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Dielectric Studies of $Ba_{0.06}(Na_{1/2}Bi_{1/2})_{0.94}TiO_3$-$Ba(Fe_{1/2}Ta_{1/2})O_3$Solid Solution

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Abstract
In this work, the effects of addition of $Ba(Fe_{1/2}Ta_{1/2})O_3$ (BFT) on the dielectric behaviour of $Ba_{0.06}(Na_{1/2}Bi_{1/2})_{0.94}TiO_3$ (BNBT) ceramics have been reported. The lead-free (1-x)BNBT-xBFT (where $0 \leq x \leq 1.0$) solid-solutions were synthesized by traditional ceramics fabrication technique followed by sintering. Further, the dielectric measurement of solid solution was performed.

Keywords: BNBT, Calcination, Perovskite, Dielectric

1. Introduction

Bismuth-based compounds have emerged as promising alternatives to lead-based materials due to environmental concerns. Bismuth-based compounds often exhibit similar or even superior properties to lead-based materials, making them attractive alternatives across various fields. Among the Bi-based systems, (1-x)$(Bi_{1/2}Na_{1/2})TiO_3$-$x$$BaTiO_3$ is considered to be one of the potential non-lead candidates for dielectric and/or piezoelectric applications. It exhibits a rhombohedral-tetragonal morphotropic phase boundary (MPB) around $0.06 \leq x \leq 0.08$ with remarkable dielectric, piezoelectric and electromagnetic properties.

In recent years, complex perovskites with nominal chemical formula $A(Fe_{1/2}B_{1/2})O_3$ (A = Ba, Sr, and Ca; B =Nb, Ta and Sb) have attracted much attention because of their giant dielectric (10^3-10^5) response over a wide temperature and frequency interval[1-6].$Ba(Fe_{1/2}Ta_{1/2})O_3$ (BFT), in particular, is considered to be one of the interesting materials among them.

The solid solution of $Ba_{0.06}(Na_{1/2}Bi_{1/2})_{0.94}TiO_3$ -$x$$Ba(Fe_{1/2}Ta_{1/2})O_3$ combines different perovskite compounds and offers the potential for tailoring its properties by varying the composition. This material can be of interest for applications in areas such as magnetoelectric devices, sensors, and actuators, where multiferroic behaviour and tunable properties are desired. The addition of $Ba(Fe_{1/2}Ta_{1/2})O_3$ (BFT) to $Ba_{0.06}(Na_{1/2}Bi_{1/2})_{0.94}TiO_3$ (BNBT) introduces iron (Fe) and tantalum (Ta) into the crystal structure. This substitution can lead to changes in the material's properties, such as its dielectric, ferroelectric, ferromagnetic, and multiferroic behaviour. The "*x*" in the formula represents the degree of substitution of BFT in the solid solution, indicating that the amount of BFT can vary.

Therefore, it is of interest to study the electrical properties of $Ba_{0.06}(Na_{1/2}Bi_{1/2})_{0.94}TiO_3$-$Ba(Fe_{1/2}Ta_{1/2})O_3$ ceramic system as a function of frequency and temperature. Accordingly, the present work reports the effects of BFT addition on the dielectric properties of BNBT ceramics.

