

# Dielectric Relaxor Behaviour of $(\text{Ba}_{0.77}\text{Ca}_{0.23})\text{TiO}_3$ Ceramic Due to the Effect of $\text{Pr}^{3+}$ Doping on the B-Site Lattice

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**Abstract:** *BaTiO<sub>3</sub> is the earliest known high dielectric constant material. The ability of this material to form solid solutions with different dopants (both iso and alio valent) makes it versatile for various applications. In this present study, BaTiO<sub>3</sub> is first modified with Ca<sup>2+</sup> and then end material is modified with Pr<sup>3+</sup> at the B site lattice. Here it is tried to search for new lead-free ferroelectric material and improve their properties. For this purpose, the barium calcium titanates (BCT) as a host and the rare earth element Pr<sup>3+</sup> as an activator were used to fabricate a multifunctional material. The end material is found to be in a single phase and the morphology says they are homogeneous and dense, also there is no evidence of secondary phase from the ceramograph. Lastly, the dielectric study is carried out and observed that, T<sub>c</sub> increases to 320.5 °C and observed to be a relaxor ferroelectric. Here in this paper, some structural parameters and dielectric constants of praseodymium modified barium (calcium) titanate ceramics is presented.*

**Keywords:** Structure, Rietveld analysis, Relaxor ferroelectric, Dielectric constants

## 1. Introduction

Barium titanate ( $\text{BaTiO}_3$ ) is an important ceramic material for electronic and memory device applications. The ability of the material to form solid solutions with different dopant ions makes the material versatile for various applications [1]. The solid solubility of the material depends on the site of substitution, charge compensation mechanism involved and solid solubility limit of the dopant. Dopant in the  $\text{BaTiO}_3$  ceramics can occupy either octahedrally coordinated  $\text{Ti}^{4+}$  site or dodecahedrally coordinated  $\text{Ba}^{2+}$  site. There are several reports on the different A site and B site dopants modifying the electrical properties of  $\text{BaTiO}_3$  ceramic [2-6]. In recent years a considerable amount of work has been done on the ferroelectric properties of  $\text{BaTiO}_3$ . In addition to  $\text{BaTiO}_3$ , there are other different perovskites which have been reported as ferroelectrics [7]. On heating,  $\text{BaTiO}_3$  undergoes a ferroelectric/paraelectric phase transition to the cubic polymorphism at a Curie temperature,  $T_c$  of 120 °C. The substitution of iso-valent cations, such as  $\text{Ca}^{2+}$  alter its lattice constant and thereby its dielectric properties. Iso valent substituted  $\text{BaTiO}_3$  are the potential candidate for various electronic applications and have been actively studied. Among them Ca doped  $\text{BaTiO}_3$  are considered to be one of the important candidate for lead free electro optic modulators and memory devices [8]. Solid solutions of  $\text{Ba}_{1-x}\text{Ca}_x\text{TiO}_3$  (for  $x=0.05$  to 0.9) has been studied widely and relaxor dielectric nature is analysed. The material shows single phase upto  $x=0.3$  in different [9] study and thereafter it becomes diphasic.  $\text{Ca}^{2+}$  has a smaller ionic radius than  $\text{Ba}^{2+}$ . However, it has been pointed out that the Ca ion in  $\text{Ba}_{1-x}\text{Ca}_x\text{TiO}_3$  might have greater atomic polarisability, thereby intensifying the interactions between the Ti ions and thus compensate for the decrease of  $T_c$ .

Substitution of aliovalent cations in  $\text{BaTiO}_3$  resulted in significant change in electrical properties because of their

acceptor/ donor behaviour [10]. Trivalent rare earth cations have moderate atomic radii between  $\text{Ba}^{2+}$  and  $\text{Ti}^{4+}$  ions and ability to substitute both A and B site and so widely used to modify  $\text{BaTiO}_3$ . [11-16].

