



Microwave dielectric properties of $Zn_{1-x}Cu_xWO_4$ ($x= 0, 0.03$)/Polymer Composites



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HIGHLIGHTS

- Novel $Zn_{0.97}Cu_{0.03}WO_4$ excels in microwave composites, offering wireless communication efficiency at a low cost.
- Dielectric properties measured (4-8) GHz via Vector Network Analyzer using transmission/reflection method.
- Optimal outcomes achieved with 10% Vf. $Zn_{0.97}Cu_{0.03}WO_4$ /epoxy & polyurethane composites, ensuring peak performance.

ABSTRACT

The rapid growth in wireless telecommunication systems has spurred a strong interest in lightweight, compact, and cost-effective materials. One promising solution for achieving these characteristics is using polymer matrix ceramic-reinforced composites. In this context, complex oxide materials, specifically $Zn_{1-x}Cu_xWO_4$ (where $x=0$ and $x=0.03$), were used to reinforce three different polymer matrices: epoxy, polyurethane, and silicone rubber. The primary objective was to create composites with low loss factors and high dielectric constants, essential qualities for telecommunications systems. To create the complex oxides, the solid-state reaction method was employed. In particular, the X-ray diffraction (XRD) analysis confirmed that $Zn_{0.97}Cu_{0.03}WO_4$ exhibited a monoclinic phase structure similar to that of $ZnWO_4$. Moreover, the impact of substituting copper ions on the dielectric properties was thoroughly examined through Rietveld refinement of X-ray diffraction data. Subsequently, composite materials were prepared using a simple hand-mixing method with 5% and 10% volume fractions of $Zn_{1-x}Cu_xWO_4$ ($x=0$ and $x=0.03$) integrated into the three different polymer matrices. To this end, the study extensively analyzed how the filler content and its concentration influenced the dielectric properties of these composites. The dielectric properties were characterized within the C-band frequency range, specifically from 4 to 8 GHz, utilizing transmission/reflection measurements with a vector network analyzer (VNA). The results confirm the potential enhancement of $Zn_{0.97}Cu_{0.03}WO_4$ /polymer composites compared to $ZnWO_4$ /polymer composites. More precisely, 10% Vf. $Zn_{0.97}Cu_{0.03}WO_4$ /epoxy composite and 10% Vf. $Zn_{0.97}Cu_{0.03}WO_4$ /polyurethane composite showed satisfactory properties with ($\epsilon_r= 1.36 \times 10^2$, $\tan\delta= 4.72 \times 10^{-1}$ at 7.1 GHz) and ($\epsilon_r= 1.18 \times 10^2$, $\tan\delta= 4.72 \times 10^{-1}$ at 7.1 GHz), respectively.

ARTICLE INFO

Handling editor: Emad A. Yousif

Keywords:

Composites
Loss factor
Dielectric constant
 $Zn_{1-x}Cu_xWO_4$ ceramics

1. Introduction

Microwave dielectrics, which find applications within the frequency range of 300 MHz to 300 GHz, are captivating materials due to their interaction with electromagnetic waves. Upon exposure to these waves, these materials generate polarization through the oscillating electric fields of high-frequency waves [1]. The behavior of electromagnetic wave propagation within a material hinges on its distinct characteristics. Variations in wavelength and velocity of electromagnetic waves transpire within a material medium, governed by its dielectric properties. Classification of microwave materials is often considered based on their unique properties or the structural makeup of the material, be it chemical or physical. Crucial attributes defining a material's performance at microwave frequencies encompass its complex permittivity and complex permeability. When seeking the most fitting material for a given application, decisions are heavily influenced by the material's electrical and/or magnetic traits and its inherent loss factors. Dielectric and magnetic losses, commonly depicted through loss tangents, are tied to the operating frequency. Consequently, thorough material characterization at microwave frequencies proves essential to pinpointing materials best suited for a specific purpose. The material selection process also considers application-specific factors, such as mechanical strength, weight, cost-effectiveness, availability, and the material's chemical and thermal stability [2]. In general, high-frequency operating

electronic devices, such as cellular phones, fast computers, integrated circuits, satellite base stations, auto anti-collision systems, radar systems, electronic navigation technologies, microstrips antennas, and capacitors [3-6], among others, are evolving rapidly. Hence, they require materials that combine both easy fabrication and mechanical strength with good dielectric properties (increasing dielectric constant of materials with retaining low dielectric loss) [7,8]. Combining mechanical properties and dielectric is not easy to obtain in single-component materials. Pure polymers are simple to produce into mechanical solid components, but the dielectric constant is generally low [9]. Furthermore, standard materials with high dielectric constant, such as ceramics, are brittle and demand high temperatures to be processed [10]. On the other hand, modern circuit integration technologies are generally incompatible with that high processing temperature. To solve this problem, a high dielectric constant material, which is mechanically strong and processable at suitable temperatures, can be used. In this regard, a huge interest has been developed in composite materials, like ceramic/polymer composites, which can include the required components properties [11,12]. High-dielectric constant ceramic/polymer matrix composites have successfully replaced traditional high-dielectric materials, such as ceramics [13]. The literature review on ceramics [14-17] highlights a notable discrepancy in the reported values of dielectric parameters for ceramic materials. As far as we know, the dielectric properties of $Zn_{1-x}Cu_xWO_4$ (where $x=0$ and $x=0.03$)/polymer composites have not been studied. This paper represents a substantial advancement in the realm of dielectric materials for wireless communication systems. It effectively fills critical gaps in the existing literature by enhancing dielectric properties, emphasizing cost-efficiency, and providing a frequency-specific analysis within the C-band range. By meticulously optimizing composite properties and underscoring the need for practical and economical solutions, this research offers valuable insights and tangible contributions to wireless telecommunications. The work paves the way for developing reliable and cost-effective materials to drive innovation and progress in wireless communication technologies.

The present study aims to fabricate cost-effective microwave materials with optimum properties. More specifically, epoxy/ $Zn_{1-x}Cu_xWO_4$ ($x=0, 0.03$), polyurethane / $Zn_{1-x}Cu_xWO_4$ ($x=0, 0.03$), and silicone rubber / $Zn_{1-x}Cu_xWO_4$ ($x=0, 0.03$) composites were considered in the current study. The phase content of complex oxides was characterized by XRD and microwave dielectric properties of composites that were investigated at the C band frequency. The effect of copper ions on the microwave dielectric properties was explored, and the effect of filler content and its concentration of composites were studied.

