

RESEARCH ARTICLE

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STUDY OF THE STRUCTURE AND ELECTRIC PROPERTIES OF (NA_{0.5}BI_{0.5})_{1-x}MGX[(TI_{0.8}ZR_{0.2})_{0.9}(NB_{2/3}ZN_{1/3})_{0.1}] O₃ CERAMICS

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ABSTRACT

This study focuses on the compound (Na_{0.5}Bi_{0.5})_xMg_{1-x}[(Ti_{0.8}Zr_{0.2})_{0.9}(Nb_{2/3}Zn_{1/3})_{0.1}]O₃ (BNTZZN-x%Mg), which was synthesized using the molten salt method for x = 0.00, 0.02, 0.06, and 0.08. The resulting samples were calcined at 900 °C for 4 hours and subsequently sintered at 1150 °C. The measured densities were found to be close to the theoretical values. X-ray diffraction (XRD) analysis confirmed the formation of a pure perovskite phase. Scanning electron microscopy (SEM) images revealed a slight variation in grain size with increasing Mg content. Overall, the material exhibited relatively high purity and density. Dielectric properties were measured on the single-phase ceramic, and their evolution was studied over a temperature range of 27 °C to 227 °C across various frequencies.



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I. INTRODUCTION

Piezoelectric materials play a vital role in numerous technological applications, including actuators, sensors [1] [2], aerospace systems, precision machinery, as well as in both military and civilian sectors [3] [4]. Among these, lead-based piezoelectric ceramics, particularly Pb (Zr,Ti)O₃ (PZT), have been extensively studied and widely utilized [3]. However, a major drawback of PZT is its high lead content—up to 60% by weight [5] [6]. Lead is known to be toxic and poses significant risks to both human health and the environment. As a result, lead-free piezoelectric materials have gained significant attention as potential alternatives to lead-based ceramics [7]. In line with this shift, European regulations currently restrict the use of most lead-containing substances in electronic and electrical devices [6]. Despite ongoing efforts, no single lead-free composition has yet been identified that can fully replace PZT, making the search for suitable alternatives an active area of research [8]. In recent years, several promising lead-free materials have been developed for actuator applications [3]. Among these, materials based on Bi_{0.5}Na_{0.5}TiO₃ (BNT), first discovered by Smolensky in 1961 [9] [10], have attracted considerable interest. BNT crystallizes in a perovskite structure (ABO₃), exhibiting rhombohedral symmetry (space group R3c) at room temperature, which transitions to tetragonal and then cubic phases at elevated temperatures ranging from 300 °C to 540 °C [11] [12]. Currently, ceramics based on Na_{0.5}Bi_{0.5}TiO₃ (BNT) are gaining significant attention due to their environmental benefits and wide range of potential applications [13]. BNT-based solid solutions have attracted considerable interest, particularly for energy storage applications [14]. In recent years, various approaches involving ionic doping have been employed to synthesize BNT-based ceramics [3]. Numerous studies have aimed to enhance the functional properties of these materials and to reduce their sintering temperatures through binary or ternary solid solutions, or by incorporating sintering aids [15]. In this study, we synthesized the compound (Na_{0.5}Bi_{0.5})_xMg_{1-x}[(Ti_{0.8}Zr_{0.2})_{0.9}(Nb_{2/3}Zn_{1/3})_{0.1}] O₃ using the molten salt method. The resulting solid solution, BNTZZN-x%Mg, was structurally and dielectrically characterized with the aim of developing a lead-free piezoelectric material capable of effectively replacing PZT ceramics in specific applications.

II. EXPERIMENTAL

II.1 PREPARATION OF CERAMICS:

Each study has own preparation process. The molten salt method was selected (MSS) used to prepare solid solution BNTZZN-x%Mg such as x= 0.00, 0.02, 0.06 and 0.08. The molten salt method is used as a medium for the preparation of complex oxides from their constituent materials (oxides and carbonates) [16] The role of the molten salts is:

- Increase the reaction rate and lower the reaction temperature. Increase the degree of homogeneity (distribution of constituent elements in the solid solution).
- Particle size control.
- Particle shape control.
- Agglomeration state control [16].

