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Structural and Electrical Studies of Bfn-St40 Nanoceramics Processed By Ball Milling

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Abstract: ***0.60 $Ba(Fe_{0.5}Nb_{0.5})O_3$ -0.40 $SrTiO_3$ nanoceramic sample was prepared by solid-state reaction method through Ball milling after annealing at high temperature 1200°C.The phase purity and crystalline have been studied by using the X-ray diffraction (XRD) technique. The various structural parameters have been calculated by Rietveld refinement method using cubic symmetry with Pm-3m space group. The microstructural analysis has been carried out by field emission scanning electron microscopy (FESEM). Two distinct peaks obtained in The temperature dependent dielectric constant and dielectric loss could be explained by the Maxwell Wagner relaxation model for different temperature relaxation region.***

Key terms:- 0.60 $Ba(Fe_{0.5}Nb_{0.5})O_3$ -0.40 $SrTiO_3$ nanoceramic, Rietveld refinement method, FESEM, polaronic effect, Maxwell Wagner relaxation model, Cole–Cole plot
Keywords: Include at least 5 keywords or phrases

I. INTRODUCTION

Worldwide ceramics market is dominated by electronic-ceramics known as (electro-ceramics),this field has been recognized as one of the most exhilarating field in material science. According to need of different technological applications electro-ceramics can be configured with synthesis of different materials. Large share of the ceramic materials, used for technological applications, is dominated by oxides based dielectrics .There is always a demand of new electro-ceramic materials with superior electric properties to meet the fast growing technological necessity. Ceramic industries are by far the fast growing and one of the vast scope industry established by humankind. Yet, there are only a half dozen specific methods to get ceramic phases that dominate synthetic method of such materials not only in terms of their production but also in terms of technological significance .In the ceramics family among thousands of complex structures there are only a dozen or so structures which dominated all. Among these are the A_2BX_4 structure, spinel; and the ABX_3 structure, perovskite are most important. Perovskite having single structure which, with different synthetic methods ,can produce improbably wide array of phases with different properties.[1]

Perovskites are ternary oxides of general formula ABO_3. More generally, the perovskite formula is ABX_3, where the anion X can be O, N, or halogen. The A site cations with co-ordination number 12 are typically large ions such as Ba^{2+},Sr^{2+}, , Rb^{+}, or a lanthanide 3+ ion, and the B ions with co-ordination number 6 are smaller transition metal ions such as Ti^{4+}, Nb^{5+}, Ru^{4+}, etc. Russian mineralogist, L. A. Perovski (1792-1856) was discovered $CaTiO_3$ as a mineral after his name this structure of this material was known as Perovskite structure. The perovskite family consists of a variety of materials ranging from ferroelectrics ($PbTiO_3$ and $BaTiO_3$), microwave dielectrics or dielectric resonators ($BaMg_{1/3}Ta_{2/3}O_3$ and $BaZn_{1/3}Ta_{2/3}O_3$),), superconductors ($Ba1\text{-}xKxBiO_3$) non-linear optical behavior materials ($KNbO_3$)., piezoelectrics ($PbZr_{1-x}TixO_3$), relaxor ferroelectrics ($PbMg_{1/3}Nb_{2/3}O_3$), multiferroics ($BiFeO_3$) and colossal magneto resistance materials ($La_{1-x}Ca_xMnO_3$).

It is observed from the literature that a non-lead ternary perovskite barium iron niobate, $Ba(Fe_{1/2}Nb_{1/2})O_3$ (abbreviated hereafter as BFN) ceramic exhibits very high dielectric constant ($\approx 10^4$–10^5). The structural, microstructural, and IR spectroscopic studies of BFN (a non- ferroelectric) were first reported by Reaney et al. [2]. Saha and Sinha [3, 4] reported that the BFN is relaxor ferroelectric material characterized by a broad dielectric transition. Raevski et al. [5] have elucidated that the Maxwell– Wagner mechanism is responsible for the nature of high values of the dielectric permittivity in BFN and similar ceramics. Afterwards, several attempts have been made to study the dielectric behavior of BFN [6–11]. In a most recent study, it is demonstrated that the value of ε_r can be increased above 10^5 by considering the suitable sintering conditions [12].

