Synthesis, Microstructure and Electromagnetic Performance of Ni$_x$Zn$_{1-x}$Fe$_2$O$_4$ Ferrites with Different Ni/Zn Ratios Prepared by a Novel Molten Salt Method

Pao Yang$^{1,2}$, Hongbin Qi$^1$, Zhijian Peng$^{1,*}$, Xiuli Fu$^{2,*}$

$^1$School of Science, China University of Geosciences, Beijing 100083, P.R. China

$^2$School of Science, Beijing University of Posts and Telecommunications, Beijing 100876, P.R.China

pengzhijian@cugb.edu.cn (Z.J. Peng); xiulifu@bupt.edu.cn

Abstract. Ni$_x$Zn$_{1-x}$Fe$_2$O$_4$ ferrites ($x=0.0$~$1.0$ with a step of $0.1$) were prepared by a novel molten salt method. The effects of Ni/Zn ratio on the microstructure and electromagnetic performance of the acquired ferrites were investigated. All the samples are composed of a pure ferrite with cubic spinel structure, and their microstructures are quite homogeneous. With the increase of Ni content, the lattice constant of the samples decreases, the grain size fluctuates in a very narrow range around $2.0 \mu m$, and the saturation magnetization firstly increases and then decreases, presenting the maximum value with $x=0.6$, while the coercivity basically displays an inverse trend to the saturation magnetization. In the frequency range from $1$ M to $1$ GHz, the initial permeability of the acquired NixZn$_{1-x}$Fe$_2$O$_4$ ferrites decreases, the cut-off frequency increases, and the magnetic loss decreases with increasing Ni content.

1. Introduction

Soft magnetic ferrites are a group of non-metallic magnetic materials with vast variety of applications in modern society, including digital communications, household appliances, computers and many other fields [1]. They are characteristic of high electrical resistivity (much higher than those of metal soft magnetic materials), and low coercivity [2,3]. Therefore, the eddy current loss of soft ferrites as magnetic cores is impressively low, which is very beneficial for their high frequency performances.

Currently, typical soft magnetic ferrite products are made from Mn-Zn, Ni-Zn, Mg-Zn, Li-Zn ferrite materials and the similar. Specifically, with the development of modern communications technology, ferrite devices are required to be used in higher frequency [4], for which Ni-Zn ferrites stand out in various ferrite products because of their unique high frequency (10-500MHz) characteristics [5]. Therefore, to explore the factors influencing the high frequency performance of Ni-Zn ferrites is a hot topic in this field.

As is well-known, Ni-Zn ferrites have a spinel configuration based on a face-centered cubic lattice of the oxygen ions. In such structure, there are two kinds of interstitial positions for metal ions, in which one is the tetrahedral interstitial consisting of four oxygen atoms (A sites), and the other is octahedral interstitial of six oxygen atoms (B sites)[6]. Accordingly, the metallic ions in the tetrahedron and octahedron of the spinel structure form two sets of sublattices with opposite magnetic moments. The total magnetic moment of the ferrite is equal to the magnetism difference between A and B sites, and the coercivity ($H_C$) and Curie temperature ($T_C$) of ferrites also depends on the
interaction between the metallic ions at the two groups of sites. Thus, through adjusting the composition of the spinel ferrites and their occupancy relationship at A and B sites, magnetic materials with different magnetic properties can be obtained [7], which can further manipulate their high frequency performance. For instance, zinc ferrite exhibits normal spinel structure, which behaves like an antiferromagnet below the Neel temperature of 10 K and as a paramagnet at room temperature [8]. The saturation magnetization ($M_s$) of pure multidomain bulk Ni ferrite can reach 55 emu/g and that of the nanocrystalline Ni ferrite is only 25.4 emu/g at 300 K [9,10]. Liu et al. [11] indicated that the nanocrystalline Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ calcined at 1050 ºC could present a high $M_s$ of 75.6 emu/g. Hossain et al. [12] prepared Ni$_{1-x}$Zn$_x$Fe$_2$O$_4$ (x=0.2, 0.4) samples, revealing that the ferrite with a higher zinc content (Ni$_{0.6}$Zn$_{0.4}$Fe$_2$O$_4$) had a lower activation energy and $T_C$, but higher $M_s$, electrical conductivity and initial permeability. And Kumar et al. [13] reported that the $M_s$ of Ni$_{0.8}$Zn$_{0.2}$Fe$_2$O$_4$ could reach 96% of the theoretical value when the samples were calcined at 1200 ºC.

