Improvement of Microwave Dielectric Properties by Impact of Sodium Ion Doping in Complex Oxides for Polymer Composites

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The research presented in this study focuses on the dielectric properties of complex oxide-filled epoxy, polyurethane, and silicone rubber composites. Specifically, we investigated various compositions of (1-x) $CaWO_4$-xNa_2WO_4 (x = 0, 0.2, 0.4), in addition to $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$ and $Bi_2Mo_{0.5}W_{0.5}O_6$. These complex oxide materials were meticulously synthesized through a solid-state reaction, and their distinct phases were confirmed via rigorous X-ray diffraction (XRD) characterization .We then proceeded to create the composites by manually blending these complex oxides with epoxy, polyurethane, and silicone rubber matrices. The central focus of the study was to examine the impact of varying volume fractions of the complex oxide fillers on the dielectric properties of the composites in the frequency range of 4 – 8 GHz.Our observations revealed a direct correlation between the content of the complex oxide filler and the dielectric properties, including the dielectric constant and dielectric loss. Specifically, the addition of 10 % by volume fraction of (1-x) $CaWO_4$-xNa_2WO_4 (x = 0, 0.4), $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$, or $Bi_2Mo_{0.5}W_{0.5}O_6$ fillers to the respective polymer matrices led to a significant enhancement in both the dielectric constant and the loss tangent .Promising results were particularly evident in three composite formulations: the 10 % 0.4Na_2WO_4-0.6$CaWO_4$/silicone rubber composite demonstrated a dielectric constant (ε_r) of 2.02×10^2 and a loss tangent (tanδ) of -4.07×10^{-1} at 7.1 GHz; the 10 % $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$/epoxy composite exhibited a dielectric constant of 1.37×10^2 and a loss tangent of -6.43×10^{-1} at 7.22 GHz; and the 10 % $Bi_2W_{0.5}Mo_{0.5}O_6$/polyurethane composite displayed favorable properties with a dielectric constant (єr) of $8.37x10^1$ and a loss tangent (tanδ) of -3.4×10^{-1} at 7.12 GHz.These results suggest the suitability of these composite materials for a wide range of microwave technology applications, including wireless communication, radar systems, and various microwave devices.

Keywords: complex oxides, solid-state reaction method, dielectric properties, composites, wireless communication.

1. INTRODUCTION

Researchers have shown significant interest in dielectric materials with high performance and low cost [1 – 3]. Polymer-based dielectric composites have attracted significant interest among these materials due to their additional advantages, such as small elastic modulus and ease of processing [4 – 6]. In this regard, one common method to harness the benefits of both high dielectric constant ceramics and low dielectric loss polymers is by blending ceramic fillers into a polymer matrix, resulting in ceramic/polymer composites [7 – 9]. Microwave dielectric materials have a vital role in diverse applications, including wireless communication, radar systems, and microwave devices [10 – 12]. In this study, a specific class of microwave dielectric materials has gained attention, namely (1-x) $CaWO_4$-xNa_2WO_4 (where x = 0, 0.2, and 0.4) reinforced with epoxy, polyurethane, or silicone rubber. Additionally, another material of interest is $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$ or $Bi_2Mo_{0.5}W_{0.5}O_6$ reinforced with epoxy, polyurethane, or silicone rubber. These composite materials are formed by incorporating the mentioned ceramic powders, such as $CaWO_4$, 0.8$CaWO_4$-0.2Na_2WO_4, 0.6$CaWO_4$-0.4Na_2WO_4, $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$, or $Bi_2Mo_{0.5}W_{0.5}O_6$, into a polymer matrix consisting of epoxy, polyurethane, or silicone rubber. Several factors affect the fundamental characteristics of composite materials, comprising relative permittivity, thermal conductivity, and coefficient of thermal expansion. These influential factors encompass the number of components or phases, the filler's volume fraction, the inherent properties of each phase, the fabrication technique, and the interactions occurring between the filler and the matrix [13, 14].

The purpose of this reinforcement is to combine the desirable dielectric properties of the ceramics with the mechanical flexibility and processability of the polymers. In this context, by varying the composition of the ceramic powders and the polymer matrix, the dielectric properties of these composites can be tailored to meet specific requirements. The choice of epoxy, polyurethane, or silicone rubber as the polymer matrix allows for versatility in terms of mechanical properties and processing techniques. These microwave dielectric composites offer advantages such as a high dielectric constant, a low loss factor, and an improved mechanical strength compared to those of pure ceramics. These properties make them suitable for applications in microwave technology, where efficient energy storage and transmission are essential. In this regard, extensive research has been conducted on the microwave characteristics of $CaWO_4$. Accordingly, it has been found that $CaWO_4$ can be produced with a low ε_r of 9.7 while maintaining a high-quality factor (Q_f) of 44,200 GHz [15].