2. Experimental procedure

2.1 Preparations of materials

The solid-state reaction method synthesized the $Zn_{1-x}Cu_xWO_4$ ($x=0, 0.03$) complex oxides. High-purity CuO (99%), ZnO (99%), and WO_3 (99%) powders were accurately measured in stoichiometric proportions. These powders were then subjected to a milling process for six hours using an electrical blender. This milling aimed to ensure a thorough and uniform mixing of the powders. Subsequently, the resulting mixtures were calcined at a temperature of 900°C for six hours. This calcination step is a crucial part of the synthesis process, contributing to the formation of the complex oxides. After calcination, the powders were finely ground to achieve a homogeneous and well-mixed powder composition. These complex oxides were then incorporated into the polymer matrices (epoxy, polyurethane, and silicone rubber) to create the desired composite materials for further analysis and characterization.

2.2 Characterization

X-ray diffraction (XRD) analysis was conducted using a Lab 6000 XRD instrument equipped with $CuK\alpha$ radiation manufactured in Japan to assess the raw materials' phase compositions and the complex oxides. The scan rate was set at 0.2°, covering a 2theta angle range from 10° to 90°. The average particle diameters of the oxides were determined through AFM measurements, yielding values of 47.78 nm for ZnO, 44.07 nm for WO_3 , and 75.69 nm for CuO. The dielectric properties of the polymer-ceramic composites were assessed utilizing the Transmission/Reflection method within the frequency range of 4–8 GHz. This technique involves the measurement of both the reflected signal (S_{11}) and the transmitted signal (S_{21}). A network analyzer (MS4642A, 20 GHz) produced in the USA was employed to conduct these measurements. Subsequently, the scattering parameters, S_{11} and S_{21} , were processed to derive dielectric properties using the mathematical Nicholson-Ross-Weir (NRW) method. [18,19].

3. Results and discussion

Figure 1a shows the x-ray diffraction patterns of $ZnWO_4$ and $Zn_{0.97}Cu_{0.03}WO_4$ complex oxides. The diffraction pattern represents the monoclinic phase of $ZnWO_4$ according to the standard PDF (.96-156-7217). Specifically, Figure 1a shows that the peaks of $Zn_{0.97}Cu_{0.03}WO_4$ ceramic are shifted to higher 2theta values. In other words, one or more lattice parameter was shrunk due to Cu^{2+} substitution. Accordingly, the cell volume was decreased, which can be attributed to the fact that the radius of copper ion is smaller than that of zinc ion, possibly leading to lattice shrinkage. The XRD results indicate that adding copper ions (Cu) to $Zn_{1-x}Cu_xWO_4$ did not significantly alter the crystal structure, as both materials maintained a monoclinic phase. This suggests that introducing copper primarily influences the material's electronic properties rather than its crystallographic arrangement. Maintaining a similar crystal structure while modifying other properties is advantageous in material design. It allows the creation of materials with tailored electronic characteristics without compromising their fundamental structure. Further analysis, such as Rietveld refinement of the XRD data, may reveal subtle structural changes or lattice parameter variations between $Zn_{0.97}Cu_{0.03}WO_4$ and $ZnWO_4$, which can provide additional insights into the impact of copper on the materials' properties. The refined $Zn_{0.97}Cu_{0.03}WO_4$ powder is shown in Figure 1b, and the refined lattice parameter of $Zn_{0.97}Cu_{0.03}WO_4$ is shown in Table 1. The volume of the lattice of $Zn_{0.97}Cu_{0.03}WO_4$ is smaller than that of $ZnWO_4$. Accordingly, the bond in the new lattice is shorter

and stronger than that in the pure phase. The dielectric response of a material, characterized by its permittivity (ϵ_r), is represented as a complex value $\epsilon_r = (\epsilon' - i \cdot \epsilon'')$. The real part (ϵ') signifies lossless dielectric interaction, while the imaginary part (ϵ'') represents lossy dielectric interaction. This study focuses on how the dielectric constant (ϵ') and loss tangent ($\tan \delta$) change with frequency, filler content, and concentration. Figures 2 to 5 reveal the variations in dielectric constant and $\tan \delta$ for the composites at different frequencies. It's evident that the dielectric constant increases slightly with frequency for all composites. However, the real impact is observed with filler content and concentration variations.

The Composites with a 10% volume fraction of $Zn_{1-x}Cu_xWO_4$ ($x=0.03$) ceramics demonstrate substantial improvements in dielectric properties. This enhancement is attributed to the Copper ions in $Zn_{0.97}Cu_{0.03}WO_4$ having higher polarizability than zinc ions in $ZnWO_4$. Furthermore, Cu ions can lead to stronger electric field interactions and improved charge storage capacity within the composite. This increased ability to store electric charge contributes to a higher dielectric constant. The composite's dielectric constant is influenced by interfacial polarization, which occurs at the interfaces between the filler and the polymer matrix. At 10%, filler exhibits a significant increase in the interfacial area, leading to enhanced interfacial polarization and denser composites, which resulted from the increase in the dielectric properties of a 10% volume fraction of $Zn_{1-x}Cu_xWO_4$, there was a higher concentration of filler particles in the polymer matrix. This increased filler content generally leads to a higher dielectric constant due to the greater number of filler particles contributing to charge storage [20-22], as depicted in Figure 3.

The $Zn_{1-x}Cu_xWO_4$ ($x=0.03$)/epoxy composites shown in Figure 2b may exhibit distinct dielectric characteristics in the 4–8 GHz frequency range. In contrast to $ZnWO_4$ as shown in Figure 2a, $Zn_{0.97}Cu_{0.03}WO_4$ is a different kind of complex oxide and can have different dielectric properties. Therefore, the dielectric constant changes in the response of $Zn_{1-x}Cu_xWO_4$ ($x=0.03$)/epoxy composites at this frequency range may vary. In this context, a notable finding is the considerable discrepancy between $Zn_{0.97}Cu_{0.03}WO_4$ and $ZnWO_4$ peak values of the dielectric constant at various frequencies. In particular, $Zn_{0.97}Cu_{0.03}WO_4$ shows a peak value of 17 at 5.38 GHz and 19.7 at 6.4 GHz compared to that of $ZnWO_4$, which reaches a peak value of 28 at 5.5 GHz. This difference in the two materials' dielectric behavior across a relatively narrow frequency range may have an impact on how they are used in different microwave devices. The responses of $Zn_{1-x}Cu_xWO_4$ ($x=0.03$)/polyurethane and $ZnWO_4$ /polyurethane composites to the electromagnetic wave in the frequency range of 4–8 GHz are very different. With a peak dielectric constant of 5 at 7.8 GHz, for $Zn_{1-x}Cu_xWO_4$ ($x=0.03$)/polyurethane composite. As opposed to this, the $ZnWO_4$ /polyurethane composite exhibits a dielectric constant value of 6.3 at 4.5GHz and 6.48 at 5.482GHz as shown in Figure. 2c and d. The response of $Zn_{1-x}Cu_xWO_4$ ($x=0.03$)/silicone rubber and $BaWO_4$ /silicone rubber composites to the electromagnetic wave at a frequency range of 4-8 GHz is shown in Figure 2 e, f, which displays a specific dielectric constant property. The peak is observed for the silicone rubber/ $ZnWO_4$ composite at 5.58 GHz with a dielectric constant of 19. In contrast, peaks may be seen in the silicone rubber/ $Zn_{0.97}Cu_{0.03}WO_4$ composite at 5.34 GHz with dielectric constant of 9.39.

