Electric field induced phase transformation, electromechanical and multiferrite enhancement via doping in BFO

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**Abstract**

Polycrystalline BiFeO3(BFO) based ceramics B(.95)L(0.05)F(1-x)TxO3, (x=0.05, 0.1, 0.15) were prepared via simple solid state route with shorttime sintering temperature. The short time sintering and low A-site doping showed a significant role towards the controlling of Bismuth volatilization, oxygen vacancies [[1](#_ENREF_1), [2](#_ENREF_2)]and improving the overall behavior of BFO based ceramics.The suppressed inhomogeneous spin structure is observed due to structural distortion upon increasing Ti concentration on Fe site. The larger remnant polarization (Pr 20µc/cm2) and remnant magnetization (Mr 0.20 emu/g), were obtained for BLFO-Ti10%. The origin of the enhanced macroscopic magnetization in BLFTO ceramics is analyzed and discussed by considering the doping induced suppression of cycloidal spin structure The BLFO-Ti15% ceramics is found to be the induced phase boundarycomposition where colossal response is always expected by the external stimuli such as temperature, electric and magnetid field. We observed both the electric field induced phase transition for BLFO-Ti15% ceramics at above room temperature and the associated electromechanical response (i.e. the enhanced dielectric constant). Both the reduced leakage current measurement for BLFO-Ti15% ceramics and reduced dielectric loss upon structural phase transition confirms that the observed enhancement in dielectric constant is not due to leakage.

**INTRODUCTION**

Single-phase multiferroicmaterials exhibit more properties such as ferroelectricity, ferroelasticity and ferromagnetism etc. The coupling between magnetic and ferroelectric domains results in the linear magnetoelectric phenomena and offers applications in many fields including memory media, spintronics, sensors and magnetic reading[1-2]. However the magnetoelectric effect (ME) is not large enough in magnitude due to the exclusive between ferroelectricity and ferromagnetism [3]. In general, Curie temperatures of ferroelectric and ferromagneticfor a lot of multiferroric materials are lower than room temperature, this strongly limits their potential applications. A few of multiferroric materials simultaneously possess both phase transition temperatures from ferroelectric to paraelectric and ferromagnetic /antiferromagnetic to paramagnetic above room temperature. Some researchers found the candidate as BiFeO3(BFO) withperovskite structure. The antiferromagnetic Neel temperature for BFO is about 643K and ferroelectric Curie temperatureis about 1103K[4,5]. Because of its small band gap about 2.5 eV, which helps BFO to enhance its visible region absorbance,BFO may be applied as optoelectronic devices and photocatalyc materials.[appending some references] Beside all these potential applications the electric field induced structural transitions in BFO which leads the double-hooped Ɛ-E curve to quadruple-hooped Ɛ-E curves, which help the scientists to discover new piezoelectric materials with enhanced properties at the first order structural transition[7].

