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Low-Loss Co_2-Y Ferrites with Added CuO Sintered in Air for High Frequency Application

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Abstract

The sintering characteristics of hexagonal Co_2-Y-type ferrite, $Ba_2Co_2Fe_{12}O_{22}$, with the addition of 0.6 wt% CuO, were studied in order to allow for preparation in air, as opposed to the conventionally recommended O_2, for industrial production. The dependence of the resistivity, ρ magnetic loss, tanδ, and the permeability, μ, at 1 GHz on the sintering temperature was investigated. A low tanδ of 0.05 with a μ of 2.7 at a frequency of 1 GHz, along with a high ρ (up to 7 × 10^4 µm), were attained under sintering at 1170˚C in air, which were the same features as those of samples sintered at 1200˚C in O_2. The dependence of tanδ on grain diameter was also examined, and it was determined that a small grain size (less than 2 µm) is preferable for low tanδ.

Keywords

Ferrite, Hexagonal, Y-Type, Sintering, Low Loss

1. Introduction

Ferrites and other magnetic materials have been widely used as the key elements in microwave devices [1]-[3] such as isolators, phase shifters [4] [5], and circulators [6]. Traditional ferrites, known as spinel types, such as Ni-Zn ferrites have been known to exhibit high permeability in the frequency range up to a few hundred MHz because of restriction by Snoek's law [7] [8]. Therefore, hexagonal ferrites are expected to be promising candidates for expansion of the device frequency to the GHz range [9]. Z- and Y-type ferrites show soft magnetic characteristics with moderate relative permeability, μ, up to 1 GHz [10]-[12]. The former is denoted as $Ba_3Me_2Fe_{24}O_{41}$, while the latter is labeled $Ba_2Me_2Fe_{12}O_{22}$, where Me represents a divalent metal ion from the first transition series or, alternatively, it may represent Zn or Mg. In particular, the Y-type ferrite has a high Curie temperature

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[11] and higher magnetic resonance than the Z-type, despite a low μ [13] [14]. Therefore, it is more applicable to high frequency devices in the GHz range.

Recently, telecommunication devices applied to mobile phones have broadened their market extensively. For these applications, antennas are essential, and miniaturization of these devices is therefore necessary. Ferrites possess permeability as well as permittivity, and are considered as a candidate material for chip antennas [15]-[19], because the wavelength is reduced proportionally according to $1\sqrt{\mu\varepsilon}$ where ε is the relative permittivity. The Y-type ferrite, $Ba_2Co_2Fe_{12}O_{22}$Y (Co_2-Y), with a high Curie temperature of ~330˚C [11], is a promising material for this application.

We have demonstrate that Co_2-Y modified by the addition of 0.6 wt% CuO exhibited a moderate μ of ~2.7 and low magnetic loss, tanδ, of 0.05, even at 1 GHz [20]. This material has been prepared by means of a conventional powder metallurgical process, and sintering has been conducted under the conventionally recommended oxygen atmosphere. However, sintering of the Co_2-Y in air would be preferable for industrial production because of cost effectiveness. To date, some studies have been conducted on Y-type $Ba_2Zn_{2-2x-2y}Co_{2x}Cu_{2y}Fe_{12}O_{22}$ ($0 \leqq x \leqq 0.1$) sintering in air [14] [21]. In addition, Co_2-Y (with no added CuO) sintering in air has also been reported [22], which showed a high resistivity of 5×10^4 Ωm but did not show values of tanδ. The industrially favorable sintering conditions (in air) of 0.6 wt% CuO added to Co_2-Y will be presented in this study in order to attain the same characteristics as the samples sintered in O_2 in our early study mentioned above. The effective factor of tanδ will also be discussed.

2. Experimental

Samples of Co_2-Y ferrite were prepared as a stoichiometric composition by means of conventional powder metallurgy. Raw material powders of Fe_2O_3, $BaCO_3$, and Co_3O_4 were well-mixed using ball-milling and calcinated at 1000˚C for 2 h in air. The calcinated powders were ground, with the addition of 0.6 wt% CuO powders and of a 1 wt% PVA binder, followed by compacting into predetermined shapes at a pressure of 20 MPa and then sintered at 1200˚C for 3 h under atmospheres with varying oxygen content (balance: nitrogen).