Moreover, the properties of Ni-Zn ferrites could be greatly influenced by the preparation methods. The maximum $M_s$ of Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ ferrites prepared by traditional solid-state sintering could reach 78.44 emu/g [14]. The sol-gel method has advantages of easily controlling the stoichiometry of the products and producing ultrafine particles with a narrow size distribution in a relatively short processing time at very low temperature [15]. Zhang et al. [16] obtained Ni-Zn ferrites with different compositions by sol-gel auto-combustion, indicating that the Zn$_{0.3}$Ni$_{0.7}$Fe$_2$O$_4$ ferrites annealed at 900 ºC would present the maximum $M_s$ of 83.2 emu/g. And Jiang et al. [17] reported the synthesis and mechanism of Ni-Zn ferrite powders by a molten salt method. The reported molten salt method could make the sintering of the ferrite ceramics more easily and the grain size of the products smaller. However, to the best of our knowledge, there is no report on the effects of Ni/Zn ratio on the microstructure and electromagnetic performance of ferrites prepared by molten salt methods.

Therefore, in this work, Ni$_x$Zn$_{1-x}$Fe$_2$O$_4$ (x = 0.0–1.0 with a step of 0.1) powders were first synthesized by a novel molten salt method at 500 ºC via utilizing analytical-grade metal oxides powders as the raw materials. Then, the samples were sintered into ceramics at 1200 ºC. Finally, the microstructure and electromagnetic performance of the obtained Ni$_x$Zn$_{1-x}$Fe$_2$O$_4$ ferrites with different Ni/Zn ratios were systematically investigated.

2. Experimental procedures

2.1. Samples preparation

Ni$_{0.5}$Zn$_{0.5}$Fe$_2$O$_4$ (x=0.0–1.0 with a step of 0.1) ferrites were prepared by a molten salt method from analytically pure iron (III) nitrate nonahydrate, nickel (II) nitrate hexahydrate, zinc (II) nitrate hexahydrate, and 99.0% KOH.

During typical processing, the stoichiometric metal nitrates were first weighted and dissolved in ethanol. The molar ratio of KOH to the nitrates was 8:1. Then the resultant mixtures were transferred into a planetary ball mill with high-resistance TZP ZrO$_2$ balls as the grinding media. For the milling, the mass ratio of the raw material, ethanol and grinding media was 1:5:4, the volume of the mill was 1L, the rotational speed was 250 rpm and the milling time was 6 h. After milling, the resultant slurries were dried in an open oven at 100ºC for 2 h. After drying, the resultant powder chunks were ground into powder with an agate mortar. And the obtained powders were then calcined at 500ºC in a muffle furnace for 3 h. The calcined powders were milled for 2 times with the same ball milling method mentioned above. After the final milling, the resultant slurries were cleaned with deionized water by centrifuging for 4 times. The collected precipitates were then dried in the oven at 100ºC for 3 h. After drying, the powder chunks were ground with 0.5 wt.% PVA as binder, and thieved into fine powders. The finally collected powders were pressed into pellets (6 mm in diameter and 1 mm in thickness, or 12 mm in diameter and 1 mm in thickness) and toroidal rings (20 mm in outer diameter, 10 mm in inner diameter and 3 mm in thickness). Finally, all the samples were sintered at 1200ºC in a muffle furnace for 4 h and then cooled down naturally to room temperature.
2.2. Materials characterization
The phase of the samples were detected by X-ray diffraction (XRD, D/max-RB, Japan, Cu Kα, λ=1.5418 Å) in a continuous scanning mode at a speed of 6°/min in the 2θ range of 20°–80°. The collected patterns were then analyzed by MDI Jade 5.0 software. The microstructure was examined on the polished surface of the samples by scanning electron microscopy (SEM, SU8020). With the obtained images, the grain size was then evaluated by Nano Measurer1.2 software. The apparent density (ρ) was measured by Archimedes method in accordance with the international standard ISO18754, which could be calculated by,

\[ \rho = \frac{\rho_w m_1}{m_1 - m_2} \]  

where \( m_1 \) and \( m_2 \) are the mass of the samples in air and in water with a unit of gram (g), respectively; \( \rho_w \) is the density of water (g/cm³) at the temperature for measurement. The relative density of the specimens could then be defined by,

\[ \text{Relative density} = \frac{\text{Apparent density}}{\text{Theoretical density}} \times 100\% \]  

in which the theoretical density was calculated by

\[ \rho = \frac{8M}{Na^3} \]  

where \( M \) is the molecular weight (g) of the ferrite, \( a \) is its lattice constant (Å) and \( N \) is Avogadro’s number (per mol).

