

Enhancement in magnetic, piezoelectric and ferroelectric properties on substitution of titanium by iron in barium calcium titanate ceramics



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ABSTRACT

Lead free polycrystalline ceramics with compositional formula $\text{Ba}_{0.90}\text{Ca}_{0.10}\text{Ti}_{1-3x/4}\text{Fe}_x\text{O}_3$ (BCT), ($x=0.000, 0.005, 0.010, 0.015$ and 0.020) were prepared by the solid state reaction method. The effect of substitution of Fe^{3+} ion at Ti-site on the ferroelectric and piezoelectric properties of BCT ceramics was studied. Remanant polarisation (P_r) and saturation polarisation (P_s) show an increasing trend while the reverse trend was observed for coercive field (E_c) with Fe^{3+} substitution. The values of the piezoelectric charge coefficient (d_{33}) and piezoelectric coupling coefficient (k_p) was found to increase with increase in Fe content. Ceramic sample with $x=0.02$ was found to have a maximum value of d_{33} (130 pC/N) and k_p (29%). The prepared ceramic samples show magnetic properties as confirmed by recorded MH loops. On substitution of Fe^{3+} ions, the ferroelectric BCT ceramics show clear transformation of the diamagnetic nature of BCT ceramics to weak ferromagnetism.

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1. Introduction

Polycrystalline ferroelectric ceramics are of great interest because of their ease of preparation and achievement of various desired properties by compositional modifications as compared to single crystals [1]. Piezoelectric materials have wide applications as sensors, actuators, charge storage devices, multilayer ceramic capacitors, high-frequency (GHz) bypass capacitors, infrared detectors and FRAMs [2,3]. Piezoelectric sensitivity is high for lead based ferroelectrics but because of toxicity of lead, there is a thrust on research of lead free materials. Barium titanate (BT) has been widely studied lead free ferroelectric and piezoelectric material but they have low ferroelectricity and piezoelectricity as compared to lead based materials [4]. It is often possible to enhance piezoelectric sensitivity by varying the composition to obtain “soft” ceramics that tend to have more temperature dependence, hysteresis and aging of properties [5]. Ca substituted Barium titanate (BCT) is widely used as a ceramic capacitor material due to improvement in temperature dependency and dielectric properties [6–10]. These materials are promising candidates for application in optical connectors, storage devices, photorefractive and advanced laser systems [11]. Furthermore, substitution of Ca^{2+} for Ba^{2+} in BT reduces the possibility of formation of non-ferroelectric undesirable hexagonal phase [12]. Transition metal doped Barium

titanate ceramics are widely investigated as single phase multi-ferroic materials, having more than one ferroic orders. There is a great advantage of doping of transition metal ions in a ferroelectric perovskites due to availability of both electric polarisation and magnetisation for storing data. Their magnetic ordering can be controlled by applied field, polarisation or lattice strain. Ferromagnetism in Barium titanate with substitution of transition metals has been well reported in literature [13–17]. Among transition metals, Fe^{3+} is a good choice for substitution of Ti^{4+} in BT to tailor various properties. An appreciable change has been observed in structural and dielectric properties on substitution of Ti ions by Fe ions in $\text{Ba}_{0.90}\text{Ca}_{0.10}\text{TiO}_3$ ceramics [10]. The aim of present study is to investigate the influence of substitution of Ti ions by Fe ions on piezoelectric, ferroelectric and ferromagnetic properties of $\text{Ba}_{0.90}\text{Ca}_{0.10}\text{TiO}_3$ ceramics.

2. Experimental

Polycrystalline ferroelectric ceramics with compositional formula $\text{Ba}_{0.90}\text{Ca}_{0.10}\text{Ti}_{1-3x/4}\text{Fe}_x\text{O}_3$ ($x=0.000, 0.005, 0.010, 0.015$ and 0.020) were prepared by solid state reaction method with raw materials BaCO_3 , CaCO_3 , TiO_2 , and Fe_2O_3 [10]. Morphology and microstructural uniformity of the samples was analysed using Scanning Electron Microscopy (SEM, Carl ZEISS Supra 55). Polarisation (P) versus electric field (E) hysteresis loops were recorded at 50 Hz from 30 to 120 °C using modified Sawyer-Tower circuit. For measuring piezoelectric properties, samples were poled at