The resistivity, ρ, was measured for samples whose dimensions were 13 mm in diameter and 3 mm in thickness. An electrode was then printed on both sides of the samples as a silver paste. The permeability and permittivity frequency response were characterized by means of a network analyzer (Agilent E8364A) and a coaxial airline fixture (KANTOH E.A.D. Co. Model: CSH2-APC-7) up to 18 GHz, after the Nicolson-Ross method [23]. Ring shaped samples (ID: 3.0 mm, OD: 7.0 mm, thickness: 3.5 mm) were used in this characterization.

The sample densities were determined by means of Archimedes' method, while their morphologies were investigated using a scanning electron microscope (SEM) (Hitachi S-800). The grain diameters were defined as the average diagonal length of approximately 30 grains, orienting the hexagonal shape towards the top, in the SEM images.

3. Results and Discussion

The Co_2-Y samples with the added 0.6 wt% CuO were prepared in various O_2 volume configurations at a sintering temperature of 1200˚C. The O_2 volume fraction varied from 15% - 100%. The dependence of μ, tanδ at 1 GHz, the density, and ρ, on the O_2 volume fraction are shown in **Figure 1(a)** and **Figure 1(b)**, respectively. Both μ and tanδ decrease with increasing O_2 volume fraction. The lowest tanδ (at 0.05) with a μ of 2.7 was achieved at a full O_2 atmosphere (100%), while the atmospheric case, *i.e.*, sintering in air (20% O_2), exhibited a high tanδ of 0.15 along with a high μ of ~4. It can be seen that the density increases while ρ decreases with decreasing O_2 volume fraction. A high density of 5.24×10^3 kg/m^3 and a low ρ of 1.2×10^4 Ωm were achieved in air. Low tanδ is required for energy-conversion devices such as inductors and antennas. High ρ is also required, because winding coils or printed electrodes make contact with ferrites directly. These characteristics were not attained when sintering at 1200˚C conducted in air, as shown in **Figure 1**. Therefore, the sintering temperature in air must be considered.

The dependence of ρ on the sintering temperature in O_2 and in air is shown in **Figure 2**, where open and black circles denote sintering in O_2 and in air, respectively. ρ decreases with sintering temperature for both atmospheres, although the change in air is steeper than that in O_2. A resistivity greater than 7×10^4 Ωm in the case of sintering in O_2 is reached below 1180˚C in air.

The change in μ and tanδ at 1 GHz due to sintering temperature in O_2 and in air are shown in **Figure 3(a)** and

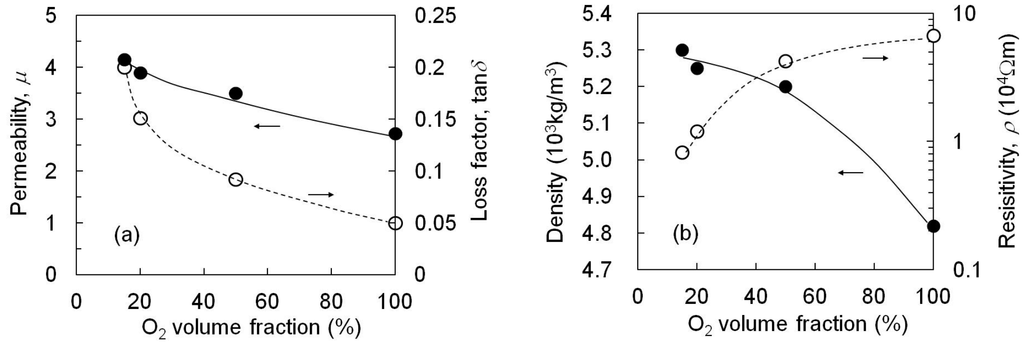

Figure 1. Dependence of (a) μ and tanδ, and (b) density and ρ, on O_2 volume fraction at sintering.