The magnetic performance tested in this work includes some static and dynamic magnetic properties. The static magnetic properties of the samples were measured by Lake Shore 7307 vibration sample magnetometer system (VSM), in which the hysteresis loop of the ferrites was recorded under a maximum magnetic field of 10 kOe. From the recorded hysteresis loop, the \( M_s \) and \( H_c \) values of the samples could be calculated. The dynamic magnetic properties of the samples including initial permeability and magnetic loss were measured by Agilent E4294A impedance analyzer under a frequency ranged from 1 MHz to 1000 MHz. All the above tests were carried out at room temperature.

3. Results and discussion
3.1. Compositional and structural properties
Figure 1 shows the XRD results of the obtained samples. The full-scan patterns (Figure 1a) reveal that all the samples with different Ni/Zn ratios present the same cubic spinel structure (JCPDS no.46-0115) for typical Ni-Zn type ferrite phase without other impurities, indicating that pure ferrites have been successfully prepared. According to Bragg equation, on the basis of the 2θ value of the (113) diffraction peak around 35.2°, the lattice constants of the obtained ferrites were calculated, and the results are listed in Table 1. To make it more clearly, an elaborated observation on the diffraction peak around 35.2° is also presented in Figure 1b. It is seen that with the increase of Ni contents, the (113) diffraction peak shifts to the direction of high 2θ value, which means that the lattice constant of the ferrites decreases. The reason for such phenomena is that the radius of Ni²⁺ ion (0.78 Å) is smaller than that of the replaced Zn²⁺ one (0.82 Å) in the designed ferrites of this work [18].

Typical SEM images on the polished surface of the obtained ferrites with different Ni/Zn ratios are displayed in Figure 2. It can be seen that with the powders prepared by the proposed molten salt method, all the sintered ferrites would present a very homogeneous microstructure with high densification, and the grain size fluctuates in a very narrow range around 2.0 μm (also see Table 1). This result reveals that the molten salt method can improve the sinterability of the prepared powder, due to the enhanced fluidity and diffusivity of the particles during sintering reaction [19]. Moreover,
because the applied molten salt could penetrate between the resultant powders and prevents the interconnection of the powder particles, there would be less agglomeration in the powders prepared by the molten salt method. Therefore, the proposed molten salt method is an effective method to obtain high-density Ni-Zn type ferrites. However, with the increase of Zn content, the lattice constant of the ferrites gradually increases, the lattice expands, the powder activity increases, and the grain growth may be accelerated, which is generally conducive to obtaining larger grains [20].

![Figure 1. XRD patterns of the prepared Ni$_x$Zn$_{1-x}$Fe$_2$O$_4$ (x=0.0-1.0 with a step of 0.1) calcined at 1200°C: (a) full patterns, and (b) an elaborated observation on the diffraction peak around 35.2°.](image)