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Additionally, by doping $CaWO_4$ with 0.5 wt.% Bi_2O_3 and 9 wt.% HBO_3, it was possible to sinter the material at a lower temperature of 850 °C, resulting in an enhancement Q_f of 70,000 GHz [16]. As another material, $CaMoO_4$, which also has a scheelite structure, exhibited a Q_f of 55,000 GHz and εr of 11.7 when subjected to sintering at 1100 °C with a slight excess of Mo [17]. Particularly, Na_2WO_4 ceramic was successfully synthesized using the reactive sintering procedure. The study investigated its composition of phase, microwave dielectric properties, and microstructure through X-ray diffraction, a vector network analyzer, and scanning electron microscopy. Based on the results, the ceramic exhibited good microwave dielectric properties with a ε_r of approximately 3.45, a quality factor (Q_f) of around 38,244 GHz (at 13.64 GHz), and a temperature coefficient of the resonant frequency of about 42 ppm/K [18].

In this study, the researchers investigated the sintering behaviors and the structures of (1-x) $CaWO_4$-x$YLiF_4$ ceramic with varying compositions of ($0.02 \leq x \leq 0.10$). The resulting ceramic with a composition of $x = 0.04$ exhibited optimized microwave dielectric characteristics, including a relative permittivity of 10.5, a quality factor (Q_f) of 73,000 GHz, and a temperature coefficient of the frequency of 37.7 ppm/°C [19]. In particular, the researchers prepared $(Li_{0.5}Bi_{0.5})(W_{1-x}Mo_x)O_4$ ceramics using the solid-state reaction technique. Consequently, at $x = 0.3$, a wolframite solid solution was formed, while for $x = 0.4$ and $x = 0.6$, both wolframite and scheelite phases were observed in the XRD analysis. The $(Li_{0.5}Bi_{0.5})$-$(W_{0.6}Mo_{0.4})O_4$ ceramic exhibited excellent microwave dielectric properties with a dielectric constant of 31.5, a Q_f value of 8500 GHz (at 8.2 GHz), and a temperature coefficient value of +20 ppm/°C [20].

As far as we are know, there has been no previous investigation or research into dielectric properties of 0.6$CaWO_4$-0.4Na_2WO_4, and $Bi_2Mo_{0.5}W_{0.5}O_6$ powders or its utilization within 0.6$CaWO_4$-0.4Na_2WO_4, or $Bi_2Mo_{0.5}W_{0.5}O_6$ polymer composites. Also, use $CaWO_4$, 0.8$CaWO_4$-0.2Na_2WO_4, and $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$ as fillers in polymer composites.

This paper marks a significant stride in the field of dielectric materials tailored for wireless communication systems. It adeptly addresses crucial voids in current literature, with a distinct focus on bolstering dielectric characteristics, highlighting cost-effectiveness, and furnishing an in-depth, frequency-specific scrutiny of the C-band spectrum. Through meticulous fine-tuning of composite attributes and a resounding emphasis on the demand for pragmatic and budget-friendly solutions, this study delivers invaluable insights and concrete contributions to the wireless telecommunications domain. This research charts a course for the creation of dependable and economically viable materials, poised to propel innovation and advancements in wireless communication technologies.

The primary goal of this research was to develop cost-effective microwave materials with optimal properties. More specifically, (1-x) $CaWO_4$-xNa_2WO_4 (x = 0, 0.2, and 0.4),$(Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4)or(Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4)$/polymer composites were created through a simple hand-mixing method, and the X-ray diffraction was employed to assess the phase composition of complex oxides, while the dielectric characteristics of composites were investigated within a frequency range of 4 – 8 GHz. The study focused on understanding the effect of Na ions on the structure and the microwave dielectric characteristics. Moreover, the study delved into the influence of the filler and the concentration within the composites.

2. EXPERIMENTAL PROCEDURE

In this work, ceramic-polymer composite materials were synthesized by mixing the polymer (epoxy, polyurethane, or silicone rubber)(made in China) based on the desired composite volume with CW, 0.8CW-0.2NW, 0.6NW-0.4NW, NBMW, or BMW (5 % or 10 %) volume fraction. Particularly, the (1-x) $CaWO_4$-xNa_2WO_4 (x = 0, 0.2, and 0.4) ceramics were synthesized using the solid-state reaction technique. Firstly, WO_3 (99.9 %), Na_2CO_3 (99 %), and CaO(99.9 %)powders were weighed in stoichiometric proportions and then milled for four hours in an electrical blender. Subsequently, the mixtures were calcined at 650 °C for four hours. The powders after calcination were finely ground for four hours. The appropriate amount of Na_2CO_3, Bi_2O_3 (99.5 %), MoO_3 (99.99 %), and WO_3 powders were accurately weighed according to the stoichiometric ratio required for $(Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4)$ complex oxide. Similarly, the appropriate amounts of Bi_2O_3, MoO_3, and WO_3 powders were weighed and then mixed for three hours for the synthesis of $(Bi_2W_{0.5}Mo_{0.5}O_6)$ ceramic. The mixtures were calcined at 650 °C for two hours. Then, the calcined powders were re-milled for three hours. After that, the compositions of oxides were analyzed using the XRD analysis with CuKα. In the sequel, the microwave dielectric characteristics of the composites were determined using the Transmission/Reflection line method within the frequency range of 4 – 8 GHz. This method entails measuring both the reflected signal (S_{11}) and the transmitted signal (S_{21}). For these measurements, a vector network analyzer was utilized. In addition, the scattering parameters, S_{11} and S_{21}, were then transformed into dielectric properties utilizing the mathematical Nicholson-Ross-Weir (NRW) method [21 – 23].