Figure 3 b, d and f shows the response of the epoxy/10% $Zn_{0.97}Cu_{0.03}WO_4$ composite, the 10% $Zn_{0.97}Cu_{0.03}WO_4$ /polyurethane composite, and the 10% $Zn_{0.97}Cu_{0.03}WO_4$ /silicone rubber to the frequency range of 4-8 GHz, displaying specific results. The dielectric constant for these composites specifically displayed unique peaks at various frequencies, including 136 at 7.1 GHz, 118 at 7.1 GHz, and 24 at 6.78 GHz, respectively. These composites may display frequency dependent behavior based on the observed fluctuation in the dielectric constants at various frequencies. In dielectric materials, where the dielectric constant fluctuates with the frequency of the applied electromagnetic field, this phenomenon is common. These fluctuations are a result of the internal dipoles of the composite reacting to the shifting electric field. While, Figure 3 a,c and e shows the response of epoxy/10% $ZnWO_4$ composite, the 10% $ZnWO_4$ /polyurethane composite, and the 10% $ZnWO_4$ /silicone rubber to the electric field. In this regard, the dielectric constant for these composites at different frequencies, including 4.11 at 7.4 GHz for epoxy/10% $ZnWO_4$ composite, 29.9 at 7.02 GHz and 14.3 at 7.54 GHz for 10% $ZnWO_4$ /polyurethane composite, 18.9 at 4.86GHz, 23.9 at 6.86 GHz, and 27 at 6.98 GHz for 10% $ZnWO_4$ /silicone rubber.

The behavior of the 10% $Zn_{0.97}Cu_{0.03}WO_4$ /epoxy composite, with a dielectric constant reaching 136 at 7.1 GHz, indicates the frequency-dependent nature of dielectric behavior. At different frequencies, the dielectric response varies due to the mobility of polar groups in the polymer chains. In the case of high-frequency applications, the mobility of polar groups in the polymer may not contribute significantly to the dielectric constant. Therefore, the dielectric constant primarily depends on the ceramic filler and its concentration [11]. The interaction and compatibility between the filler ($Zn_{1-x}Cu_xWO_4$) and the polymer matrix are essential. Effective coupling between the filler and the matrix can enhance dielectric properties. The $Zn_{0.97}Cu_{0.03}WO_4$ may form stronger bonds or interactions with the polymer matrix. Epoxy has been recognized for its superior compatibility and robust interactions with inorganic fillers like $Zn_{0.97}Cu_{0.03}WO_4$, surpassing silicone rubber or polyurethane. These intensified interactions promote better dispersion and more efficient bonding between the filler and the polymer matrix, ultimately improving dielectric properties, facilitating better charge storage, and resulting in a higher dielectric constant. This insight is crucial for designing materials optimized for specific frequency bands [23,24]. The dielectric loss, represented as the tangent of the loss angle ($\tan \delta$), is an essential material parameter that should ideally be minimized. Figure 4 compares the loss tangent ($\tan \delta$) for the three types of matrices reinforced with a 5% volume fraction of $Zn_{1-x}Cu_xWO_4$ ($x=0, 0.03$). On the other hand, Figure 5 shows the loss tangent for the three polymers loaded with a 10% volume fraction of $Zn_{1-x}Cu_xWO_4$ ($x=0, 0.03$) complex oxide. In particular, the dielectric loss decreases with increasing frequency and becomes frequency-independent at higher frequencies, as demonstrated in Figures 4 and 5. The high dielectric loss at lower frequencies can be attributed to charge carrier accumulation at the interfaces. The filler content and its concentration can influence the loss tangent of these composites Figure 4 a,c, and e shows the loss tangent of epoxy/5% $ZnWO_4$ composite, the 5% $ZnWO_4$ /polyurethane composite, and the 5% $ZnWO_4$ /silicone rubber, including 0.12 at 5.58GHz for epoxy/5% $ZnWO_4$, 1.08 at 4.5GHz and 1.4 at 5.48 for 5% $ZnWO_4$ /polyurethane composite, and 1.1 at 5.58 GHz for 5% $ZnWO_4$ /silicone rubber. Figure 4 b,d, and f shows the loss tangent of the epoxy/5% $Zn_{0.97}Cu_{0.03}WO_4$ composite, 5% $Zn_{0.97}Cu_{0.03}WO_4$ /polyurethane composite, and the 5% $Zn_{0.97}Cu_{0.03}WO_4$ /silicone rubber composite, these composites have different loss tangent at various frequencies, including 0.2 at 5.38 GHz and 0.336 at 6.4GHz for epoxy/5% $Zn_{0.97}Cu_{0.03}WO_4$ composite,