| $x$ | Apparent density (g/cm$^3$) | Relative density (%) | Lattice constant a = b = c (Å) | Grain size(μm) |
|-----|----------------------------|----------------------|--------------------------------|----------------|
| 0.0 | 5.049                      | 93.35                | 8.449                          | 1.94           |
| 0.1 | 5.244                      | 96.99                | 8.425                          | 2.07           |
| 0.2 | 5.249                      | 97.37                | 8.424                          | 1.80           |
| 0.3 | 5.227                      | 97.22                | 8.419                          | 1.88           |
| 0.4 | 5.220                      | 97.36                | 8.398                          | 2.24           |
| 0.5 | 5.259                      | 98.37                | 8.397                          | 2.14           |
| 0.6 | 5.147                      | 98.44                | 8.388                          | 1.93           |
| 0.7 | 5.247                      | 98.70                | 8.376                          | 1.75           |
| 0.8 | 5.198                      | 99.00                | 8.372                          | 1.76           |
| 0.9 | 5.092                      | 95.38                | 8.356                          | 1.66           |
| 1.0 | 5.143                      | 95.78                | 8.345                          | 2.12           |
Table 1 displays the apparent and relative densities of the acquired specimens. It can be seen that both the apparent density and relative density present the trend of first rising and then decreasing. At x=0.5, the apparent density reaches the maximum of 5.259 g/cm$^3$, and the relative density reaches the maximum of 99.00% at x=0.8. This trend can be attributed to the fact that the increase of Ni content in a small range in the present ferrites will facilitate the movement of grain boundaries, thus promoting the densification of the samples during sintering [21]. However, when the Ni content is too high, the densification of the samples would be hindered due to the lowered activity of the powders [20]. Resultantly, the porosity in the samples increases, and the apparent and relative densities of the samples decrease.
3.2. Magnetic performance

Figure 3 illustrates the recorded hysteresis loops for the as-obtained ferrites. The calculated basic magnetic parameters ($M_s$ and $H_c$) are illustrated in Figure 4 as a function of the Ni content. It can be seen from Figure 4 that, the $M_s$ of the obtained ferrites increases first and then decreases with the increase of Ni content. It is well known that, the $M_s$ of a ferrite can be described by the following equation[22],

$$M_s = |M_A - M_B|$$

in which $M_A$ and $M_B$ denote the magnetic moments of A and B sites, respectively. The present Ni-Zn type ferrites are of pure cubic spinel structure. In this structure, the A sites are occupied by Zn$^{2+}$ and Fe$^{3+}$ ions, while the B sites are occupied by Ni$^{2+}$ and Fe$^{3+}$ ions. The magnetic moment of Zn$^{2+}$ and Ni$^{2+}$ ions is 0 and 2.0 $\mu_B$, respectively, while that of Fe$^{3+}$ ions is 5.0 $\mu_B$. And the net magnetic moment of the ferrites is the sum of different magnetic moments in tetrahedron A and octahedron B sites. In this study, the pure zinc ferrite is of a normal spinel structure. The non-magnetic Zn$^{2+}$ ions occupy the A sites while all the magnetic Fe$^{3+}$ ions are evenly distributed in the A and B sites. Therefore, it does not show any magnetism. After Zn$^{2+}$ ions are replaced by Ni$^{2+}$ ones, all the magnetic Ni$^{2+}$ ions will occupy the B sites, while little Fe$^{3+}$ ions might migrate into A sites. Resultantly, the obtained ferrites will present certain magnetism due to the higher magnetic moment of B site than that of A site. In an appropriate range, with increasing Ni content, the magnetism of the ferrites will increase. For example, when $x$=0.2 and below, the magnetism of the samples are very weak. That is why the data of the samples with $x$=0.2 and below are not shown in the figure. When Ni$^{2+}$ ions further substitute for Zn$^{2+}$ ions ($0.2 < x \leq 0.5$), the increased amount of magnetic Ni$^{2+}$ ions will further increase the super-exchange between A and B sites, which will ultimately lead to the increase of $M_s$, although the simultaneously appeared migration of more Fe$^{3+}$ ions into A sites will result in an inverse effect. However, with further increase of Ni content ($x > 0.5$), more Fe$^{3+}$ ions will migrate into the A sites from the B sites, because the substituted Ni$^{2+}$ ions have a stronger preference to inhabit B sites than Fe$^{3+}$ ions. As a result, while the magnetic moment of B sites increases, the magnetic moment of A sites also increases. Finally, the $M_s$ value of the ferrites ($x > 0.5$) decreases.

Figure 3. Magnetic hysteresis loops of the prepared Ni$_x$Zn$_{1-x}$Fe$_2$O$_4$ ($x=0.0–1.0$ with a step of 0.1).
Figure 4. Saturation magnetization and coercivity of the prepared 
Ni$_x$Zn$_{1-x}$Fe$_2$O$_4$ ($x=0.0-1.0$ with a step of 0.1).