Based on different properties ferroelectric materials may be divided in to two different classes, one is normal (classical) and the other is diffuse (relaxor) ferroelectrics [17]. Relaxor ferroelectrics are crystalline materials and have been widely studied in recent years due to their remarkable properties and wide prospects of practical applications [18-19]. The relaxor ferroelectric generally belongs to the family of disordered materials, and, in particular to the group of mixed cations ferroelectric of a perovskite structure with general chemical formula  $\text{AA}'\text{BB}'\text{O}_3$ . Maximum dispersion in relaxor ferroelectric is believed to originate from polar nano regions (PNR) inside which a parent ferroelectric-like order exists.

The most intensively studied relaxors are lead based system, such as (PMN), (PMN-PT), (PST), (PLZT) etc. [20-21]. But, due to toxicity and high volatility of lead these materials are undesirable. With the recent growing demand of global environmental protection, lead-free and heavy metal free materials have attracted much attention. Researchers have shown their interest in the solid solution system of perovskite  $\text{BaTiO}_3$  like, BZT, BCT, BMT, BST etc. with different concentrations of dopants and their combinations i.e. double perovskite system. In the present study an attempt is made to study the relaxor behaviour of the effect of Praseodymium at the solid solubility limit of  $\text{Ba}_{0.77}\text{Ca}_{0.23}\text{TiO}_3$  ceramic.

## 2. Materials and Methods

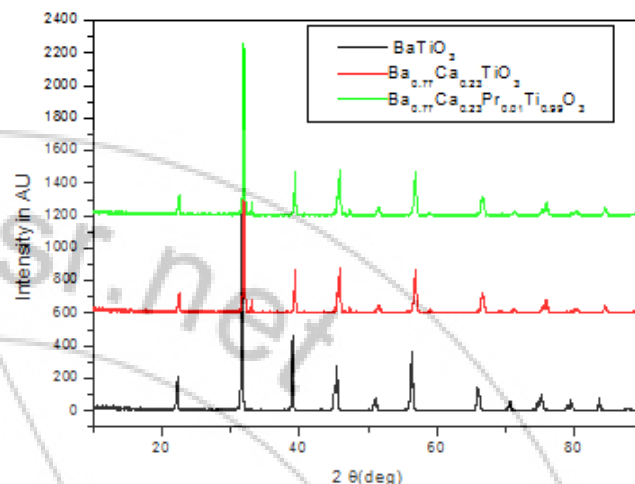
Pr modified (BaCa)  $\text{TiO}_3$  ceramics were prepared just above its solid solubility limit i.e.  $\text{Ba}_{0.77}\text{Ca}_{0.23}\text{TiO}_3$  by conventional

solid state reaction (SSR) technique. Stoichiometric proportionate high purity powders of BaCO<sub>3</sub> (99.9% pure Merck), CaCO<sub>3</sub> (99.9% pure Merck), TiO<sub>2</sub> (99.9% pure Merck) and Pr<sub>6</sub>O<sub>11</sub> (99.9% pure Merck) were ground for more than 6 h with acetone medium in an agate mortar for homogeneous mixing of powders. The composite powder of the compounds was conventionally calcined at 1280 °C for 4 h in an electrical furnace. Then the calcined powders were once again thoroughly mixed and ground for 2 h, mixed with 2wt.% of polyvinyl alcohol (PVA) as binder and pressed into disk-shaped pellets of 10 mm diameter and finally the green ceramics were sintered at 1320 °C for 6 h. The ceramics were structurally characterized by an x-ray diffractometer. The x-ray powder diffraction profiles of the samples were recorded using graphite filtered Cu K $\alpha$  radiation from a highly stabilized and automated x-ray generator operated at 30kV and 20 mA. For this experiment, 0.1° divergence slit, 1mm receiving slit were used. The step scan data of step size 0.01° and step scan 0.6s were recorded for the angular range 10-90°. The microstructure study was carried out by simultaneous structure and microstructure refinement using MAUD (1.4) Rietveld software. Scanning electron microscopy (SEM) of Jeol JSM 6480LV was used for the observation of the microstructure of the ceramics. An electron beam of 10 kV and 57  $\mu$ A was used for the SEM study. The average grain size was estimated through scanning electron microscope. For dielectric measurement of ceramic samples, both sides of the samples were polished for smooth surfaces to a thickness of 0.7 mm. For electrical measurement electrodes were made by applying silver paint on both sides of the specimen and annealed at 350 °C for 5min. Dielectric measurement was carried out over the frequency range 1 kHz to 1MHz using an LCR meter connected to PC. The dielectric data was collected at an interval of 5 °C while heating at a rate of 0.5 °C per min.