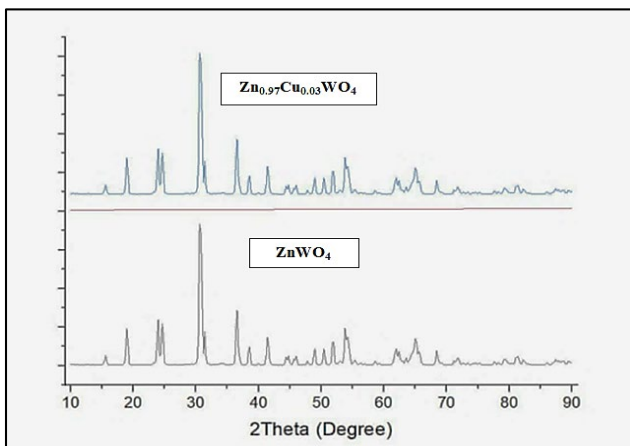
0.329 at 7.86 GHz for 5% $Zn_{0.97}Cu_{0.03}WO_4$ / polyurethane composite, and 0.445 at 5.34 GHz for 5% $Zn_{0.97}Cu_{0.03}WO_4$ / silicone rubber. Figure 5 a, c, and e shows the loss tangent of epoxy/10% $ZnWO_4$ composite, 10% $ZnWO_4$ /polyurethane composite, and the 10% $ZnWO_4$ /silicone rubber, including -4.43 at 7.4 GHz for epoxy/10% $ZnWO_4$ composite, 0.85 at 7.02 GHz and -4.8 at 7.54 GHz for 10% $ZnWO_4$ / polyurethane composite, and -1.27 at 4.86 GHz, 0.0955 at 6.86 GHz, and 0.371 at 6.98 GHz for 10% $ZnWO_4$ /silicone rubber. Figure 5 b, d, and f shows the loss tangent of the epoxy/10% $Zn_{0.97}Cu_{0.03}WO_4$ composite, 10% $Zn_{0.97}Cu_{0.03}WO_4$ /polyurethane composite, and the 10% $Zn_{0.97}Cu_{0.03}WO_4$ / silicone rubber composite, including -1.38 at 5.7 GHz, 0.472 at 7.1 GHz, and 0.474 at 7.76 GHz for epoxy/10% $Zn_{0.97}Cu_{0.03}WO_4$ composite, 1.06 at 5.76 GHz, and 0.47 at 7.1 GHz for 10% $Zn_{0.97}Cu_{0.03}WO_4$ / polyurethane composite, and -0.662 at 4.84 GHz, 0.602 at 5.44 GHz, and 0.608 at 6.78 GHz for 10% $Zn_{0.97}Cu_{0.03}WO_4$ / silicone rubber composite. The findings are succinctly presented in Table 2.

Table 1: The data of crystallographic from Rietveld refinement for $Zn_{1-x}Cu_xWO_4$ ($x=0.03$)

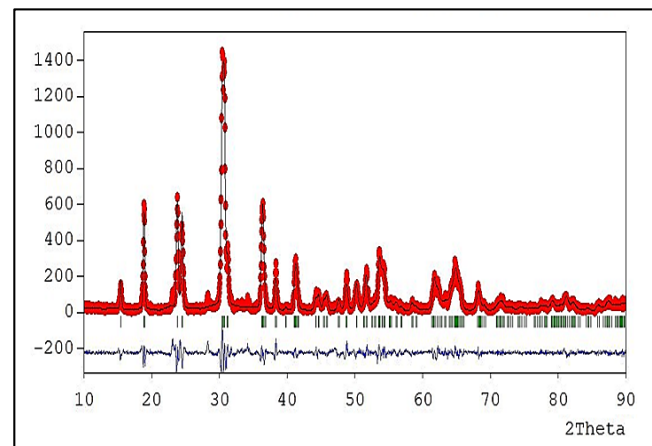
x value	0	0.03
a Å	4.698	4.698
b Å	5.722	5.719
c Å	4.928	4.931
V Å ³	140.0877	132.485

Table 2: Summary of results

Polymer-ceramic	Frequency (GHz) as a function of filler volume fraction (%)		Dielectric constant as a function of filler volume fraction (%)		Loss tangent as a function of filler volume fraction (%)	
	5%	10%	5%	10%	5%	10%
	Epoxy/ $ZnWO_4$	5.58	7.4	28	4.11	0.12
Polyurethane / $ZnWO_4$	4.5	7.02	6.3	29.9	1.08	0.85
	5.48	7.54	6.48	14.3	1.4	-4.8
Silicone Rubber/ $ZnWO_4$	5.58	4.86	19	18.9	1.1	-1.27
		6.86		23.9		0.0955
Epoxy/ $Zn_{0.97}Cu_{0.03}WO_4$		6.98		27		0.371
	5.38	5.7	17	11.3	0.2	-1.38
	6.4	7.1	19.7	136	0.336	0.472
		7.76		26		0.474
Polyurethane/ $Zn_{0.97}Cu_{0.03}WO_4$	7.86	5.76	5	11.6	0.329	1.06
		7.1		118		0.47
Silicone Rubber/ $Zn_{0.97}Cu_{0.03}WO_4$	5.34	4.84	9.39	22.4	0.445	-0.662
		5.44		6.65		0.602
		6.78		24.5		0.608



(a)



(b)

Figure 1: (a) The X-ray diffraction patterns of $(Zn_{1-x}Cu_x)WO_4$ ($x=0, 0.03$) complex oxides, (b) the refinement pattern of $Zn_{0.97}Cu_{0.03}WO_4$ complex oxide using the fullprof software

