Optimization of $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ ceramics with promising magneto-dielectric properties for VHF antenna miniaturization

Zhuohao Xiao^{*,†,||}, Chuanhu Wang[‡], Lie Liu[§], Zhihong Yang[¶]
and Ling Bing Kong^{*,†,***}

^{*}*School of Materials Science and Engineering, Jingdezhen Ceramic Institute
Jingdezhen 333001, Jiangxi, P. R. China*

[†]*School of Materials Science and Engineering, Nanyang Technological University
50 Nanyang Avenue, Singapore 639798*

[‡]*Department of Material and Chemical Engineering
Bengbu University, Bengbu 233030, Anhui
P. R. China*

[§]*General Test Systems Inc., Shenzhen 518102, Guangdong, P. R. China*

[¶]*College of Materials Science and Technology
Nanjing University of Aeronautics and Astronautics
Nanjing 211100, P. R. China*

^{||}xiaozhuohao@126.com
^{***}elbkong@ntu.edu.sg

Received 6 December 2017; Revised 14 December 2017; Accepted 19 December 2017; Published 17 January 2018

Magnetic, dielectric and DC conductive properties of $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ (with $x = 0-0.20$ at an interval of 0.05) ferrite ceramics were studied, in order to develop magneto-dielectric materials with almost equal values of relative permeability and permittivity, for the miniaturization of HF (3–30 MHz) and VHF (30–90 MHz and 100–300 MHz) antennas. The ferrite ceramics were prepared by using the conventional two-step sintering process. The real part of relative permeability is increased almost linearly with increasing concentration of Zn, while that of relative permittivity keeps nearly unchanged. It is found that promising magneto-dielectric materials, with close values of real permeability and permittivity over 30–90 MHz (VHF), can be obtained for the samples at Zn concentrations between $x = 0.05$ and $x = 0.10$.

Keywords: Magneto-dielectric materials; complex permittivity; complex permeability; antenna miniaturization; ferrite ceramics.

1. Introduction

High frequency (HF, 2–30 MHz) and very high frequency (VHF, 30–90 MHz and 100–300 MHz) antennas usually have quite large sizes physically.^{1,2} It is thus always desired to realize miniaturization of HF and VHF antennas for more convenient practical applications. However, while physical dimensions of the antennas are decreased, their electrical performances should not be compromised.^{3,4} Antenna miniaturization can be made by using materials with high dielectric permittivity.^{1–4} One issue regarding the use of high permittivity materials would be presence of highly localized electric field near by, thus leading to significant reduction in antenna efficiency and narrowed band performances. Meanwhile, a large mismatch in impedance was generated, due to the application of the high permittivity materials, which tremendously damaged the matching requirement for antenna designs. In order to address these problems, it has been proposed to load magneto-dielectric materials that possess

same values of real permeability and permittivity. This is because magneto-dielectric materials can be used to reduce the physical size of an antenna by a factor of $n = \sqrt{\mu'_0\mu'\varepsilon'_0\varepsilon'} = n_0\sqrt{\mu'\varepsilon'}$ (where μ' and ε' are real parts of permeability and permittivity, and $n_0 = \sqrt{\mu'_0\varepsilon'_0}$ is the refraction index of free space). Furthermore, because the impedance of the magneto-dielectric materials, which is given by $Z = \sqrt{\mu'_0\mu'/\varepsilon'_0\varepsilon'} = \eta_0\sqrt{\mu'\varepsilon'} = \eta_0$ (with $\mu'_0 = \varepsilon'_0$), is very close to that of free space (η_0), the antenna performances will not be affected.^{1,2}

However, such materials have rarely been found in nature and much less information on this type of materials can be found in the literature.^{1,2} Among various types of materials, ferrites are potentially capable of achieving such behavior, since they have both magnetic and dielectric properties. In our previous study, we explored composites comprising of resin epoxy as holding matrix and ferrite powders as

^{||,***}Corresponding authors.