Although BFO has received the great attention from the investigators due to its potential applications, BFO has also some inherent problems, such as the pure phase synthesis, a large leakage current, the lower magnetoeletric coupling coefficient and a huge difference in ferroic transition temperaturesTC and TN. [8-11]The low resistivity and the high value of leakage current badly effect the ferroelectric properitesof BFO. It has been confirmed that BFO has an incommensurate spiral spin structure with a period of 62 nm[12]. This spiralspin structure from transition metal cation Fe3+results in the antiferromagnetic spins of BFO, the cancellation of net magnetization due to averaging over the period, that further prohibits the linear ME effect in BFO bulk ceramics. Different research groups tried different doping strategies to restrain the spiral spin structure, reduceleakage currents,enhance the ME effect and improve themultiferroic properties in BFO. Among all these the co-substitutions of Bi3+ ions by Nd3+, La3+ and Fe3+ ions partially by V5+, Mn4+ and Ti4+ have been proven to be effective for enhancing resistivity and thus improving the overall behavior of BFO[13-15]. The La3+ substitution on A-site is favorable for perovskite structurestability while on the other hand Ti4+ ions can reduce the charge defects and increasethe electrical resistivity of BFO [16-17]. It has been also found that high magnetic field [18] and doping [19,20] can suppress the cycloid spin structure and can release the hidden ferromagnetism and ME effect [21] in BFO ceramics. So far a comprehensive and overall picture on the substitutional effect of A and B site ions in BFO bulk ceramics is still missing. A worth mentioning consequence of the chemical substitution in BFO ceramics is the phase transition of parent compound from rhombohedral ferroelectric structure to another structure in the presence of external stimuli such as electric field, magnetic field and temperature. At the phase boundary the enhanced piezoelectric coefficient and dielectric constant are expected. The high leakage current might be the reason that prohibits the electric field induced structural phase transition and the associated enhanced functionality across the boundary. There are some reports available on co-substituted Sr2+, Ti4+, and Ca2+ and Ti4+ BFO [22,23], about the reduced leakage current and improved dielectric and magnetic properties, but the ferroelectric polarization was not included in these reports, it may because of the heavy doping on Bi3+ site and longtime sintering under high temperature that results in the Bi volatilization, which can affect the ferroelectric properties as the ferroelectric polarization in BFO is mainly contributed from Bi3+[24]. Therefore it is interesting to investigate the overall behavior of BFO using the idea of short time sintering, lightly A-site substitution by La3+ and partially B-site substitution by Ti4+ and in response expect some new features such as ferroelectric enhancement and electric field induced structural phase transition and the associated enhanced electromechanical properties.

In this worka pure phase BFO: La, Ti ceramic is obtained by the light Asite doping and short time sintering under high temperature. The origin of enhanced ferromagnetic response via doping in BFO,the chemical pressure induced phase boundaries and the electromechanical enhancement upon electric field induced structural transitionare discussed in detail.

**Experimental**

B(0.95)L(0.05)F(1-x)TxO3, (x=0.05, 0.1, 0.15) bulk ceramic samples were prepared via simple solid state reaction route with short time sintering using high puritypowders of La2O3, Bi2O3, Fe2O3, and TiO2 as reagents. Thesepowders were carefully weighed in stoichiometric proportion, grinded (or stirred) thoroughly in ethanol medium to obtain the homogeneous mixture. The first heat treatment was performed at 800 oC for 30 minutes witha heating rate of 5 oC/ min. The calcined powder was grinded again for 2 hours in order to obtain the more homogeneous mixture and then dried. The grindedpowders were then pressed into pellets under a pressure of about 5ton/ inch2 with 1 mm thickness and 10 mm diameter. Finally, the pellets were sintered at about 850 oC for 15 minutes withthesameheating rate. The crystal structure and phase purity of the samples were characterized by x-ray diffraction (XRD) (Rigaku smartlab) withCu Kα radiation. Field emissionscanning electron microscope (FESEM) was used to examine the microstructure of the surface. Dielectric characterization of the ceramic samples was carried out on Agilent 4294A precision impedance analyzer (Agilent Technologies, USA). The magnetic hysteresis and ferroelectric hysteresis loops of BLFTO bulk ceramics were measured at room temperature by SQUID-VSM (Quantum Design, USA) and Radiant Technologies Precision Premier II (Radiant Tech., USA), respectively. FTIR spectra were recorded by Bruker tensor 37 FTIR. Differential scanning calorimetry (DSC,Discovery DSC from TA instrument) measurements were recorded from room temperature to 510 oC at heating and cooling rate of 10 oC/ min. The oxidation state of Fe ions in BFO ceramics were examined by x-ray photoelectron spectroscopy (XPS, ESCALAB 250).