### 3. Results and Discussion

Figure 1 gives the x-ray diffractogram of BaTiO<sub>3</sub>, Ba<sub>0.77</sub>Ca<sub>0.23</sub>TiO<sub>3</sub> and Ba<sub>0.77</sub>Ca<sub>0.23</sub>Pr<sub>0.01</sub>Ti<sub>0.99</sub>O<sub>3</sub> ceramics. X-ray the fingerprint of the material shows that, the material is in single phase and there is no evidence of any secondary

phase. The x-ray parameters are given in table-1. The x-ray micrographs of these materials are then subjected to Rietveld analysis for their structure and microstructure refinement. Output of this refinement is listed in table-2, which represents the different refined parameters and the lattice parameters.



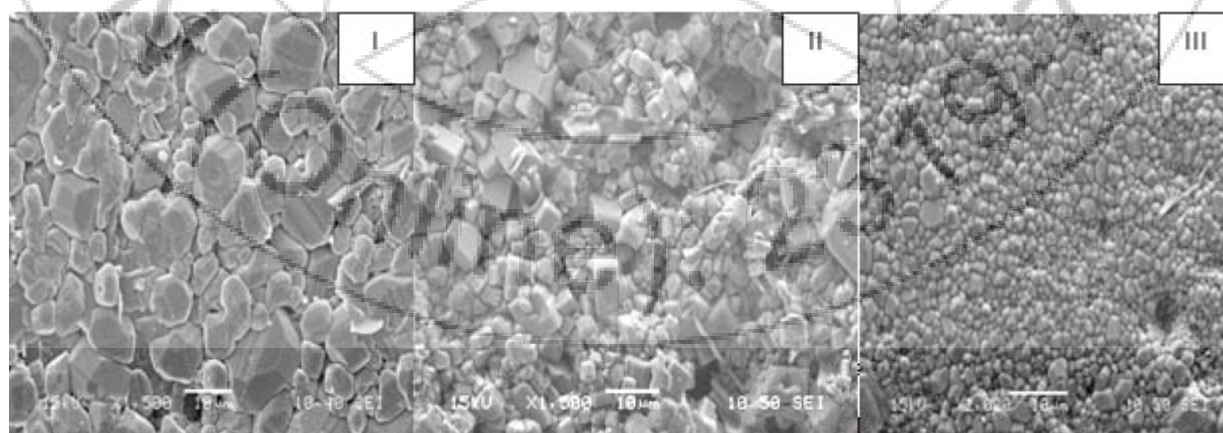
**Figure 1:** gives the x-ray diffractogram of BaTiO<sub>3</sub>, Ba<sub>0.77</sub>Ca<sub>0.23</sub>TiO<sub>3</sub> and Ba<sub>0.77</sub>Ca<sub>0.23</sub>Pr<sub>0.01</sub>Ti<sub>0.99</sub>O<sub>3</sub> ceramics.

**Table 1:** Microstructural parameters of (I) BaTiO<sub>3</sub>, (II) Ba<sub>0.77</sub>Ca<sub>0.23</sub>TiO<sub>3</sub> and (III) Ba<sub>0.77</sub>Ca<sub>0.23</sub>Pr<sub>0.01</sub>Ti<sub>0.99</sub>O<sub>3</sub> ceramics.

Sample	2 $\theta$ (deg)	d <sub>hkl</sub> (nm)	FWHM (nm)	D <sub>hkl</sub> (nm)	lattice strain ( $\epsilon$ )	plastic strain (e)
I	31.63	0.2827	0.0243	34.02	0	0.88
II	31.89	0.2804	0.0267	30.92	0.008	0.87
III	31.56	0.2833	0.0384	21.50	0.002	0.88

**Table 2:** Rietveld Refined parameters of (I) BaTiO<sub>3</sub>, (II) Ba<sub>0.77</sub>Ca<sub>0.23</sub>TiO<sub>3</sub> and (III) Ba<sub>0.77</sub>Ca<sub>0.23</sub>Pr<sub>0.01</sub>Ti<sub>0.99</sub>O<sub>3</sub> ceramics.