In this research work we first time report about Synthesis of 0.60 $Ba(Fe_{0.5}Nb_{0.5})O_3$ -0.40 $SrTiO_3$ Nanoceramic processed by solid stste reaction method through ball milling.

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II. EXPERIMENTAL PROCEDURES

Complex perovskite oxides 0.60Ba(Fe0.5Nb0.5)O3-0.40SrTiO3 hereafter abbreviation as BFN-ST40 was prepared by a mixed oxide preparation route. High $BaCO_3$(Merck,99.101%),$SrCO_3$(Loba chemie 98%),Fe_2O_3(Merck ≥90%),TiO_2(Merck ≥98.5%) and Nb_2O_5(Loba chemie ,99.9%). were used for the preparation of BFN-ST40 ceramics. These chemicals were taken in stoichiometric ratio, and mixed in the presence of air for 2 hrs. and in acetone for 6 h using mortar and pastle. The finely mixed powder of BFN-ST40 was calcined at 1200°C for 8 h. The calcined powder of above mentioned ceramics were regrinded through 250 rpm for 20 Hrs using Retsch Ball Mill in ethanol medium to obtain nano-ceramics. The structure and quality of the obtained powder was checked with The X-ray powder diffraction pattern of the sample is taken at room temperature using a X-ray powder diffract meter (Rigaku Miniflex, Japan) using Cukα radiation (λ = 1.5418 Å) in a wide range of Bragg angles 2θ (20° ≤ 2θ ≤ 80°) with scanning rate 2°/min. Powder is used to make pellet of diameter ~10 mm and thickness 1 - 2 mm using polyvinyl alcohol as binder. The pellets were sintered at 1250°C for 5 h and then brought to room temperature under controlled cooling. The frequency dependence of the capacitance and conductance is measured using an LCR meter in the temperature range from 40°C to 400°C and in the frequency range from 1 KHz to 1 MHz .

III.STRUCTURAL ANALYSIS

Fig. III.1 shows the XRD patterns of the 0.60Ba($Fe_{0.5}Nb_{0.5}$)O_3-0.40$SrTiO_3$ i.e. BFN-ST40 nanoceramics powder conventionally sintered at 1200 °C for 8 hrs. Sharp and distinct XRD peaks were obtained for the given sample, which suggest the good crystalline and homogenous nature of the BFN-ST40 nanoceramic samples.

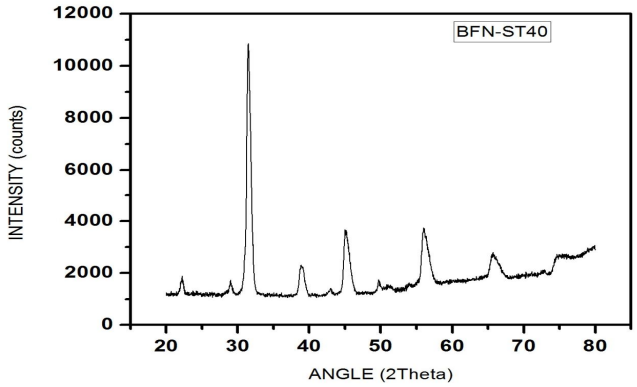

Fig. III.1. XRD pattern of 0.60 Ba ($Fe_{0.5}Nb_{0.5}$)O_3 -0.40 $SrTiO_3$Nanoceramic system

IV.MORPHOLOGICAL ANALYSIS

Fig. IV.1 shows the FESEM micrographs of the sintered BFN-ST40 nanoceramics. The sintered BFN-ST40 nanoceramics show dense and almost non-uniform microstructures.There is good crystallinity can be confirm with help of FESEM image present here.

