Low-Loss Z-type Hexaferrite ($\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$) for GHz Antenna Applications

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Abstract

We report a low magnetic loss $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ (Co_2Z) hexaferrite for use in gigahertz (GHz) antennas. Acid-etching was very effective in removal of unwanted Y-type hexaferrite ($\text{Ba}_2\text{Co}_2\text{Fe}_{12}\text{O}_{22}$) from **calcined** Co_2Z powder. It is found that the **calcined** and acid etched (AE) Co_2Z hexaferrite shows a low magnetic loss tangent ($\tan \delta_\mu$) of 0.012 and 0.037 at 1 and 2 GHz, respectively. These low $\tan \delta_\mu$ are attributed to removal of Y type hexaferrite, which possesses a lower anisotropy field (H_k) than W-type hexaferrite ($\text{BaCo}_2\text{Fe}_{16}\text{O}_{27}$). The figure of merit (FOM) of the AE Co_2Z hexaferrite is 141.7 and 48.7 at 1 and 2 GHz, respectively. These FOM are much higher than the FOM of previously reported low-loss magnetic materials. Therefore, the AE Co_2Z hexaferrite can be a good candidate for GHz antenna application in the ultra-high frequency (UHF) band.

Keywords:

Low magnetic loss; Z-type hexaferrite; GHz antenna application; Acid etching

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1. Introduction

Multi-functional mobile devices require a large number of radio frequency (RF) components, including antennas in the limited space. Accordingly, the size of antenna needs to be reduced without any performance degradation. In response to this, ferrites have been widely studied and used as substrates of GHz antennas. This is because ferrites possess both permeability and permittivity, and are good electrical insulators [1]. However, ferrite is magnetically lossy at gigahertz (GHz) frequencies, which is undesired for antenna applications. We have reported that the experimental radiation efficiency (RE) of a dual polarized hexaferrite antenna increases more or less linearly to 77% from 66% at 2.4 GHz as the magnetic loss tangent ($\tan \delta_\mu = \mu''/\mu'$) decreases to 0.05 from 0.11 at 2.4 GHz [2].

In order to further reduce the magnetic loss tangent at GHz, ferrites with a high magneto-crystalline anisotropy field (H_k) and permeability have been searched. This is because the larger H_k , the higher the ferromagnetic resonant frequency (f_{FMR}) according to Eq. (1), thereby, a higher operation frequency leading to a low magnetic loss.

$$f_{FMR} = (\gamma / 2\pi) \times (H + H_k), \quad (1)$$

where γ is the gyromagnetic ratio and H is the applied dc magnetic field. Also, the device miniaturization factor (n) increases with the relative permeability (μ_r) by Eq. (2) [3],

$$n = \sqrt{\epsilon_r \mu_r}, \quad (2)$$

where ϵ_r is the relative permittivity and μ_r is the relative permeability. Thus, high H_k hexaferrites have received great attention [4-9]. It has been reported that Y-type ($\text{Ba}_{1.5}\text{Sr}_{0.5}\text{Co}_2\text{Fe}_{12}\text{O}_{22}$) hexaferrite shows 2.3 of real part (μ') of complex permeability and 0.095 of magnetic loss

tangent ($\tan \delta_\mu$) at 2 GHz [9]. Moreover, $\text{Zn}_5\text{Ni}_{75}\text{F}_{20}$ /polymer (ER 182) composite has μ' of 3.24 and $\tan \delta_\mu$ of 0.063 at 1 GHz [10]. However, these losses are too high for GHz antenna applications. Z-type ($\text{Ba}_3\text{Co}_{1.4}\text{Cu}_{0.6}\text{Fe}_{24}\text{O}_{41}$) hexaferrite/polymer composite shows low $\tan \delta_\mu$ of 0.025 at 1 GHz, but the composite suffers from a relatively low μ' of 1.36 [11]. Antenna performance is in part determined by the permeability and quality factor (Q -factor = $\mu'/\mu'' = 1/\tan \delta_\mu$) of a magnetic material, i.e. the figure of merit ($\text{FOM} = \mu' \times Q$ -factor). Therefore, high μ' and low magnetic loss of a substrate are desired to achieve high radiation efficiency and miniaturization of GHz antenna.