Sample	Refined Parameters				Lattice Parameters (nm)		
	Rwp	Rpb	Rp	Rwpb	a	b	c
I	0.88	0.93	0.84	1.05	0.505	0.505	0.515
II	0.79	0.73	0.69	0.78	0.396	0.396	0.397
III	0.80	0.77	0.71	0.88	0.145	0.145	1.698



**Figure 2:** SEM micrograph of (I) BaTiO<sub>3</sub>, (II) Ba<sub>0.77</sub>Ca<sub>0.23</sub>TiO<sub>3</sub> and (III) Ba<sub>0.77</sub>Ca<sub>0.23</sub>Pr<sub>0.01</sub>Ti<sub>0.99</sub>O<sub>3</sub> ceramics.

Fig. 5 shows the temperature dependent dielectric constant ( $\epsilon'$ ) of bulk Ba<sub>0.77</sub>Ca<sub>0.23</sub>Pr<sub>0.01</sub>Ti<sub>0.99</sub>O<sub>3</sub> ceramics at different frequencies (1 kHz to 1MHz). The figure shows, the value of temperature dependence permittivity increases gradually to a

maximum value ( $\epsilon_m$ ) with increase in temperature up to the transition temperature and then decreases smoothly indicating a phase transition. The maximum of dielectric permittivity,  $\epsilon_m$ , and the corresponding maximum

temperature  $T_m$ , depend upon the measurement frequency. The magnitude of dielectric constant decreases with increase in frequency and the maxima are shifting towards higher temperature. This indicates that the dielectric polarization is of relaxation type in nature. At 1 kHz, the dielectric maximum ( $\epsilon_m$ ) of  $Ba_{0.77}Ca_{0.23}Pr_{0.01}Ti_{0.99}O_3$  ceramic and the observed high value of  $T_m$  is due to the higher grain size. It is reported that as the grain size decreases, the maximum dielectric constant and transition temperature decrease [22]. The broadness or diffusiveness occurs mainly due to compositional fluctuation and structural disordering in the arrangement of cation in one or more crystallographic sites of the structure. This suggests a microscopic heterogeneity in the compound with different local Curie points. The nature of the variation of dielectric constant and non-polar space group suggest the ferroelectric phase transition of the material.

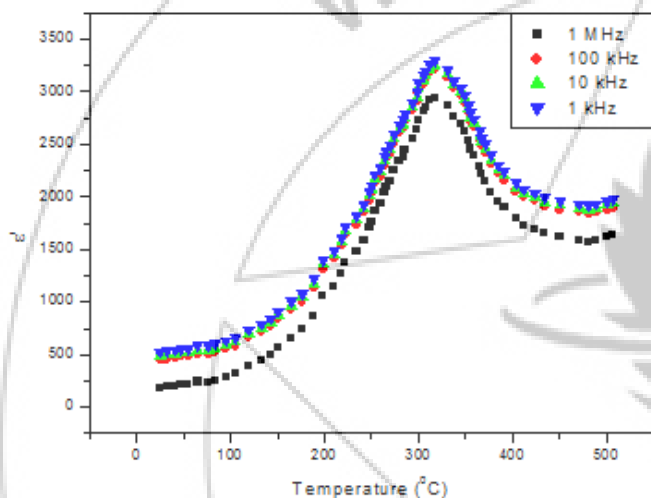


Figure 3: Dielectric constant ( $\epsilon'$ ) Vs. Temperature ( $^{\circ}C$ ) of  $Ba_{0.77}Ca_{0.23}Pr_{0.01}Ti_{0.99}O_3$  ceramics.

It is known that the dielectric permittivity of a normal ferroelectric above the Curie temperature follows the Curie–Weiss law described by

$$\epsilon = \frac{C}{T - T_0}, \quad (T > T_C)$$

where  $T_0$  is the Curie–Weiss temperature and  $C$  is the Curie–Weiss constant.