The basic oxides used in the synthesis of our samples are MgO, TiO₂, ZrO₂, Bi₂O₃, Na₂CO₃, Nb₂O₅, and ZnO. The molten salt (MSS) method was used to prepare a solid solution (BNTZZN-x%Mg). In our study, BNT-type ceramics were synthesized from a mixture of oxides and carbonates. The oxides and carbonates required to synthesize a mixture of about 15g shall be weighed according to the stoichiometric quantities provided and calculated according to the reaction equation as shown in Table1, using an electronic balance. Then, the precursor was ground in a porcelain mortar for 03 hours. after that, we added (KCl-NaCl) and grind the sample for 02 hours. This operation aimed at promoting the homogeneity of the mixture to obtain powder with the finest particle size. The powder was burned at 900°C for 4h in a programmable oven (N-Abbertherm-D) under ambient temperature rise of 2°C/min to constant temperature of 4h. The resulting materials are cooled slowly. Then, we grind again for 5h. The aim of the regrinding is to reduce the agglomerates formed during calcination, in order to homogenize the powder, and increase its reactivity. After that, the powder obtained is compacted in the form of pellets of 1,5g and a diameter of about 13 mm. Our ceramics are sintered in an aluminum crucible at 1150°C by natural sintering under air. The temperature rises at a speed rate of 2°C/min up to the set temperature. Therefore, the temperature is stable for 4 hours. Finally, the temperature is descent to the ambient.

II.2 CHARACTERIZATION OF CERAMICS:

The spectra of our sintered ceramics were obtained at room temperature on a vertical diffractometer, «BRUKER-AXE type D8» from the University of Biskra laboratory. Profiles lines were measured using an automatic point-by-point counting system with a 0.02° steps. All diffraction patterns were recorded in the angle range 0° < 2θ < 80°, which may be sufficient to identify the different phases. We note that the analysis used for indexing different lines by X'Pert HighScore. The density of the sintered samples was calculated from the sample dimensions and weight. Microscopic images of our samples were performed using a scanning electron microscope type JEOL JSM-6390lv from the University of Biskra laboratory. It is controlled by a computer. The purpose of these analyses is to study the morphology of our samples after sintering. Dielectric measurements were carried out in the laboratory of physical engineers' the University of Tiaret. The measurement of the dielectric properties was performed on a single-phase ceramic sample, and the evolution of these properties were investigated at temperatures ranging from 27°C to 227°C at different frequencies. It was investigated using an automatic (LCR) counter combined with a computer controlled temperature chamber at 100Hz, 500Hz, 1KHz, 10KHz, and 100KHz.

III. RESULT AND DISCUSSION

III.1 DENSITY

Density is a physical quantity that affects the structure and electrical properties of ceramics. The relative density D (compactness) is given by equation (1) of the material NBMTZNZ sintered at 1150 °C:

As:

$$D = \frac{\rho_{\text{exp}}}{\rho_{\text{th}}} \quad (1)$$

ρ_{exp} : Experimental density calculated from the mass and the volume of sintered ceramics pellets.

ρ_{th} : Theoretical density obtained from X-ray diffraction database providing the mesh parameters [17].

Table 1 shows the density values. These values range from 5.26 to 5.53 g/Cm³ or from 88.58% to 89.77% so that the theoretical value is 6.12, 6.15 and 6.16g/Cm³. Thus, the experimental values are close to the theoretical values. This note means that the ceramic is very dense with less porosity [18] BNTZZN-x%Mg has been well prepared, so the choice of molten salt method was adequate.

Table1: Density (g/Cm³)

X	0.00	0.02	0.06	0.08
$\rho_{\text{exp}}(\text{g/Cm}^3)$	5.66	5.43	5.26	5.53
$\rho_{\text{th}}(\text{g/Cm}^3)$	6.12	6.13	6.15	6.16
D (%)	92.48	88.58	85.52	89.77

Source: Authors, (2026).