3. RESULTS AND DISCUSSION

The X-ray diffraction pattern displayed in Fig. 1 a for (1-x) CW-xNW (x = 0, 0.2, 0.4) complex oxides exhibits a strong agreement with the findings previously reported by Tao Sun et al. [24]. Moreover, all the observed peaks can be accurately matched with the standard PDF card [96-721-9230 and 96-591-0225], which confirms the successful formation of two different phases of $CaWO_4$ and Na_2WO_4. More precisely, the $CaWO_4$ phase adopts a tetragonal scheelite structure with the I41/a space group. The corresponding cell parameters are determined as follows: a = b = 5.243 Å, c = 11.373 Å, and V = 312.12 $Å^3$. On the other hand, the Na_2WO_4 phase adopts a cubic spinel structure with the Fd-3m space group. The parameters of the cell for this phase are a = b = c = 9.130 Å and V = 761.0 $Å^3$. The XRD patterns of $(Na_{0.5}Bi_{0.5})$ $(Mo_{0.5}W_{0.5})$ (x = 0.5) and $Bi_2Mo_{0.5}W_{0.5}O_6$ complex oxides are presented in Fig. 1 b

and c, respectively. All the peaks are identifiable based on standard PDF cards (96-200-0339 and 96-156-7272), respectively. For $(Na_{0.5}Bi_{0.5})(Mo_{(1-x)}W_x)$ (x = 0, 0.5) with tetragonal scheelite structure with the I41/a space group, parameter of the cell are a = b = 5.27850 Å, c = 11.64100 Å, and V = 324.34 Å^3. The $Bi_2Mo_{0.5}W_{0.5}O_6$ includes an orthorhombic structure (double perovskite) (P c a 21(29) space group), and the parameters of the cell are a = 5.47180, b = 16.32530, c = 5.46160, V = 470.64 Å^3, a = 5.48220 A°, b = 16.19860 A°, c = 5.50910 A°, and V = 488.3 Å^3.

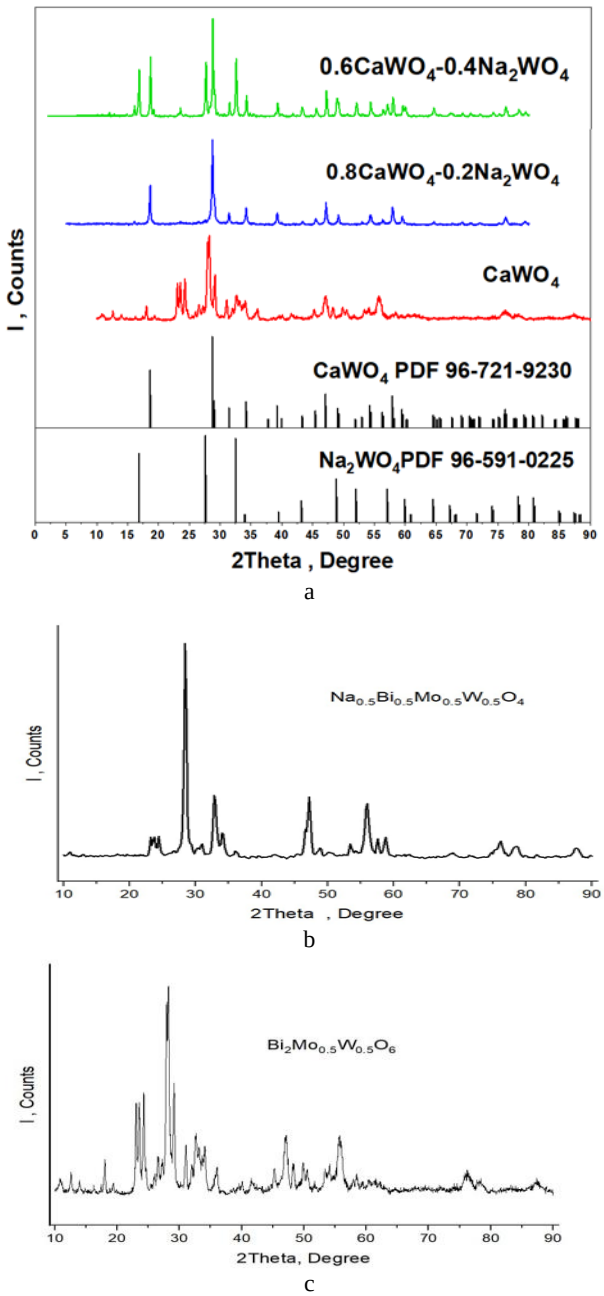

Fig. 1. a – the X-ray diffraction patterns of (1-x) CW-xNW (x = 0, 0.2, 0.4) ceramics; b – the XRD pattern of $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$ ceramic; c – the XRD pattern of $Bi_2W_{0.5}Mo_{0.5}O_6$ ceramic