**Results and Discussion**

Figure 1 shows the XRD patterns for all B(0.95)L(0.05)F(1-X)TxO3, (X=0, 0.05, 0.1, 0.15) samples at room temperature. The impurity phasessuch as Bi2Fe4O9 andBi25FeO40observed by Kumar et al [XXX smart material, 2015] do not present in the current BFO sample.The strong and shape diffraction peaks reveal good crystallization for all samples.The splitting peaks around 320 and 390 for BLFO are response to thedistorted rhombohedral R3c structure.[XXX] The magnified patterns in the vicinity of 320 indicate that the peaks are well separated initially in BLFO samples, then gradually move and overlap into a single broadening peak with the increasing Ti concentration as seen in inset of Fig.1.This result is consistent with the early reports[33,34]. The peak redshift indicates the stronger distortion with increasing Ti. The distorted rhombohedral structure finally changes to anorthorhombicfor 15% Ti substitution. This kind of structural distortion also results inthe spontaneous polarization and the ferroelectric polarization enhancements.[33]

The SEM images in Fig. 2show the increasingdensity and stable grain size with Ti substitution in BLFO ceramic samples.

Ti4+ ions play the role of donor in BFO due to its higher valence as compared to Fe3+. The substitution of Ti4+ in BLFO samples need charge compensation that may be achieved by filling oxygen ion vacancies [38], which further results in the reduction of Fe 2+ ions (to be shown below in xps results ) with the increasing concentration of Ti substitution. The reduction in grain size by Ti substitution in BFO based ceramic samples can be interpreted by the suppression of oxygen ion vacancy concentration, which result in slower oxygen ions motion and consequently lower grain growth rate [39]. In addition to that the calcinations and sintering for a short period of time also suppressed the generation of oxygen ion vacancies.

The hopping electron mechanism between two Fe ions dominates the electronic conduction in BFO ceramics. The oxygen ion vacancies in the lattice act as a bridge between Fe2+ and Fe3+. The transition of Fe3+ to Fe2+ can be represented by the following equation.[35]

 (1)

To observe the significant role of Ti4+ substitution in suppressing oxygen vacancies and thus result in the reduction Fe2+ ions with increasing Ti content we used the XPS spectra for investigation. Fe 2p XPS spectra of Bi0.95La0.05Fe(1-x)TixO3 (x=0.05, 0.1, 0.15) bulk ceramics are shown in Fig.3(a). The expected binding energy of Fe 2p3/2 is 710.7eV and 709.3eVfor Fe3+ and Fe2+, respectively. An asymmetric broadband at about 710eV can be seen for all the BFO based ceramics, which confirms the oxidation state of Fe in our ceramic samples is the presence of both Fe2+ and Fe3+ ions. Because it cannot be avoid during synthesis of BFO based materials by high annealing process [36]. The standard Lorentzian fitting method is used to decomposethe Fe 2p3/2 into two sub bands, as shown in Fig. 3(b) for all Bi0.95La0.05Fe(1-x)TixO3 (x=0.05, 0.1, 0.15) bulk ceramics. The inset of Fig. 3(b) shows the content of Fe2+of iron ions in percentage. The decreasing of Fe2+ ions with the increasing Ti content shows the suppression of oxygen vacancies. The XPS strongly support the FESEM measurement, i.e., the decrease in grain size with Ti substitution suppress the oxygen ion vacancies that result in slower oxygen ions motion and consequently lower grain growth rate[37].

**Leakage current**

Figure 4 shows the leakage current for different Ti substituted BLFTO ceramic samples. Generally, the high leakage current in BFO is because of the presence of charged defects governed by Fe2+ ions, oxygen vacancies and bismuth volatilization [40-41]. Therefore, the high leakage currents reduce the electrical resistivity of the BFO ceramics and hamper its dielectric and ferroelectric properties. Here in our case the significant reduction in the leakage current with increasing Ti content clearly reflects the importance of Ti substitution towards the improved overall behavior of BFO based ceramics.