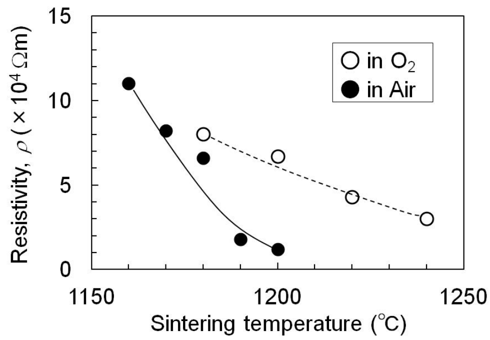

Figure 2. Dependence of ρ on sintering temperature in O_2 and in air.

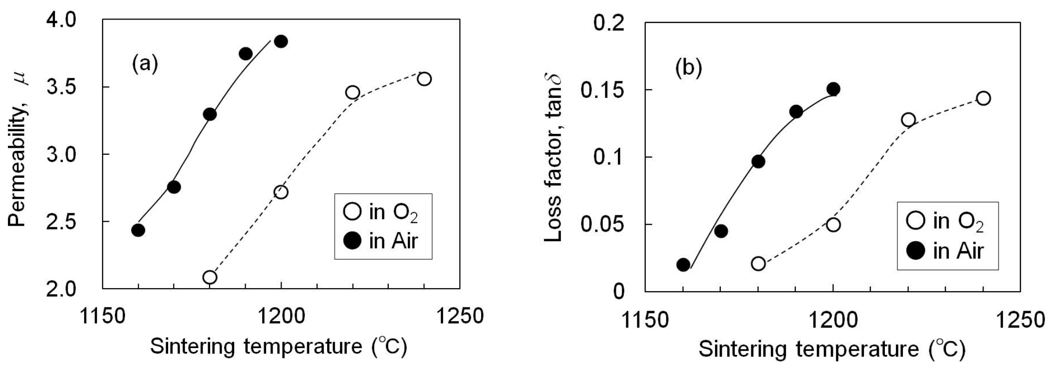

Figure 3. Change in (a) μ and (b) tanδ at 1 GHz due to sintering temperature in O_2 and in air.

Figure 3(b), respectively. μ and tanδ increase with sintering temperature for both atmospheres, and the behavior in the air case are shifted to a lower temperature than that of the O_2 case. The target characteristics, namely, μ of 2.8 with a low tanδ (less than 0.05 at 1 GHz), are attained at 1170˚C in the case of sintering in air, which is almost identical to the sample sintered at 1200˚C in O_2. It was proven that the crystal structure of this sample was that of a Co_2-Y ferrite using X-ray diffraction as shown in **Figure 4**, where no spinel phase was detected as in [21].

The fractured surfaces of the samples with identical characteristics and sintered in different atmospheres are shown in **Figure 5**. Here, **Figure 5(a)** is an image of a sample sintered at 1200˚C in O_2, while **Figure 5(b)** shows a sample sintered at 1170˚C in air. The density of the former is 4.82×10^3 kg/m^3 and that of the latter is 4.90×10^3 kg/m^3, respectively. Both samples have the same morphological aspect with small grains isolated by fine pores. In addition, the majority of the grains have a pseudo-hexagonal platelet shape with thin thickness. The grain sizes, defined as the diagonal length of the hexagonal faces, are estimated as being 2.0 μm in the O_2 case and 1.8 μm for the air case from these images. It is assumed that the same magnetic characteristics are attributed to the same morphological nature in both samples.

The relationship between grain diameter and tanδ is shown in **Figure 6**. All samples fabricated in this study are plotted on this figure regardless of sintering conditions. The value of tanδ increases with grain diameter, and rises abruptly from 0.05 to 0.15 around a grain diameter of ~2 μm. It has been reported previously that energy dissipation in inductors composed of NiZn ferrites is affected by grain size, and that the dissipation was minimized at a single domain size of 2 - 3 μm [24]. The magnetization process is classified into a spin rotational mode or a magnetic domain wall motion depending on magnetic domain sizes. The rotational mode dominates in the case of small grain diameters equal to magnetic single domain sizes, while larger grain sizes lead to two- or multi-domain structures.

