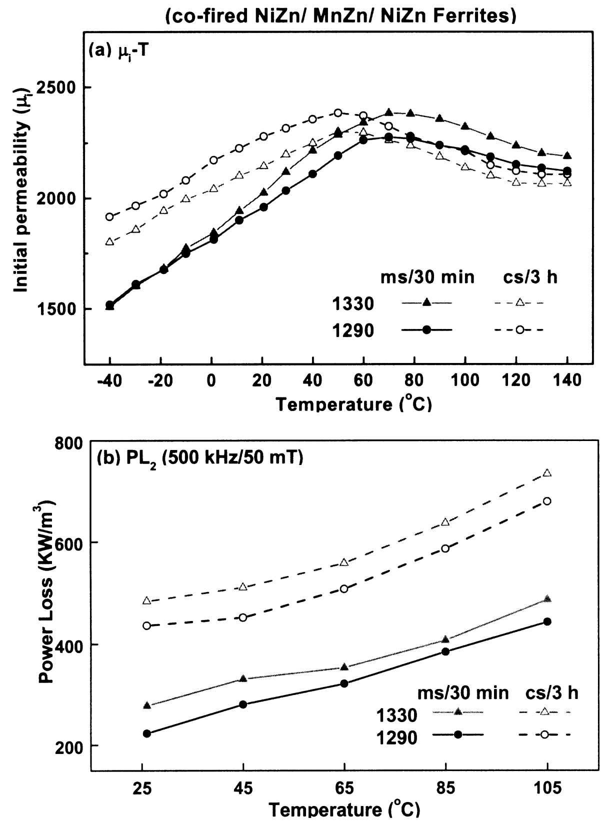

Fig. 4. Temperature dependence of (a) initial permeability, μ_i, and (b) power loss at 500 kHz/50 mT for MnZn/NiZn-composite ferrites.

the MnZn-ferrites, since the proportion of the NiZn-ferrites is only about 28.57 vol.% for the sandwiched samples. The pronounced shifting in T_{smp}-value and significant decrease in $(\mu_i)_{max}$-value are very similar with the phenomena when the MnZn-ferrites were subjected to external or internal stress.[12] Therefore, the modification on co-fired MnZn-ferrites indicated in Fig. 4(a) is attributed to the induction of internal stress onto MnZn-ferrites, which were sandwiched in-between two NiZn-ferrites. The internal stress is presumably resulted from the difference in densification rate (or thermal expansion coefficient) between MnZn-ferrites and NiZn-ferrites.[10]

The co-firing process markedly increases the power loss (P_L) of the cs-materials, but only moderately modifies the P_L-value for ms-materials [Fig. 4(b)]. The dramatic increase in power loss is, again, ascribed to the induction of internal strain on MnZn-ferrites sandwiched in between the NiZn-ferrites. The degradation on magnetic properties of the MnZn-ferrites due to co-firing process is less significant when they were microwave sintered. One of the probable factors is that the ms-samples possess smaller grain size, which renders the MnZn-ferrite portion of the co-fired samples less sensitive to the internal strain induced.

The electrical properties of the sandwich structured MnZn/NiZn-composite ferrite was measured with the frequency sweeping from 100 Hz to 40 MHz. Fig. 5, the complex-impedance ($R-X$) plots of the MnZn/NiZn-ferrites, shows that a semi-circle is observed for all the R–X plots. To understand the electrical characteristics thus measured, both the MnZn-ferrite and NiZn-ferrite segments are simulated by a parallel combination of resistor and capacitor. The intercepts of the R–X plots at DC end represent the summation of resistance of MnZn-ferrites segments and NiZn-ferrite segments, whereas the

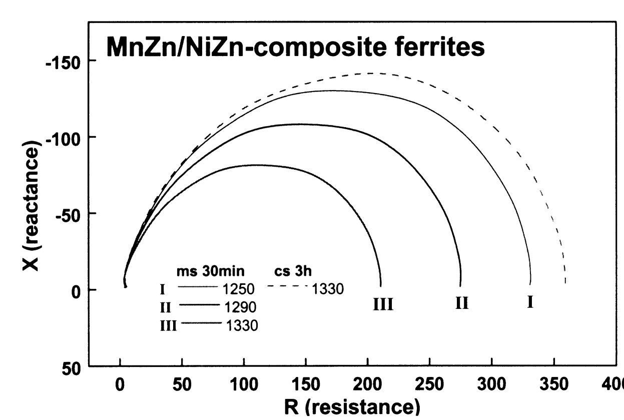

Fig. 5. Complex impedance (R–X) plots of MnZn/NiZn-composite ferrites.

intercepts of these plots at very high frequency (40 MHz) represent the case where the high resistance segments in MnZn/NiZn-composite ferrite were short circuited. Fig. 5 shows that the DC resistance of MnZn/NiZn-composite materials is very close to those for pure MnZn-ferrites although the DC resistance of NiZn-ferrites is very large ($R_{NiZn} \equiv 7.5 \times 10^5$ Ω). The implication of these results is that pronounced interdiffusion between MnZn- and NiZn-ferrites might have occurred. Such a interdiffusion process lower the electrical resistance of NiZn-ferrites and is presumably the prime factor degrading the magnetic properties of MnZn-ferrites.

4. Conclusion

The effect of co-firing process on the characteristics of MnZn-ferrite materials was systematically examined. The degradation of the magnetic properties is attributed to the strain induced in the MnZn-ferrites. The decreases in initials permeability (μ_i) and the increases in power loss (P_L) is less extent for the materials densified by the microwave sintering process. Such a phenomenon is attributed to a smaller grain size in MnZn-ferrites and a sharper interface between MnZn-ferrite to NiZn-ferrite, when the materials were microwaves sintered.

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