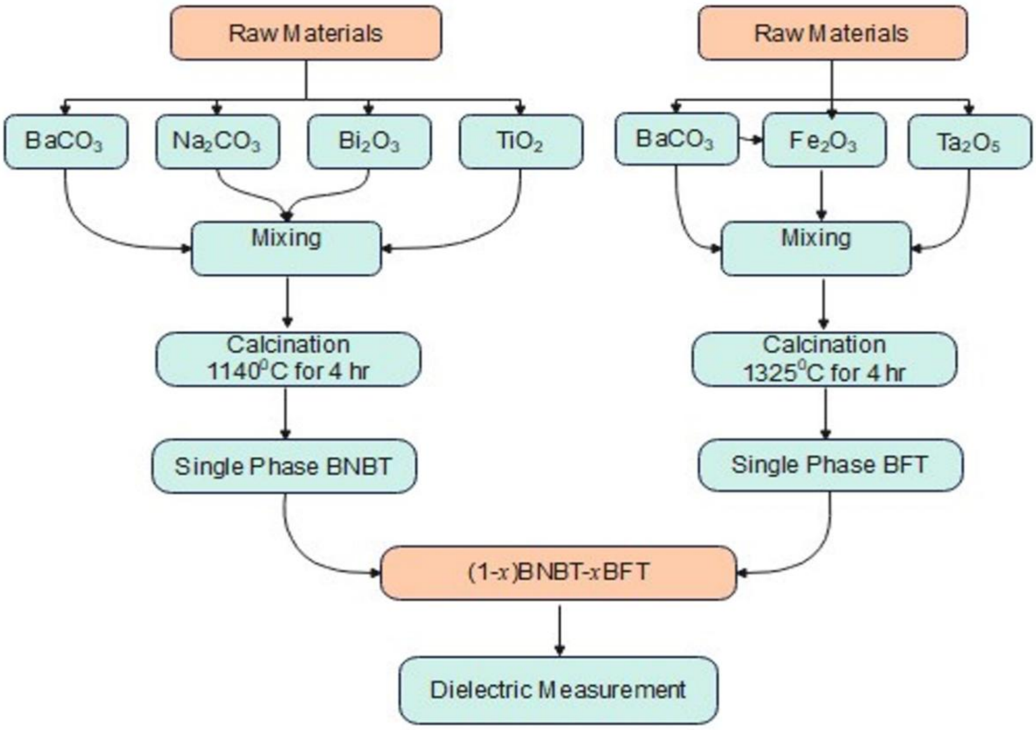
2. Experimental

The polycrystalline $Ba_{0.06}(Na_{1/2}Bi_{1/2})_{0.94}TiO_3$ and $Ba(Fe_{1/2}Ta_{1/2})O_3$ powders were prepared separately by solid-state reaction technique using high purity (>99.9%) carbonates/oxides, i.e. $BaCO_3$, Na_2CO_3, Bi_2O_3, TiO_2, Fe_2O_3 and Ta_2O_5. Following thermochemical reactions were carried out in an air atmosphere at 1140°C and 1325°C, respectively

$$0.06BaCO_3+\frac{0.94}{4}Na_2CO_3+\frac{0.94}{4}Bi_2O_3+TiO_2 \rightarrow Ba_{0.06}(Na_{1/2}Bi_{1/2})TiO_3+0.295CO_2(g)\uparrow$$

$$BaCO_3+\frac{1}{4}Fe_2O_3+\frac{1}{4}Ta_2O_5 \rightarrow Ba(Fe_{1/2}Ta_{1/2})O_3+CO_2(g)\uparrow$$

BNBT was then doped with varying percentages of BFT. A series of $(1-x)Ba_{0.06}(Na_{1/2}Bi_{1/2})_{0.94}TiO_3$–$xBa(Fe_{1/2}Ta_{1/2})O_3$ (x = 0, 0.10, 0.25, 0.50, 0.75, 0.90 and 1.0) samples were compacted into thin (1.5 mm) cylindrical disks with an applied uniaxial pressure 5 Tons. The samples were finally sintered between 1180 and 1350^0C for 4 h. The circular surfaces of the disks were covered with thin silver paste layers and fired at 500^0 C for 30 min, which act as the electrodes for the electrical measurements. Dielectric measurements were carried out using a computer-controlled LCR Hi-Tester (HIOKI 3532-50, Japan) interfaced with a microprocessor controlled dry temperature controller (DPI-1100, Sartech Intl., India) at a heating rate of 4^0C/min. The schematic of the synthesis and characterization process is shown in figure 1.