III.2 X-RAY ANALYSIS (DRX)

The majority of diffraction peaks observed in the synthesized samples were indexed based on the cubic perovskite structure of $\text{Na}_{0.5}\text{Bi}_{0.5}\text{TiO}_3$, using reference pattern 01-089-3109. Within the 2θ angle ranges of (21–22), (31–33), (39–41), (45–47), (51–53), (58–59), (68–69), and (76–78), distinct peaks corresponding to the perovskite phase were identified, associated with the following Miller indices: (100), (110), (111), (200), (210), (211), (220), (221), and (310). Minor secondary phases—specifically, traces of pyrochlore—were detected using the X'Pert HighScore software, and matched to the ASTM card 98-009-9437, as illustrated in Figure 1[13].

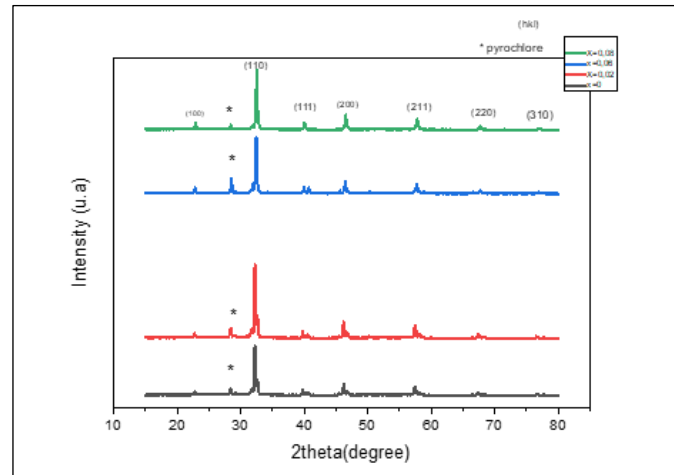


Figure 1: X-ray indexed diffraction patterns.

Source: Authors, (2026).

The stability of the perovskite structure (ABO_3) is characterized by a tolerance factor (t) that determines the density and freedom of movement of the cations A and B. According to Goldschmidt, the structure is stable if $0.88 < t < 1.09$, but exhibits a structural stability distortion beyond this range. The tolerance factor (t) can be determined by the following formula:

$$t = \frac{\sum R_a + R_o}{\sqrt{2(\sum R_b + R_o)}} \quad (2)$$

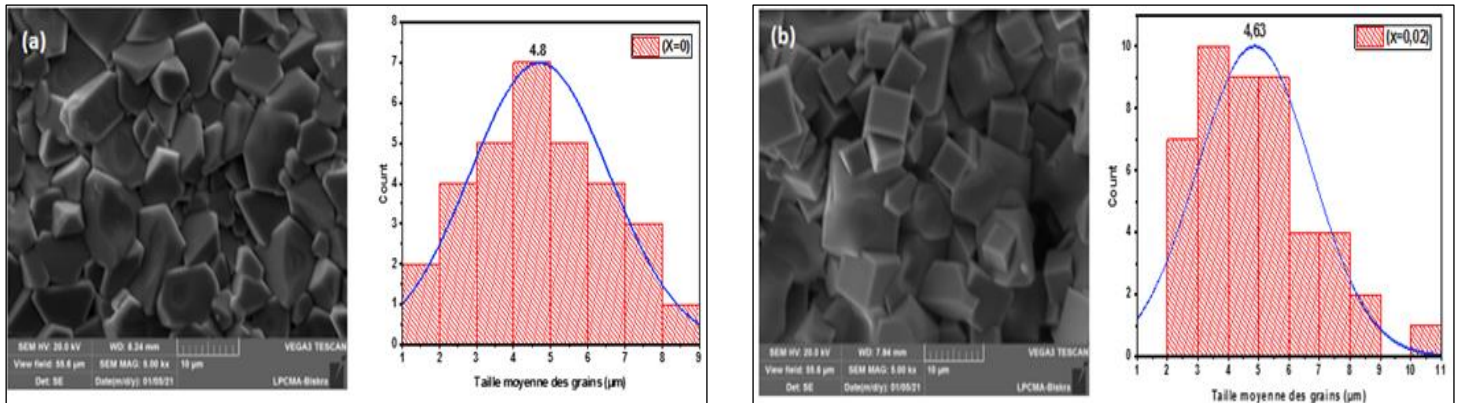
Where: $\sum R_a$, $\sum R_b$ and R_o are respectively the ionic radii of the cations, respectively; A, B and Oxygen [17-21] The perovskite structure is more stable when the tolerance factor (t) is close to 1. The involved bonds play a strong ionic role [14].

