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Improvement on magnetic power loss of MnZn-ferrite materials by V_2O_5 and Nb_2O_5 co-doping

S.H. Chen [a], S.C. Chang [a], C.Y. Tsay [a], K.S. Liu [a], I.N. Lin [b,*]

[a] Department of Materials Science and Engineering, National Tsing-Hua University, Hsinchu 300, Taiwan, ROC

[b] Materials Science Center, National Tsing-Hua University, Hsinchu 300, Taiwan, ROC

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Abstract

Simultaneous incorporation of V_2O_5 and Nb_2O_5 dopants into low loss MnZn-ferrites markedly improves the power loss characteristics of the materials, provided no abnormal grain growth phenomenon was induced. The finer the grain size is, the smaller the power loss. The beneficial effect of V_2O_5 and Nb_2O_5 co-doping is presumed to be the reduction on the eddy current loss for the MnZn-ferrite materials. However, the prime factor reducing the power loss in high frequency regime (3 MHz) is the suppression on residual power loss of the materials. The mechanism for the decrease in the residual power loss, in addition to the reduction on grain size (GS < 2.27 μm) and the increase in grain boundary resistance (r_{gb} > 70 Ω), is probably the increase in the uniformity of grain size distribution.

Keywords: Co-doping; Ferrites

1. Introduction

MnZn-ferrite materials possess marvelous magnetic properties such as high initial permeability (μ_i), large magnetic induction (Bs) and high electrical resistivity (ρ) and are widely applied as core materials for transformers in switching mode power supplies (SPS) or DC-to-DC converters.[1,2] The operation frequency of these power supplies has been increased markedly from 100 kHz to 10 MHz for the purpose of reducing the size of the magnetic components, in accordance with the trend for the miniaturization of electrical systems.[3–6] Therefore, how to lower the power loss factor of MnZn-ferrite materials at such a high operation frequency is urgently needed.

The conventional approach for reducing the power loss factor for the MnZn-ferrite materials is to lower the eddy current loss of the materials, since this loss mechanism is one of the most important factors limiting the application frequency for the materials.[7–9] The CaO and SiO_2 additives, which result in an insulating layer surrounding the grain boundaries significantly, improve the power loss of the materials via the increase in grain boundary resistance. However, uniform distribution of such a high resistance layer is needed to effectively reduce the eddy currant loss of the materials. Various kinds of additives, such as HfO_2, Ta_2O_5, V_2O_5[9–12] have been reported to be beneficial for improving the microstructure uniformity of the CaO-SiO_2 incorporated MnZn-ferrite materials. The details on how the addition of these additives modify the characteristics of CaO-SiO_2 incorporated MnZn-ferrite materials is still not well understood. In this paper, we systematically examined the effect of V_2O_5 or Nb_2O_5 addition on the microstructure and power loss of the materials. The correlation between these characteristics is discussed.

2. Experimental

The MnZn-Ferrite materials with the nominal composition, $(Mn_{0.73}Zn_{0.21}Fe_{0.06})Fe_2O_4$, which contains 200 ppm SiO_2 and 500 ppm CaO, were prepared from high purity Fe_2O_3, Mn_3O_4 and ZnO via mixed oxide process. The powder mixtures were calcined at 1000°C for 2 h, and then pulverized down to ~1 μm size. In the V_2O_5

* Corresponding author. Tel.: +886-3-572574; fax: +886-3-571 6997.

E-mail address: inlin@mx.nthu.edu.tw (I.N. Lin).

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based samples, 0.03 wt.% of Nb_2O_5, in addition to 0.1 wt.% V_2O_5, was mixed with calcined MnZn-ferrite powders, followed by spray drying process. These materials are designated as V_2O_5-riched samples. For comparison, the Nb_2O_5-based MnZn-ferrite powders, which contain 0.03 wt.% V_2O_5, in addition to 0.1 wt.% Nb_2O_5, were prepared by similar process and were designated as Nb_2O_5-riched samples.