3. Result and Discussion

3.1Dielectric Studies

Dielectric properties of solid materials are of immense importance due to their potential applications in the area of solid-state electronics and electrical engineering [7-10]. These applications are mostly concerned with the properties like: dielectric constant, loss tangent, dielectric strength, *etc.* Study of dielectric properties of a material as a function of frequency and temperature helps in understanding various polarization mechanisms present in them [11]. The selection of a particular dielectric material for specific application is generally depending on its ability to operate under usual environmental conditions, suitable thermal expansion and dimensional characteristics.

3.2 Dielectric Constant as a function of frequency

Figs. 2 and 3 show the variation of the real part (ε') and imaginary part (ε'') of the dielectric constant with frequency at various temperatures for the entire series of (1-x)$Ba_{0.06}(Na_{1/2}Bi_{1/2})_{0.94}TiO_3$-$x$ $Ba(Fe_{1/2}Ta_{1/2})O_3$; x = 0, 0.10, 0.25, 0.50, 0.75, 0.90, and 1.00. For x=0and x=0.05 the dielectric constant initially decreases and then increases with an increase in frequency, especially at low temperatures. $\frac{d\varepsilon'}{df}$ is steeperin the low-frequency region and at low temperatures. For the imaginary part of the dielectric constant, it is observed that ε'' is almost frequency independent in the low-frequency region (< 10kHz) for x=0and x=0.05. For $x \geq 0.25$ it is observed that with the increase in frequency both ε' and ε'' in general decreases.A relatively high dielectric constant at low frequencies is a characteristic of a dielectric material. This may be due to the space charge contribution. At very low frequencies, dipoles follow the field and we have $\varepsilon' \approx \varepsilon_s$. As the frequency increases dipoles begin to lag behind the field andslightlydecrease. When frequency reaches the characteristic frequency, the dielectric constant drops (relaxation process), and at very high frequencies, dipoles can no longer follow the field and $\varepsilon' \approx \varepsilon_\infty$. Further, $\varepsilon'' - f$ plots find aminimum after 300^0C, which shifts towardthe lower frequency side with increasing temperature. Thiscould be due to the resonance effect [12]. The values ε' and ε'' are, respectively, found to be 3699 and 10,788 at1 kHz.

