

Electromagnetic Interference Shielding Properties of $\text{SrFe}_{12-2x}(\text{Cu}_{0.5}\text{Co}_{0.5}\text{Ti})_x\text{O}_{19}$ Hexaferrite

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Abstract

Electromagnetic (EM) pollution has become one of the most severe forms of pollution for humans and electronic devices. In order to protect humans and electronic devices from electromagnetic interference (EMI), the material that could shield the EMI should be considered. Hexagonal ferrites are considered as promising candidates for EMI shielding materials. This work fabricated Cu, Co, and Ti co-doped $\text{SrFe}_{12}\text{O}_{19}$ ($\text{SrFe}_{12-2x}(\text{Cu}_{0.5}\text{Co}_{0.5}\text{Ti})_x\text{O}_{19}$) samples to investigate EMI shielding properties. Among the samples, the doped ones exhibited better EMI shielding performance than the undoped one, which could be judged through total shielding efficiency (SE_T). In addition, the samples exhibited the best shielding performance in the frequency range of 8–12 GHz (X-band), with the best value of $SE_T = 4.20$ dB for the $x = 0.20$ sample, which could block ~62% of the incident microwave. The $x = 0.05$ sample also exhibited similar values with $SE_T = 4.17$ dB. The enhancement of SE_T values for doped samples compared to the undoped one could be attributed to the increase in conductivity caused by doping. In addition, the enhancement of SE_T could also be attributed to the effects of the magnetic dipoles combined with the electric dipoles of ferrites.

Keywords- $\text{SrFe}_{12}\text{O}_{19}$, Transitional metal, Doping, EMI shielding, Shielding efficiency.

1. Introduction

Recently, humans have faced massive electromagnetic (EM) pollution due to the large-scale use of electronic communication devices. EM pollution is created from EM waves due to the charged particles oscillating (Marin et al., 2008; Park et al., 2009; Zeng & Xu, 2010). EM pollution can seriously affect nearby electronic devices and human health, which need a prime concern as shielding. This shielding could be called electromagnetic interference (EMI) shielding, which could be achieved through electrical conductive shielding and/or partially imparting both magnetic permeability and electrical conductivity (Jan et al., 2017; Saboor et al., 2016; Saboor et al., 2018; Saboor et al., 2019). Two mechanisms in EMI shielding, reflection and absorption, prevent incident waves and absorb waves, then generate heat energy

inside the shielding materials, respectively. The total amount of reflected and absorbed energy could be considered total shielding effectiveness. The first candidate for EMI shielding is metals or alloys (Sivaprakash et al., 2023), which still have severe drawbacks, such as corrosion, complicated processing, and high weight. Therefore, more potential candidates should be considered.

Ferrites, including spinel ferrite, garnet, and hexaferrite, could be promising candidates (Oh et al., 2023). Among ferrites, M-type strontium hexaferrite ($\text{SrFe}_{12}\text{O}_{19}$) has intriguing properties of large magnetization, large magneto-crystalline anisotropy, excellent high-frequency response, and high stability, which could be used in many practical applications such as magnetic recording media, permanent magnets, and microwave absorbers (Chitra et al., 2014; Harris et al., 2009; Luo et al., 2014). Doping on the $\text{SrFe}_{12}\text{O}_{19}$ host lattice would effectively improve their EMI shielding properties (Lu et al., 2024; Mohammed et al., 2019). This work would investigate the EMI properties of Cu, Co, and Ti co-doped on $\text{SrFe}_{12}\text{O}_{19}$ samples to determine which concentration would be the best among the dopings. The doping concentration was started at 0, with small increments of doping concentration to achieve the pure phase for as many samples as possible. The sintering temperature was selected as low as possible to achieve samples with small grain sizes.