Table 2: factor tolerance values.

X	Tolerance factor
0.00	0.9225
0.02	0.9138
0.06	0.8964
0.08	0.8748

Source: Authors, (2026).

Figure .2 presents the SEM micrographs along with the histograms of the BNTZZN-x%Mg samples after sintering at 1150 °C. For each of the five samples, a uniform distribution of cubic-shaped grains is observed across the entire surface of the compositions. The ceramic micrographs reveal a satisfactory material density, very low intergranular porosity, and a narrow grain size distribution within each sample. The histograms obtained using ImageJ software [15] allowed the determination of an average grain size ranging from 3.07 μm to 4.80 μm for the four samples.



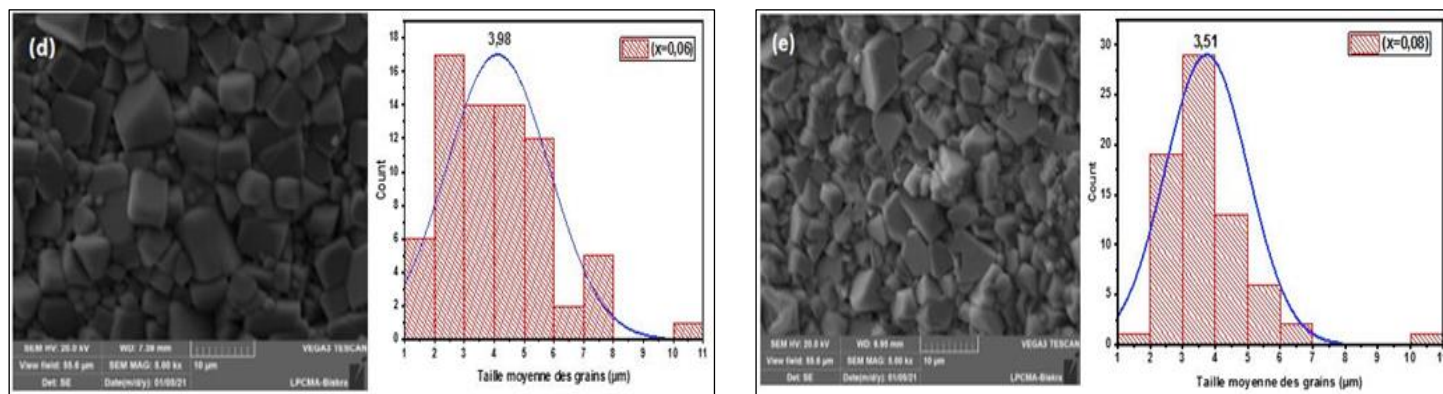


Figure 2: SEM images with histograms of the BNTZZN-x%Mg ceramic compositions showing the average grain size.

Source: Authors, (2026).

Table 3: Average grain size.

X	0.00	0.02	0.06	0.08
Grain size (μm)	4.80	4.630	3.984	3.514

Source: Authors, (2026).