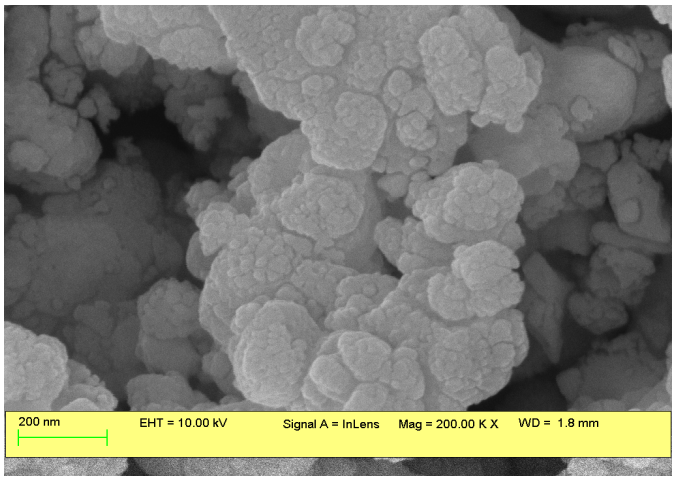

Fig. IV.1. FESEM image of 0.60 Ba($Fe_{0.5}Nb_{0.5}$)O_3 -0.40 $SrTiO_3$Nanoceramic system

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V. DIELECTRIC STUDIES

A. Frequency dependent dielectric behavior of BFN-ST40 Nanoceramics

Fig. V.1 (a) to (b) and (c) to (d) show frequency dependence of real (ε') and imaginary (ε") parts of the permittivity (ε*(ω) =ε'-jε") for BFN-ST40 nanoceramics respectively. The ε' decreases with the increase in frequency for the present sample and convergence at higher frequencies and attain a value of ~100 at 1 MHz, which signify behavior of high dielectric constant materials [13]. The dielectric loss (ε") in a material is a combination of both ac relaxation processes and dc conduction [14]. Generally, the relaxation processes, observed in the case of dipolar materials, have a dispersive sharp curve in the ε' spectrum form alongwith a relaxation peak in the ε" spectrum [15]. Such type of dielectric behavior appear due to the inhomogeneity in the materials [16], internal barrier layer capacitor [17] and metal insulator interfacial effects [18].

