A case study on the electric and dielectric response of ferroelectrics

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Abstract. In this work, the electrical properties of ferroelectric compounds have been reviewed. But the main focus is on dielectric properties of ferroelectrics. Though ferroelectrics are well-known for ferroelectric devices, it is identified that ferroelectrics also have great diversity as dielectric materials. So we have gathered different published articles, and required information from some internet sources. The main theme of this paper is to report the work done on dielectric materials. The dielectric properties presented include the values of dielectric constant at room temperature, maximum dielectric constant, Curie temperature (Tc), etc. The electrical properties reported include output voltage, short circuit current/current density, power density, etc. Also, a basic understanding on the dielectric behavior of ferroelectrics has been presented.

Keywords: Dielectrics; Ferroelectrics; Electrical Properties; Ceramics and Films;

1. Introduction

The materials reviewed in this work are well-known ferroelectric materials. Because, they show large values of polarization, and this polarization can be switched from one state to the other state. Ferroelectric is a material that exhibit some amount of spontaneous electric polarization, even in the absence of alternating electric field. This property was discovered by Valasek in 1920, in Rochelle salt [1]. One can refer to the published papers, for better understanding on the history of ferroelectricity [2-8]. The ferroelectrics include barium titanate (BaTiO₃), bismuth ferrite (BiFeO₃), calcium titanate (CaTiO₃), lead titanate (PbTiO₃), etc. These compounds contain different crystals within them, where the arrangement/orientation of dipoles within a crystal is said to be unvaried. These crystal regions are generally known as domains. The separation between the domains is usually known as domain walls. On the application of alternating electric field externally, the domains tend to follow the field and orient accordingly. On the reverse application of the same field, domains switch to a new direction with respect to the field orientation. At a specific temperature, called as Curie temperature (Tc), the ferroelectricity appear to cease. Because, the thermal heat stimulates the orientation of dipoles to align in the direction of field [1]. So, the temperature at which all the possible dipoles orient in the direction of field is known as Tc. Below and above Tc, the material is said to be ferroelectric and paraelectric respectively. Hence, Tc helps to identify the phase transition (from one to another state). This phase transition actually represents how the material is transforming its state, behavior/properties, with respect to an external stimulus such as heat/light, mechanical stress, etc. Since the behavior of ferroelectrics is dependent on constant or variable temperature, the ferroelectrics can be taken as a system of thermodynamic. Just like for the thermodynamic systems, phase transitions are categorized into two types for ferroelectric systems. One is the first order ferroelectric phase transition and other is the second order ferroelectric phase transition. This division is based on the variation of polarization parameter, as a function of temperature [1, 9].

1st order ferroelectric transition: The polarization of a ferroelectric decreases, with increase in temperature, and shows a discontinuity at specific temperature i.e. Tc. (refer figure 1)

2nd order ferroelectric transition: The polarization of a ferroelectric decreases continuously, with increase in temperature, and shows a discontinuity at specific temperature i.e. Tc. (refer figure 1)
Figure 1. Types of Phase Transitions of Ferroelectrics

Actually, the concept of phase transitions of a thermodynamic system is basically discussed in terms of the type of transition. Because the transition can be either continuous or discontinuous. Based on the variation of free energy of a thermodynamic system, with respect to a varying thermodynamic quantity, phase transitions are categorized into first order and second order. But they were found to be inefficient in explaining the behavior of free energy. For example, the heat capacity of a ferromagnetic transition tends to infinity. Then based on latent heat, new and classy definitions have been developed. According to the modern definitions, a transition that is engaged by latent heat is known as first order phase transition. Examples include freezing, boiling, etc. The phase transitions that are said to be continuous, are known as second order phase transitions. Examples include the ferromagnetic transition, phase transition of superconductors, etc [1, 9]. The transition behavior of both ferromagnet and ferroelectric are similar to each other. For ferroelectrics, when the temperature is incremented, the polarization is reduced and become negligible at $T_c$.