2. Experimental Details

$\text{SrFe}_{12-2x}(\text{Cu}_{0.5}\text{Co}_{0.5}\text{Ti})_x\text{O}_{19}$ samples were fabricated from SrCO_3 , Fe_2O_3 , CuO , Co_3O_4 , and TiO_2 (purchased from Sigma-Aldrich with high purity) using a combination of ball milling and heat treatment methods. Six samples had concentrations of $x = 0, 0.05, 0.10, 0.15, 0.20$, and 0.25 . The fabrication process details were presented in our previous works (Tho et al., 2022; Xuan et al., 2024). The details of the physical properties of these samples were also investigated in our priority works (Tho et al., 2022; Xuan et al., 2024). In this work, the EMI shielding properties were investigated through S-parameters (S_{11} , S_{21} , S_{12} , and S_{22}), which were recorded from a vector network analyzer (VNA, Keysight Model PNA-X N5242A). Before measuring S-parameters, the samples were well mixed with paraffin with a volume ratio of 40:60. Then the mixtures were molded into toroidal shapes with inner and outer diameters of 3.04 and 7.00 mm and a thickness of 3.0 mm.

3. Results and Discussion

Figure 1 shows XRD patterns of $\text{SrFe}_{12-2x}(\text{Cu}_{0.5}\text{Co}_{0.5}\text{Ti})_x\text{O}_{19}$ samples with different doping concentrations. Structural properties of $x = 0$ – 0.15 samples have been detailed and discussed in our previous reports (Tho et al., 2022; Xuan et al., 2024). Only the $x = 0$ and $x = 0.05$ samples exhibited the pure phase of M-type hexaferrite with the space group of $P6_3/mmc$. The $x = 0.10$ and $x = 0.15$ samples showed impurity phases of SrFe_2O_4 (marked by *) and $\alpha\text{-Fe}_2\text{O}_3$ (denoted by #), besides the main phase of M-type hexaferrite. Consequently, the $x = 0.20$ and $x = 0.25$ samples also exhibited the purity phases of SrFe_2O_4 and $\alpha\text{-Fe}_2\text{O}_3$, where the percentage of impurity phases was higher than those of the $x = 0.10$ and 0.15 samples. We believe the samples with high doping concentrations required higher sintering temperatures than those with undoped/low doping concentrations. Thus, the impurity phases occurred in these samples. We also believe the pure phase could be achieved for the samples with high doping concentration at appropriate sintering temperatures. However, the samples were sintered at the same temperature to compare other properties easily.

Figure 2 shows SEM images of $\text{SrFe}_{12-2x}(\text{Cu}_{0.5}\text{Co}_{0.5}\text{Ti})_x\text{O}_{19}$ samples, where the morphological properties were significantly changed by the change in doping concentrations. The samples consisted of polygonal particles that were inhomogeneous in shape and size. With the increase in doping concentration, the particles agglomerated to form larger clusters, increasing the average grain size.

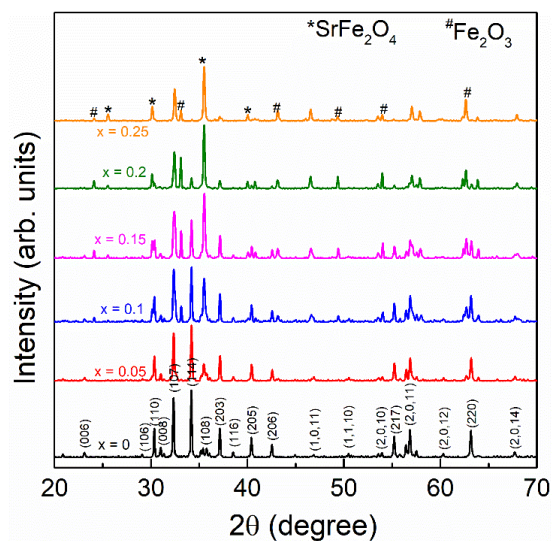


Figure 1. XRD patterns of $\text{SrFe}_{12-2x}(\text{Cu}_{0.5}\text{Co}_{0.5}\text{Ti})_x\text{O}_{19}$ hexaferrite samples (Tho et al., 2022).