The samples of toroidel geometry (8 mm I.D.×16 mm O.D.×6.75 mm high) with 3.0 g/cm^3 green density, were debindered at 400°C for 2 h and then sintered at 1025–1175°C in controlled atmosphere for 5 h. In order to maintains the Fe^{2+}/Fe^{3+} ratio obtained at sintering temperature, the oxygen partial pressure of the sintering atmosphere was varied with the temperature during sintering and cooling periods (J/min) in accordance to the formula:

$$\text{Log}\ (PO_2) = -14540\ /T(\text{K})\ + 10 \quad (1)$$

The heating process was controlled at 10°C/min in nitrogen atmosphere. The power loss of the samples were measured by Iwatsu 8623 B-H analyzer from 100 kHz to 3 MHz at $Bm=20$ mT. The frequency dependence of inductance and impedance ($Z=R+jX$) were measured by HP 4194A gain phase analyzer. The microstructures of the samples were examined by using optical microscopy.

3. Results and discussion

Incorporation of V_2O_5 and Nb_2O_5 into the MnZn-ferrite materials, which already contain 200 ppm SiO_2 and 500 ppm CaO additives, markedly enhances the densification kinetics for the materials.[13] Fig. la shows that the density of the samples increases monotonously with the sintering temperature. Typical granular structures examined using SEM is illustrated in Fig. 2a, showing that most of the samples contain fine grains with very uniform size distribution. Abnormal grain growth phenomenon is induced when the high Nb_2O_5-riched samples were sintered at 1150ºC and higher temperature, as illustrated in Fig. 2b. In these samples, the small grains remained as a size finer then 3 μm before they were consumed by the abnormally growing large grain. The variation of grain size with sintering temperature are shown in Fig. 1b, indicating that the grain size of Nb_2O_5-riched samples is larger than that of the V_2O_5-riched sample, when the materials were sintered at 1050–1100°C.

The power loss (PL) of the MnZn-ferrite materials, which is the most important magnetic properties of concern, varies with the sintering temperature markedly. The sintering temperature dependence of the power loss for the blank, V_2O_5-riched and Nb_2O_5-riched samples

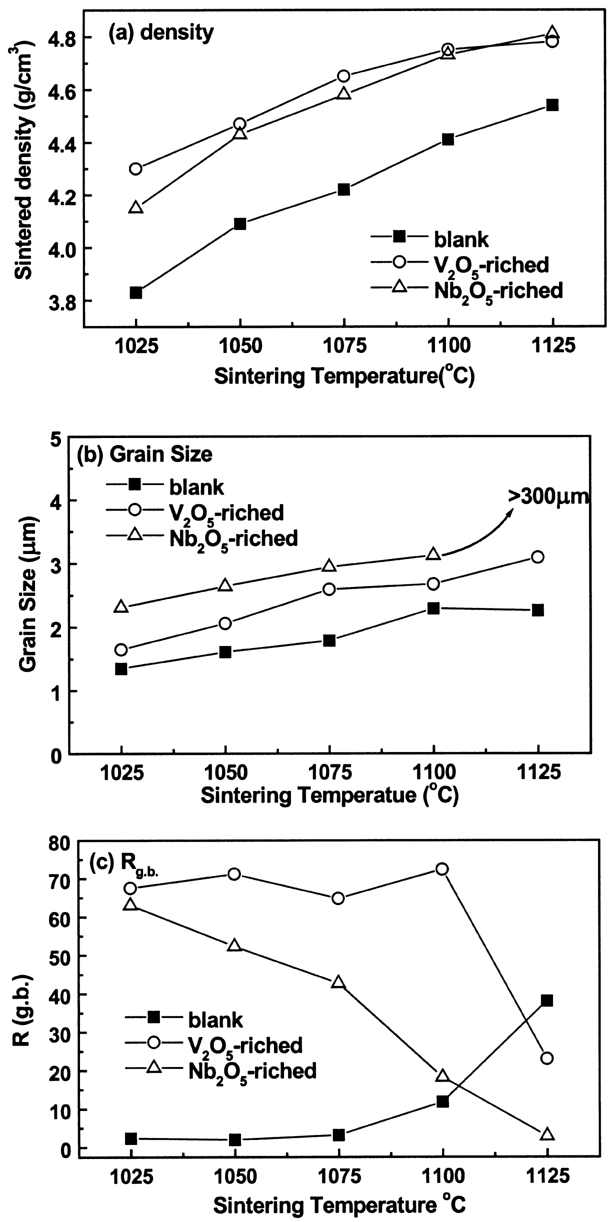

Fig. 1. Variation of (a) sintered density, (b) grain size and (c) grain boundary resistance of MnZn-ferrite materials with sintering temperature (blank materials contain 200 ppm SiO_2 and 500 ppm CaO; V_2O_5-riched materials are blank materials incorporated with 0.1 wt.% V_2O_5 and 0.03 wt.% Nb_2O_5; Nb_2O_5-riched materials are blank materials incorporated with 0.1 wt.% Nb_2O_5 and 0.03 wt.% V_2O_5).