inclusions. The composites obtained with ferrite powders of specific compositions possessed almost equal values of permeability and permittivity of about 5, with magnetic loss tangent of ~ 0.02 and dielectric loss tangent of ~ 0.05 , over 3–30 MHz.^{5,6} In reality, it is very hard to decrease the dielectric loss tangent to be less than 10^{-2} , owing mainly to the fact that the polymer matrix generally has a relative high intrinsic dielectric loss tangent. Moreover, the low loss frequency range cannot be over 30 MHz. Therefore, bulk ceramic materials have been proposed, starting with magnesium ferrite and lithium ferrite. Although both ferrites are good candidates to achieve magneto-dielectric materials,^{7–9} they are only suitable for HF band. In this regard, it is still necessary to develop such kind of materials for high frequencies (VHF).

This paper presents preparation and characterization of $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ ceramics, in order to optimize their magnetic properties to obtain desired magneto-dielectric materials. Although Ni–Zn ferrite ceramics have been studied for other applications, no report has been found to explore their magneto-dielectric properties. In this work, it is found that close values of permeability and permittivity 30–90 MHz can be possibly achieved by $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ ceramics at Zn concentration between $x = 0.05$ and $x = 0.10$.

2. Experimental

The ferrite composition was $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ (with $x = 0-0.20$ at an interval of 0.05). Commercially available Fe_2O_3 (99% purity, Aldrich Chemical Company Inc., USA), NiO (99+% purity, Aldrich Chemical Company Inc., USA), ZnO (98% purity, Aldrich Chemical Company Inc., USA), Co_3O_4 (99+% purity, Aldrich Chemical Company Inc., USA) and MnO_2 (98% purity, Aldrich Chemical Company Inc., USA) powders, were used as starting materials. In addition, 3 wt.% Bi_2O_3 was used as sintering aid to ensure that all the samples could be fully densified at temperatures of $< 1200^\circ\text{C}$. The oxides with different compositions were thoroughly mixed by using a planetary ball mill without the presence of any additive. The mixed samples were then calcined at 1000°C for 2 h. The calcined samples were pulverized, compacted and finally sintered at 1150°C for 2 h.

Two types of samples, namely disk (diameter of ~ 10 mm and thickness of ~ 1.5 mm) and coaxial cylinder (outer diameter of ~ 20 mm, inner diameter of ~ 10 mm and thickness of ~ 2.5 mm), were prepared. Disk samples were used for measurement of DC resistivity and permittivity, while toroidal samples were used for measurement of permeability.

Phase compositions of the mixed, milled, calcined and sintered samples were examined by using a Philips PW 1729

type X-ray diffractometer (XRD) with $\text{Cu K}\alpha$ radiation at a step of 0.05° and a scan rate of $4^\circ/\text{min}$. Microstructure, grain size and grain morphology of the sintered samples were observed by using a JEOL JSM-6340F type field emission scanning electronic microscope (FESEM). Densities of the ferrite ceramics were calculated with the masses and dimensions of the samples.

The DC resistances of the disk samples were measured by using a multimeter, based on which resistivities were calculated with their dimensions. Complex permeability and permittivity of the ceramics were measured with the two types of samples by using an Agilent E4991A RF impedance/materials analyzer over 1 MHz–1 GHz. For dielectric permittivity measurement, the samples were uniformly coated with an air-drying silver paste to minimize the contact resistance.

3. Results

Figure 1 shows the XRD patterns of the $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ powders after calcination at 1000°C for 2 h. Single phase ferrite with spinel structure has been obtained for all compositions. Figure 2 shows SEM images of the $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ ceramics after sintering at 1150°C for 2 h. XRD patterns indicated that the phase compositions of the sintered ceramic samples were almost the same as those of the calcined powders, showing crystalline stability of the ferrites against processing temperature. Meanwhile, due to its small quantity as a sintering aid, Bi_2O_3 was not detected in the XRD patterns. The samples exhibited a similar microstructure, consisting of round grains with pores located only at grain boundaries. The grain sizes of the ceramics estimated from the SEM images are found to be 1–3 μm , having a slight increase with increasing concentration of Zn. Densities of all samples are higher than 95% of their theoretical densities. DC resistivities of the samples are listed in Table 1, which are comparable with the reported values for Ni–Zn ferrites.