III.3 DIELECTRIC MEASUREMENTS

Figure.3 illustrates the evolution of the dielectric constant of the BNTZZN-x%Mg material for various values of x (0, 0.02, 0.06, and 0.08) as a function of temperature (between 300 and 500 K), at different frequencies (100 Hz, 500 Hz, 1 kHz, 10 kHz, and 100 kHz). The $\epsilon_r(T)$ curves exhibit a similar trend, increasing with temperature until reaching a maximum, then decreasing. However, these curves differ in the position and intensity of their peak, which corresponds to the Curie temperature (T_C) [15] [16]. In the ferroelectric region, the dielectric constant ϵ_r increases with temperature, which means that the material possesses an electric dipole moment even in the absence of an external electric field [17]. The dielectric constant reaches a maximum at the Curie temperature, which corresponds to the transition of the material from the ferroelectric phase to the paraelectric phase. Beyond this temperature, the ceramics lose their ferroelectric properties and transition to a high-symmetry phase, leading to a decrease in dielectric permittivity [16] [18]. It is observed that the high values of dielectric permittivity for each composition appear at 100 Hz. The dielectric constant (ϵ_r) increases as the frequency decreases and becomes nearly negligible at high frequency values (100 kHz) for all compositions. The dielectric constant is frequency-dependent, particularly at low frequencies—a phenomenon known as low-frequency dielectric dispersion. A strong low-frequency dielectric dispersion has also been previously reported in the BNT material [19]. At temperatures above T_m and at low frequencies, an increase in dispersion is observed, caused by a rise in electrical conductivity. The dielectric peak broadens with increasing frequency, a phenomenon attributed to the structural disorder of the system. The real part of the dielectric constant reflects the loss less elastic response of the material to an applied alternating electric field. At temperatures above T_m and low frequencies, this dispersion becomes more pronounced due to the increased electrical conductivity.

The broadening of the dielectric peak with rising frequency further indicates structural disorder within the system [20]. Some authors explain the maximum value of the dielectric constant at room temperature and low frequencies by the presence of different types of polarization [15]. The polarization of dielectric ceramics varies significantly, mainly due to the different polarization mechanisms present in the various compositions. The polarization in dielectric ceramics primarily results from the displacement of electrons, ions, and chemical defects under the influence of an electric field, including electronic polarization, ionic polarization, orientational polarization, and space charge polarization. It can be classified into displacement polarization and relaxation polarization [21]. Moreover, ceramics with small, uniformly distributed grain sizes and a high density of grain boundaries generally exhibit higher dielectric breakdown strength (E_b) [22]. Factors affecting E_b also include temperature, humidity, the type of voltage (DC, AC, or pulse), test duration, and the thickness of the dielectric layer. In a specific experiment, E_b can be improved by increasing resistivity, reducing porosity, increasing density, and decreasing grain size. Dielectric loss refers to the energy consumed by dielectric materials due to heating under the influence of an electric field, and it is represented by $\tan \delta$. A higher $\tan \delta$ indicates not only increased energy consumption but also a rise in the temperature of the dielectric material. Therefore, a low $\tan \delta$ signifies better performance of the dielectric material and greater application value. In dielectric ceramics, $\tan \delta$ is generally attributed to conductivity loss, orientational and relaxation polarization loss, as well as structural loss [21]. Figure .4 illustrates the evolution of dielectric losses of the studied ceramic materials for various x values (0, 0.02, 0.04, 0.06, and 0.08) as a function of temperature (between 300 and 500 °C) and frequency (100 Hz, 500 Hz, 1 kHz, 10 kHz, and 100 kHz).