are illustrated in Fig. 3. These figures indicate that the power loss (PL) of the samples is maintained at low level as long as the grain size is small. The power loss characteristics degrade substantially whenever the abnormal grain growth phenomenon is triggered. Higher density usually leads to lower power loss for the materials, provided that no abnormal grain growth phenomenon has occurred. It should be noted that all the 1025°C-sintered samples show large power loss properties regardless of V_2O_5 (Nb_2O_5) content in the samples, which is owing to

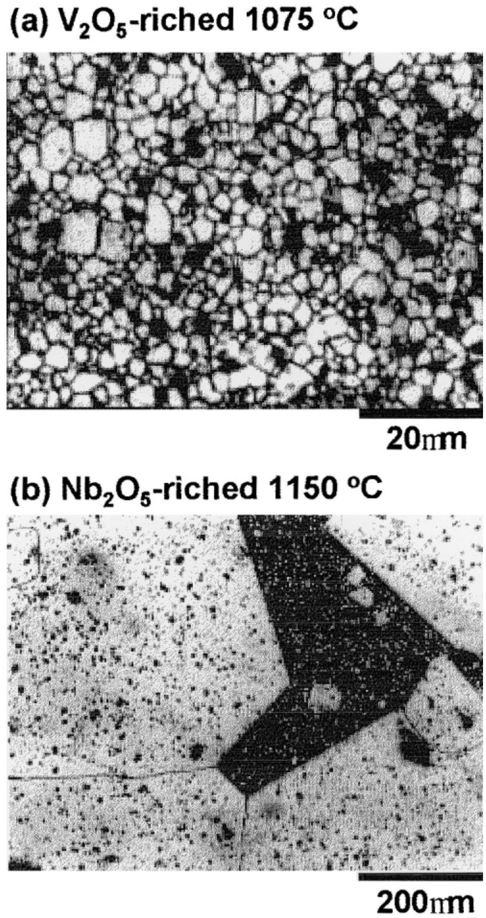

Fig. 2. Typical SEM micrographs of (a) fine grain and (b) duplex microstructured MnZn-ferrite materials.