In this paper, we report a low magnetic loss $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ (Co_2Z) hexaferrite with an 142 of FOM for use in the ultra high frequency (UHF) band antennas. Low magnetic loss tangent and high ferromagnetic resonant frequency of the Co_2Z hexaferrite are explained by ferrite phases.

2. Experimental

The conventional solid state reaction method was used to synthesize Co_2Z hexaferrite powders. A mixture of $\alpha\text{-Fe}_2\text{O}_3$, BaCO_3 , and Co_3O_4 was wet-milled with a high energy ball milling (Spex Sample Prep 8000D) for 4 h. The wet-ball milled powder was **calcined** at 1080 °C to form intermediate phases, including M and Y-phases [12,13]. Then, the **calcined** powder was wet ball-milled for 6 h, followed by **re-calcination** at 1300 °C to obtain Co_2Z hexaferrite phase. One of the **re-calcined** powders was etched in diluted hydrochloric acid at 60 °C for 30 min to remove unwanted ferrite phases. Then, both the **as-recalcined** and acid-etched (AE) Co_2Z powders were subjected to wet-ball milling for 2 h to decrease the particle size, which lead to increase in f_{FMR} [14]. A mixture of **as-recalcined** (or AE Co_2Z) powder and 2 wt% polyvinyl alcohol (PVA) was pressed into toroidal rings (inner diameter: 3.2 mm; outer diameter: 7.0 mm;

Fig. 1. XRD patterns on **as-recalcined** and acid-etched $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ (Co_2Z) hexaferrite powders.

thickness: 2 mm) and sintered at 950 °C for 2 h in air to characterize dynamic magnetic properties.

We have used an X-ray diffractometer (XRD) and vibrating sample magnetometer (VSM) to characterize the **as-recalcined** and AE Co_2Z powders and sintered samples for crystalline phases and static magnetic properties, respectively. Dynamic magnetic properties of both non-acid-etched (NAE) and AE Co_2Z sintered toroidal rings were characterized by an impedance/material analyzer (IA: Agilent E4991A) in the range of 100 MHz – 2 GHz. In addition, a vector network analyzer (VNA: Agilent N5230A) was used in the range of 2 to 10 GHz. The microstructure of **as-recalcined** and AE Co_2Z powders were observed by a field emission scanning electron microscope (FE-SEM: JEOL 7000 FE).

3. Results and discussion

Figure 1 shows XRD patterns of **as-recalcined** and AE Co_2Z powders. The diffraction peaks are indexed to the Z-type, Y-type ($\text{Ba}_2\text{Co}_2\text{Fe}_{12}\text{O}_{22}$), and W-type ($\text{BaCo}_2\text{Fe}_{16}\text{O}_{27}$)

Fig. 2. SEM images of acid-etched $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ (Co_2Z) hexaferrite particles.

hexaferrites. Both **as-recalcined** and AE Co_2Z powders show a major phase of Co_2Z hexaferrite. Reportedly, Z-type hexaferrite contains both Y- and W-type hexaferrite phases [15-17]. However, the Y-type phase disappears after acid-etching, while the W-phase still exists as shown in Fig. 1. This indicates that the Z- and W-phase were not decomposed. The AE Co_2Z particles show hexagonal pores after removal of Y-phases as shown in Fig. 2, but no pore was observed in **as-recalcined** Co_2Z particles. This is possibly attributed to the lower activation energy of the Y-phase than the Z- and W-phases [18].