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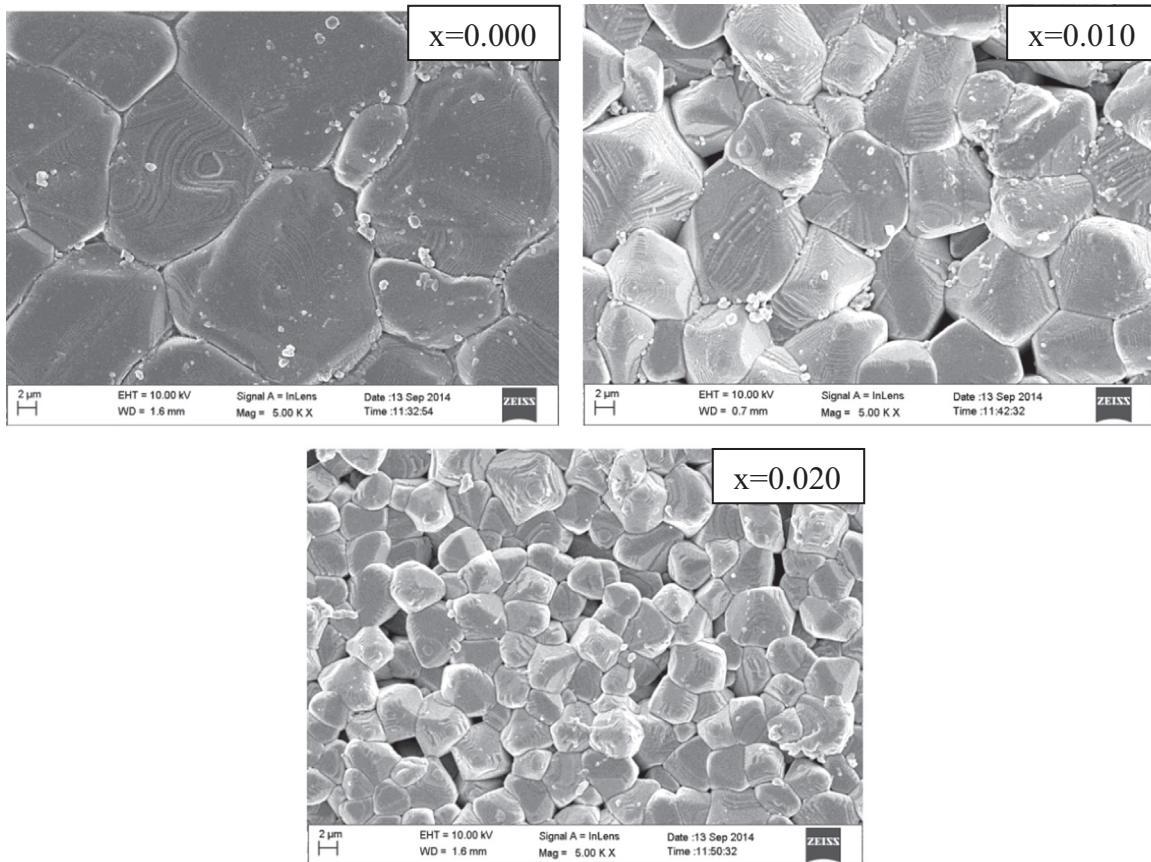


Fig. 1. SEM micrographs of samples with $x=0.000$, 0.010 and $x=0.020$.

Table 1

Coercive field (E_c), Remanant polarisation (P_r), Saturation polarisation (P_s), Piezoelectric charge coefficient (d_{33}), electromechanical coupling coefficient (k_p) and grain size of $\text{Ba}_{0.90}\text{Ca}_{0.10}\text{Ti}_{1-3x/4}\text{Fe}_x\text{O}_3$ ceramic for different values of x .

x	E_c (kV/cm)	P_r ($\mu\text{C}/\text{cm}^2$)	P_s ($\mu\text{C}/\text{cm}^2$)	d_{33} (pC/N)	k_p (%)	Grain size (μm)
0.000	5.39	7.295	16.114	107	20.784	18.71
0.005	5.01	7.312	18.235	109	21.438	12.41
0.010	4.98	7.452	18.602	119	22.291	9.92
0.015	4.89	8.006	18.935	122	26.497	6.08
0.020	4.27	9.359	20.831	130	29.692	4.30

85 °C in silicon oil by applying an electric field of 10 kV/cm. The longitudinal piezoelectric charge coefficient d_{33} was measured using Piezometer system of Concord Electroceramics Ind., India. Magnetic properties of the samples have been studied using Vibrating Sample Magnetometer (VSM, Microscience E-29).

3. Results and discussion

The morphology of the prepared polycrystalline ceramics was studied using Scanning Electron Microscope (SEM) at room temperature (30 °C). SEM micrographs for $x=0.000$, 0.010 and 0.020 are shown in Fig. 1. The average grain size for each sample was calculated and given in Table 1. It is observed that the average grain size decreases with increase in Fe content which indicates that present sintering conditions favour slow kinetics of inter-diffusion. Fe substituted ceramics were also found to have uniform grain size distribution and regular morphologies [18,19].

PE hysteresis loops for all the samples at 50 Hz are shown in

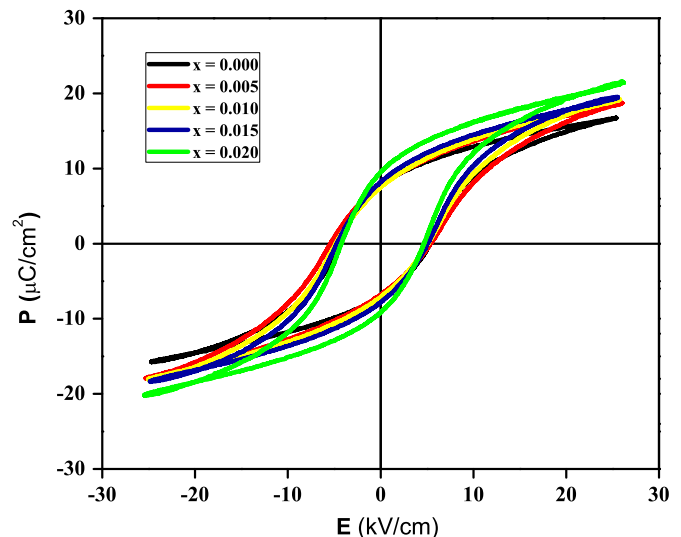


Fig. 2. PE hysteresis loops at room temperature for all samples.

Fig. 2. Ferroelectric properties were found to have strong dependency on Fe content. However, they are also influenced by various factors such as phase transition, particle size, ionic radii, temperature, and polarisability [20]. From the PE loops P_r , P_s and E_c were recorded and given in Table 1. It can be observed that P_r and P_s increase with increase in Fe content while E_c shows the reverse trend. The increase in P_r indicates enhancement in polarisation value by substitution of Ti^{4+} ions by Fe^{3+} ions. This might be attributed to the fact that Fe–O bonds are less ionic than Ti–O bonds [21]. While the decrease in E_c with Fe content may be

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