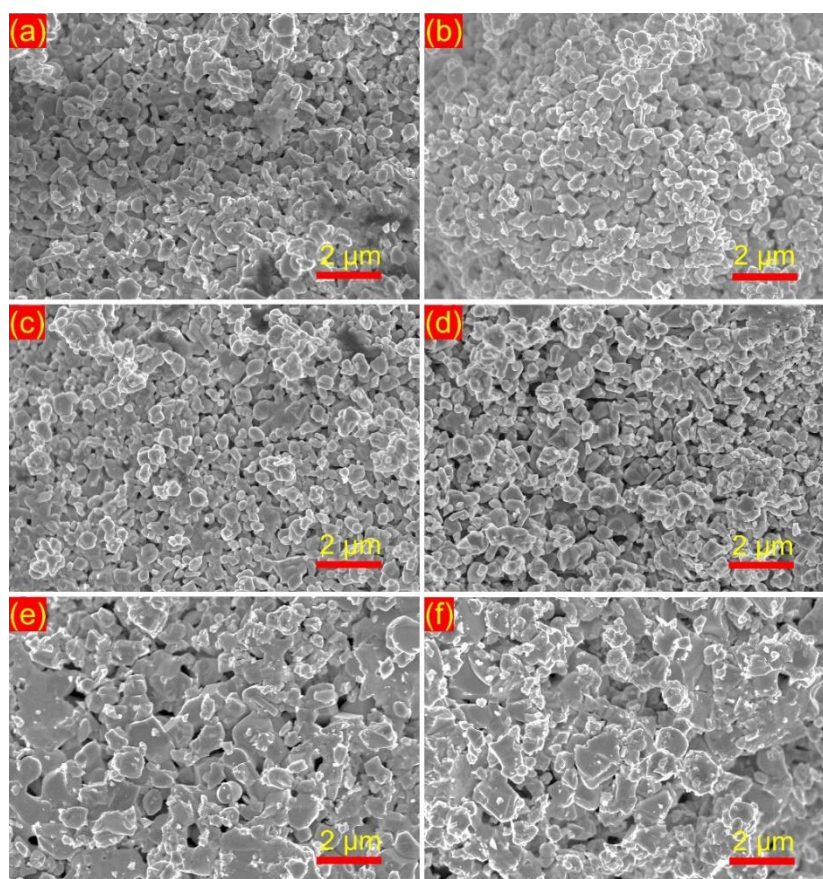


Figure 2. SEM images of $\text{SrFe}_{12-2x}(\text{Cu}_{0.5}\text{Co}_{0.5}\text{Ti})_x\text{O}_{19}$ hexaferrite samples: (a) $x = 0$, (b) $x = 0.05$, (c) $x = 0.10$, (d) $x = 0.15$, (e) $x = 0.20$, and (f) $x = 0.25$ (Tho et al., 2022; Xuan et al., 2024).

Figure 3 shows the S_{11} parameter and reflection coefficient curves as a function of frequency. In the S_{11} parameter curves, the $x = 0$ sample had the lowest values in the 2–10 GHz range, and the $x = 0.15$ sample mostly had the lowest values in the 10–18 GHz range (as shown in **Figure 3(a)**). Actually, the $x = 0.25$ sample had some points that showed the lowest S_{11} values in the frequency at 18 GHz. Overall, the S_{11} curves had low values at two ends and high values at the centered frequencies, meaning that the microwave energy was mostly absorbed at two ends and mainly reflected at the centered frequency. From the S_{11} data, the reflection coefficient (R) could be calculated through the equation $R = |S_{11}|^2 = |S_{22}|^2$. The R values could be divided into two ranges with opposite tendencies, which were similar to the S_{11} values' tendency. The R values increased with the frequency increase in the 2–10 GHz range, and the $x = 0.25$ sample had the highest R -value. Meanwhile, the R -value decreased with the frequency increase in the 10–18 GHz range, and the $x = 0.10$ sample had the highest value in this range (as shown in **Figure 3(b)**). The lower R values at the two ends could originate from ferromagnetic resonance frequency (f_{FMR}) at these frequencies (Lee et al., 2023). According to the EMI shielding theory, the high R -value could result from the sample's high electrical conductivity (Shakir et al., 2022). With the increased doping concentration, the DC conductivity increased in the 2–10 GHz frequency range, thus increasing the reflection coefficient. With the frequency higher than 10 GHz, the DC conductivity decreased, leading to a decrease in R values.