It is found that the ferrite ceramics have similar complex relative permittivity curves although their compositions are

Fig. 1. XRD patterns of the $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ powders synthesized at 1000°C for 4 h.

Fig. 2. SEM images of the $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ ceramics sintered at 1150°C for 2 h (a) $x = 0$, (b) $x = 0.05$, (c) $x = 0.10$ and (d) $x = 0.20$.

different. Figure 3 shows representative complex relative permittivity curves of the ferrite ceramics ($x = 0.01$). Both the real part and imaginary part of relative permittivities keep almost constant over the frequency range studied. The dielectric loss tangent maintains at sufficiently low levels, which is essential for practical applications. The real parts of relative permittivity of the samples are also included in Table 1. The values are comparable with those reported in the literature.^{10–12}

Table 1. Physical and electrical properties of the $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ ceramics.

x	Density (g/cm^3)	DC ρ (Ω cm)	ϵ' at 30 MHz
0	4.81	5.3×10^8	7.74
0.05	4.76	6.7×10^8	7.39
0.10	4.78	6.6×10^8	7.56
0.15	4.75	6.8×10^8	7.54
0.20	4.71	5.9×10^8	7.55

Complex relative permeability spectra of the ferrite ceramics at various concentrations of Zn are shown in Fig. 4. The real part of relative permeability (30 MHz) as a function of concentration of Zn is plotted in Fig. 5. The real permeability μ' of the ceramics increases almost linearly with

Fig. 3. Representative complex permittivity curves of the $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ ceramics ($x = 0.01$).

Fig. 4. Complex permeability curves of the $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ ceramics with different contents of Zn.

Fig. 5. Real part of relative permeability (at 10 MHz) of the $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ ceramics as a function of Zn content.

increasing concentration of Zn. The complex permeability spectra of the ceramics exhibit a common feature. The real permeability μ' remains almost constant up to a certain frequency, beyond which μ' begins to decrease. The resonance has been attributed to spin rotation in the ferrites.

Magneto-dielectric properties of the samples with $x = 0.05$ and $x = 0.10$ are shown in Figs. 6–8, over 30–90 MHz (VHF). The impedance of the sample at $x = 0.05$ is ~ 0.95 ,

Fig. 6. Impedance curves of the $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ ceramics with $x = 0.05$ and $x = 0.10$ over 30–90 MHz.

Fig. 7. Magnetic loss tangents of the $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ ceramics with $x = 0.05$ and $x = 0.10$ over 30–90 MHz.

Fig. 8. Dielectric loss tangents of the $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ ceramics with $x = 0.05$ and $x = 0.10$ over 30–90 MHz.

while that of the sample at $x = 0.10$ is ~ 1.07 – 1.08 , both of which are close to the impedance of free space. The magnetic loss tangent of both samples is less than 0.015 over the frequency range. The small resonances observed on the magnetic loss tangent curves are derived from the complex relative permeability curves, which is seemingly caused by the attachment of instrument used because it appears for all measurements. Therefore, it is not an intrinsic property of the ferrite materials. The dielectric loss tangent of the samples is relatively low. Especially above 50 MHz, the loss tangent is below 10^{-2} , which is satisfactory for practical applications.