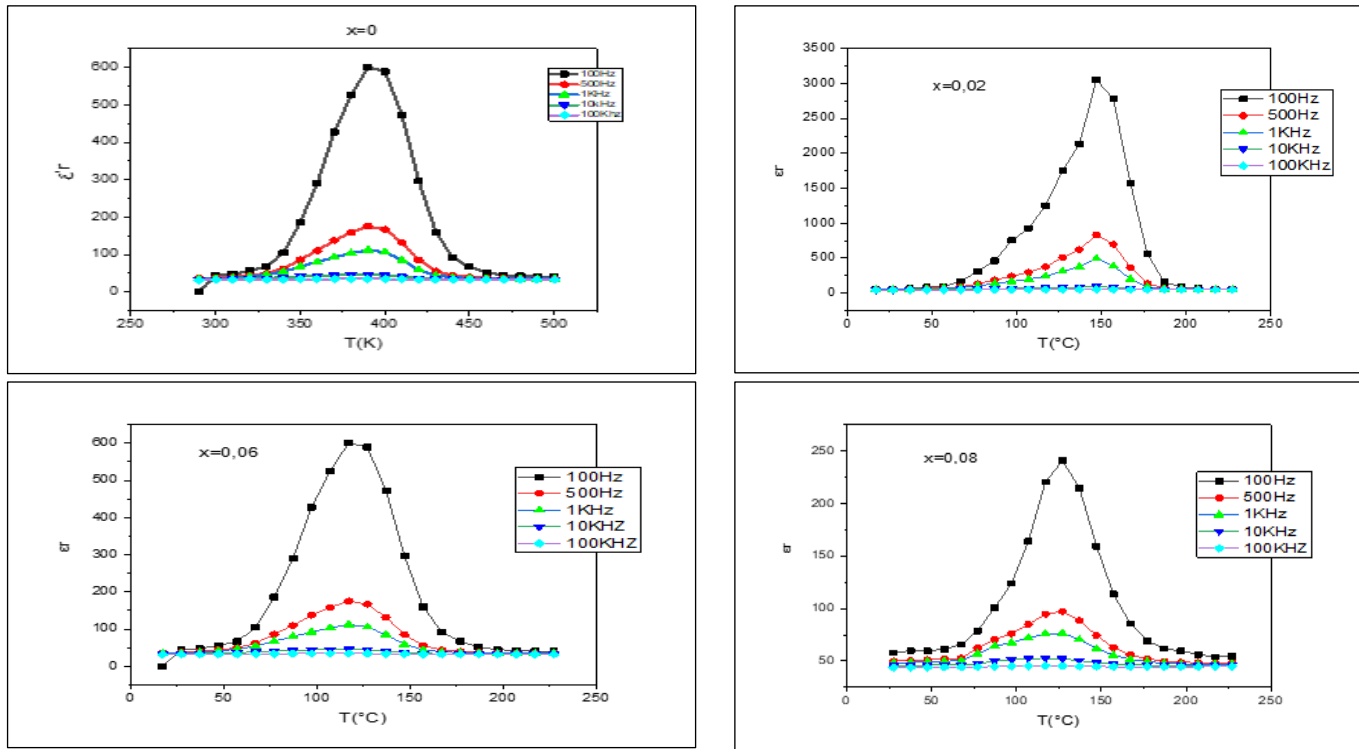


Figure 2: Evolution of the dielectric constant for the BNTZZN-x%Mg compositions as a function of temperature and frequency.
Source: Authors, (2026).

It is clearly observed that the loss angle increases with rising temperature, reaching a maximum at a certain point, resulting in a dielectric loss peak for the sample, followed by a decrease. This decrease, caused by the continued rise in temperature, deteriorates the material's properties related to domain wall motion [15]. The higher the permittivity, the greater the losses. The role of intrinsic losses in the overall loss process increases with the material's permittivity. For high-quality ferroelectrics, in the paraelectric state or near the transition, intrinsic losses may even dominate the total dielectric losses [17]. The analysis of the data in Table .4 reveals a non-monotonic influence of the magnesium content (X) on the tangent loss coefficient ($\tan \delta$) of the BNTZN-xMg material. Indeed, although a general decreasing trend in $\tan \delta$ is observed with increasing X, significant fluctuations appear, particularly at low X values. This complexity suggests that the mechanisms responsible for dielectric losses are sensitive to compositional variations, even at low magnesium concentrations. Given that all other elements are kept constant, it is reasonable to attribute these variations primarily to the effect of magnesium doping [23] [24].