In this regard, the high dielectric constant of a material signifies its capacity to store electrical energy when an electric field is applied. In the case of the mixed compound xNa_2WO_4-1-$xCaWO_4$ (x = 0.2, 0.4) compared to $CaWO_4$, the higher dielectric constant at 6.5 – 7.5 GHz can be attributed to the presence of Na ions in the complex oxides, as shown in Fig. 2, Fig. 3 and Fig. 4. To this end, the dielectric constant of a material depends on its electronic and atomic structures. When an electric field is applied, the electric dipoles in the material tend to align with the field, which results in the storage of electrical energy. The ability of a material to align these dipoles depends on factors such as the magnitude and the mobility of the charge carriers' ions or the present electrons. In the case of $0.2Na_2WO_4$-$0.8CaWO_4$, the introduction of sodium ions in the complex oxide leads to an increase in the number of charge carriers compared to pure $CaWO_4$. On the other hand, sodium ions have a smaller atomic size compared to Ca ions, and they have higher mobility due to their lower charge density. These factors contribute to a higher dielectric constant in the mixed compound. Additionally, the presence of different types of ions in a compound can induce polarization effects. The sodium and calcium ions in the mixed compound may exhibit different polarizability, leading to enhanced dipole moments and an increased dielectric constant compared to those of pure $CaWO_4$.

The percentage of the filler material in the composite plays a crucial role in determining the dielectric constant as shown in Fig. 2 – Fig. 7. In this case, the 10 % composition contains higher filler content than that of the 5 % composition. As the filler content increases, there is a higher chance for the filler particles to come into contact with each other, creating a continuous conductive network. This conductive network enhances the overall dielectric constant of the composite. In this regard, the interface between the polymer matrix and the filler material can contribute to the dielectric properties of the composite. When an electric field is applied, charges can accumulate at the interface, resulting in interfacial polarization. This polarization effect increases the overall dielectric constant of the composite. Moreover, the presence of higher filler content in the 10 % composition provides more opportunities for interfacial polarization, leading to a higher dielectric constant [25 – 27].

Fig. 5, Fig. 6 and Fig. 7 show that the high dielectric constant of $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$ compared to that of $Bi_2W_{0.5}Mo_{0.5}O_6$ can be attributed to several factors, including the presence of different elements and the crystal structures. Particularly, $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$ contains Na, Bi, Mo, and W, while $Bi_2W_{0.5}Mo_{0.5}O_6$ consists of only Bi, Mo, and W. In fact, the inclusion of Na in $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$ may contribute to its higher dielectric constant, as different elements can interact differently with electric fields, leading to enhanced polarization effects. The crystal structure plays a crucial role in determining the dielectric properties of a material. $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$ belongs to the scheelite family, which is known for its high dielectric constant. On the other hand, $Bi_2W_{0.5}Mo_{0.5}O_6$ is a complex perovskite. The distinct crystal structures of these compounds can result in variations in their dielectric responses.

In particular, the best result was for 10 % $0.4Na_2WO_4$-$0.6CaWO_4$/silicone rubber composite, 10 % $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$/epoxy, and 10 % $Bi_2W_{0.5}Mo_{0.5}O_6$/polyurethane due to the compatibility between the matrix and the filler. The compatibility between

the filler material and the polymer matrix is crucial for achieving a high dielectric constant. If the filler and the polymer have a good affinity for each other, this promotes better dispersion of the filler particles within the polymer matrix, and this uniform dispersion enhances the overall dielectric properties of the composite.

The lower loss tangent of $0.4Na_2WO_4$-$0.6CaWO_4$ compared to $CaWO_4$ and $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$ compared to $Bi_2W_{0.5}Mo_{0.5}O_6$, as shown in Fig. 8 – Fig. 13, can be attributed to the presence of Na and the combined effect of the composition and the crystal structure. Consequently, the inclusion of Na in the compound can alter its dielectric properties. In this regard, Na ions have higher mobility, which can lead to reduced losses due to less friction and energy dissipation during the polarization process. Both $0.4Na_2WO_4$-$0.6CaWO_4$ and $CaWO_4$ have a scheelite-type crystal structure. However, the presence of Na in $0.4Na_2WO_4$-$0.6CaWO_4$ may induce structural changes or lattice distortions that can affect the polarization behavior and reduce the tangent loss. The distinct crystal structures of $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$ and $Bi_2W_{0.5}Mo_{0.5}O_6$ can result in variations in their dielectric responses. Furthermore, the specific arrangement of atoms and ions in the crystal lattice can affect the polarization behavior and reduce the tangent loss. The positive tan δ (dielectric loss) value is attributed to normal relaxation processes. However, the occurrence of a negative value is unconventional. Axelrod et al. demonstrated that negative tan δ is a result of the sample releasing more energy than it absorbs [28]. The findings are succinctly presented in Table 1.