4. Discussion

All samples are nearly fully densified mainly at the sintering temperature of 1150°C. The use of this relatively low sintering temperature in the present study effectively avoided the formation of Fe^{2+} as mentioned above. The presence of Fe^{2+} can badly worsen the electrical and dielectric properties of ferrite ceramics, due to the conduction caused by the electron hopping between Fe^{2+} and Fe^{3+} . This is because the transfer of electrons from Fe^{2+} ion to Fe^{3+} ion occurs within the octahedral sites, without changing the energy state of the crystal related to the transition. It was reported that the

presence of 0.3% Fe^{2+} content in a ferrite ceramic can reduce the DC resistivity by a factor of more than two orders of magnitude. In this respect, to obtain fully dense ferrite ceramics with sufficiently high resistivity, sintering aids are usually used to lower the sintering temperature.^{13–16}

The presence of Fe^{2+} in ferrite materials always contributes to high permittivity because Fe^{2+} has a larger polarization than Fe^{3+} . Fe^{3+} ion has a stable d -shell configuration with a spherical symmetry of the charge cloud, due to its five d -electrons distributed according to the Hund's rule, whereas Fe^{2+} ion has an extra electron as compared to Fe^{3+} , which disturbs the symmetry of charge electron cloud.¹⁷ As a result, the presence of Fe^{2+} increases the polarization in ferrites and thus ferrites containing a larger number of Fe^{2+} ions are likely to exhibit a higher permittivity. However, the high permittivity of ferrites due to the formation of Fe^{2+} is undesired because such a high permittivity is inevitably accompanied by a poor DC resistive property and hence high dielectric loss tangent as discussed below.

High frequency permittivity of ferrite crystals is mainly contributed by the atomic and electronic polarization in the ceramic grains. The permittivity of polycrystalline ferrite ceramics can be readily interpreted by the Maxwell–Wagner effect,¹⁸ where ferrite ceramics are considered to be comprising of conductive grains separated by layers (grain boundaries) of lower conductivity. Therefore, the permittivity values of ferrite ceramics are affected by a number of factors, such as microstructure, grain size, density and impurities. SEM examination and density measurement results showed that the ferrite ceramics have a similar microstructure. On the other hand, the components of the Ni–Zn ferrite ceramics are from the same transitional metallic group. In this regard, it is understandable that the samples used in the present study have close values of relative permittivity.

Dielectric loss tangent of polycrystalline ferrite ceramics results from the lag in polarization versus the alternating electric field, which is closely related to various factors, including the presence and type of impurities, imperfection in the ferrite structure and microstructures (porosity).¹⁹ However, the most significant contribution to dielectric loss tangent comes from the conduction loss, due to the electron hopping between Fe^{2+} and Fe^{3+} ions, especially at low frequencies. In this respect, the increase in permittivity due to the presence of Fe^{2+} ions is generally undesired, because it is always accompanied by an extremely high dielectric loss tangent. Usually, Fe^{2+} was formed in ferrite ceramics, as the sintering temperature is over 1200°C. Because of the use of Bi_2O_3 as sintering aid, the Ni–Zn ceramics could be well sintered at 1150°C. As a consequence, the formation of Fe^{2+} was effectively avoided. Therefore, dielectric loss tangent of the ferrite ceramics in this study was maintained to be sufficiently low, as required for the design of antennas.

Permeability of ferrite materials is determined by the saturation magnetization and coercivity, via the relation