The abrupt increase in tanδ by more than 0.1 beyond a diameter of 2 μm could be attributed to the switching of the magnetization mode from the spin rotational mode to the magnetic domain wall motion. The critical size of a magnetic single domain could be estimated as being approximately 2 μm in the Co_2-Y ferrite. Therefore, controlling the grain size has an effect on reducing tanδ.

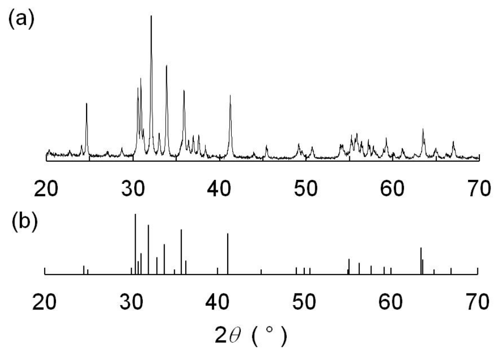

Figure 4. X-ray diffraction profiles of (a) the sample sintering at 1700˚C in air, and (b) calculated standard Y-type (CuKμ, λ = 0.15405 nm).

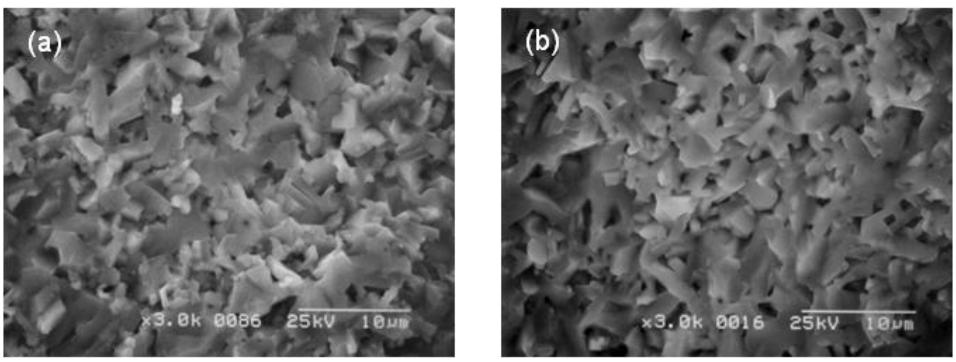

Figure 5. Fractured surface of the samples sintered at (a) 1200˚C in O_2 and (b) 1700˚C in air. Grain sizes were estimated as being ~2.0 μm and ~1.8 μm, respectively.

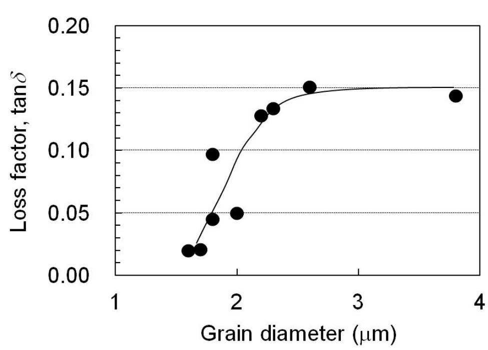

Figure 6. The relationship between grain diameter and tanδ.

It is apparent that the Co_2-Y ferrite with low tanδ even at 1 GHz fabricated in air is favorable for industrial production, and is a promising magnetic material for application to high frequency devices.

4. Conclusion

The sintering characteristics of the hexagonal Co_2-Y-type ferrite, $Ba_2Co_2Fe_{12}O_{22}$, with the addition of 0.6 wt% CuO in air were examined in order to apply the ferrite to industrial production. It was found that sintering at 1170˚C in air resulted in a low tanδ of 0.05, with a high μ of 2.7 at 1 GHz and a high ρ of 7×10^4 μm, identical to a sample sintered at 1200˚C in the conventionally recommended oxygen atmosphere. The relationship between grain size and tanδ was also examined. It was found that tanδ was dependent on grain size, and that a size of less than 2 μm is preferable for reducing tanδ, which suggests the ferrite can be applied to microwave devises with low energy dissipation.

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