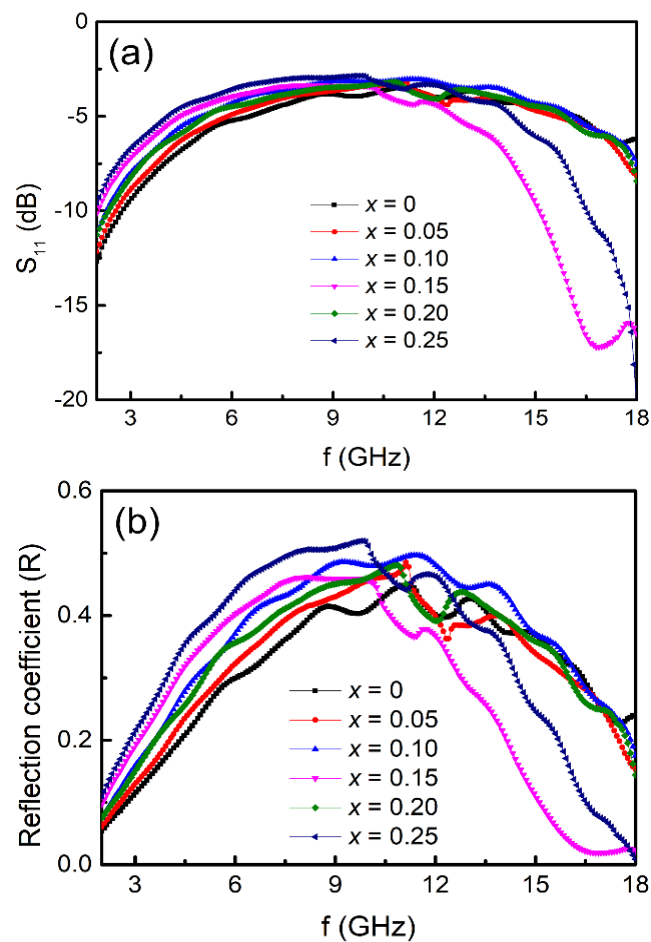


Figure 3. Frequency dependence of (a) the S_{11} parameter and (b) the R -coefficient of SrFe_{12-2x}(Cu_{0.5}Co_{0.5}Ti)_xO₁₉ hexaferrite samples.

Figure 4 shows the S_{21} parameter and transmission coefficient curves as a function of frequency. In opposite tendencies with the S_{11} parameter, the S_{21} parameter decreased with the increase of frequency in the range of 2–10 GHz and then increased with the increase of frequency in the range of 10–18 GHz (as shown in **Figure 4(a)**). In detail, the $x = 0$ sample mostly showed the highest S_{21} value in the frequency range of 2–10 GHz, and the $x = 0.25$ sample showed the lowest value in this frequency range. In the frequency range of 10–18 GHz, the $x = 0.20$ sample showed the lowest value, and the highest values were achieved for the $x = 0.15$ sample (in the frequency range of 10–16 GHz) and the $x = 0.25$ sample (in the frequency range of 16–18 GHz). The transmission coefficient (T) could be calculated from S_{21} values through the equation $T = |S_{21}|^2 = |S_{12}|^2$. The T tendencies of the samples, of course, were similar to those of the S_{21} ones, where T values were highest for the $x = 0$, $x = 0.15$, and $x = 0.25$ samples in the range of 2–10, 10–16, and 16–18 GHz, respectively (as shown in **Figure 4(b)**).