With the increase of Ni content, the $H_C$ of ferrites basically displays an inverse trend to the $M_s$ (also see Figure 4). The $H_C$ is the measure of the reverse magnetic field applied on a magnetic material to drive the magnetic flux return to zero, which represents the ability of magnetic materials to remain magnetized after magnetization [23]. The $H_C$ of a ferrite can be described by [24],

$$H_c = P_c \sqrt{AK_1}$$

(5)

where $P_c$ is a dimensionless factor, $A$ is the exchange constant, $K_1$ is magneto-crystalline anisotropy constant and $D$ is the grain size. It can be seen that the $H_C$ is inversely proportional to $M_s$ and $D$. In the present ferrites, when Ni ions enter into the ferrite lattice, the magneto-crystalline anisotropy constant of the sample increases, because Ni ferrite has larger magneto-crystalline anisotropy constant than zinc ferrite. Moreover, the magneto-crystalline anisotropy constant is also determined by the densification of the samples. A higher densification means less porosity in the sample, which will result in a lower magneto-crystalline anisotropy constant. In the present samples, with the increase of Ni content, the relative density of the samples first increases and then decrease; thus the corresponding magneto-crystalline anisotropy constant decreases initially and increases later. Consequently, the magneto-crystalline anisotropy constant totally varies in a very limited range. Meanwhile, the grain size of the ferrites fluctuates in a very narrow range around 2.0 $\mu$m. On the other hand, $M_s$ changes in a very large range from 0 to approximately 80 emu/g, which is thus the dominant factor for the change of $H_C$. Therefore, it is expected that the $H_C$ of the present ferrites basically displays an inverse trend to the $M_s$[25]. With the increase of Ni content, because the $M_s$ firstly increases and then decreases, so the $H_C$ initially decreases and then increases.

The relationship between permeability and frequency is displayed in Figure 5. The real part of permeability represents ($\mu'$) the ability to store energy (Figure 5a), while the imaginary part ($\mu''$) reflects the permeability loss ($\mu=\mu'+j\mu''$) (Figure 5b). In this study, the test frequency is in the range of 1-1000 MHz, but the data of the samples with $x$ from 0 to 0.2 fall into a straight line before 1MHz, which is not within the frequency range discussed. Therefore, they are not presented in this figure. From Figure 5a, the cut-off frequency of the obtained ferrites was calculated, and the results are illustrated in Figure 6 as a function of the Ni content. It can be seen that the initial permeability of the
obtained ferrites with increasing Ni contents increases first and then decrease later. It is well known that in the sintered polycrystalline ferrite samples, there are two magnetization processes: wall-shift and domain-shift [26], which can be described by,

$$\mu_j = 1 + X_{dom} + X_{spin}$$  \hspace{1cm} (6)

in which, $X_{dom}$ denotes the domain wall susceptibility and $X_{spin}$ is the inherent spin susceptibility. The values of the $X_{dom}$ and $X_{spin}$ can be calculated by the following equations,

$$X_{dom} = \frac{3\pi M_s^2 D}{4\gamma}$$ \hspace{1cm} (7)

$$X_{spin} = \frac{2\pi M_s^2}{K_1}$$ \hspace{1cm} (8)

Figure 5. Frequency dependence of the permeability for the prepared Ni$_x$Zn$_{1-x}$Fe$_2$O$_4$ ($x=0.0-1.0$ with a step of 0.1): (a) real part and (b) imaginary part.
in which, \( D \) is the mean grain size, \( \gamma \) is the domain wall energy, and \( K_1 \) is the magnetocrystalline anisotropy constant. It is seen that a higher \( M_s \) will result in a larger permeability. And generally speaking, for the samples with larger grains, the domain walls are easy to move during magnetization or demagnetization. These samples are expected to have a lower \( H_C \) and a higher permeability, which can also be achieved for the samples with low magnetocrystalline anisotropy constant. The initial permeability of the present Ni-Zn ferrites with \( x \) between 0.3 and 0.4 are much higher than that of other samples. With the increase of Ni content, the initial permeability decreases gradually.