$\mu \propto M_s/H_C$. Spinel ferrite has a general formula $\text{M}^{2+}\text{O}\cdot\text{Fe}_2^{3+}\text{O}_3$, where M^{2+} is a divalent metallic ion. NiFe_2O_4 and ZnFe_2O_4 are typical *reverse* and *normal* spinels, respectively. Therefore, $\text{Ni}_{1-x}\text{Zn}_x\text{Fe}_2\text{O}_4$ can be expressed as $(\text{Zn}_x\text{Fe}_{1-x})[\text{Ni}_{1-x}\text{Fe}_{1+x}]\text{O}_4$, where () and [] represent the a (tetrahedral) and b (octahedral) sites, respectively. Since the a site is spin-up and b site is spin-down, the net spin polarization of $\text{Ni}_{1-x}\text{Zn}_x\text{Fe}_2\text{O}_4$ will be increased due to the substitution of nonmagnetic ion Zn^{2+} for magnetic ion Ni^{2+} . In this case, it seems that the magnetization should be increased with increasing content of Zn. However, the magnetization is not always increased with the Zn concentration. This is because more Zn ions at the a site will weaken the exchange interaction between the a and b sites. As a result, an anti-parallel coupling of magnetic ions at the a and b sites cannot be sustained, thus leading to a canted structure at the b sites. Therefore, the magnetization falls at a certain value of Zn concentration. This value is in between those of the samples with $x = 0.4$ and $x = 0.5$,²⁰ which is much higher than the levels in our present study. It is also known that the introduction of Zn in NiFe_2O_4 reduces the magnetocrystalline anisotropy, which results in a reduction in coercivity, thus leading to high permeability. On the other hand, due to the decreased magnetocrystalline anisotropy, the natural resonance frequency decreases with Zn concentration.²⁰ Therefore, it is expected that magnetic permeability of the $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ ceramics should be qualitatively increased with increasing concentration of Zn, as clearly demonstrated in Figs. 4 and 5.

As mentioned earlier, it is desired to have magneto-dielectric materials for the design of low frequency antennas, especially for HF (2–30 MHz) and VHF (30–90 MHz and 100–300 MHz) bands, because these conventional antennas have rather large physical sizes. Our results show that $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ ferrites with compositions between $x = 0.05$ and $x = 0.10$ have $\epsilon' \approx \mu'$ over 30–90 MHz. Ni–Zn ferrite ceramics have been extensively studied for various applications. However, no report has been found for their magneto-dielectric properties in the open literature. Therefore, this study has explored a new application for Ni–Zn ferrite ceramics.

5. Conclusions

Spinel ferrite, $\text{Ni}_{0.95-x}\text{Zn}_x\text{Co}_{0.05}\text{Fe}_{1.90}\text{Mn}_{0.02}\text{O}_4$ (with $x = 0-0.2$), were synthesized and characterized, in order to optimize magnetic properties, so that it is possible to develop magneto-dielectric materials with almost equal values of permeability and permittivity for the miniaturization of HF and VHF antennas. It is concluded that such properties can be achieved for the compositions between $x = 0.05$ and $x = 0.10$. This result expanded the application area of Ni–Zn ferrite ceramics.

Acknowledgment

This work was supported by the Natural Science Fund of China (51762023), the Jiangxi Association for Science and Technology, the Jiangxi Provincial Department of Education, and the Training Program of Outstanding Young Scientists in Jiangxi Province (20171BCB23070).