**Dielectric properties**

The frequency dependent dielectric constant (Ɛ) and loss tangent (tanδ) for all BLFTO samples are shown in Fig. 5. The higher value of dielectric constant (Ɛ) at lower frequency and remain fairly constant at higher frequency could be explained by the dipole relaxation phenomena, wherein at higher frequencies of applied field dipoles are unable to follow the field reversal while at lower frequencies the dipoles have enough time to follow the applied field[22]. These dipoles may arise from oxygen vacancies and localized charges. The oxygen vacancies are mainly due to the transition of Fe from Fe3+ to Fe2+ and volatilization of Bi that can be shown by rewriting eq. (1) and another eq. (2)

 (1)

 (2)

Here it is worth mentioning that we tried to control Bi volatilization by short time sintering instead of heavy A-site doping because heavy doping on A-site may result in vanishing ferroelectric polarization (to be discussed below). The creation of oxygen ion vacancies can increase the probability of hopping electrons conduction mechanism, that definitely result in larger value of dielectric constant and dielectric loss of BFO based ceramics. With Ti substitution in BLFO ceramics shown inFig.5, the larger value of dielectric constant and small dispersion at lower frequencies accompanied by the reduced dielectric loss with the increasing concentration of Ti substitution clearly reflect the significance of Ti substitution in BFO based ceramics. This can be explained as Ti4+ ions in BLFO ceramics requires charge compensation which can be achieved by 1) decreasing the cation valence i.e., reducing the Fe2+ ions. 2) Suppressing oxygen vacancies. Therefore for the requirement of charge compensation with Ti substitution in BLFO ceramics is expected to reduce the oxygen ion vacancies and thus reduce the associated leakage current that leads to the increase of frequency independent region. This observation is in complete agreement with both leakage current and XPS measurement of our BLFTO ceramics.

Figure 6 shows the temperature dependence of dielectric constant and tangent loses of B(0.95)L(0.05)F(1-X)TO3, X=0.5, 0.1, 0.15 ceramics. The anomaly reported for Y-doped [42], Mn-doped [43] and La and Pb co-doped [44] BFO, which has attributed to the transient interaction between oxygen ion vacancies and Fe3+/Fe2+ redox couple [42] has not been observed for our BLFTO samples. This clearly shows the significance of Ti substitution in BFO based ceramics in improving the dielectric properties. Also for Sr2+, Ti4+ and Ca2+, Ti4+ codoped BFO ceramics the anomaly has been observed, the reason is that the divalent substitution needs charge compensation which results in oxygen vacancies. At this point of view our approach towards controlling Bi volatilization by shorttime sintering instead of heavy doping on A-site is the best choice in improving the dielectric properties of BLFTO ceramic samples. Most important observation here is the electric field induced transition for BLFO-Ti15% ceramic samples. Such kind of electric field induced transition has already reported earlier for BFO [45]. The difference in energy per unit volume (∆U) between the two structures is [45]:

 (3)

Daisuke et al, [45] reported the difference in energy for transformation has been provided only in the form of electric field, but in our case the temperature is another candidate along with electric field to induce such transformations. The disturbance and sharp change in Ɛ-E curve has been observed for our BLFO-Ti15% sample at 359K and 365K respectively, which shows the presence of temperature along with electric field for structure phase transformation in 15-BLFTO ceramics. Such kind of behavior for Ɛ-E curve has also been seen for temperature dependent dielectric measurement of NaNbO3 [46]. This kind of electric field induced phase transformation results in the enhancement of electromechanical response such as enhancement of piezoelectric coefficient and dielectric constant at the structural phase transition. We have shown the electric field induced phase transition and the associated enhanced dielectric constant for BLFO-Ti15% bulk ceramics(induced phase boundary composition) shown in Fig. 5(a) . The leakage current also found to reduce for BLFO-Ti15% ceramics, this with addition to the reduction in the dielectric loss across the boundary shown in Fig.5(b)further confirms that the enhancement in the dielectric constant is not due to leakage. The resulting enhancement in the piezoelectric properties due to such kind of electric field induced transformations has been reported for Pb-based ferroelectric materials [47-51].