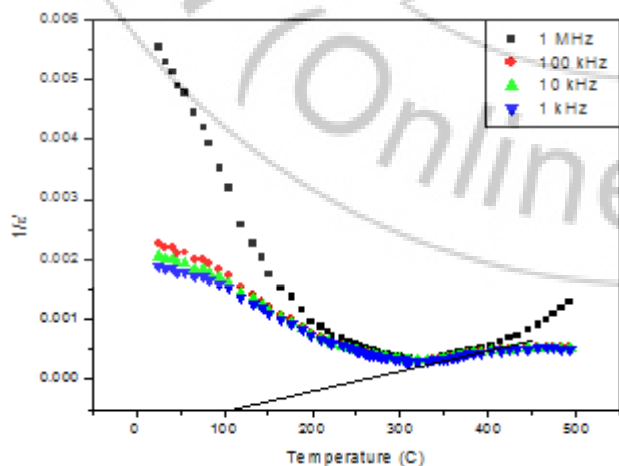


Figure 4: Inverse dielectric constant ( $1/\epsilon'$ ) Vs. temperature ( $^{\circ}C$ ) of  $Ba_{0.77}Ca_{0.23}Pr_{0.01}Ti_{0.99}O_3$  ceramics at different frequencies.

Fig. 4 shows the plot of inverse dielectric constant versus temperature at different frequencies. The different parameters obtained at 10 kHz are listed in Table-2. The parameter  $\Delta T_m$ , which describes the degree of the deviation from the Curie–Weiss law, is defined as

$$\Delta T_m = T_{CW} - T_m.$$

where  $T_{CW}$  denotes the temperature from which the permittivity starts to deviate from the Curie–Weiss law and  $T_m$  represents the temperature of the dielectric maximum. The Curie temperature is determined from the graph by extrapolation of the reciprocal of dielectric constant of the paraelectric region and the values obtained are given in Table-2.

The diffuse characteristics of ferroelectric–paraelectric phase transition are known to deviate from the typical Curie–Weiss behavior and can be described by a modified Curie–Weiss relationship [23–24],

$$\frac{1}{\epsilon} - \frac{1}{\epsilon_m} = \frac{(T - T_m)^\gamma}{C}$$

where  $\gamma$  and  $C$  are assumed to be constant. The parameter  $\gamma$  gives information on the character of the phase transition: For  $\gamma = 1$ , a normal Curie–Weiss law is obtained, and  $\gamma = 2$  describes a complete diffuse phase transition [25].

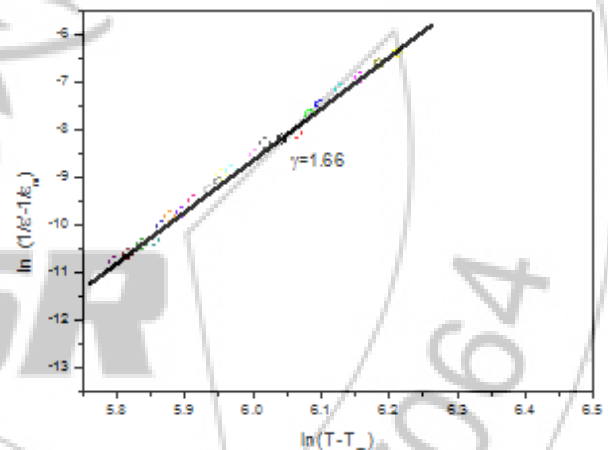
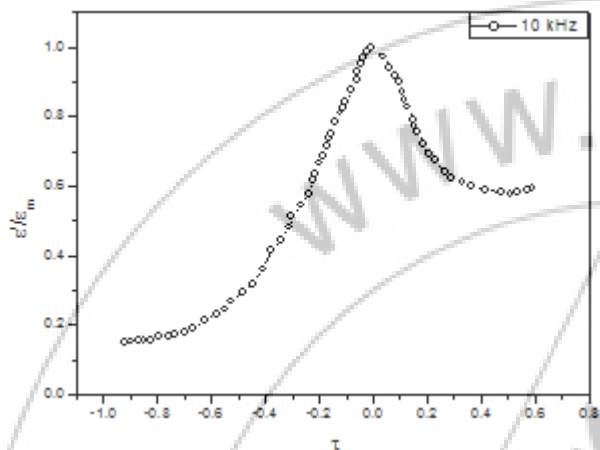