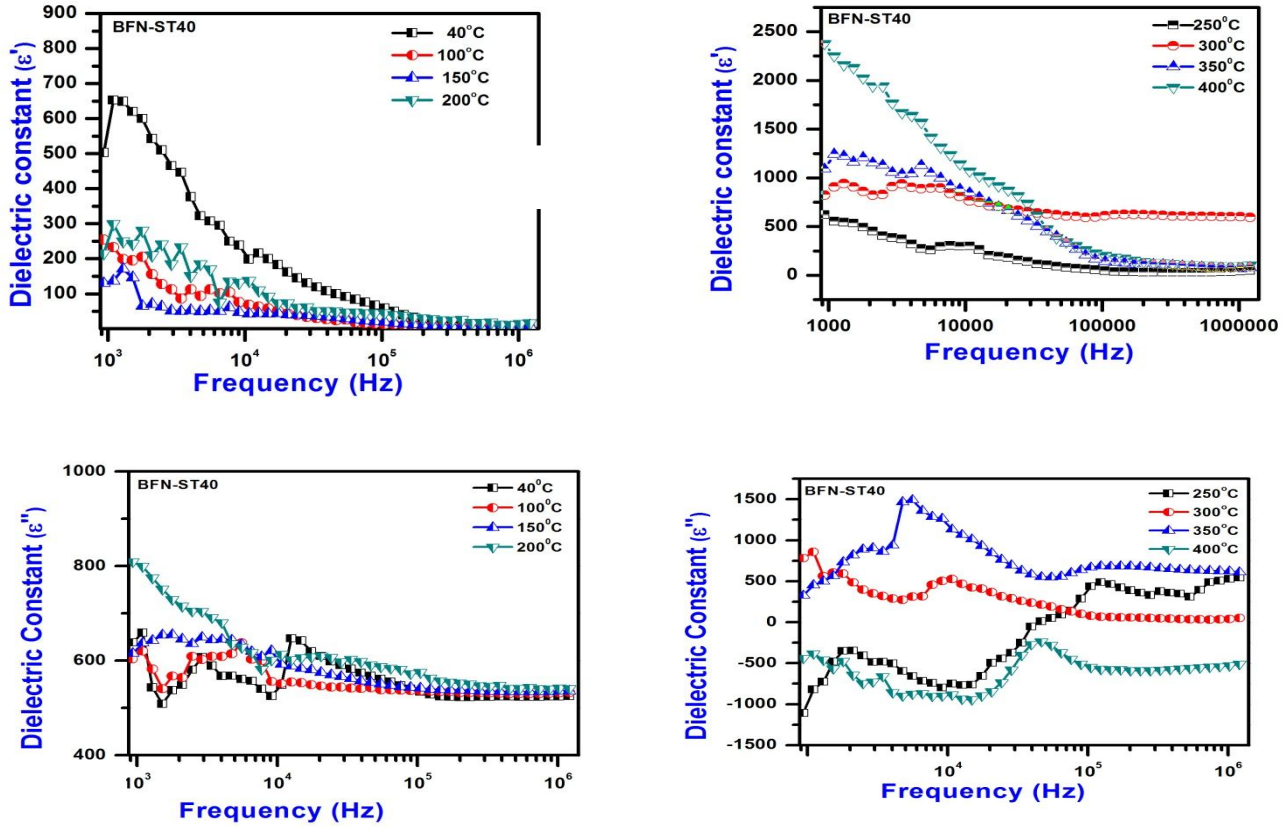

Fig.v.1 (a) and (b) shows Frequency dependence behavior of real part of Dielectric Constant .Fig. 6.4 (c) to (d) shows Frequency dependent behavior of imaginary part of Dielectric Constant ,at different temperatures.

It is well known that different types of relaxation processes fall in different frequency regions. At room temperature, the relaxation processes like space charge polarization, Maxwell-Wagner polarization show their loss peak below < 10^5 Hz, whereas loss peak due to electronic and hopping polarizations are observed at or above 10^5 Hz [19]. This suggests that Maxwell-Wagner polarization contributing to the BFN-ST40 nanoceramics in temperature range 40 to 200°C. But at 250°C it shows Hopping or electronic Polarization.Generally, oxygen vacancies are produced in the perovskite oxide materials processed at high temperature and in normal atmosphere [20] It causes distortion in Oxygen stoichiometry of the material and leads to some cationic as well as anionic defects in it.In the BFN-ST40 nanoceramic system both the transition metals, iron (Fe) and niobium (Nb), have more than one oxidation states (Fe^{2+}, Fe^{3+}, Nb^{4+}, and Nb^{5+}). Electrons get independent during the ionization of oxygen vacancies finely trapped by the Fe^{3+} and Nb^{5+} cations, which promote electronic relaxation between the Fe^{3+} & Fe^{2+} or Nb^{5+} & Nb^{4+} ions. Similarly, electronic relaxation can also exists via oxygen vacancy as a medium in the form of Fe^{2+}-O - Fe^{3+} [21]. The relaxation processes in BFN-ST40 nanoceramics, shown in Fig.v.1 (b), occurred in the different frequency regions. Since, the dielectric loss peak was observed around 10^5Hz at temperature range 250-400°C for the sample; therefore the possible origin of the relaxation process in this sample may be due to the electronic relaxation of oxygen vacancies or relaxation between Fe^{3+}& Fe^{2+} or Nb^{5+}& Nb^{4+} ions. Whereas, the relaxation

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in temperature range 40-200°C was of Maxwell- Wagner type of relaxation. This confirmed that the processing phenomena have great influence not only on the microstructures but also on the dielectric behavior of the HDC materials.Fig. v.2 (a) and (b) confirm variation in imaginary part of dielectric constant accompanied by variation in loss tangent as a function of frequency. At room temperature and low frequency there is low dielectric loss which can be due to processing phenomena of present sample but temperature >350°C ,loss tangent increases it may be due to oxygen vacancies.