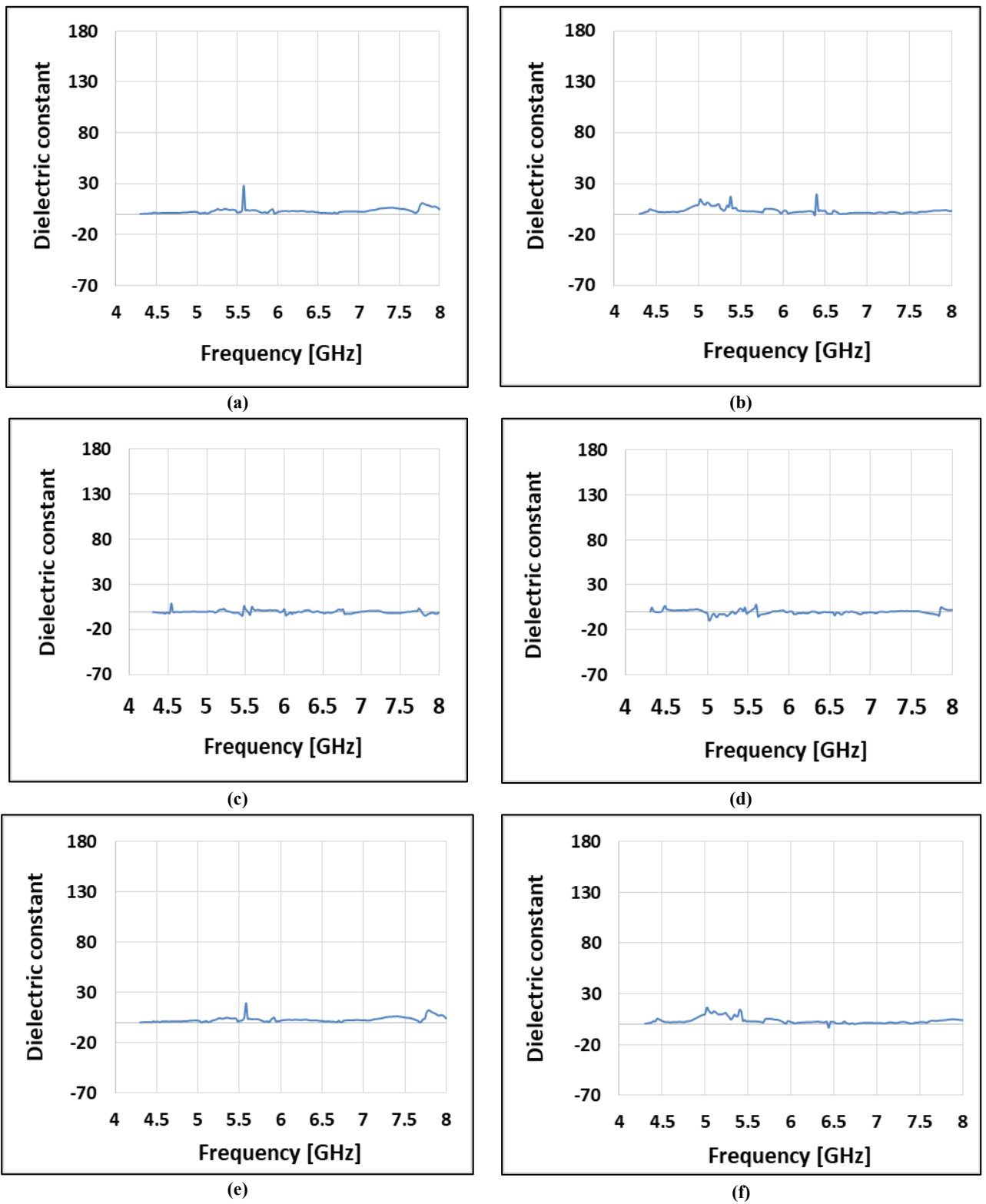


Figure 2: The dielectric constant calculated in the C-band frequency for (a) 5% volume fraction $Zn_{1-x}Cu_xWO_4(x=0)$ /epoxy composite (b) 5% volume fraction $Zn_{1-x}Cu_xWO_4(x=0.03)$ /epoxy composite (c) 5% volume fraction $Zn_{1-x}Cu_xWO_4(x=0)$ /polyurethane composite (d) 5% volume fraction $Zn_{1-x}Cu_xWO_4(x=0.03)$ / polyurethane composite (e) 5% volume fraction $Zn_{1-x}Cu_xWO_4(x=0)$ /silicone rubber composite (f) 5% volume fraction $Zn_{1-x}Cu_xWO_4(x=0.03)$ / silicone rubber composite

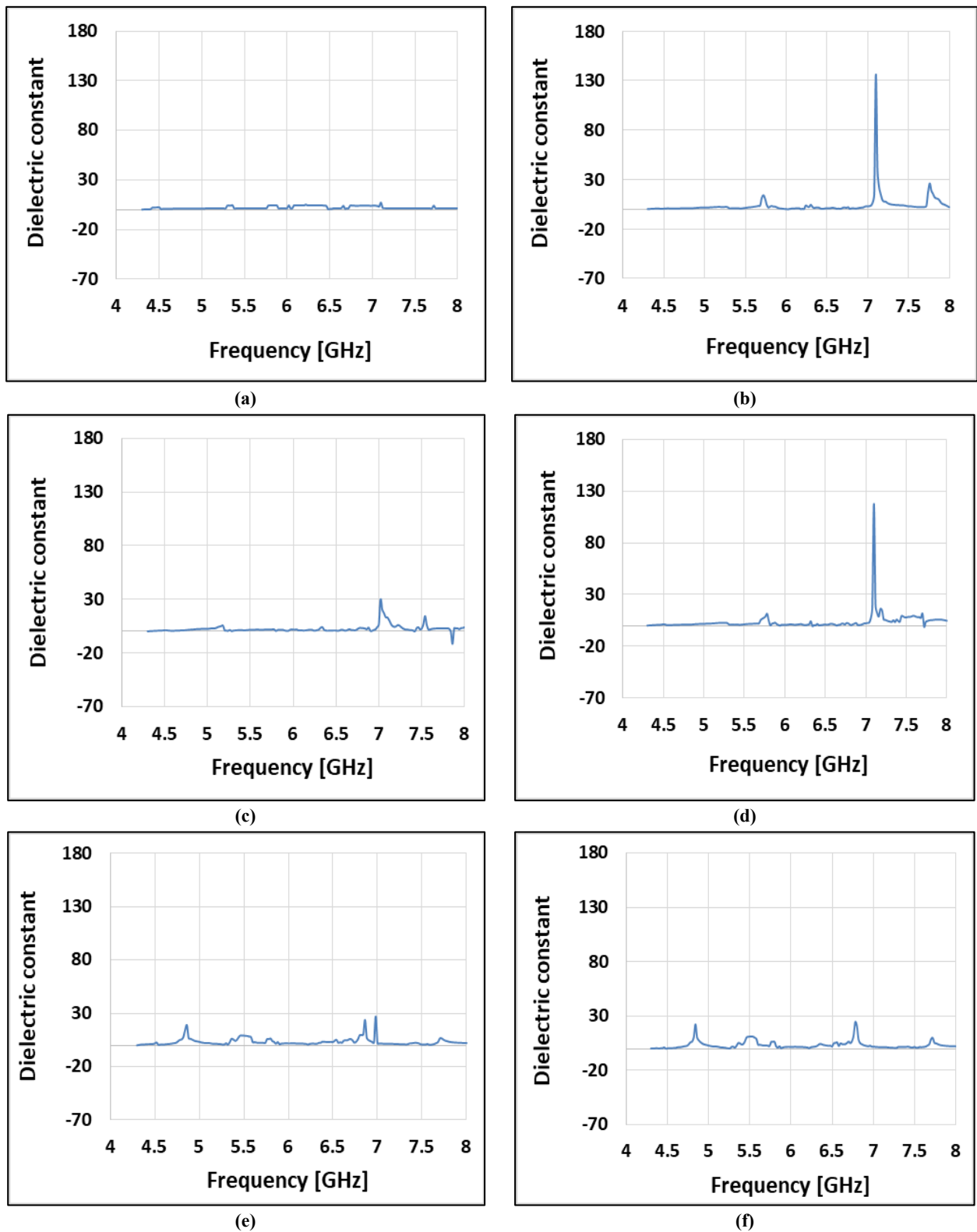


Figure 3: The dielectric constant calculated in the C-band frequency for (a) 10% volume fraction $Zn_{1-x}Cu_xWO_4(x=0)$ /epoxy composite (b) 10% volume fraction $Zn_{1-x}Cu_xWO_4(x=0.03)$ /epoxy composite (c) 10% volume fraction $Zn_{1-x}Cu_xWO_4(x=0)$ /polyurethane composite (d) 10% volume fraction $Zn_{1-x}Cu_xWO_4(x=0.03)$ / polyurethane composite (e) 10% volume fraction $Zn_{1-x}Cu_xWO_4(x=0)$ / silicone rubber composite (f) 10% volume fraction $Zn_{1-x}Cu_xWO_4(x=0.03)$ / silicone rubber composite