Figure 5:  $\ln(1/\epsilon - 1/\epsilon_m)$  vs.  $\ln(T - T_m)$  of  $Ba_{0.77}Ca_{0.23}Pr_{0.01}Ti_{0.99}O_3$  ceramics at 1 kHz.

Fig. 5 shows the plot of  $\ln(1/\epsilon - 1/\epsilon_m)$  vs.  $\ln(T - T_m)$ . The values of  $\gamma$  was found to be 1.66 at frequency 10kHz (Table-2). Thus, it is clear that, there is a clear diffuse phase transition. The diffuse phase transition and deviation from Curie–Weiss type behaviour may be assumed due to disordering. The diffusiveness occurs mainly due to the compositional fluctuation and structural disordering in the arrangement of cations in one or more crystallographic sites of the structures. The randomly distributed electrical field in a mixed oxide system is reported is to be the main reason leading to the relaxor behavior. Aliovalent cations incorporated in a perovskite lattice have been reported to serve as donors or acceptors, which could affect the electrical characteristics greatly, even though the solubility remained at the trace level.

**Table 3:** Temperature dependency dielectric parameters of the composition Ba<sub>0.77</sub>Ca<sub>0.23</sub>Pr<sub>0.01</sub>Ti<sub>0.99</sub>O<sub>3</sub> ceramics at 10kHz.

Parameters Sample	$\epsilon_m$	$T_m$ (°C)	$T_0$ (°C)	$T_{cw}$ (°C)	$\Delta T_m$ (°C)	$\gamma$	$\Delta T_{relax}$ (°C)
Ba <sub>0.77</sub> Ca <sub>0.23</sub> Pr <sub>0.01</sub> Ti <sub>0.99</sub> O <sub>3</sub>	3200.716	318.7578	230	320.5	1.74	1.66	0

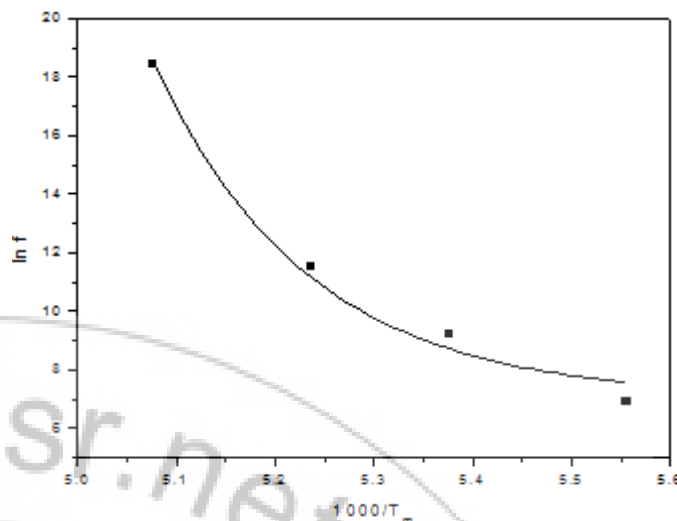


**Figure 6:** Reduced dielectric constant ( $\frac{\epsilon}{\epsilon_m}$ ) as a function of reduced temperature ( $\tau$ ) at 10 kHz.