In comparison to the unfilled polymer counterparts, as shown in Fig. 2 – Fig. 13, there is a substantial enhancement in the dielectric properties of the composites due to different things. Ceramics often have a high intrinsic dielectric constant because they are insulators with a large number of electric dipoles. When these ceramics are incorporated into a polymer matrix, they introduce additional polarizability to the composite. This leads to an increased dielectric constant because polarization is a key factor in determining the dielectric behavior of materials.

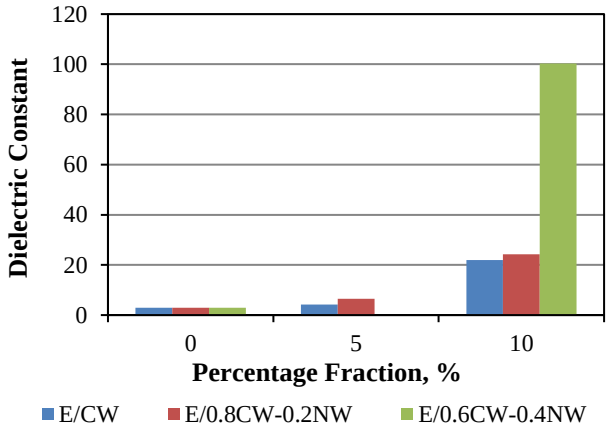

Fig. 2. The dielectric constant calculated in the frequency range of 7 – 7.98 GHz for $CaWO_4$/epoxy composite, $0.8CaWO_4$-$0.2Na_2WO_4$ /epoxy composite and $0.6CaWO_4$-$0.4Na_2WO_4$ /epoxy composite as a function of volume fraction of fillers

At the interface between ceramic particles and the polymer matrix, there can be localized electric fields due to differences in permittivity. This interfacial polarization effect can significantly contribute to an enhanced dielectric constant .The presence of two or more materials with significantly different dielectric constants in a composite can lead to the MWS effect (Maxwell-Wagner-Sillars). This effect arises from charge accumulation at interfaces and contributes to higher dielectric constants in composites [29, 30].

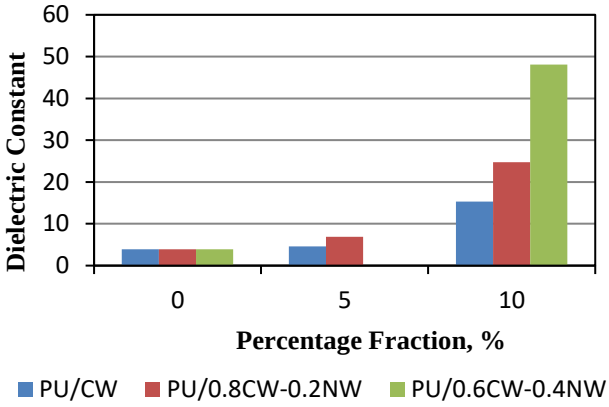

Fig. 3. The dielectric constant calculated in the frequency range of 7 – 7.98 GHz for $CaWO_4$/polyurethane composite, $0.8CaWO_4$-$0.2Na_2WO_4$ /polyurethane composite and $0.6CaWO_4$-$0.4Na_2WO_4$/ polyurethane composite as a function of volume fraction of fillers

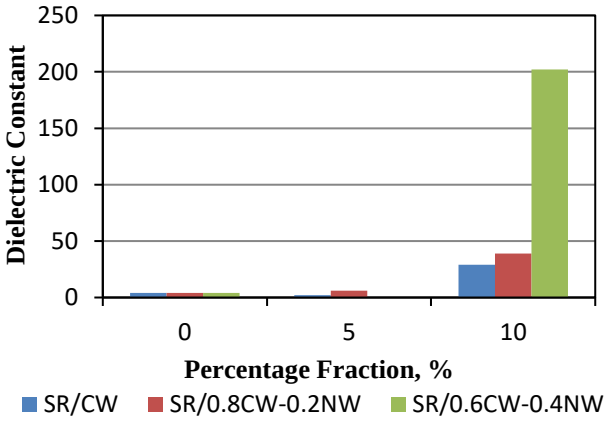

Fig. 4. The dielectric constant calculated in the frequency range of 7 – 7.98 GHz for $CaWO_4$/ silicone rubber composite, $0.8CaWO_4$-$0.2Na_2WO_4$ /silicone rubber composite and $0.6CaWO_4$-$0.4Na_2WO_4$/ silicone rubber composite as a function of volume fraction of fillers

Based on the previous discussion, the present research indicates that a variety of variables affect the capacity of materials to conduct electricity, such as frequency, polymer matrix type, and complex oxide. As a result, selecting an appropriate frequency range and composite material when building electrical components or devices is vital for assuring the optimal performance for a variety of applications.