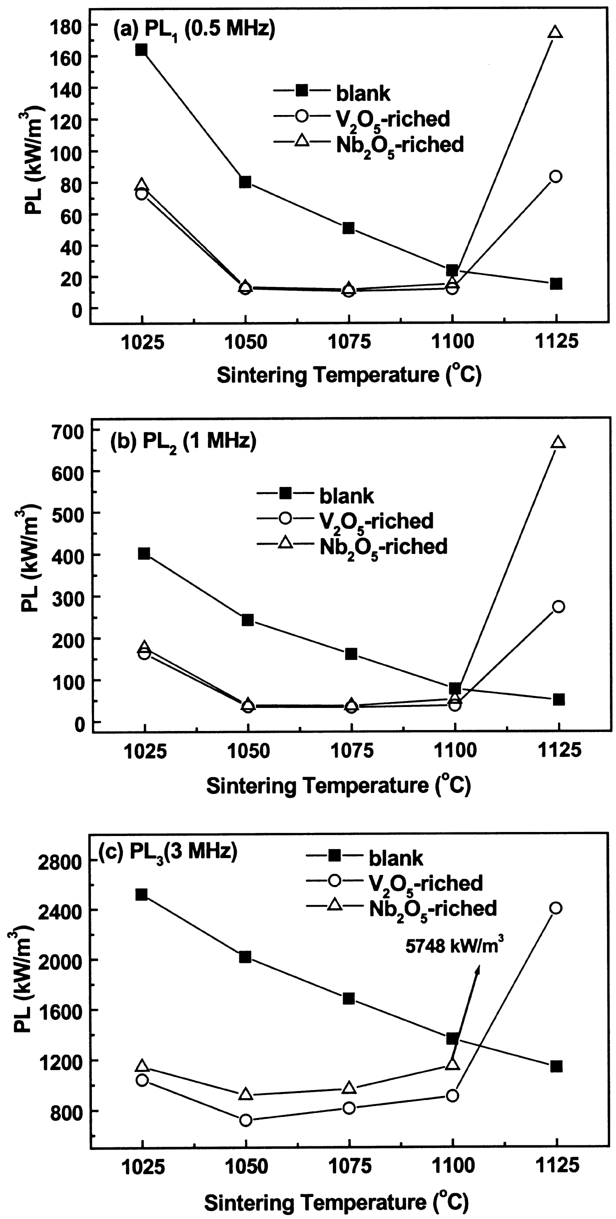

Fig. 3. Variation of power loss of MnZn-ferrite materials with sintering temperature (blank materials contain 200 ppm SiO_2 and 500 ppm CaO; V_2O_5-riched materials are blank materials incorporated with 0.1 wt.% V_2O_5 and 0.03 wt.% Nb_2O_5; Nb_2O_5-riched materials are blank materials incorporated with 0.1 wt.% Nb_2O_5 and 0.03 wt.% V_2O_5).

the presence of secondary phase due to insufficient sintering.

Moreover, the Nb_2O_5-riched samples possess slightly larger grain size than the V_2O_5-riched samples. Slightly larger grain size insignificantly alters the low frequency power loss characteristics, PL_1 (0.5 MHz) and PL_2 (1 MHz), for the MnZn-ferrite materials, but results in about 10% increase in the high frequency power loss, PL_3 (3 MHz), for the materials. Detailed examination indicates that the granular size distribution of the Nb_2O_5-riched samples is less uniform than that of the V_2O_5-riched samples. Multi-domain structure may occur for the large grains, which is the probable mechanism leading to higher power loss for the large grain samples.

To understand how the grain growth phenomenon degrades the power loss properties, the grain and grain boundary resistances were analyzed by using complex impedance technique. For the first, the electrical properties of the materials were modeled by series combination of grain and grain boundary *R–C* lump circuits. The reactance (X) of the samples was plotted against their resistance (R) over wide range of frequency regime. The grain resistance (r_g) is then approximated as the resistance (R) at high frequency end of *R–X* plots and the grain boundary resistance (r_{gb}) is approximated as the difference between DC and high frequency resistance, viz.

$$r_g = R_{dc},\ \ r_{g.b.} = R_{dc} - R_\infty \qquad (2)$$

The results of analyses are shown in Fig. 1c, indicating that grain boundary resistance (r_{gb}) is markedly lowered when the grains grow to a size larger than 2.5 μm. It should be noted that the reduction on grain boundary resistance due to gain growth occurs more prominently for the materials containing Nb_2O_5 species. Such a

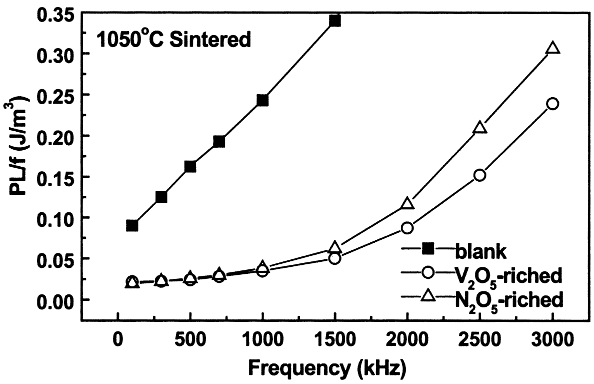

Fig. 4. Typical frequency dependence of power loss of MnZn-ferrite materials (blank materials contain 200 ppm SiO_2 and 500 ppm CaO; V_2O_5-riched materials and blank materials incorporated with 0.1 wt.% V_2O_5 and 0.03 wt.% Nb_2O_5; Nb_2O_5-riched materials are blank materials incorporated with 0.1 wt.% Nb_2O_5 and 0.03 wt.% V_2O_5).

phenomenon is in accord with the assumption that V_2O_5-species tend to remain at grain boundary region, acting as liquid phase sintering aids, whereas Nb_2O_5-species tend to dissolve in the grains, acting as aliovalent substitution dopants.