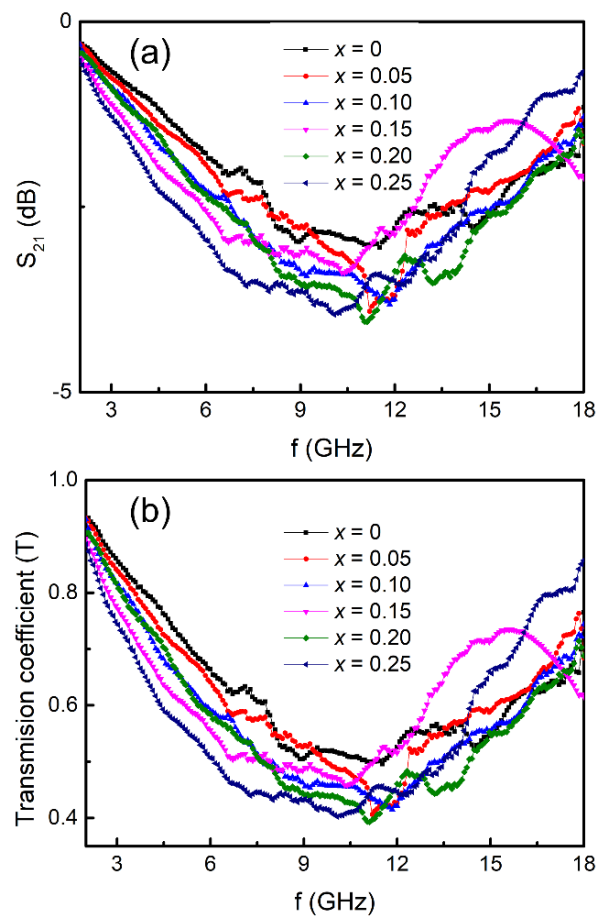


Figure 4. Frequency dependence of (a) the S_{21} parameter and (b) the T -coefficient of SrFe_{12-2x}(Cu_{0.5}Co_{0.5}Ti)_xO₁₉ hexaferrite samples.

After calculating the R and T coefficients, the absorption coefficient (A) could be calculated through the equation $A = 1 - R - T$. **Figure 5** shows the A -coefficient as a function of frequency, where the A values were mainly below 0.1 in the frequency range of 2–8 GHz and relaxation around 0.1 for the frequency range of 8–18 GHz, except for the $x = 0.15$ sample. The high A values of the $x = 0.15$ sample in the frequency range of 14–18 GHz implied the high absorption capability of the $x = 0.15$ sample in that frequency range.

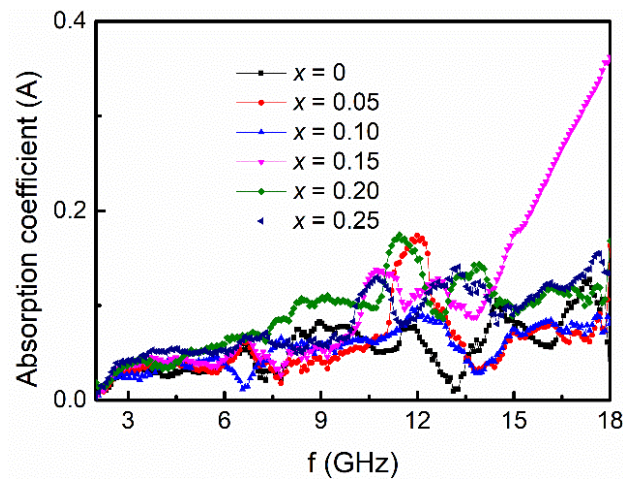


Figure 5. Frequency dependence of the A -coefficient of SrFe_{12-2x}(Cu_{0.5}Co_{0.5}Ti)_xO₁₉ hexaferrite samples.

After calculating the R coefficient, the reflection shielding efficiency (SE_R) could be calculated through the equation $SE_R = -10 \log(1 - R) = -10 \log(1 - |S_{11}|^2)$ (Khan et al., 2022; Singh et al., 2008). **Figure 6** shows SE_R curves as a function of frequency, which had similar tendencies to the R -coefficient. The highest SE_R values could be obtained for the $x = 0.25$ sample, followed by the $x = 0.10$ sample.

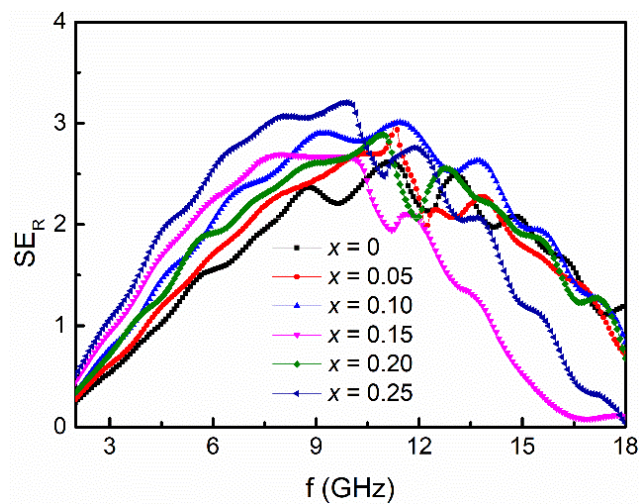


Figure 6. Frequency dependence of SE_R of SrFe_{12-2x}(Cu_{0.5}Co_{0.5}Ti)_xO₁₉ hexaferrite samples.