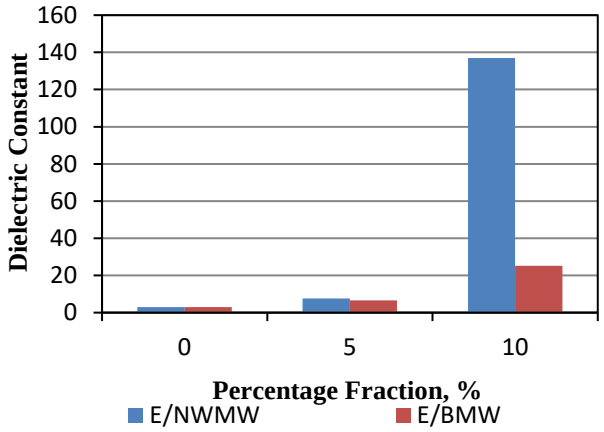

Fig. 5. The dielectric constant calculated in the frequency range of 7 – 7.98 GHz for $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$/epoxy composite, and $Bi_2Mo_{0.5}W_{0.5}O_6$/epoxy composite as a function of volume fraction of fillers

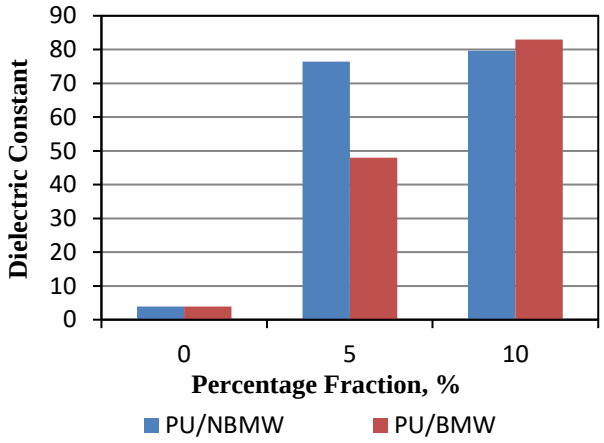

Fig. 6. The dielectric constant calculated in the frequency range of 7 – 7.98 GHz for $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$/polyurethane composite, and $Bi_2Mo_{0.5}W_{0.5}O_6$/polyurethane composite as a function of volume fraction of fillers

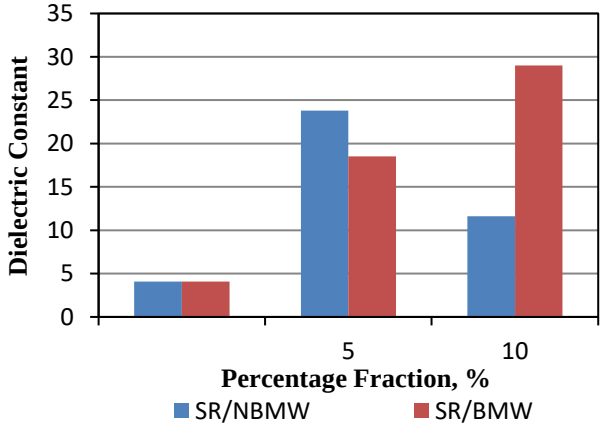

Fig. 7. The dielectric constant calculated in the frequency range of 7 – 7.98 GHz for $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$/silicone rubber composite, and $Bi_2Mo_{0.5}W_{0.5}O_6$/silicone rubber composite as a function of volume fraction of fillers

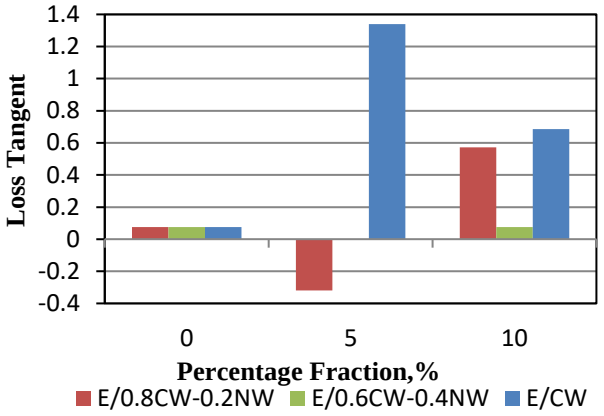

Fig. 8. The loss tangent calculated in the frequency range of 7 – 7.98 GHz for $CaWO_4$/epoxy composite, 0.8$CaWO_4$-0.2Na_2WO_4/epoxy composite and 0.6$CaWO_4$-0.4Na_2WO_4/epoxy composite as a function of volume fraction of fillers

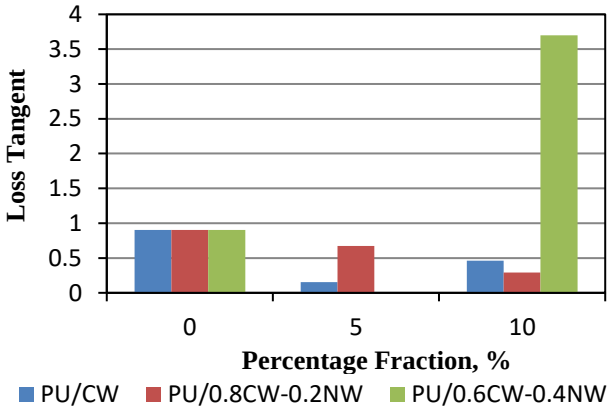

Fig. 9. The loss tangent calculated in the frequency range of 7 – 7.98 GHz for $CaWO_4$/polyurethane composite, 0.8$CaWO_4$-0.2Na_2WO_4/ polyurethane composite and 0.6$CaWO_4$-0.4Na_2WO_4/ polyurethane composite as a function of volume fraction of fillers