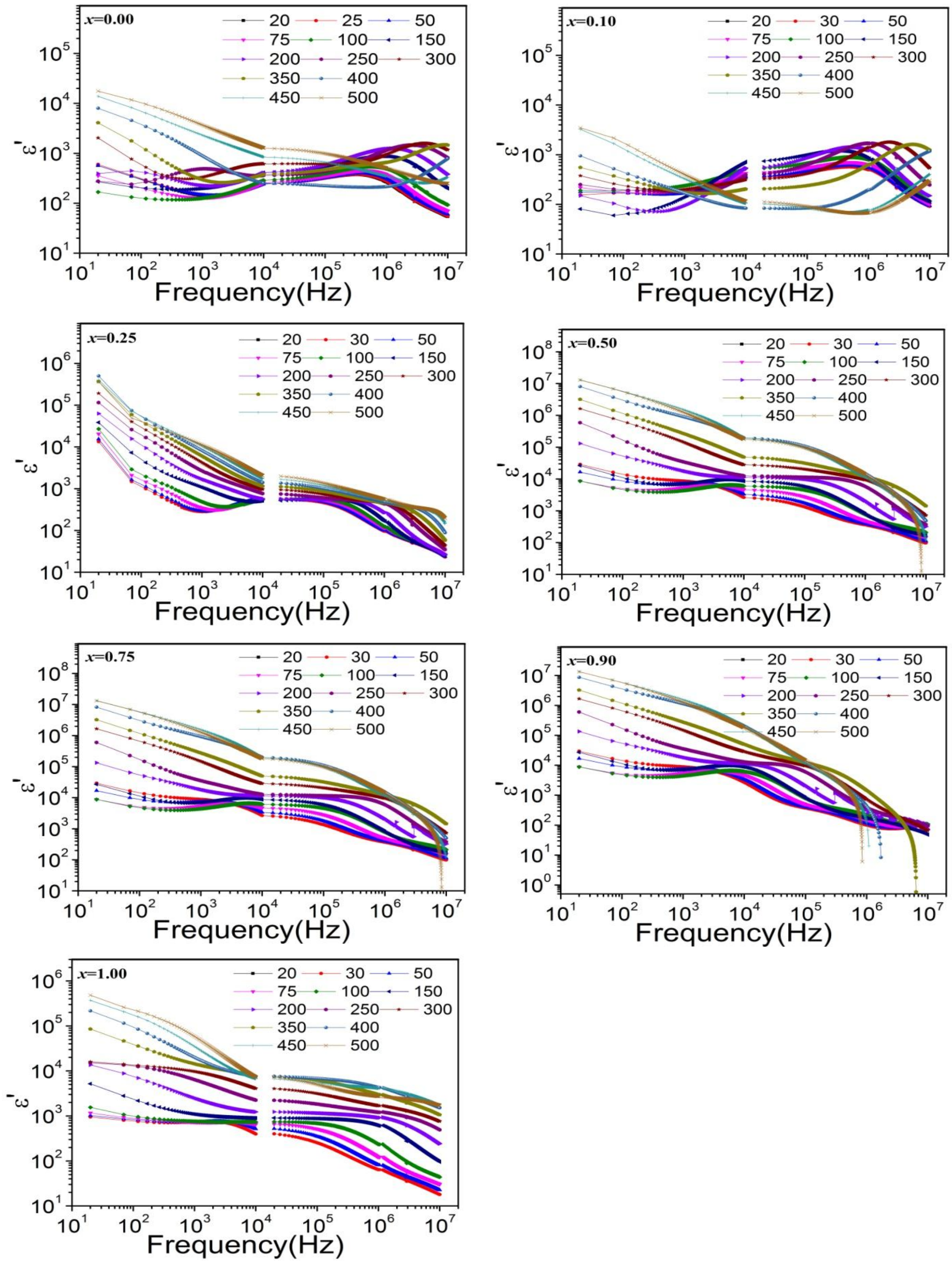


Figure 2: Frequency dependence of real part of the dielectric constant of (1- x)$Ba_{0.06}(Na_{1/2}Bi_{1/2})_{0.94}TiO_3$-$x$$Ba(Fe_{1/2}Ta_{1/2})O_3$ ceramics.