(a) (b)

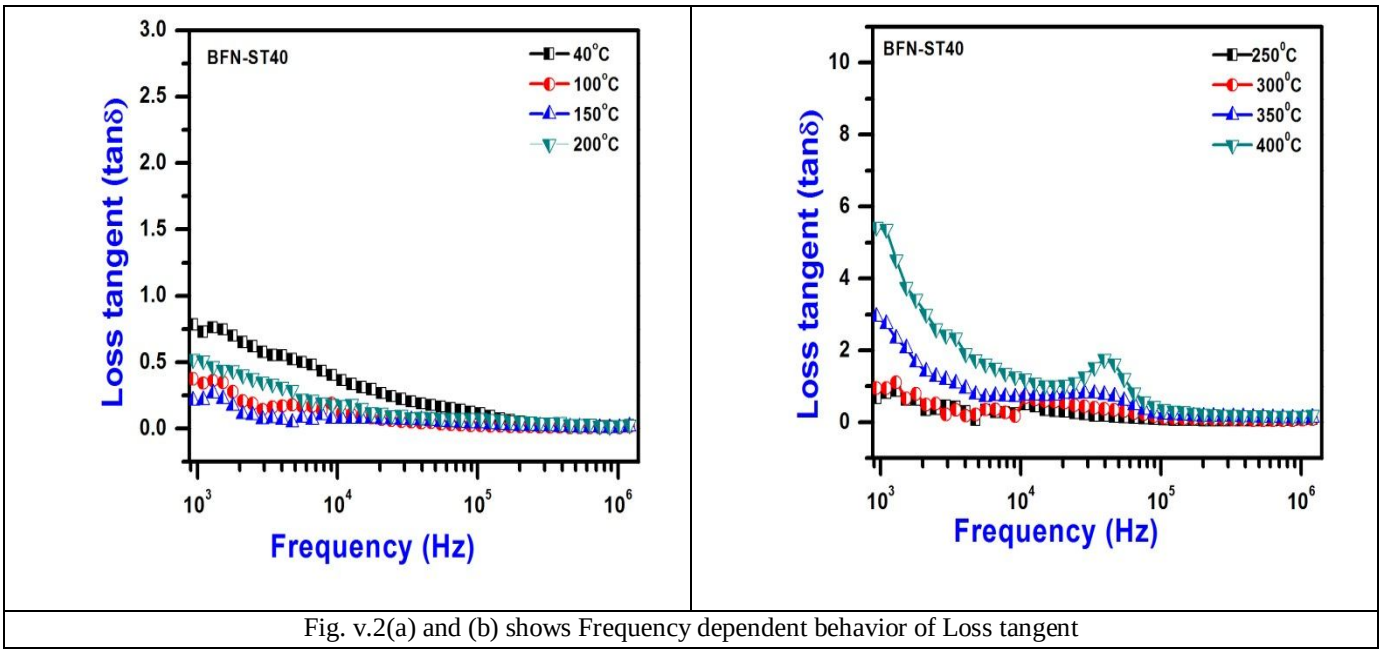
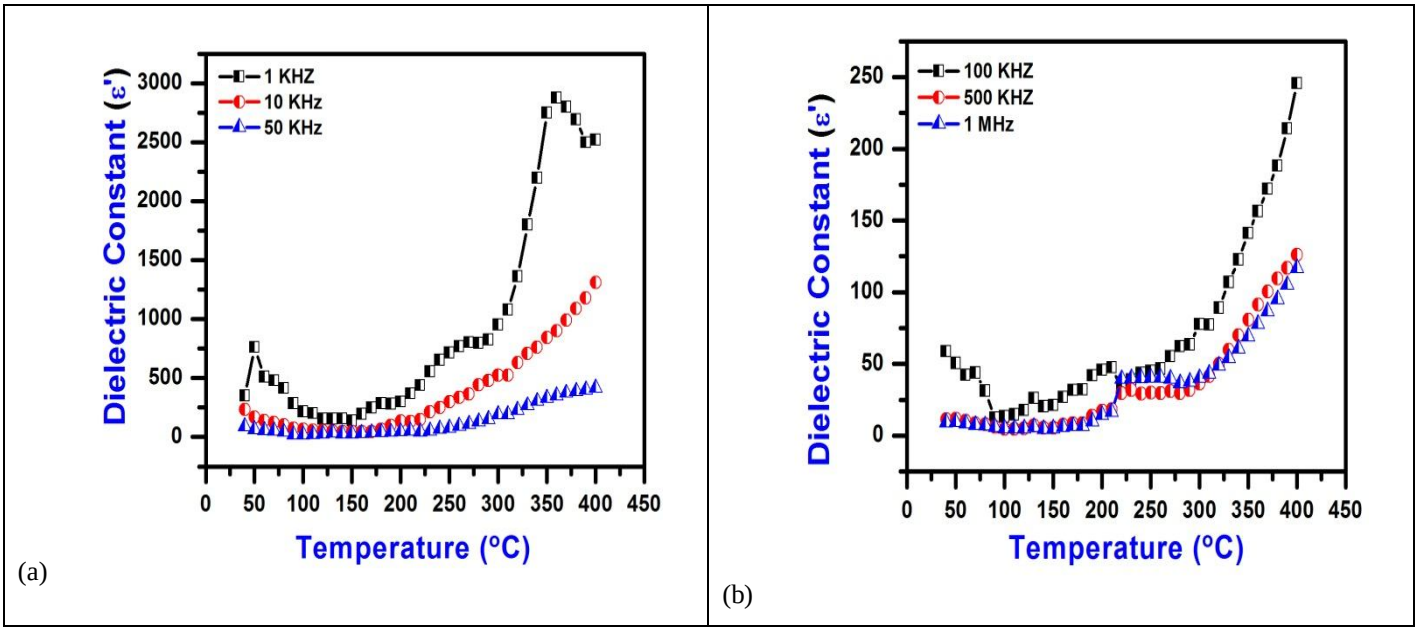