Magnetic hysteresis loops of **as-recalcined** and AE Co_2Z hexaferrite powders are shown in Fig. 3. Both **as-recalcined** and AE Co_2Z powders are magnetically soft. The AE Co_2Z powder has a higher saturation magnetization (σ_s) of 55 emu/g than 51 emu/g of the **as-recalcined** powder. This is because Y-type hexaferrite has a lower σ_s of 34 emu/g (Co_2Y) as compared to Z-type (50 emu/g) [1] and W-type (~ 78 emu/g) hexaferrites [19]. Fig. 4 shows the magnetic hysteresis loops of NAE and AE sintered Co_2Z samples. By comparing Fig. 3 and 4, it is noticed that the σ_s remains unchanged after sintering both **as-recalcined** and AE powder, but the intrinsic coercivity (H_{ci}) of the NAE and AE sintered hexaferrite increased to 398.5 from 18.9 Oe and to

Fig. 3. Magnetic hysteresis loops of the **as-recalcined** and acid-etched $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ (Co_2Z) hexaferrite powders.

613.8 from 8.4 Oe, respectively. An increase in the H_{ci} is attributed to the smaller grain size and the increase in stress and surface anisotropy during the milling process [14]. In order to obtain the H_k and high field differential susceptibility (χ_p), the M - H curves in Fig. 4 were fitted in the range of 5 – 10 kOe with Eq. (3), which is the law of approach to saturation (LAS) [19,20]. In the case of hexagonal symmetry, the LAS can be expressed as

$$M = M_s \left(1 - A/H - H_k^2 / 15H^2 \right) + \chi_p H, \quad (3)$$

where A is the inhomogeneity parameter and H is the magnetic field. At a large magnetic field, contributions from A/H vanish in theory [19,20]. The static magnetic properties and fitted parameters are given in Table 1. The fitted H_k of NAE and AE sintered Co_2Z hexaferrites are 6.16 and 8.01 kOe, respectively. Therefore, the anisotropy constant (K) of the AE Co_2Z sintered

Table 1

Summary of magnetic properties for non-acid-etched (NAE) and acid-etched (AE) $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ (Co_2Z) sintered samples.

Sample	σ_s (emu/g)	H_c (Oe)	H_k (kOe)	χ_p ($\times 10^{-4}$ emu/g)
NAE Co_2Z	49	398.5	6.16	1.36
AE Co_2Z	55	613.8	8.01	2.13

hexaferrite is higher than that of the NAE Co_2Z hexaferrite. This can be explained by the larger K of W-type ($-3.5 \times 10^6 \sim -5 \times 10^6$ erg/cm³) hexaferrite than that of Y-type (-2.6×10^6 erg/cm³) hexaferrite [16].

With regard to dynamic properties, Fig. 5 shows the measured frequency dependence of the complex permeability in a frequency range from 100 MHz to 10 GHz (100 MHz – 2 GHz: IA; 2 – 10 GHz: VNA). The μ' of NAE and AE Co_2Z sintered samples are 1.9 and 1.7 at 100 MHz, respectively. The f_{FMR} of AE Co_2Z sample appears at 9.6 GHz, which is higher than

Fig. 4. Magnetic hysteresis loops of the non-acid-etched (NAE) and acid-etched (AE) $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ (Co_2Z) sintered samples.

Fig. 5. Frequency dependence of the measured complex permeability of the non-acid-etched (NAE) and acid-etched (AE) $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ (Co_2Z) sintered samples: measured with the (a) impedance/material analyzer and (b) vector network analyzer.

Fig. 6. Frequency dependence of the measured magnetic loss tangent of the non-acid-etched (NAE) and acid-etched (AE) $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ (Co_2Z) sintered samples.

6.2 GHz of NAE Co_2Z hexaferrite as indicated in Fig. 5(b). This is because the f_{FMR} is proportional to H_k according to Eq. (1). The μ' of AE Co_2Z hexaferrite is lower than that of NAE Co_2Z hexaferrite. This is because the increase in H_k of AE Co_2Z hexaferrite is higher than the increase in M_s , thereby, μ' is decreased by acid-etching according to $\mu' = 1 + (2M_s/3H_k)$ [21,22].