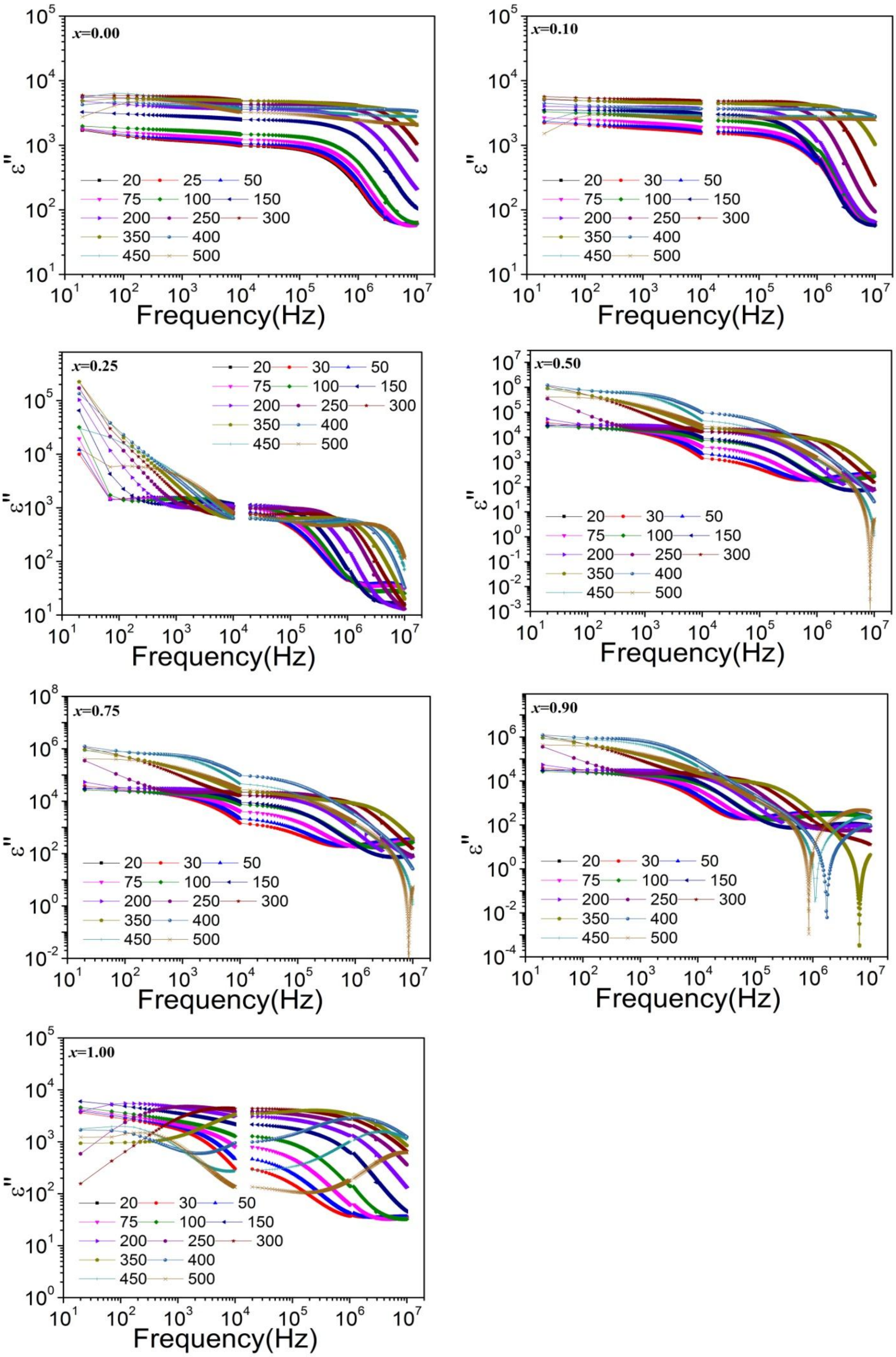


Figure 3: Frequency dependence of imaginary part of the dielectric constant of $(1\text{-}x)Ba_{0.06}(Na_{1/2}Bi_{1/2})_{0.94}TiO_3\text{-}xBa(Fe_{1/2}Ta_{1/2})O_3$ ceramics.