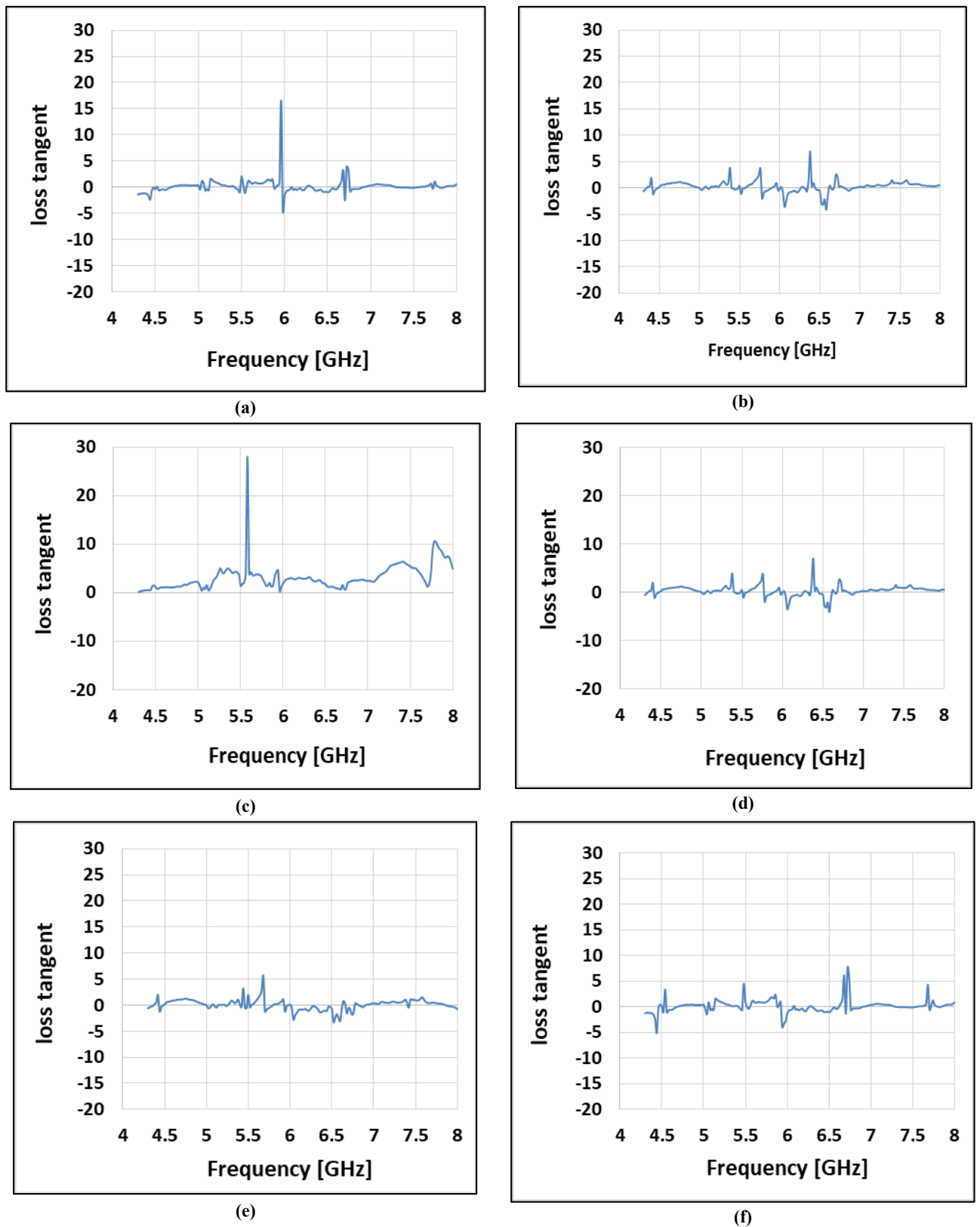


Figure 4: The tangent loss calculated in the C-band frequency for (a) 5% volume fraction $Zn_{1-x}Cu_xWO_4(x=0)$ /epoxy composite (b) 5% volume fraction $Zn_{1-x}Cu_xWO_4(x=0.03)$ /epoxy composite (c) 5% volume fraction $Zn_{1-x}Cu_xWO_4(x=0)$ /polyurethane composite (d) 5% volume fraction $Zn_{1-x}Cu_xWO_4(x=0.03)$ /polyurethane composite (e) 5% volume fraction $Zn_{1-x}Cu_xWO_4(x=0)$ /silicone rubber composite (f) 5% volume fraction $Zn_{1-x}Cu_xWO_4(x=0.03)$ /silicone rubber composite