The mechanism that the V_2O_5 and Nb_2O_5 incorporation modifies the power loss characteristics of the MnZn-ferrtie materials can be understood, using the Wijn's model. In this model, the power loss of the materials is presumed to be contributed by magnetic hysterasis loss (*Ph*), eddy current loss (*Pe*) and residual loss (*Pr*), viz.

$$\mathrm{PL} = \mathrm{Ph} + \mathrm{Pe} + \mathrm{Pr} \qquad (3)$$

The magnetic hysteresis increases with operating frequency linearly, $Ph = Kh \times f$, whereas the hysteresis loss increases with frequency quadratically, $Pe = Ke \times f^2$. The coefficients of magnetic hysteresis loss (*Kh*) and eddy current loss (*Ke*) can be approximated as the intercept and slope of $PL/f - f$ plots, since

$$PL/f = Kh + Ke \times f + (Pr/f) \qquad (4)$$

Typical $PL/f - f$ plots are illustrated in Fig. 4 for the materials sintered at 1050°C. The *Kh* and *Ke* values can be estimated from PL/f-f plots for all the materials. The magnetic hysteresis loss ($Ph = Kh \times f$) and eddy current loss ($Pe = Ke \times f^2$) can then be calculated and residual loss (*Pr*) can be derived from Eq. (3). The power loss processes (*Ph*-, *Pe*- and *Pr*- values) for the best MnZn-ferrite materials measured at 0.5, 1.0 or 3.0 MHz operating frequencies were listed in Table 1a–c, respectively.

The power loss is mainly contributed by the magnetic hysteresis loss (*Ph*) in low operating frequency regime (0.5 MHz, Table 1a). The relative contribution of eddy current loss (*Pe*) increases with operating frequency, surpassing the magnetic hysteresis loss (*Ph*) at 3 MHz operating frequencies (Table 1b and c). The residual power loss (*Pr*), which is negligibly small, at 0.5 and 1 MHz operating frequencies, predominates the power loss at high operating frequencies (3 MHz). The *Pr*-value contributed more than 80% of total power loss at 3 MHz (Table 1c).

Table 1
The variation of magnetic power loss (*Ph*), eddy current power loss (*Pe*) and residual power loss (*Pr*) of V_2O_5-riched and Nb_2O_5-riched MnZn-ferrites with sintering temperature

	V_2O_5-riched[a]		Nb_2O_5-riched[a]	
	1050°C	1075°C	1050°C	1075°C
(a) (0.5 MHz)				
Ph[b]	9.95	7.95	8.7	7.6
Pe[b]	2.5	3.0	4.25	4.0
Pr[b]	~0	~0	~0	~0
(b) (1 MHz)				
Ph[b]	19.9	15.9	17.4	15.2
Pe[b]	10.0	12.0	17.0	16.0
Pr[b]	4.9	5.1	3.9	5.8
(c) (3 MHz)				
Ph[b]	59.7	47.7	52.2	45.6
Pe[b]	90.0	108.0	153	144
Pr[b]	567.3	655	712	774

[a] V_2O_5-riched MnZn-ferrite materials contain 0.1 wt.% V_2O_5 and 0.03 wt.% Nb_2O; Nb_2O_5-riched MnZn-ferrite materials contain 0.1 wt.% Nb_2O_5 and 0.03 wt.% V_2O_5.
[b] All the power losses are in kW/m^3.

4. Conclusion

Effect of V_2O_5 and Nb_2O_5 co-doping on the power loss characteristics of MnZn-ferrtie materials were systematically examined. The sintered density of the materials increases monotonously with the sintering temperatures. The grain size remains at small value (GS < 3.24 μm) for all samples except for those samples which contain large proportion of co-dopants and were sintered at too high temperature. All the large grain samples (GS > 300 μm) exhibit high power loss. Only the small grain materials (GS < 3.24 μm) own small power loss properties and the finer the grain size is the better the *PL*-values. Complex impedance analysis reveals that the prime mechanism is the increase in grain boundary resistance.

Acknowledgements

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