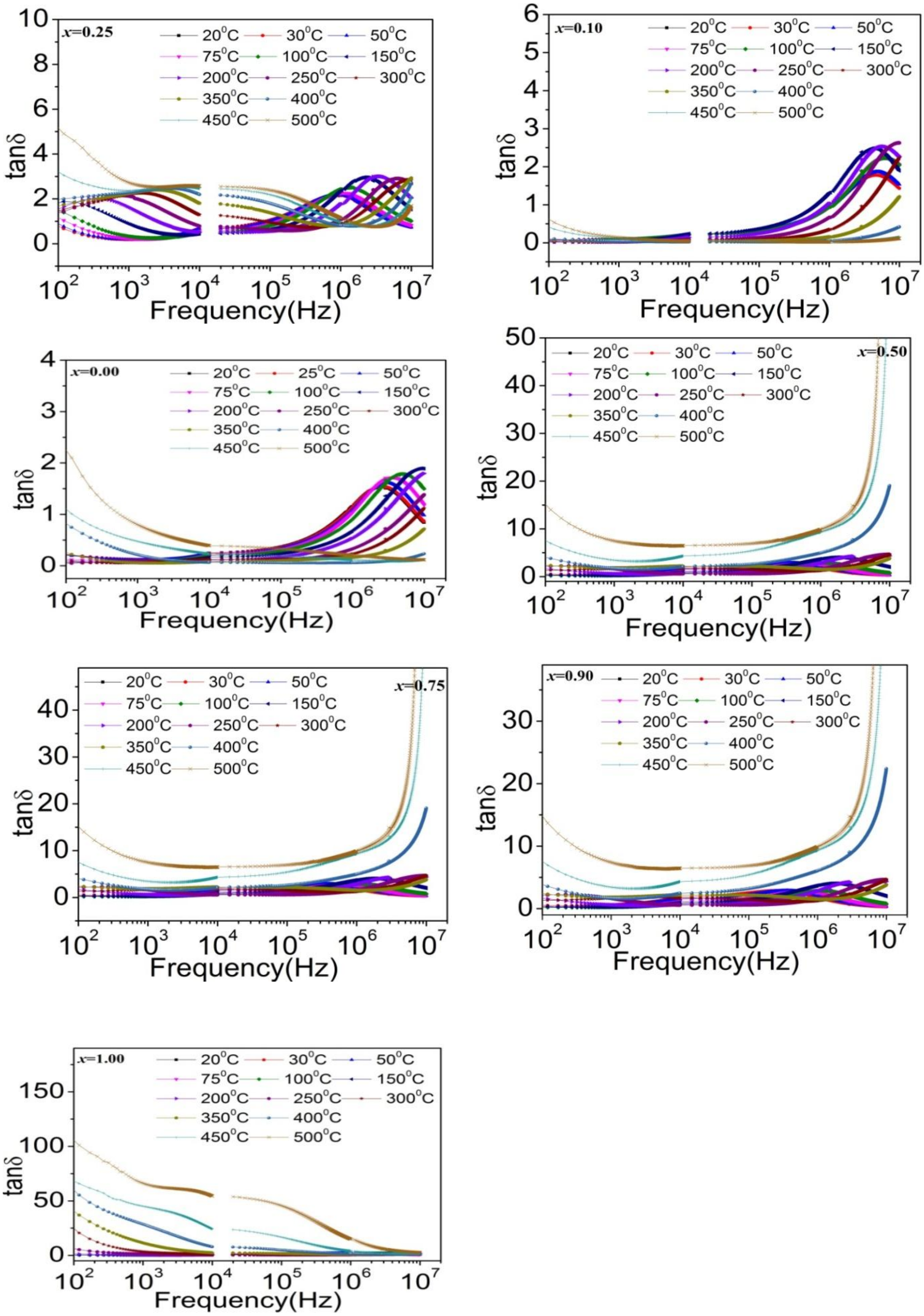


Figure 3: Frequency dependence of tangent loss of $(1\text{-}x)Ba_{0.06}(Na_{1/2}Bi_{1/2})_{0.94}TiO_3$-$xBa(Fe_{1/2}Ta_{1/2})O_3$ ceramic .

3.3 Tangent loss as a function of frequency

There is a peak in the tanδ-*f* curve for $x = 0$, 0.05 and 0.10 but the peak and its amplitude have no significant change with an increase in BFT concentration. The tanδ peak almost disappears for $x \geq 0.50$. The value at room temperature was found to be in the range of 0.1 to 1 at 1KHz for the compounds under investigation. The low value can be advantageous for many electronic/microelectronic applications.

4. Conclusion

Perovskite type lead-free ceramics, $(1-x)Ba_{0.06}(Na_{1/2}Bi_{1/2})_{0.94}TiO_3–xBa(Fe_{1/2}Ta_{1/2})O_3$ ($0 < x < 1.0$) were synthesized using the solid state reaction method. The dielectric property of obtained solid solutions was performed. The value of tanδat room temperature was found to be in the range of 0.1 to 1 at 1KHz for the compounds under investigation. The low value can be advantageous for many electronic/microelectronic applications.

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