Frequency and temperature dependent dielectric studies of BaTi$_{0.96}$Fe$_{0.04}$O$_3$

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Abstract. A Finest possible sample of 4% Iron doped BaTiO$_3$ (BTO) with possible tetragonal structure via a solid state route was prepared. Prepared sample was characterized by X-ray diffraction (XRD) using Bruker D8 Advance XRD instrument, the value of 2θ is in between 20° to 80°. Detailed analysis of dielectric constant, dielectric loss, ac conductivity and electrical modulus at various range of frequency and temperature have been done of 4% Fe doped BTO was recorded on hp-Hewlett Packard 4192 A, LF impedance, 5Hz-13Hz analyser.

1. Introduction
Barium titanate is a ferroelectric ceramic material with ABO$_3$ type perovskite structure [1]. It exhibits ferroelectric properties at and above room temperature, and finally because it can be easily prepared and used in the form of ceramic polycrystalline samples. Due to its high dielectric constant and low loss characteristics, barium titanate has been used in applications, such as capacitors and multilayer capacitors (MLCs) [2]. Doped barium titanate has found wide application in semiconductors, PTC thermistors and piezoelectric devices, and has become one of the most important ferroelectric ceramics.

2. Synthesis of iron doped barium titanate
We started from highly pure fine powdered samples of BaCo$_3$, TiO$_2$ and Fe$_3$O$_4$ for bulk sample of doped BaTiO$_3$. All these were mixed in the calculated percentage ratio. Samples were ground for 8-hours and heat treatment was given at 900°C for 24-hours.

3. Experimental technique
3.1. X-ray Diffraction
The X-ray was produced using a sealed tube and the wavelength of X-ray was 0.15nm (Cu-Kα). XRD pattern of the Fe-doped BaTiO$_3$ at 900°C. The XRD data has concluded that all major peaks of Fe-BTO are matching with reported XRD data of pure BTO, and sample is in single phase. Which shows doping is perfect. Fig 1 shows that all the peaks in the pattern are matching and it’s showing purely tetragonal single phase crystal of the BaTiO$_3$ ceramics. The single phase crystal of the BaTiO$_3$ ceramics are tetragonal symmetry in the space group P4mm and pattern matched with the standard pattern PCPDF, no. 81-2203.
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Figure 3.1. XRD pattern of BaTi$_{0.96}$Fe$_{0.04}$O$_3$ doped BTO at 900$^\circ$C

3.2. Dielectric constant measurement

3.2.1. Dielectric constant

First a pallet of 12mm diameter of bulk sample was prepared and then coated both sides of the sample with low temperature silver paste. Put the sample in between two electrodes in furnace and obtain the value of dielectric constant with variation of temperature manually. In fig 3.2.1(a) after doping Fe in pure BTO, on increasing the temperature on higher side, the transition temperature was shifted from 120$^\circ$C to 118$^\circ$C. Figure shows the temperature dependence of dielectric constant in Fe ion substituted at the Ti site of BTO. The dielectric studies indicated a shift in the transition temperature ($T_c$) towards the lower temperature side.

![Dielectric Constant vs Temperature](image_url)

**Figure 3.2.1(a).** Temperature dependent Dielectric Constant.

Figure 3.2.1(b) & (c) shows a strong increase of $\varepsilon''$ (also $\varepsilon$) at low frequency regime is attributed to interfacial polarization. The magnitude of $\varepsilon'$ slowly decreases with increase of frequency before leaving off a constant value at higher frequency. This is a commonly observed feature in ceramic materials.
3.2.2. Dielectric loss

The dielectric loss (tan δ) is an important parameter that determines the suitability of a dielectric material in microelectronics. The Fe doped BaTiO₃ sample have shown thermal activated sharp loss peak. The peak position of sample BaTiO₃ shift towards higher frequency with the increase in temperature. The high dielectric loss at lower frequency in Fe doped BaTiO₃ sample is the effect of interfacial loss but dielectric loss is below 0.1 at higher frequency.

Figure 3.2.2. Variation of Dielectric loss (tan δ) with frequency at selected temperature.

Figure 3.3.3. Variation of σ'(f) at selected temperature.
3.3.3. AC conductivity

Fig 3.3.3 shows the frequency dependence of the real part ($\sigma'$) of ac conductivity at selected temperatures in the range 29°C to 130°C for BTFO-bulk sample. BTFO sample showed nearly a frequency independent conductivity at lower frequencies. BTFO showed a conductivity minimum in between the frequency independent and frequency dependent conductivity regimes. This minimum becomes prominent at higher measurement temperature.

3.3.4. Electrical modulus

The main advantage of applying modulus formalism is to suppress the electrode effect on electric conductivity contribution BTFO-bulk sample. Fig 3.3.4(a) and (b) shows sharp increases of M' below 100Hz followed by a slow increase with frequencies and finally saturated above 100KHz. The imaginary part of modulus (M'') shows a sharp peak at lower frequency and the peak position shift to higher frequencies with the increase of measurement temperature.

![Figure 3.3.4 (a). Frequency dependent modulus (M') at different temperature for BTFO-bulk sample.](image1)
![Figure 3.3.4 (b). Frequency dependent modulus (M'') at different temperature for BTFO-bulk sample.](image2)

4. Discussion

The XRD pattern showed that this material crystallizes in a perovskite-type single phase tetragonal structure with space group p4mm. In dielectric constant measurement, the transition temperature of Fe-BTO is occurring at 118°C. The reported value of transition temperature of pure BTO is 120°C. The high dielectric loss at lower frequency in Fe doped BaTiO$_3$ sample is the effect of interfacial loss but dielectric loss is below 0.1 at higher frequencies. The conductivity of BTFO sample showed nearly a frequency independent conductivity at lower frequencies. The imaginary part of modulus (M'') shows a sharp peak at lower frequency (below 100Hz) and the peak position shifts to higher frequency with the increase of measurement temperature. In conductive relaxation process the real part of modulus (M') generally exhibits a sharp monotonic increase with frequency.

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References

[1] Jona F and Shirane G 1993 Ferroelectric Crystals (New York: Dower publications)
[2] Maga D, Igor P and Sergei M 2000 J. Mater. Chem. 10 941
[3] Jona A, Kundu T K, Pradhan S K and Chakrvorty D 2005 J. Appl. Phys. 97 44311
[4] Ismailzade I H and Ismailov R M 1980 Phys. Stat. Sol. (a) 59 K 191
[5] Smolenski G A and Isupov V A 1954 Dokl. Akad. Nauk. SSSR 9 653