References

- ¹A. Saini, A. Thakur and P. Thakur, Miniaturization and bandwidth enhancement of a microstrip patch antenna using magneto-dielectric materials for proximity fuze application, *J. Electron. Mater.* **46**(3), 1902 (2017).
- ²K. Buell, H. Mosallaei and K. Sarabandi, A substrate for small patch antennas providing tunable miniaturization factors, *IEEE Trans. Microw. Theory Tech.* **54**, 135 (2006).
- ³F. Ferrero, A. Chevalier, J. M. Ribero, R. Staraj, J. L. Mattei and Y. Queffelec, A new magneto-dielectric material loaded, tunable UHF antenna for handheld devices, *IEEE Antennas Wirele. Propag. Lett.* **10**, 951 (2011).
- ⁴J. Y. Lee, J. Y. Lee, K. H. Min and Y. H. Cheon, Miniaturized antennas with reduced hand effects in mobile phones using magneto-dielectric material, *IEEE Antennas Wirele. Propag. Lett.* **13**, 935 (2014).
- ⁵H. Su, X. L. Tang, H. W. Zhang, Y. L. Jing and F. M. Bai, Low-loss magneto-dielectric materials: Approaches and developments, *J. Electron. Mater.* **43**(2), 299 (2014).
- ⁶L. B. Kong, Z. W. Li, G. Q. Lin and Y. B. Gan, Ni-Zn ferrite composites with almost equal values of permeability and permittivity for low frequency antenna design, *IEEE Trans. Magn.* **43**(1), 6 (2007).
- ⁷L. B. Kong, Z. W. Li, G. Q. Lin and Y. B. Gan, Magneto-dielectric properties of Mg-Cu-Co ferrite ceramics: II. Electrical, dielectric and magnetic properties, *J. Am. Ceram. Soc.* **90**(7), 2104 (2007).
- ⁸L. B. Kong, Z. W. Li, G. Q. Lin and Y. B. Gan, Electrical and magnetic properties of magnesium ferrite ceramics doped with Bi₂O₃, *Acta Mater.* **55**(19), 6561 (2007).
- ⁹L. B. Kong, M. L. S. Teo, Z. W. Li, G. Q. Lin and Y. B. Gan, Development of magneto-dielectric materials based on Li-ferrite ceramics: III. Complex relative permeability and magneto-dielectric properties, *J. Alloys Compd.* **459**(1–2), 576 (2008).
- ¹⁰P. A. Miles, W. B. Westphal and A. von Hippel, Dielectric spectroscopy of ferromagnetic semiconductors, *Rev. Mod. Phys.* **29**(3), 279 (1957).
- ¹¹S. M. Hoque, Md. A. Choudhury and Md. F. Islam, Characterization of Ni-Cu mixed spinel ferrite, *J. Magn. Magn. Mater.* **251**, 292 (2002).
- ¹²K. O. Low and F. R. Sale, Electromagnetic properties of gel-derived NiCuZn ferrites, *J. Magn. Magn. Mater.* **246**, 30 (2002).
- ¹³J. J. Shrotri, S. D. Kulkarni, C. E. Deshpande, A. Mitra, S. R. Sainkar, P. S. Anil Kumar and S. K. Date, Effect of Cu substitution on the magnetic and electrical properties of Ni-Zn ferrite synthesized by soft chemical method, *Mater. Chem. Phys.* **59**, 1 (1999).
- ¹⁴A. Barba, C. Clausell, C. Feliú and M. Monzó, Sintering of (Cu_{0.25}Ni_{0.25}Zn_{0.50})Fe₂O₄ ferrite, *J. Am. Ceram. Soc.* **87**(4), 571 (2004).
- ¹⁵J. Y. Hsu, W. S. Ko and C. J. Chen, The effect of V₂O₅ on the sintering of NiCuZn ferrite, *IEEE Trans. Magn.* **31**(6), 3994 (1995).
- ¹⁶M. Drofenik, A. Znidarsic and D. Makovec, Influence of the addition of Bi₂O₃ on the grain growth and magnetic permeability of MnZn ferrites, *J. Am. Ceram. Soc.* **81**(11), 2841 (1998).
- ¹⁷M. T. Johnson and E. G. Visser, A coherent model for the complex permeability in polycrystalline ferrites, *IEEE Trans. Magn.* **26**(5), 1987 (1990).
- ¹⁸M. George, S. S. Nair, K. A. Malini, P. A. Joy and M. R. Anantharaman, Finite size effects on the electrical properties of sol-gel synthesized CoFe₂O₄ powders: Deviation from Maxwell-Wagner theory and evidence of surface polarization effects, *J. Phys. D, Appl. Phys.* **40**, 1593 (2007).
- ¹⁹J. I. Powar, S. R. Sawant, S. A. Patil, R. N. Patil and R. N. Karekar, Abrupt behaviour of dielectric constant and loss tangent of MgMn mixed ferrites, *Mater. Res. Bull.* **17**(3), 339 (1982).
- ²⁰J. L. Mattei, E. Le Guen, A. Chevalier and A. C. Tarot, Experimental determination of magnetocrystalline anisotropy constants and saturation magnetostriction constants of NiZn and NiZnCo ferrites intended to be used for antennas miniaturization, *J. Magn. Magn. Mater.* **374**, 762 (2015).