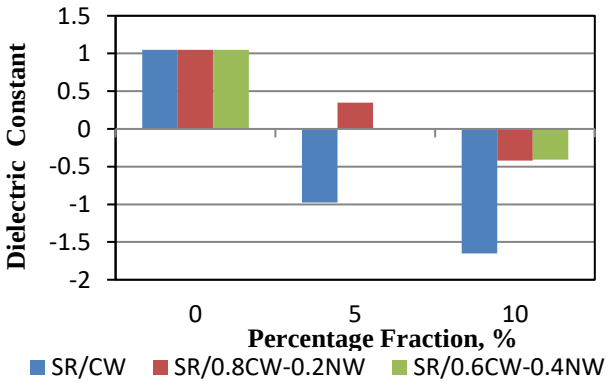

Fig. 10. The loss tangent calculated in the frequency range of 7 – 7.98 GHz for $CaWO_4$/silicone rubber composite, 0.8$CaWO_4$-0.2Na_2WO_4/ silicone rubber composite and 0.6$CaWO_4$-0.4Na_2WO_4/ silicone rubber composite as a function of volume fraction of fillers

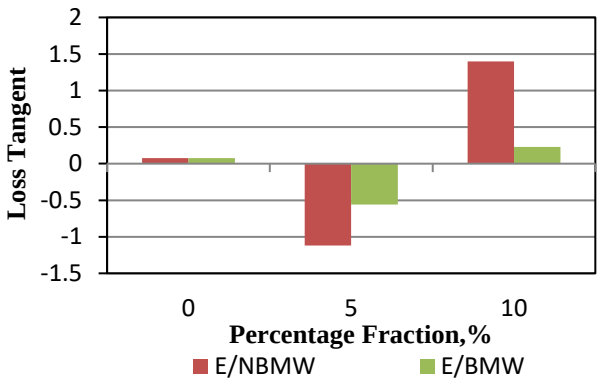

Fig. 11. The loss tangent calculated in the frequency range of 7 – 7.98 GHz for $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$/epoxy composite, and $Bi_2Mo_{0.5}W_{0.5}O_6$/epoxy composite as a function of volume fraction of fillers

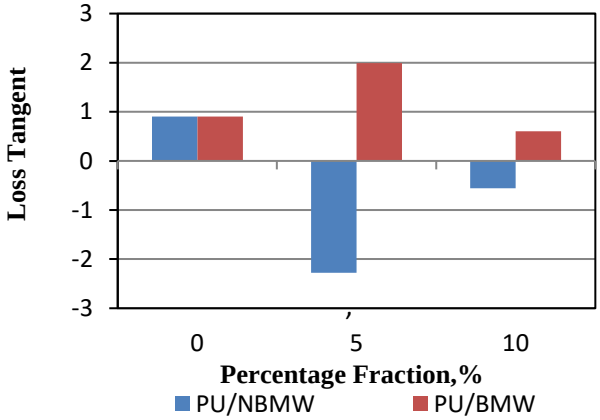

Fig. 12. The loss tangent calculated in the frequency range of 7 – 7.98 GHz for $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$/polyurethane composite, and $Bi_2Mo_{0.5}W_{0.5}O_6$/ polyurethane composite as a function of volume fraction of fillers

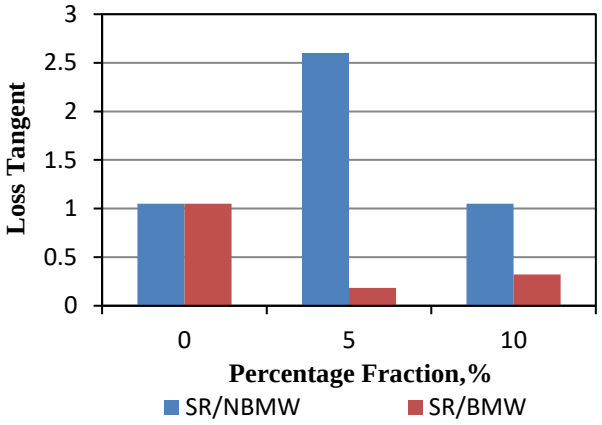

Fig. 13. The loss tangent calculated in the frequency range of 7 – 7.98 GHz for $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$/silicone rubber composite, and $Bi_2Mo_{0.5}W_{0.5}O_6$/silicone rubber composite as a function of volume fraction of fillers