When the \( \mu' \) drops to 1/2, the corresponding frequency is the cut-off frequency of the ferrite \( (f_r) \). The \( f_r \) denotes the upper limit for the use of a soft magnetic ferrite. It can be seen from Figure 6 that among all the obtained ferrites, the \( f_r \) of the samples with \( x=0.3 \) and 0.4 are lower, although their initial permeability is higher. When the content of Ni gradually increases, the \( f_r \) value of the obtained ferrites will increase. The height of \( f_r \) depends mainly on the relaxation and resonance of domain wall displacement and the natural resonance caused by the domain rotation. The relationship between \( f_r \) and material parameters can be described by Snock formula [27],

\[
(\mu_t - 1)f_r = \frac{1}{2\pi} \gamma \cdot M_S
\]

(9)

where \( \mu_t \) is the initial permeability of the ferrite, \( f_r \) is the cut-off frequency, and \( \gamma \) is a measure determined by the intrinsic properties of materials. The Snock formula gives a theoretical limit for the high frequency permeability of soft magnetic materials. Because ZnO has relatively low magnetocrystalline anisotropy \( K_1 \), the addition of ZnO will increase \( \mu_t \) and decrease \( f_r \) of the ferrites, which implies that the cut-off frequency of the ferrites increases with the increase of Ni content.

![Figure 6. Cut-off frequency of the Ni\(_x\)Zn\(_{1-x}\)Fe\(_2\)O\(_4\) (\( x = 0.0 \sim 1.0 \) with a step of 0.1).](image)

The frequency dependence of the magnetic loss tangent (tan\( \delta _m \)) for the obtained ferrites is shown in Figure 7. The tan\( \delta _m \) represents the attenuation ability of magnetic properties of the ferrites against incident electromagnetic waves. The ratio of \( \mu'' \) to \( \mu' \) is often used to define the tan\( \delta _m \). As expected, the tan\( \delta _m \) of the present ferrites with different Ni/Zn ratios increases with the increase of frequency. With the increase of Ni content, under the same frequency, the tan\( \delta _m \) of the ferrites gradually decreases. Soft
magnetic ferrites will produce a variety of losses under alternating field. According to their generation mechanism, it can be sorted into eddy current loss, hysteresis loss and residual loss [28]. The eddy current loss is the power loss caused by eddy current. The main influencing factors on it are the electrical resistivity and thickness of the materials. A ferrite with high resistivity is helpful to reduce its eddy current loss. In general, reducing the coercivity and increasing initial permeability are the main ways to reduce hysteresis loss. In this work, the coercivity of the samples with \( x = 0.1 - 0.6 \) is relatively low (\( \leq 20 \) Oe), which is an ideal material with low magnetic loss. Residual losses are all losses except the eddy current loss and hysteresis loss. They mainly involve in the relaxation loss caused by size resonance, domain wall resonance and natural resonance. The residual losses at room temperature are mainly caused by the electron diffusion between \( \text{Fe}^{3+} \) and \( \text{Fe}^{2+} \). The diffusion of other ions, such as \( \text{Ni}^{3+} \) and \( \text{Ni}^{2+} \), may occur at high temperature. Therefore, preventing the diffusion of electron vacancies and ions is the main method to avoid the residual losses. In the present samples, the content of Fe is fixed, so the goal of obtaining ferrites with a lower magnetic loss is to enhance their initial permeability and decrease their coercivity. From this point of view, the samples with \( x = 0.1 - 0.6 \) all meet the requirements.

4. Conclusions

\( \text{Ni}_x\text{Zn}_{1-x}\text{Fe}_2\text{O}_4 \) \( (x = 0.0 - 1.0 \text{ with a step of 0.1}) \) powders were successfully synthesized by a novel molten salt method at a low temperature of 500 °C, and \( \text{Ni}_x\text{Zn}_{1-x}\text{Fe}_2\text{O}_4 \) ceramic ferrites were prepared by sintering the samples from the corresponding powders at 1200 °C. The main conclusions are summarized as follows.

1. All the obtained ferrites are of pure cubic spinel structure. With the increase of Ni content, the lattice constant decreases, and the density first increases and then decreases. All the samples have a quite homogeneous microstructure. Their grain size fluctuates in a very narrow range around 2.0 \( \mu \)m.

2. With increasing content of Ni, the saturation magnetization firstly increases and then decreases, presenting the maximum value with \( x = 0.6 \), while the coercivity basically displays an inverse trend to the saturation magnetization; and in the frequency range from 1M to 1GHz, the initial permeability of the acquired \( \text{Ni}_x\text{Zn}_{1-x}\text{Fe}_2\text{O}_4 \) ferrites decreases, the cut-off frequency increases, and the magnetic loss decreases.