Fig. v.2(a) and (b) shows Frequency dependent behavior of Loss tangent

B. Temperature dependent dielectric behavior of BFN-ST40 Nanoceramics

Figs. V.3 (a) & (b) show the temperature dependence of dielectric constant (ε') and dielectric loss (tanδ) at different frequencies of BFN-ST40 nanoceramic sample. Here, because of the high dielectric constant, the BFN-ST40 nanoceramic sample was taken for temperature



(a) (b)

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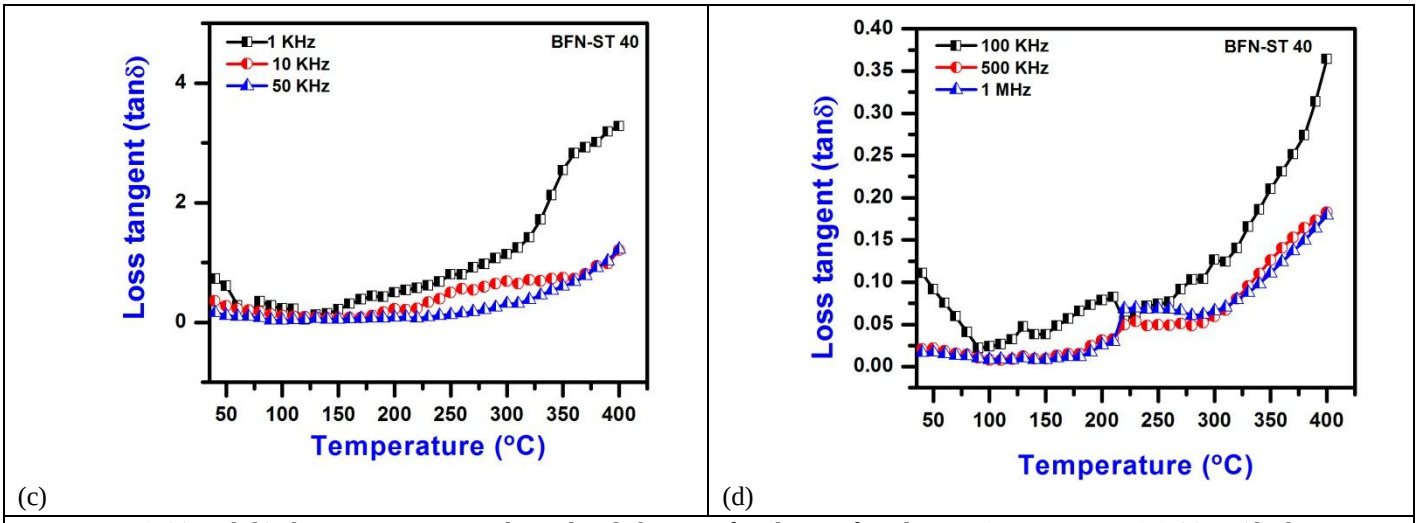