**Ferroelectric properties**

Ferroelectric polarization is one of the most promising propertiesof BFO ceramics. The ferroelectric polarization in BFO originates from A site i.e., the central cause of this promising property is the Bi3+ lone pair (6s2 orbital). However high annealing temperature during the synthesis of BFO ceramics can cause the Bi volatilization which is one of the main reason of oxygen vacancies generation, that makes BFO conductive and hamper its dielectric properties while on the other hand, the volatilization of Bi3+ means the absence of lone pairs that results in the vanishing of ferroelectric polarization in BFO. So the approach to control the Bi volatilization by heavy A-site substitution such as Sr2+, Ca2+ co-doped with Ti [22,23] in BFO ceramics can only promise the improved dielectric properties but cannot guarantee the good ferroelectric response. Even for the only B site partial substitution in BFO only produced a small value of ferroelectric polarization (0.8µc/cm2) because of its longtime annealing under high temperature which causes the Bi volatilization [52]. At this point of view, our approach of shorttime sintering with high temperature instead of heavy A-site doping by divalent ions to keep alive the ferroelectric polarization is seemed logical. Fig.7 shows the ferroelectric hysteresis loops for B(.95)L(0.05)F(1-X)TxO3 (x=0.05, 0.1) ceramics, measured at room temperature at the electric field of 100KV/cm. the ferroelectric polarizations for Ti5% and Ti10%are5µc/cm2 and 20uc/cm2, respectively. The ferroelectric enhancement for Ti10% of about 4 times as compared to Ti5% samples is attributed to the structural distortion of BLFTO ceramics shown in Fig.1 (b). The similar structural distortion and ferroelectric enhancement has been reported earlier [53]. Here the most important and worth mentioning observation is the distorted hysteresis loop for BLFO-Ti15% sample measured at room temperature and at a very low electric field. This clearly reflects that we are at the phase boundary for Ti15% concentration [54] where the colossal response is largely expected in reaction to weak external stimuli such as temperature, electric field or magnetic field which is consistent with the early reported work [47-51]. There is the region of double hysteresis loop beyond the phase boundary, but we did not get through because we were lack of high temperature, as PE loops were taken at room temperature. This observation completely agrees with our Ɛ-E curves measurements for Ti15% samples where the electric field induced transformation is in the presence of temperature, which reflects the presence of temperature is necessary for such electric field induced transformation in our case. Our this observation is also in agreement with Yang et al [54] that not only the change of concentration of dopant is required for inducing phase boundary but also strongly depend on the pathway taken by the system. Electric field induced structural transition in BLFO-Ti15% ceramics synthesis by a conventional solid state route is a significant achievement because the associated electromechanical enhancement such as enhanced dielectric constant (achieved in our case) and piezoelectric coefficient (achieved by A or B-site substitution) are very well known in perovskite ferroelectrics and has a great importance because of its enhanced functionality.

**Raman spectra**

To get the further structural informations with the increasing Ti concentration of BLFTO bulk ceramics, the Raman spectra were recorded within the wave number range 50-800 cm-1 as shown in Figure 8. the clearly resolved Raman modes of BiFeO3 and B(.95)L(0.05)F(1-x)TxO3, (x=0.05, 0.1, 0.15) are all indexed to the mode of BFO molecules. The intensity for almost all the peaks reduced for all the doped samples but no significant changes has been found for A1 (LO) mode in the Raman spectra with the increasing concentration of Ti doping on Fe site which is an agreement with the Kheral et al [ref] that doping on the Fe site produce no structural distortion about the Bi site. However, the emergence of the E-1 mode and the disappearance of A-2 mode in the doped samples clearly show the structural modifications with Ti doping on Fe site. The Raman spectra analysis is in complete agreement with our XRD measurement that the increasing concentration of Ti doping on Fe site produces structural distortion that further support our argument that the potential cause of the observation of macroscopic magnetization at room temperature for our BLFTO ceramics is the structural distortion that suppress the inhomogeneous spin structure.