![Figure 7. Frequency dependence of the magnetic loss tangent (\( \tan\delta_m \)) for the \( \text{Ni}_x\text{Zn}_{1-x}\text{Fe}_2\text{O}_4 \) (\( x = 0.0 - 1.0 \text{ with a step of 0.1}) \).](image-url)
Acknowledgements
Many thanks for the financial support to this work by National Natural Science Foundation of China (grant nos. 11674035 and 61274015) and Fundamental Research Funds for the Central Universities.

References
[1] Sugimoto M 1999 *Journal of the American Ceramic Society* **82** 269
[2] Bhise B V, Dongare M B, Patil S A and Sawant S R 1991 *Journal of Materials Science Letters* **10** 922
[3] Shokrollahi H and Janghorban K 2007 *Journal of Materials Processing Technology* **189** 1
[4] Pardavii-Horvath M 2000 *Journal of Magnetism and Magnetic Materials* **215-216** 171
[5] Tsay C Y, Liu K S, Lin T F and Lin I N 2000 *Journal of Magnetism and Magnetic Materials* **209** 189
[6] Daniels J M and Rosenемwaig A 1970 *Canadian Journal of Physics* **48** 381
[7] Costa A C F M, Tortella E, Morelli M R and Kiminami R H G A 2003 *Journal of Magnetism and Magnetic Materials* **256** 174
[8] Hastings J M and Corliss L M 1956 *Physical Review* **102** 1460
[9] Smit J and Wijin H P J 1959 *Wiley* 54
[10] Kale A, Gubbala S and Misra R D K 2004 *Journal of Magnetism and Magnetic Materials* **277** 350
[11] Liu Y and Qiu T 2013 *Chinese Physics* **12** 3837
[12] Hossain A K M A, Mahmud S T, Seki M, Kawai T and Tabata H 2007 *Journal of Magnetism and Magnetic Materials* **312** 210
[13] Kumar P S A, Shroti J J, Kulkarni S D, Deshpande C E and Date S K 1996 *Materials Letters* **27** 293
[14] Chen D G, Tang X G, Tong J J, Wu J B, Jiang Y P and Liu Q X 2011 *Solid State Communications* **151** 1042
[15] Srivastava M, Chaubey S and Ojha A K 2009 *Materials Chemistry and Physics* **118** 174
[16] Zhang H E, Zhang B F, Wang G F, Dong X H and Gao Y 2007 *Journal of Magnetism and Magnetic Materials* **312** 126
[17] Jiang H T, Wang X F, Yu C L and Wang L 2010 *Materials and Manufacturing Processes* **25** 1489
[18] Dang W G, Zhang Z M, Lin M, Fan X L, Chai G Z, Xu Y and Xu D S 2009 *Journal of Physics D: Applied Physics* **42** 125006
[19] Guo Z P, Du G D, Nuli Y, Hassan M F, Liu H K 2009 *Journal of Materials Chemistry* **19** 3253
[20] Zhou J P, Lv L and Chen X C 2010 *Journal of Ceramic Processing Research* **11** 263
[21] Gumaan M S, Shalaby R M, Yousef M K M, Ali E A M and Abdel-Hady E E 2019 *Soldering and Surface Mount Technology* **28** 0954
[22] Munir A, Ahmed F, Saqib M and Anis-Ur-Rehman M 2016 *Journal of Magnetism and Magnetic Materials* **397** 188
[23] Fu X L, Ge H L, Xing Q K and Peng Z J 2011 *Materials Science and Engineering B* **176** 926
[24] Xue D S, Chai G Z, Li X L and Fan X L 2008 *Journal of Magnetism and Magnetic Materials* **320** 1541
[25] Suzuki K, Herzer G and Cadogan J M 1998 *Journal of Magnetism and Magnetic Materials* **177-181** 949
[26] Khan Z H, Rahman M M, Sikder S S, Hakim M A and Saha D K 2013 *Journal of Alloys and Compounds* **548** 208
[27] Nakamura T 2000 *Journal of Applied Physics* **88** 348
[28] Bellad S S and Chougule B K 2000 *Materials Chemistry and Physics* **66** 58