The presence of ferroelectricity can be identified from the nature of dielectric constant of the material. The behavior dielectric constant, indeed, depends on various factors such as method of preparation, purity of raw materials, grain size, external electric field (under certain frequency, temperature and intensity), type of dopants and modifiers (and their concentration), etc [10]. The phase transition of a ferroelectric can be visualized as it is shown in figure 2. As discussed, phase transition happens at a specific temperature ($T_c$) indicating a dielectric peak. The peak represent the transition of material phase from ferroelectric to non-ferroelectric (paraelectric) phase.
Figure 2. Illustration of Phase Transition of an Ideal Ferroelectric

If the peak is very sharp (or slightly broader), then material is said to be ferroelectric. If peak is broader or wider, then material is said to be relaxor ferroelectric. At $T_c$, the value of permittivity is known as maximum dielectric constant ($\varepsilon_{\text{max}}$).

The thermal energy of atoms of ferroelectrics will increase, with increment in temperature. If the thermal energy increases continuously, this can overpower the energy barrier of polarization. When the temperature is paramount, the material become paraelectric dielectric. When the ferroelectric is in paraelectric phase, the nature of permittivity as a function of temperature can be explained by Curie Weiss law [9]:

$$\varepsilon = \frac{C}{T-T_c},$$

where $C$, $T$ and $\varepsilon$ are constant, temperature and permittivity respectively. The above expression can be further modified as follows:

$$\frac{1}{\varepsilon} = \frac{T-T_c}{C}.$$

Finally we get the modified equation as follows:

$$\frac{1}{\varepsilon} = \frac{T}{C} - \frac{T_c}{C}.$$

The magnitude of constant $C$ can be obtained from the variation of $\frac{1}{\varepsilon}$ with temperature. When the $\frac{1}{\varepsilon}$ is plotted as function of temperature (as shown in figure 3), in an ideal case, the shape of the graph will be almost linear for which the slope ($\frac{1}{\varepsilon}$) and intercept ($T^0$ is Curie temperature) can be obtained. But this $T^0$ may or may not be same from the actual $T_c$ value [9].
In this paper, we focus our review on the report of dielectric and other electrical properties of different ferroelectric materials. Ferroelectrics include barium titanate, barium zirconate, lead titanate, potassium niobate, etc.

2. Discussion and Analysis

This section provides an insight into the dielectric properties of different ferroelectric compounds.

2.1. Barium Titanate (BaTiO$_3$)

From the literature, we have identified that the BaTiO$_3$ was discovered around 1940 and 1943 [11-14]. Between 1945 and 1946, the phenomenon of ferroelectricity was discovered in the same compound [15-16]. Then the first ceramic transducer based on BaTiO$_3$ was made around 1952-1954 [17]. It is widely used in several major industrial applications such as capacitors, transducers, sensors, etc. It is an inorganic perovskite ferroelectric compound that exhibit five crystal geometrical structures like cubic, tetragonal, rhombohedral, orthorhombic and hexagonal. When the BaTiO$_3$ shows cubic phase, it means that the compound is in paraelectric state (not ferroelectric). When the compound is crystallized into other four phases, it shows the presence of polarization even in the absence of external ac electric field [1]. In BaTiO$_3$, dipole moment is created by the hopping of barium and titanium ions with respect to oxygen ions.

In one report [18], Kim and Han investigated the effect of size of grain, on the dielectric constant of BaTiO$_3$. The prepared compounds possess the size of grains ranging from 0.86 $\mu$m to 10 $\mu$m. BaTiO$_3$ of 0.86 $\mu$m grain size, exhibit dielectric constant of 4500 at room temperature. Under the same ambient temperature, the dielectric constant of 1800 has been observed for BaTiO$_3$ of 10 $\mu$m grain size. It also has been observed that, the magnitude of dielectric constant is decreased with increase in grain size. At the Curie temperature, BaTiO$_3$ of 0.86 and 10 $\mu$m grains sizes, dielectric constant is 6200 and 7000 respectively. The trend of $T_c$, dielectric constant is same as it is observed at room temperature.