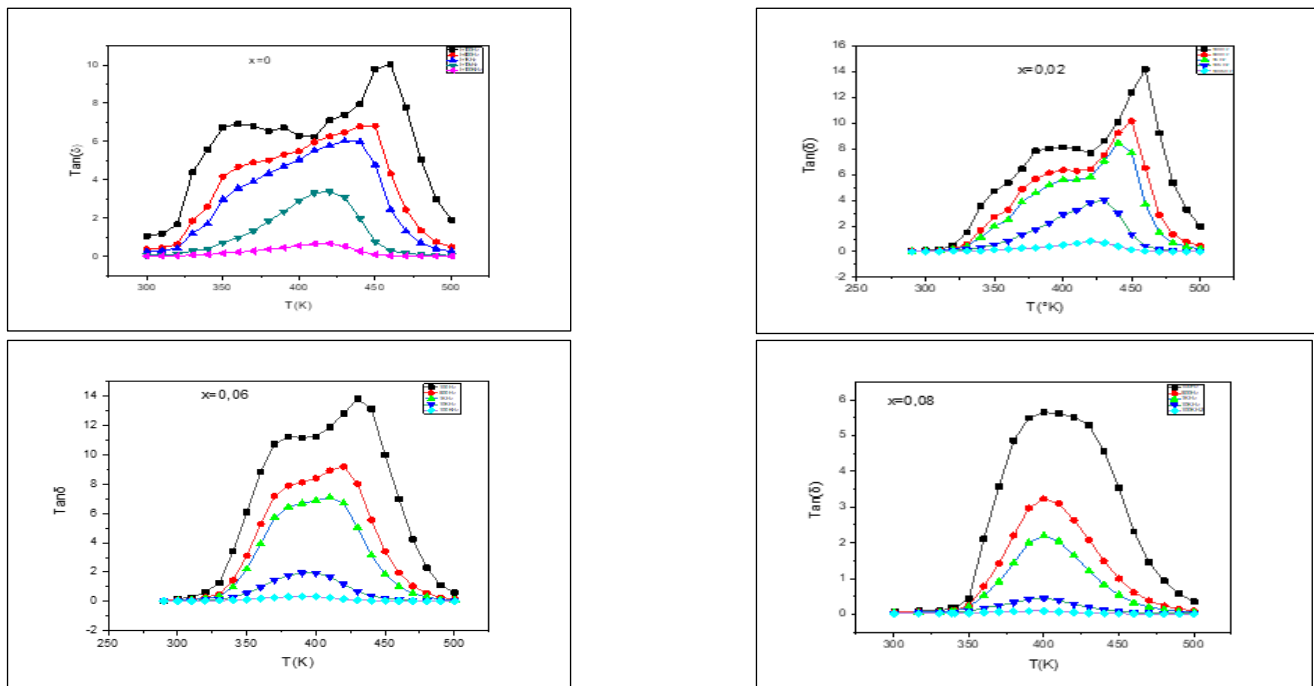


Figure 3: The evolution of dielectric loss for all compositions of BNTZZN-x Mg as a function of temperature and frequency.
Source: Authors, (2026).

Table 4: The dielectric loss values for BNTZZN-xMg.

x	Dielectric loss
0.00	10
0.02	14
0.06	14
0.08	6

Source: Authors, (2026).

IV. CONCLUSIONS

- X-ray diffraction (XRD) analysis reveals that the prepared BNTZZN-xMg materials adopt a cubic structure, with the presence of a secondary phase known as 'pyrochlore'.
- The density test shows that the densification rate of the analyzed ceramics ranges between 85.52% and 92.50%.
- The dielectric properties of the studied materials show a strong dependence on temperature, frequency, and composition. As the temperature increases, the permittivity generally rises until reaching the Curie temperature, followed by a decrease. Low-frequency dielectric dispersion is attributed to multiple polarization mechanisms and ionic conductivity.

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VI. AUTHOR'S CONTRIBUTION

Conceptualization: Ouahab Zakaria.

Methodology: Ouahab Zakaria.

Investigation: Ouahab Zakaria.

Discussion of results: Zakaria.

Writing – Original Draft: Ouahab Zakaria.

Writing – Review and Editing: Ouahab Zakaria, Ouahab Abdelwhab.

Resources: Ouahab Zakaria.

Supervision: Ouahab Abdelwhab.

Approval of the final text: Ouahab Abdelwhab.

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