Figure 6 shows the measured magnetic loss tangent ($\tan \delta_\mu = \mu''/\mu'$) of NAE and AE Co_2Z hexaferrites. The $\tan \delta_\mu$ significantly decreased after acid-etching. This can be attributed to an

Table 2

Real part of complex permeability and magnetic loss tangent of the non-acid-etched (NAE) and acid-etched (AE) $\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41}$ (Co_2Z) sintered samples.

Sample	μ'				$\tan \delta_\mu$			
	0.1	1	2	3 GHz	0.1	1	2	3 GHz
NAE Co_2Z	1.9	1.9	2.1	2.0	0.022	0.033	0.065	0.149
AE Co_2Z	1.7	1.7	1.8	1.7	0.006	0.012	0.037	0.083

Table 3

Permeability (μ'), magnetic loss tangent ($\tan \delta_\mu$), quality factor (Q -factor), and figure of merit (FOM) for previously reported low loss magnetic material at GHz.

Ref.	Material	μ'	$\tan \delta_\mu$	Q -factor	FOM	f (GHz)
[9]	$\text{Ba}_{1.5}\text{Sr}_{0.5}\text{Co}_2\text{Fe}_{12}\text{O}_{22}$	2.3	0.095	10.53	23	2
[10]	$\text{Zn}_5\text{Ni}_{75}\text{Fe}_{20}$ + polymer (composite)	3.24	0.063	15.87	51.8	1
[11]	$\text{Ba}_3\text{Co}_{1.4}\text{Cu}_{0.6}\text{Fe}_{24}\text{O}_{41}$ + polymer ER182 (composite)	1.36	0.025	40	54.4	1
		1.7	0.012	83.33	141.7	1
	This work $(\text{Ba}_3\text{Co}_2\text{Fe}_{24}\text{O}_{41})$	1.8	0.037	27.02	48.7	2

increase in the f_{FMR} . The μ' and $\tan \delta_\mu$ at different frequencies are given in Table 2. It is noted that the AE Co_2Z hexaferrite shows the lowest $\tan \delta_\mu$ above 1 GHz as compared to the reported low-loss GHz magnetic materials [9-11].

In evaluating a magnetic material as to its suitability for antenna applications, the figure of merit (FOM) is an important criterion. Therefore, the FOM should be taken into consideration. The FOM of NAE and AE Co_2Z are 60.6 and 141.7 at 1 GHz, and 32.3 and 48.7 at 2 GHz, respectively. These results show that the FOM is increased by 134 and 51 % at 1 and 2 GHz, respectively, by the acid-etching, although μ' was decreased by 0.2 and 0.3. Table 3 summarizes the dynamic magnetic properties of the previously reported low-loss materials and the AE Co_2Z . The FOM of the AE Co_2Z hexaferrite is 174 % and 160 % higher than that of the $\text{Zn}_5\text{Ni}_{75}\text{Fe}_{20}$ composite and $\text{Ba}_3\text{Co}_{1.4}\text{Cu}_{0.6}\text{Fe}_{24}\text{O}_{41}$ composite, respectively, at 1 GHz [10,11]. Moreover, the FOM of the $\text{Ba}_{1.5}\text{Sr}_{0.5}\text{Co}_2\text{Fe}_{12}\text{O}_{22}$ is 23 at 2 GHz, which is 53 % lower than the FOM of the AE

Co₂Z hexaferrite [9]. Therefore, the low loss AE Co₂Z hexaferrite is suitable for GHz antenna applications in the UHF band.

4. Conclusions

The synthesized Co₂Z hexaferrite powder was acid etched at 60 °C and wet-ball milled to obtain a low magnetic loss at GHz range. It was found that acid-etching is very effective in removal of Y-type hexaferrite phase from the Co₂Z hexaferrite sample, resulting in an increase in H_k and σ_s . Accordingly, a low $\tan \delta_\mu$ of 0.037 with relatively high μ' of 1.8 was achieved at 2 GHz by pushing the ferromagnetic resonant frequency to a frequency higher than 2 GHz. Therefore, AE Co₂Z hexaferrite can be a good candidate for GHz antenna applications.

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Figure captions

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