**Magnetic properties**

The magnetic structure of BFO is known to be G-type Antiferromagnetic with spin ordering modulated by a cycloidal over a period of 620 ± 20 Ao .[58] This cycloid structure results in the absence of macroscopic magnetization in BFO because of the averaging over the period. Many attempts have been made to suppress this cycloid spin structure by doping [59,60], strain or high magnetic field [61] that may result in the macroscopic magnetization in BFO based materials. The room temperature magnetic hysteresis loops for BLFTO ceramics measured at a maximum of 4 Tesla are shown in Fig.9 (a) which clearly reflects the ferromagnetic nature. The enhancement of macroscopic magnetization in BFO based ceramics is due to the formation of Fe2+ ions [62, 63] (which can possibly cause the double exchange mechanism between Fe2+ and Fe3+), increasing of canting angles [63] and suppressing of cycloid spin structures [64]. In order to verify the origin of the macroscopic magnetization, we need to recall the XPS measurements for BLFTO ceramic samples. As shown in the inset of fig.9 (a) the Fe2+ contents decrease with increasing Ti concentration (due to controlling the oxygen vacancies) that results in the enhancement of electric properties(dielectric and ferroelectric properties) of BLFTO ceramics, but this can possibly decrease the macroscopic magnetization. At this point of view, the potential causes for the enhanced macroscopic magnetization are the increasing canting angle and/ or the suppressing cycloid spin structure by Ti doping in BLFO ceramics. The inhomogeneous or cycloid spin structure of BFO is correlated to its R3C structure [59]. Ti doping in BLFO ceramics leads to the structural distortion (as shown by the XRD measurements in fig.1 (b) which can suppress the cycloid spin structure. The latent and unrealized magnetization released that was previously locked within the cycloid. This causes the macroscopic enhancement of magnetization for BLFTO ceramics. This observation is in complete agreement with XPS and XRD measurements. The macroscopic magnetization decreases for BLFO-Ti15% ceramics which mainly due to the monotonic decrease of Fe2+ ions (as shown in XPS measurements), enhanced second phase (shown in XRD measurements) and the increased defects. The DSC curves measured for all BLFTO ceramics are shown in Fig 9(b). The Neel temperature for BFO ceramics is 370 0C, the inset of Fig.9 (b) shows the Neel temperature for all BLFTO ceramics. The reduction in the Neel temperature TN and the enhanced macroscopic magnetization is in agreement with the early reported work [65].

**Conclusion**

B(.95)L(0.05)F(1-x)T(x)O3, (x=0.5, 0.1, 0.15) polycrystalline bulk ceramics has been prepared via solid state route with short time sintering temperature. The idea of the low A-site doping by La and short time sintering temperature was used to stabilize the phase and to overcome the Bi volatilization. The increasing Ti concentration in BLFO ceramics resulted in structural distortion and significantly decreased the grain size. The XPS measurements revealed the monotonic decrease in the formation of Fe2+ ions with Ti concentration that shows the significant decrease in the generation of oxygen vacancies during the synthesis of BLFTO bulk ceramics. The significant reduction in the leakage current and the decrease in both the dielectric constant and dielectric loss with Ti substitution in BLFO ceramics were observed. The increased frequency independent regions and the stability of dielectric constant with regard to temperature during the dielectric properties measurement is in complete agreement with the XPS results. We have reported in this paper that the potential cause of the enhanced macroscopic magnetization in BLFTO bulk ceramics is the suppressed cycloid spin structure and/or increased canting angle, both the XPS measurement (monotonic decrease in the Fe2+ ions with Ti substitution) and XRD measurement ( the structural distortion with Ti substitution) completely supports our this observation. Meanwhile, as the Ti concentration increases, the ferroelectric rhombohedral phase undergoes a phase transition, showing a distorted hysteresis loop at room temperature. Across the phase boundary, the enhanced functionality such as enhancement in piezoelectric coefficient and dielectric constant etc are expected by the external stimuli such as electric field, magnetic field and temperature. We have shown the enhancement of the dielectric constant across the phase boundary for the BLFO-Ti15% bulk ceramics at a temperature of about 360 K and at a constant electric field. Our current observation of the enhanced dielectric properties because of the electric field induced phase transition at the temperature above room temperature in BFO based bulk ceramics prepared by simple solid state route provides the platform for the search of other materials with enhanced functionality at the phase transitions.