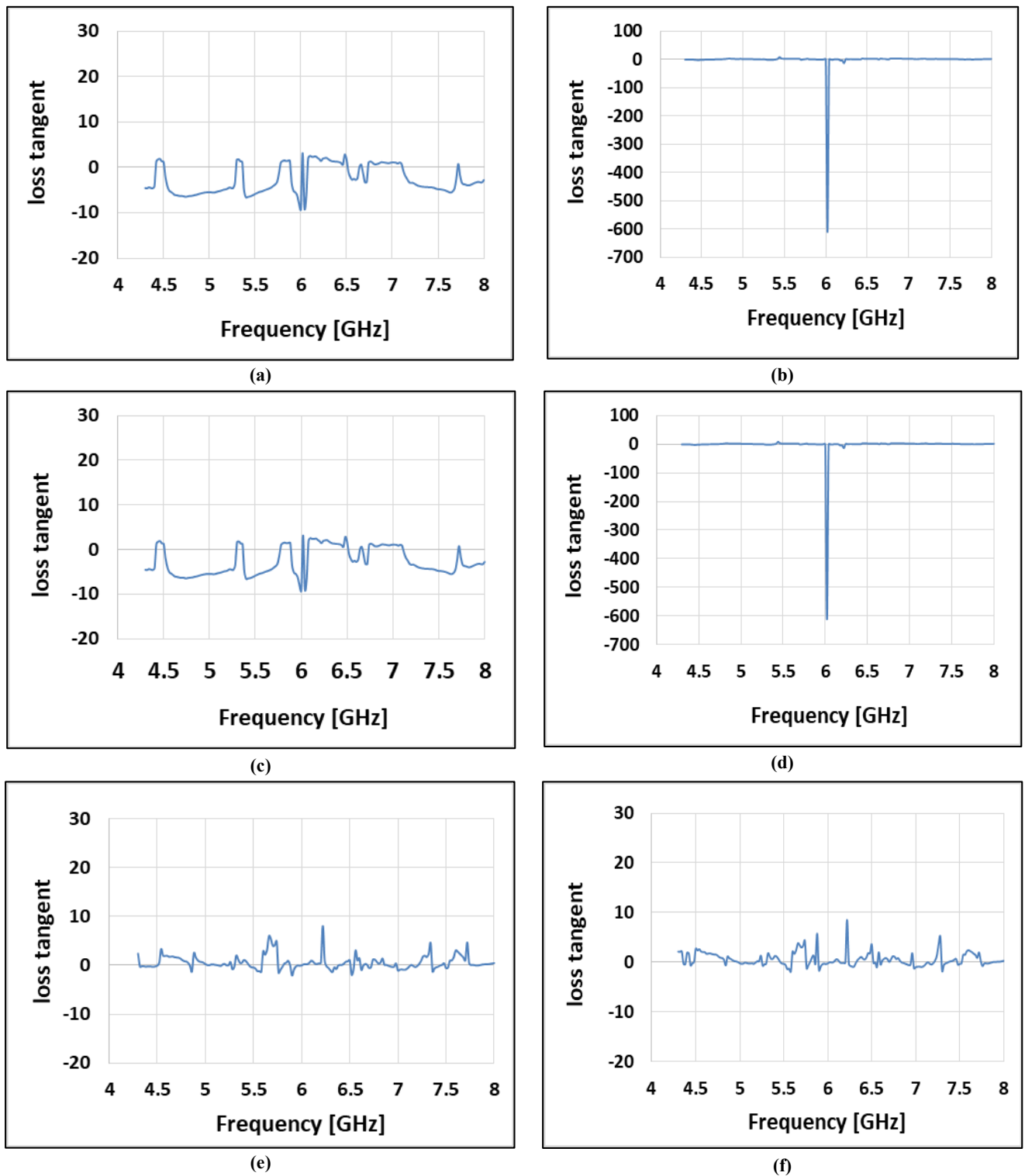


Figure 5: The tangent loss calculated in the C-band frequency for (a) 10% volume fraction $Zn_{1-x}Cu_xWO_4(x=0)$ / epoxy composite (b) 10% volume fraction $Zn_{1-x}Cu_xWO_4(x=0.03)$ /epoxy composite (c) 10% volume fraction $Zn_{1-x}Cu_xWO_4(x=0)$ /polyurethane composite (d) 10% volume fraction $Zn_{1-x}Cu_xWO_4(x=0.03)$ /polyurethane composite (e) 10% volume fraction $Zn_{1-x}Cu_xWO_4(x=0)$ /silicone rubber composite (f) 10% volume fraction $Zn_{1-x}Cu_xWO_4(x=0.03)$ /silicone rubber composite

4. Conclusion

A microwave dielectric ceramic ($Zn_{1-x}Cu_xWO_4$ ($x=0, 0.03$)) was synthesized utilizing the solid-state reaction method. The complex oxide matches $ZnWO_4$, and the riet-veld refinement results give the crystal lattice parameters. In particular, ($Zn_{1-x}Cu_xWO_4$ ($x=0, 0.03$))/Epoxy, ($Zn_{1-x}Cu_xWO_4$ ($x=0, 0.03$))/polyurethane, and silicone rubber / ($Zn_{1-x}Cu_xWO_4$ ($x=0, 0.03$)) composites have been successfully synthesized using a simple hand-mixing technique. In addition, the microwave dielectric properties of the composites were measured by the transmission/reflection method using a vector network analyzer, and the S-

parameters were converted to the dielectric properties by the Nicholson-Ross-Weir (NRW) method. The presence of ($Zn_{1-x}Cu_xWO_4$ ($x=0, 0.03$)) ceramic in three types of polymer matrices has contributed to changing the dielectric properties (the dielectric constant and the loss tangent) in the C-band frequency range of (4-8) GHz. Substituting Zn ion with copper improves microwave dielectric properties by the increased dielectric constant and the decreased loss factor for $Zn_{1-x}Cu_xWO_4$ ($x=0.03$)/polymer composites. The dielectric constant and the loss factor of the ($Zn_{1-x}Cu_xWO_4$ ($x=0, 0.03$))/polymer composites at the filler volume fraction of 10% were the best at 7-7.5 GHz and better than those of the filler volume fraction of 5%.

Author contributions

Conceptualization, L. Faeq, S. Farid and F. Hashim; data curation, L. Faeq, S. Farid and F. Hashim; writing—original draft preparation, L. Faeq, S. Farid and F. Hashim ; writing—review and editing, L. Faeq, S. Farid and F. Hashim. All authors have read and agreed to the published version of the manuscript.

Funding

This research received no specific grant from any funding agency in the public, commercial, or not-for-profit sectors.

Data availability statement

The data that support the findings of this study are available on request from the corresponding author.

Conflicts of interest

The authors declare that there is no conflict of interest.