Following SE_R , the absorption shielding efficiency (SE_A) could be calculated through the equation of $SE_A = -10 \log(1 - A_{\text{eff}})$, where A_{eff} is the effective absorbance, which could be calculated through R and T coefficients through the equation of $A_{\text{eff}} = (1 - R - T)/(1 - R)$ (Saini et al., 2009). Therefore, the SE_A equation could be rewritten as $SE_A = -10 \log(T/(1 - R))$. The SE_A curves of the samples had similar tendencies with the A -coefficient, where the $x = 0.15$ sample had high values in the frequency range of 14–18 GHz, and the $x = 0.20$ sample mostly had high values in the frequency range of 8–14 GHz (as shown in **Figure 7**). The differences in frequency at the highest values of SE_A could be attributed to the presence of impurity phases. As mentioned above, the percentages of SrFe₂O₄ and α -Fe₂O₃ in the $x = 0.20$ sample were higher than those in the $x = 0.15$ samples, leading to a decrease in frequency for maximum SE_A values. Compared to the SE_R

values, the SE_A values were much lower, meaning that reflection was the primary shielding mechanism for the shielding effectiveness of the sample. This could also be used to explain the quite noisy data from the SE_A .

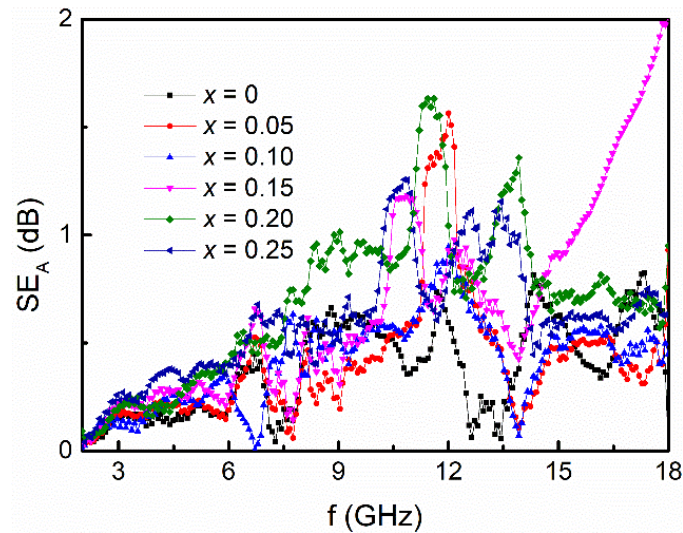


Figure 7. Frequency dependence of SE_A of SrFe_{12-2x}(Cu_{0.5}Co_{0.5}Ti)_xO₁₉ hexaferrite samples.

Figure 8(a) shows total shielding efficiency (SE_T) in the 2–18 GHz frequency range, which could be the sum of SE_R and SE_A . The detail equation is $SE_T = SE_R + SE_A = -10\log(|S_{21}|^2)$. In overall tendencies, the SE_T values increased with the increase in frequency to about 10 GHz, then decreased when the frequency increased from 10 to 18 GHz. The SE_T increased with doping elements in the 2–10 GHz frequency range among the samples. The difference in SE_T between undoped and doped samples was not visible in the 10–18 GHz frequency range. **Figure 8(b)** shows the enlarged view of **Figure 8(a)** in the X band, which confirms the most stable shielding performance of all samples in this band (8–12 GHz). Among the samples, the shielding performance of doped samples was higher than that of undoped one, where the best performances could be achieved for the $x = 0.05$, 0.20, and 0.25 samples.