In another report [19], BaTiO$_3$ ceramics were synthesized through hydrothermal technique. The ceramics of grain size ranging from 10 $\mu$m to 1 $\mu$m, have been obtained. Here also, it has been found that the magnitude of dielectric constant is decreased with increase in grain size. Hence, the relation:

![Figure 3. Curie Weiss behavior of Ferroelectric in Ideal Case (in paraelectric phase)](image-url)
grain size is inversely proportional to dielectric constant i.e. grain size $\propto \frac{1}{\text{dielectric constant}}$ (may not be true for all ferroelectrics). Due to the large values of dielectric constant, BaTiO$_3$ was first employed in capacitor application (as per the record) [1]. Because of the larger dielectric constant, the compounds may exhibit large values of capacitance, and hence are useful for capacitors. The same compound, in the ferroelectric thin film form, was synthesized in 2010 [20]. The main motive of this work is to convert the (input) mechanical energy into an (output) electrical energy. The films were prepared by depositing on a substrate i.e. Pt/Ti/SiO$_2$, through radio frequency magnetron sputtering. The structure of device is as shown in figure 4. The properties are summarized in table 1.

Table 1. Summary of Electrical Properties of BaTiO$_3$

| S.No. | Property                          | Value | Ref. |
|-------|-----------------------------------|-------|------|
| 1.    | Output voltage (V)                | 1.0   |      |
| 2.    | Output current (nA)               | 26    |      |
| 3.    | Output current density (µA/cm$^2$)| 0.19  | 20   |
| 4.    | Output power density (mW/cm$^3$)  | 7     |      |

Figure 4. Structure of BaTiO$_3$ Thin Film

In 2012, Park and others [21], have realized a nanogenerator based on nanoparticles of BaTiO$_3$. Samples were prepared through simple and low cost hydrothermal synthesis. To make nanocomposite of piezoelectric nature, nanoparticles of BaTiO$_3$ were dispersed in PDMS (polydimethylsiloxane). Final device structures, were deployed on the finger and/or feet of the human body. Due to the biomechanical movements, electric signals of the following properties (shown in table 2) have been observed. It has been mentioned that these compounds are suitable for operation of a red LED.

Table 2. Summary of Electrical Properties of BaTiO$_3$ Nanoparticles

| S.No. | Property                  | Value     | Ref. |
|-------|---------------------------|-----------|------|
| 1.    | Open circuit voltage (V)  | 3.2       | 21   |
| 2.    | Short circuit current (nA)| 250-350   |      |
Similar to the work of Park, Lin and others [22] have developed a piezoelectric nanogenerator based on BaTiO3-PDMS nanocomposite. The following electrical properties (as shown in table 3) have been observed for the as prepared nanocomposite. It has been mentioned that these compounds are suitable for operation of liquid crystal display (LCD). It is optically transparent also.

Table 3. Summary of Electrical Properties of BaTiO3-PDMS Nanocomposite

| S.No. | Property                        | Value | Ref. |
|-------|--------------------------------|-------|------|
| 1.    | Open circuit voltage (V)        | 5.5   | 22   |
| 2.    | Short circuit current (nA)      | 350   |      |

Therefore, it is worth to mention that devices based on nanoscale BaTiO3 are highly useful for energy harvesting applications. To support the aforementioned statement, we have reviewed another work [23]. Nanogenerator based on BaTiO3, prepared through template method, exhibit open circuit voltage of 6 V and short circuit current of 300 nA [23].