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Figure Captions:

Figure 1 X-ray diffraction patterns of B(.95)L(0.05)F(1-x)TxO3, (x=0.05, 0.1, 0.15) BFO bulk ceramics.Inset is the magnified patterns in the vicinity of 2θ=32°.

Figure 2 The SEM images of (a)BLFO-Ti5%, (b)BLFO-Ti10%, and (c)BLFO-Ti15%

Figure 3 (a) Fe 2*p* XPS spectra of B(.95)L(0.05)F(1-x)TxO3, (x=0.05, 0.1, 0.15) BFO bulk ceramics. (b) Lorentzian dividing of the Fe 2*p*3/2 core level for a  
B(.95)L(0.05)F(1-x)TxO3, (x=0.05, 0.1, 0.15) BFO bulk ceramics. The inset in 3(b) shows Ti composition dependent Fe2+ content.

Figure 4 Leakage current for all B(.95)L(0.05)F(1-X)TxO3, X=0.5, 0.1, 0.15, sintered at 840c0 for 15 minutes.

Figure 5 Frequency dependence of (a) the dielectric constant (Ɛ) and (b) the  
dielectric loss (tan δ) of all the samples at 573 K.

Figure 6 Temperature dependence of (a) the dielectric constant (Ɛ) and (b)  
the dielectric loss (tan δ) for all samples at 1 MHz.

Figure 7 (a) and (b) shows the PE hysteresis loops at 100KV/cm and at room temperature for 5-Ti and 10-Ti BLFTO bulk ceramics respectively. Fig.7. (c) shows the distorted hysteresis loop taken at 3.4KV/cm and at room temperature for 15-Ti BLFTO bulk ceramics which shows the induction of the phase boundaries for this composition.

Figure 8 IR absorption spectra of B(.95)L(0.05)F(1-x)TxO3, (x=0.05, 0.1, 0.15).

Figure 9 (a) Field dependence of magnetization, the inset shows the magnified M-H curves. (b) DSC signal vs. temperature traces for B(.95)L(0.05)F(1-x)TxO3, (x=0.05, 0.1, 0.15), the inset shows the Ne´el temperature of BFO as a function of Ti doping contents.



Fig.1.

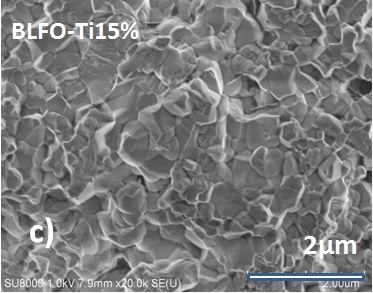
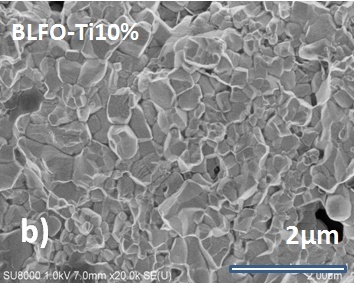
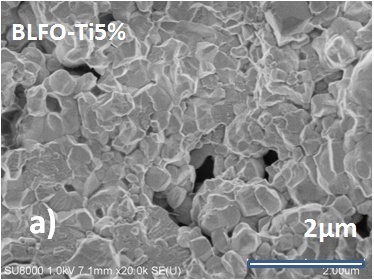


Fig.2.





FIG.3.



Fig.4.



576K 270K



576K 270K

Fig.5.



1MHz 40 Hz



1MHz 40 Hz

Fig.6.



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Fig.7.



Fig.8.





Fig.9.