- [1] H. Ohsato, Functional advances of microwave dielectrics for next generation, *Ceram. Int.*, 38 (2012) 141-146. <https://doi.org/10.1016/j.ceramint.2011.04.068>
- [2] A. Raveendran, M.T. Sebastian, S. Raman, Applications of microwave materials: A review, *J. Electron. Mater.*, 48 (2019) 2601-2634. <https://doi.org/10.1201/9781003459880-7>
- [3] E.J. Rothwell, R.O. Ouedraogo, Antenna miniaturization: definitions, concepts, and a review with emphasis on metamaterials, *J. Electromagn. Waves Appl.*, 28 (2014) 2089-2123. <https://doi.org/10.1080/09205071.2014.972470>
- [4] D.H. Kuo, C.C. Chang, T.Y. Su, W.K. Wang, Dielectric behaviours of multi-doped $BaTiO_3$ /epoxy composites, *J. Eur. Ceram. Soc.*, 21 (2001) 1171-1177. [https://doi.org/10.1016/s0955-2219\(00\)00327-7](https://doi.org/10.1016/s0955-2219(00)00327-7)
- [5] D.H. Kuo, C.C. Chang, T.Y. Su, W.K. Wang, Dielectric properties of three ceramic/epoxy composites, *Mater. Chem. Phys.*, 85 (2004)201-206. <https://doi.org/10.1016/j.matchemphys.2004.01.003>
- [6] G.C. Manika, G.C. Psarras, Barium titanate/epoxy resin composite nanodielectrics as compact capacitive energy storing systems, *EXPRESS Polym. Lett.*, 13 (2019) 749-758. <https://doi.org/10.3144/expresspolymlett.2019.63>
- [7] J. Guo, X. Zhao, T. Herisson De Beauvoir, J.H. Seo, S.S. Berbano, A.L. Baker, C. Azina, Recent progress in applications of the cold sintering process for ceramic–polymer composites, *Adv. Funct. Mater.*, 28 (2018) 1801724. <https://doi.org/10.1002/adfm.201801724>
- [8] Y. Qi, Q. Luo, J. Shen, L. Zheng, J. Zhou, W. Chen, Surface modification of BMN particles with silane coupling agent for composites with PTFE, *Appl. Surf. Sci.*, 414 (2017)147-152. <https://doi.org/10.1016/j.apsusc.2017.04.072>
- [9] R. Popielarz, C.K. Chiang, Polymer composites with the dielectric constant comparable to that of barium titanate ceramics, *Mater. Sci. Eng. B*, 139 (2007) 48-54. <https://doi.org/10.1016/j.mseb.2007.01.035>
- [10] D. Dimos, S.J. Lockwood, R.W. Schwartz, Thin-film decoupling capacitors for multichip modules, *IEEE Trans. Compon. Packag. Manuf. Technol.*, Part A: 18 (1995) 174-179. [https://doi.org/10.1016/0026-2714\(96\)84499-4](https://doi.org/10.1016/0026-2714(96)84499-4)
- [11] R. Popielarz, C.K. Chiang, R. Nozaki, J. Obrzut, Dielectric properties of polymer/ferroelectric ceramic composites from 100 Hz to 10 GHz, *Macromolecules*, 34 (2001) 5910-5915. <https://doi.org/10.1021/ma001576b>
- [12] S. Ogitani, S.A. Bidstrup-Allen, P. Kohl, Development of high-K photo-definable epoxy composites for embedded capacitors, In *Twenty Third IEEE/CPMT Int. Electron. Manufacturing Technol. Symp.*, (1998) 199-205. <https://doi.org/10.1109/iemt.1998.731076>
- [13] Z. Zhang, J. Zhang, S. Li, J. Liu, M. Dong, Y. Li, N. Lu, Effect of graphene liquid crystal on dielectric properties of polydimethylsiloxane nanocomposites, *Compos.B:Eng.*, 176 (2019) 107338. <https://doi.org/10.1016/j.compositesb.2019.107338>
- [14] R.C. Pullar, S. Farrah, N.M.N. Alford, $MgWO_4$, $ZnWO_4$, $NiWO_4$ and $CoWO_4$ microwave dielectric ceramics, *J. European Ceram. Soc.*, 27 (2007) 1059-1063. <https://doi.org/10.1016/j.jeurceramsoc.2006.05.085>
- [15] X. Lu, B. Quan, K. Zheng, P. Chu, J. Wang, G. Shen, Sc modification induced short-range cation ordering and high microwave dielectric performance in $ZnGa_2O_4$ spinel ceramics, *J. Alloys Compd.*, 873 (2021) 159758. <https://doi.org/10.1016/j.jallcom.2021.159758>

- [16] Y. Wang, J. Lv, J. Wang, F. Shi, Z. Qi, Lattice vibrational characteristics, crystal structure and dielectric properties of Ba₂MgWO₆ microwave dielectric ceramic, *Ceram. Int.*, 47 (2021) 17784-17788. <https://doi.org/10.1016/j.ceramint.2021.02.224>
- [17] E.N. Sota, F.C. Ros, J. Hassan, Synthesis and characterisation of AWO₄ (A= Mg, Zn) tungstate ceramics, *J. Phys. Conf. Ser.*, 1083 (2018) 012002. <https://doi.org/10.1088/1742-6596/1083/1/012002>
- [18] Y. Ouhassan, S. Bri, M. Habibi, Complex permittivity of FeSiAl/Al₂O₃ ceramic composite at X-band frequencies, In E3S Web of Conferences, 297 (2021) 01013. <https://doi.org/10.1051/e3sconf/202129701013>
- [19] K. Mabhouti, M. Karamirad, P. Norouzzadeh, Measurement of nickel doped zinc oxide NPs resonance frequencies and electromagnetic properties in X-Band, *Phys. B: Condens. Matter*, 602 (2021) 412532. <https://doi.org/10.1016/j.physb.2020.412532>
- [20] Z.H. Jiang, Y. Yuan, Microwave dielectric properties of SrAl₂Si₂O₈ filled polytetrafluoroethylene composites, *IOP Conf. Ser.: Mater. Sci. Eng.*, 479 (2019) 012090. <https://doi.org/10.1088/1757-899x/479/1/012090>
- [21] S.K. Yoon, H.G. Lee, W.S. Lee, E.S. Kim, Dielectric properties of ZnNb₂O₆/epoxy composites, *J. Electroceram.*, 30 (2013) 93-97. <https://doi.org/10.1007/s10832-012-9737-0>
- [22] S. Dash, R.N.P. Choudhary, A. Kumar, M.N. Goswami, Enhanced dielectric properties and theoretical modeling of PVDF-ceramic composites, *J. Mater. Sci. Mater. Electron.*, 30 (2019) 19309-19318. <https://doi.org/10.1007/s10854-019-02291-z>
- [23] N. Hasan, N.S.M. Hussain, A.A.M. Faudzi, Cured epoxy resin dielectric characterization based on accurate waveguide technique, *AIP Conf. Proc.*, 2129 (2019) 020080. <https://doi.org/10.1063/1.5118088>
- [24] S.M. Seng, K.Y. You, F. Esa, M.Z.H. Mayzan, Dielectric and Magnetic Properties of Epoxy with Dispersed Iron Phosphate Glass Particles by Microwave Measurement, *J. Microw. Optoelectron. Electromagn. Appl.*, 19 (2020) 165-176. <https://doi.org/10.1590/2179-10742020v19i2824>