In order to clarify the effects of doping on the EMI shielding properties of the SrFe_{12-2x}(Cu_{0.5}Co_{0.5}Ti)_xO₁₉, the maximum SE_T values were determined and plotted in **Figure 9**. The $x = 0$ (undoped) sample had a maximum SE_T value of 3.12 dB, the lowest among the samples. With the first doping (the $x = 0.05$ sample), the maximum SE_T value increased to 4.17 dB. Higher doping concentrations decreased the maximum SE_T values to 3.85 and 3.52 dB for the $x = 0.10$ and 0.15 samples, respectively. When the doping concentration reached 0.20, the sample achieved the best maximum SE_T with a value of 4.20 dB, which could be blocked by ~62% of the incident microwave. Then, the maximum SE_T value decreased to 4.01 dB for the $x = 0.25$ sample. It is worth noting that the SE_T value depends on complex permittivity and complex permeability (Gholampoor et al., 2017). In addition, the SE_T of EMI strongly depends on the frequency, the sample's thickness, and the material's conductivity. Thus, the best maximum SE_T values in the $x = 0.05$ and 0.20 samples could be attributed to their highest conductivity caused by doping and the formation and percentages of impurity phases. Furthermore, the enhancement of the SE_T could also be attributed to the synergistic effects of the magnetic dipoles of ferrites and electric dipoles (Kozlovskiy et al., 2021; Shlimas et al., 2021).

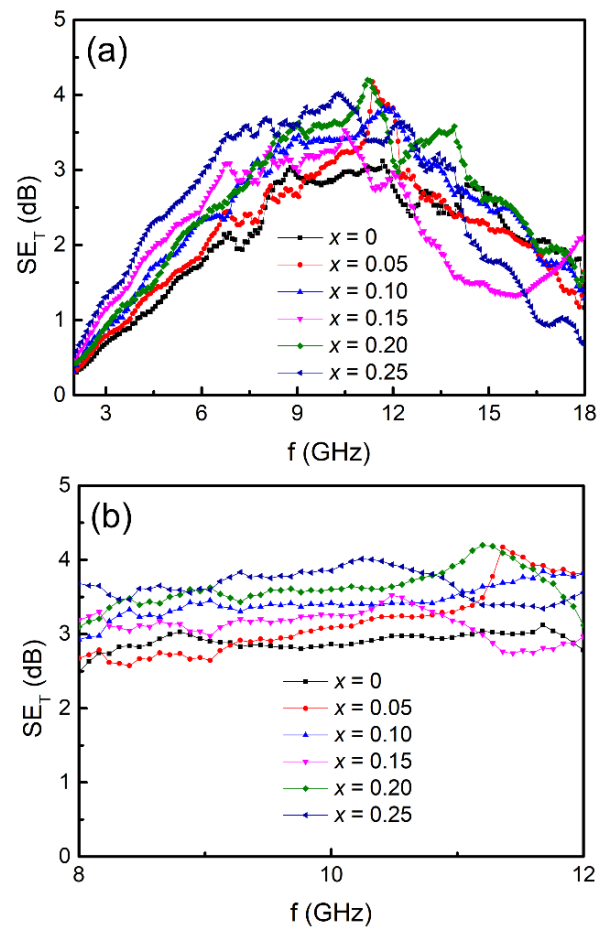


Figure 8. Frequency dependence of SE_T of SrFe_{12-2x}(Cu_{0.5}Co_{0.5}Ti)_xO₁₉ hexaferrite samples in the frequency ranges (a) 2–18 GHz and (b) 12–18 GHz (enlarged view).

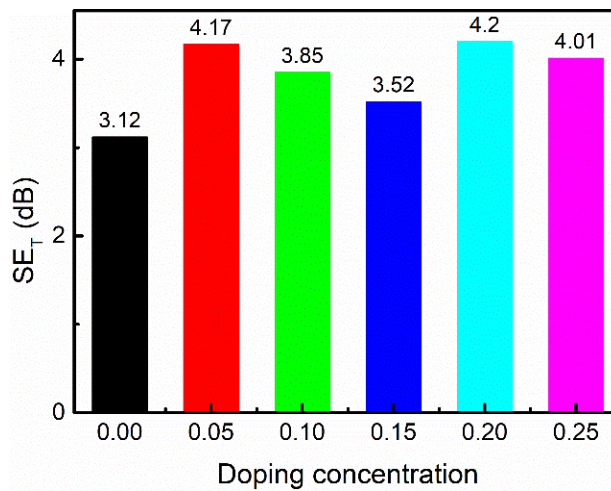


Figure 9. Maximum SE_T values for each sample of SrFe_{12-2x}(Cu_{0.5}Co_{0.5}Ti)_xO₁₉ hexaferrite.