Fig.V.3 (a) and (b) shows Temperature dependent behavior of real part of Dielectric Constant .Fig. 6.6 (c) to (d) shows Temperature dependent behavior of Loss tangent, at different frequencies.

dependent dielectric properties variation study. Two anomalies were observed in the ε' vs. temperature plot in all the samples i.e. a dielectric step was observed ~50°C and a dielectric peak was observed ~350°C for the present samples .The dielectric peak observed at low frequency (1 kHz) ~350°C diminished with the increase in frequency (1 MHz), which suggests that the relaxation process must be originating from the extrinsic factors. Most of the ferroelectrics also exhibit similar type of dielectric relaxation in the paraelectric region. The dielectric abnormalities observed in this temperature range is the result of the oxygen vacancies and the other point defects as reported earlier . Concentration of oxygen vacancies and the used processing conditions influence the relaxation process. The indexing of the calcined BFN powder pattern confirmed the centro-symmetric nature of BFN nanoceramic at room temperature. Literature reports also confirm that there is no structural phase transformations in the BFN ceramics from room temperature to 550°C . This confirmed that the origin of dielectric peak observed around 200°C was not due to any structural transformation, but due to the formation of oxygen vacancies and related to other transition metal point defects. This confirmed BFN is a non-ferroelectric ceramic.

The difference in the dielectric relaxations due to space charges present in the ferroelectric materials and the high dielectric constant (high-k) materials is that, space charges are present at the vicinity of room temperature in high-k materials , whereas effect of space charges can be observed far away from the room temperature in ferroelectric materials . The space charges created during sintering process are confined to their defect sites, which promotes electronic relaxation among the defect sites. Generally, in high-k materials, thermal fluctuations are significantly high at room temperature such that the space charges are no more confined to their defect sites but extended to the whole sample, which makes the sample semiconducting. With the application of an electric field to these high-k materials, space charges moves inside the grains and forms different conducting regions near grain boundaries. This leads to Schottky type of barriers formation at the interfaces, which leads to high dielectric constant in these non-ferroelectric materials.

The interfacial or Maxwell-Wagner polarizations can be interpreted on the basis of semiconducting grains and insulating grain boundaries. It is well known that fast cooling in normal atmosphere after sintering produces semiconducting grains and insulating grain boundary . Therefore, the Maxwell-Wagner polarization or interfacial polarization arises due to presence of different types of conducting processes present in the BFN-ST40 nanoceramics system.