Table 1. Dielectric constant and loss tangent of composites

Polymer-ceramic	Frequency, GHz		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/$CaWO_4$	7.42	4.82 5.44 6.92 4.74 7.56	4.14	11.2 20.6 61.2 16.1 21.9	1.34	0.412 -0.0136 -0.17 1.2 -0.519
Polyurethane/$CaWO_4$	7.54	4.74 7.58	3.56	16.1 15.3	0.154	1.2 0.46
Silicone Rubber/$CaWO_4$	5.72 7.12	4.82 5.42 6.24 7.1 7.4	5.91 1.99	16.4 15.9 12.9 29.1 14.5	-0.987 -0.945	-0.759 0.686 -0.491 -1.65 2.16
Epoxy/0.8$CaWO_4$-0.2Na_2WO_4	7.8	4.84 5.5 6.78 7.7	6.48	22.1 13.5 24.2 15.1	-0.319	-0.759 0.686 -0.491
Polyurethane/0.8$CaWO_4$-0.2Na_2WO_4	4.42 6.4 7.78	4.88 5.54 6.88 7.72 7.78	56.04 18.6 6.87	15.8 9.01 30.9 21.9 24.7	0.674 0.048 -0.221	-0.942 3.2 -0.336 1.13 0.294

continued on next page

continuation of Table 1

Polymer-ceramic	Frequency, GHz		Dielectric constant as a function of filler volume fraction, %		Loss tangent as a function of filler volume fraction, %	
	5 %	10 %	5 %	10 %	5 %	10 %
Silicone Rubber/0.8$CaWO_4$-0.2Na_2WO_4	5.5 7.04 7.18 7.96	4.8 5.56 6.84 6.98 7.72	6.67 6.35 6.05 5.15	15.8 8.2 28.1 39.1 7.2	0.00413 0.24 0.349 0.0228	0.774 -1.15 1.95 -0.42 3.3
Epoxy/0.6$CaWO_4$-0.4Na_2WO_4	5.72 7.1 6.4		18.4 100.3 15.9		-0.33 0.435 -0.0936	
Polyurethane/0.6$CaWO_4$-0.4Na_2WO_4	7.22 7.4 5.72		19.7 48.1 13.9		1.67 3.7 -0.137	
Silicone Rubber/0.6$CaWO_4$-0.4Na_2WO_4	7.1		202		-0.407	
Epoxy/$Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$	4.4 5.14 5.76 6.44 7.24	6.3 7.22 7.26 7.32 7.44	25.4 5.18 6 6.14 7.71	16.1 137 62.9 47 34.6	0.0899 1.2 -0.176 -0.171 -1.12	4.38 1.45 1.42 1.4 1.39
Polyurethane/$Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$	7 6.2 6.38 7.86	5.72 6.32 7.12	76.43 11.43 12.3 1187	13.25 13.7 79.64	-2.28 -0.236 -0.905 -0.469	-4.39 1.49 -0.556
Silicone Rubber/$Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$	6.4 7.02 7.54	4.84 5.5 6.78 7.7	20 23.8 12.7	22.8 11 30 11.6	1.85 0.183	1.33 1.11 -0.139 1.05
Epoxy/$Bi_2Mo_{0.5}W_{0.5}O_6$	5.24 6.76 7.66 7 6.2 6.38 7.86	5.88 7.14 7.24 7.5	7.89 8 6.64 76.43 11.43 12.3 11.87	8.01 12.5 25.1 17	0.183 0.148 0.0574 -0.561 -1.48 -1.23 -9.92	-0.39 0.6 0.229 -0.531
Polyurethane/$Bi_2Mo_{0.5}W_{0.5}O_6$	7.4 6.4	5.76 7.12 7.7	48.1 15.9	9.94 83.7 27.6	1.99 -0.942	1.84 0.997 0.602
Silicone Rubber/ $Bi_2Mo_{0.5}W_{0.5}O_6$	6.34 7.54	4.84 6.8 7.7	102 18.5	19.3 150 29	0.0824 0.183	-0.408 0.00318 0.322

4. CONCLUSIONS

In this study, different composites were created by mixing complex oxides with epoxy, polyurethane, and silicone rubber as a matrix. The composites included (1-x$CaWO_4$-xNa_2WO_4 (x = 0, 0.2, 0.4)), ($Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$), or ($Bi_2W_{0.5}Mo_{0.5}O_6$). A microwave dielectric ceramic was synthesized using a solid-state reaction method. The XRD profiles confirmed the composition of the complex oxides. Moreover, the complex permittivity of the composites was measured using a vector network analyzer, and the S-parameters were converted to relative permittivity using the Nicholson-Ross-Weir (NRW) method. In particular, the presence of Na in the composites increased their microwave dielectric properties. In addition, the permittivity of the composites was increased when the filler concentration reached a 10 % volume fraction. Specifically, at 7 – 7.5 GHz, the composites 10 % 0.4Na_2WO_4-0.6$CaWO_4$/silicone rubber, 10 % $Na_{0.5}Bi_{0.5}Mo_{0.5}W_{0.5}O_4$/epoxy, and 10 % $Bi_2W_{0.5}Mo_{0.5}O_6$/polyurethane exhibited suitable dielectric properties.

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