The broadening of the phase transition is better illustrated by plotting the reduced dielectric constant ( $\frac{\epsilon}{\epsilon_m}$ ) as a function of reduced temperature ( $\tau$ ) at different frequencies (Fig.6). The full width of the plot has very little dispersion over a wide frequency range similar to the observation made in other relaxor materials [26]. Ba<sup>2+</sup> ions are replaced by Ca<sup>2+</sup> ions in the first stage, which have a smaller ionic radius. Moreover, aliovalent substitutions cause a distortion of the lattice. The interactions between B-site ions and O<sup>2-</sup> become stronger resulting in an increase in Tc and Pr<sup>3+</sup> tends to occupy B sites rather than A-sites. Because of the larger radius, the substitution of Pr<sup>3+</sup> for Ti<sup>4+</sup> in B-sites may depress the oriented displacement of B-site ions in the oxygen octahedrons, which are responsible for the spontaneous polarization. Yet another parameter, which is used to characterize the degree of relaxation behavior in the frequency range of 100 Hz to 10 kHz, is described [27]

$$\Delta T_{Relax} = T\epsilon_m(100kHz) - T\epsilon_m(10kHz),$$

The value of  $\Delta T_{relax}$  was determined to be zero K for the present sample. The above characterization done on the basis of Curie-Weiss law and the value of empirical parameters like  $\Delta T_m$ ,  $\gamma$ , and  $\Delta T_{relax}$  suggest that the permittivity of Ba<sub>0.77</sub>Ca<sub>0.23</sub>Pr<sub>0.01</sub>Ti<sub>0.99</sub>O<sub>3</sub> ceramic follows Curie-Weiss law only at temperatures much higher than  $T_m$ . Thus, the large deviation from the Curie-Weiss type behavior, large relaxation temperature  $\Delta T_{relax}$ , and  $\gamma = 1.66$ , suggests that Ba<sub>0.77</sub>Ca<sub>0.23</sub>Pr<sub>0.01</sub>Ti<sub>0.99</sub>O<sub>3</sub> is a relaxor ferroelectric.



**Figure 7:** ln f vs. 1000/ $T_m$  of Ba<sub>0.77</sub>Ca<sub>0.23</sub>Pr<sub>0.01</sub>Ti<sub>0.99</sub>O<sub>3</sub> ceramics at 1kHz

The frequency dependency of  $T_m$  is shown in Fig. 7 as ln f vs. 1000/ $T_m$ . The observed frequency dependence of  $T_m$  was empirically evaluated using Vogel-Fulcher's relationship given as

$$f = f_0 \exp\left[\frac{-E_a}{k_B(T_m - T_f)}\right],$$

where  $f_0$  is the attempt frequency,  $E_a$  is the measurement of average activation energy,  $k_B$  the Boltzman constant,  $T_f$  the freezing temperature of polarization fluctuation and  $f_0$  is the pre exponential factor. The fitting parameters for the compositions are found to be,  $E_a = 0.1145$  eV,  $T_f = 102$  K,  $f_0 = 102.3 \times 10$  Hz. The fitting parameters having close agreement with the data of Vogel- Fulcher's relationship suggests that the relaxor behaviours in the systems are analogous to that of a dipolar glass with polarization fluctuations above a static freezing temperature.

#### 4. Conclusion

The perovskite Ba<sub>0.77</sub>Ca<sub>0.23</sub>Pr<sub>0.01</sub>Ti<sub>0.99</sub>O<sub>3</sub> ceramic was prepared through a solid state reaction route. The room temperature XRD study suggests that the composition has a single phase cubic symmetry with space group Pm-3m. The dielectric study of the compositions shows a typical relaxor like behavior. The relaxor behavior observed in this ceramics can be induced by many reasons such as microscopic compositions fluctuation, the merging of micro-polar regions into macro-polar regions, or a coupling of the order parameter and the local disorder mode through the local strain. In perovskite-type compounds the relaxor behaviour appears when at least two cations occupy the same crystallographic sites A or B. Both Ti and Pr are ferroelectrically active and these cations are off-centred in the octahedral site giving rise to a local dipolar moment. A quantitative characterization of the relaxor behavior based on the empirical parameters ( $\Delta T_m$ ,  $\Delta T_{res}$ , and  $\Delta T_{cw}$ ) confirms the relaxor behavior of Ba<sub>0.77</sub>Ca<sub>0.23</sub>Pr<sub>0.01</sub>Ti<sub>0.99</sub>O<sub>3</sub> composition. Other rare earth may be doped and different structural and electrical properties can be studied.

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