VI. CONCLUSION

Polycrystalline BFN-ST40 nanoceramic has been successfully synthesized by mechanochemical method. The present sample shows the cubic perovskite structure with *Pm*-3*m* space group. There are two peaks observed in variation of dielectric constant with

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temperature which are analogous to lower temperature relaxation and higher temperature relaxation and both of these relaxation may be result of Oxygen vacancies and Maxwell–Wagner model respectively

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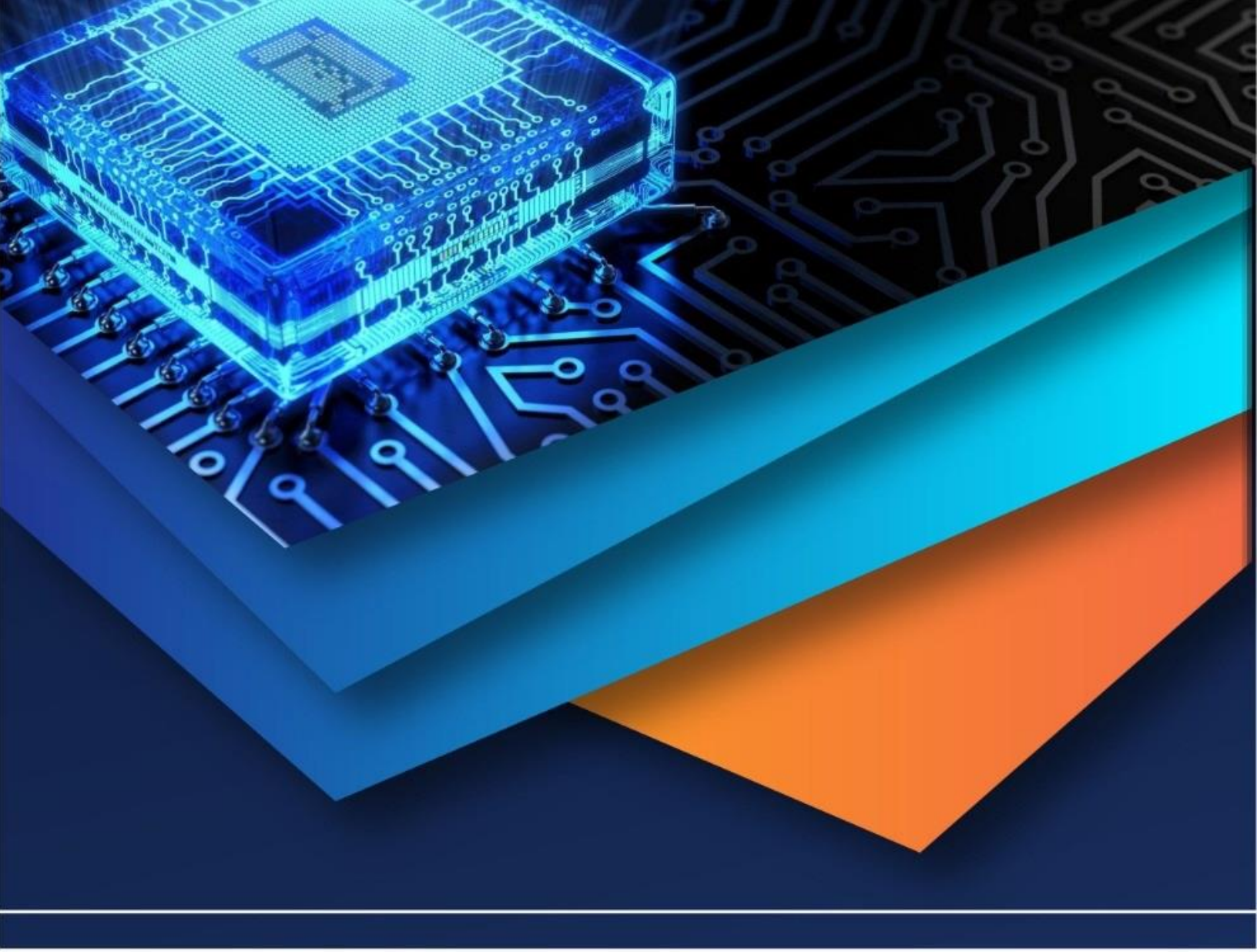

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