In order to have a comparison among some related ferrites, their EMI shielding efficiency was listed in **Table 1**. Lu et al. reported (2024) that Sr_{1-x}Sm_xFe_{12-y}Mn_yO₁₉ samples reached the best SE_T values with $x = 0.075$ and $y = 0.75$, with the SE_T values varying in the range of 0.8–1.6 dB (Lu et al., 2024). These values were much lower than the SE_T values in this work. Zn_xCu_(1-x)Fe₂O₄ nanoparticles (NPs) prepared by the sol-gel auto-combustion method achieved SE_T values of 4–9 dB (Kavas et al., 2014), which were comparable to our work. In order to achieve higher SE_T values, ferrite should be part of composites, including magnetic and dielectric components. For example, PANI/MWCNTs/Fe₃O₄, PANI@Fe₃O₄ hybrid, and Fe₃O₄@PANI NPs composites achieved SE_T values of 16, 29.3, and 35.1 dB, respectively (Cao et al., 2012; Shukla, 2019; Sun et al., 2013). Similarly, composites of electroless plated FeCo magnetic metal hollow fibers and ethylene propylene diene monomer (EPDM) could reach a SE_T value of 30 dB for a thickness of 3.0 mm (Choi et al., 2015). In this composite, the magnetic alloy of FeCo could benefit EMI shielding effectiveness.

Table 1. Comparison of the EMI shielding efficiency of some ferrite materials.

Sample	Fabrication method	Filler loading (%)	Thickness (mm)	SE_T (dB)	References
Zn _x Cu _(1-x) Fe ₂ O ₄ NPs	Sol-gel auto combustion	50%	3.0	4-9	Kavas et al. (2014)
PANI/MWCNTs/Fe ₃ O ₄	Co-precipitation	-	4.0	16	Cao et al. (2012)
FeCo/EPDM	Electroless plating process	-	3.0	30	Choi et al. (2015)
PANI@Fe ₃ O ₄ hybrid	Solvothermal	-	4.5	29.3	Shukla (2019)
Fe ₃ O ₄ @PANI NPs	Oxidation-reduction	-	1.7	35.1	Sun et al. (2013)
Sr _{1-x} Sm _x Fe _{12-y} Mn _y O ₁₉ ; $x = 0.075, y = 0.75$	Sol-gel	20%	-	~0.8	Lu et al. (2024)
		30%	-	~1.0	
		50%	-	~1.6	
SrFe _{12-2x} (Cu _{0.5} Co _{0.5} Ti) _x O ₁₉ , $x = 0$	Ball milling and heat treatment	40%	3.0	3.12	This work
SrFe _{12-2x} (Cu _{0.5} Co _{0.5} Ti) _x O ₁₉ , $x = 0.05$				4.17	
SrFe _{12-2x} (Cu _{0.5} Co _{0.5} Ti) _x O ₁₉ , $x = 0.10$				3.85	
SrFe _{12-2x} (Cu _{0.5} Co _{0.5} Ti) _x O ₁₉ , $x = 0.15$				3.52	
SrFe _{12-2x} (Cu _{0.5} Co _{0.5} Ti) _x O ₁₉ , $x = 0.20$				4.20	
SrFe _{12-2x} (Cu _{0.5} Co _{0.5} Ti) _x O ₁₉ , $x = 0.25$				4.01	

4. Conclusion

This work investigated the EMI shielding properties of SrFe_{12-2x}(Cu_{0.5}Co_{0.5}Ti)_xO₁₉ samples through scattering parameters. Reflection shielding efficiency (SE_R) mainly contributed to the total shielding efficiency (SE_T). The SE_T increased with the frequency increase in the frequency range of 2–10 GHz, then mainly decreased in the frequency range of 10–18 GHz. The maximum SE_T values of all doped samples were higher than that of the undoped one. Among the doped samples, the $x = 0.05$ and 0.20 samples showed the best SE_T with values of 4.17 and 4.20 dB, respectively. The enhancement of EMI shielding properties of the doped samples could be attributed to the increase in conductivity caused by doping and the combination effects of the magnetic dipoles of ferrites and electric dipoles. The SE_T values of our samples were better or comparable to the SE_T values of M-type hexaferrites and spinel ferrites.

Conflict of Interest

The authors confirm that there is no conflict of interest to declare for this publication.

Acknowledgments

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