2.2. Lead Zirconate Titanate (Pb (Zr, Ti)O3)

Lead zirconate titanate is most commonly signified as PZT. Among the widely used and investigated ferroelectrics, PZT is one of the important material. It belongs to perovskite class of materials i.e. PZT exhibit ABO3 structure. The visual structure can be found in one of our report [24]. The lead and zirconium/titanium ions get displaced with respect face centered oxygen ions, along a specific direction. This creates a net dipole moment in the material. In one of our recent work [25], we reported the dielectric properties of PZT of Pb (Zr0.35Ti0.65)O3 composition. The sample, PZT, has been modified by manganese (of 2% and 6%). The grain sizes and dielectric properties are summarized in table 4. Dielectric constant is shown at room temperature and at 50 kHz.

Table 4. Summary of Dielectric Properties of 35/65 PZT

| S.No. | Material                        | Grain Size (µm) | Dielectric constant | ε_{max} |
|-------|--------------------------------|-----------------|---------------------|---------|
| 1.    | Pb(Zr0.35Ti0.65)O3              | 3.3             | 165                 | 5440    |
| 2.    | Pb(Zr0.33Mn0.02Ti0.65)O3        | 1.6             | 306                 | 9223    |
| 3.    | Pb(Zr0.31Mn0.04Ti0.65)O3        | 1.3             | 320                 | 10092   |

One can observe that, the nature of dielectric constant is increased with decrease in grain size. This is the same behavior observed for BaTiO3 compound also. For the 35/65 PZT compound, modified by cerium (Ce), we have reported the bulk resistance values (shown in table 5) in one report [26].

Table 5. Bulk Resistance of 35/65 PZT at 449 °C

| S.No. | Material                        | Bulk Resistance (kΩ) |
|-------|--------------------------------|----------------------|
| 1.    | Pb(Zr0.35Ti0.65)O3              | 0.51                 |
| 2.    | Pb(Zr0.30Ce0.05Ti0.65)O3        | 35.9                 |
| 3.    | Pb(Zr0.25Ce0.10Ti0.65)O3        | 19.5                 |
| 4.    | Pb(Zr0.20Ce0.15Ti0.65)O3        | 12.1                 |

The dielectric properties of PZT of Pb (Zr0.52Ti0.48) O3 composition [24], modified by cerium, are given in table 6. Dielectric constant is shown at room temperature and at 1000 kHz.
Table 6. Summary of Dielectric Properties of 52/48 PZT, at 1000 kHz

| S.No. | Material                               | Dielectric constant | $\varepsilon_{max}$ | $T_c$ (°C) |
|-------|----------------------------------------|---------------------|----------------------|------------|
| 1.    | Pb(Zr_{0.52}Ti_{0.48})O$_3$            | 264                 | 2921                 | 377        |
| 2.    | Pb(Zr_{0.33}Mn_{0.02}Ti_{0.65})O$_3$   | 282                 | 4564                 | 375        |

The dielectric properties of PZT of Pb (Zr$_{0.35}$Ti$_{0.65}$)O$_3$ composition [27], modified by cerium, are given in table 7. The value of dielectric constant, shown in table 7, is at room temperature and at 100 kHz.

Table 7. Summary of Dielectric Properties of 52/48 PZT, at 1000 kHz

| S.No. | Material                               | Grain Size ($\mu$m) | Dielectric constant | $\varepsilon_{max}$ | $T_c$ (°C) |
|-------|----------------------------------------|---------------------|---------------------|----------------------|------------|
| 1.    | Pb(Zr$_{0.35}$Ti$_{0.65}$)O$_3$         | 1.2                 | 265                 | 2232                 | 310        |
| 2.    | Pb(Zr$_{0.30}$Ce$_{0.05}$Ti$_{0.65}$)O$_3$ | 5.5                 | 347                 | 15884                | 320        |
| 3.    | Pb(Zr$_{0.25}$Ce$_{0.10}$Ti$_{0.65}$)O$_3$ | 2.6                 | 291                 | 11841                | 326        |
| 4.    | Pb(Zr$_{0.20}$Ce$_{0.15}$Ti$_{0.65}$)O$_3$ | 3.4                 | 380                 | 8325                 | 341        |

One can infer from the above table that, the magnitude of dielectric constant is changing exactly identical to the grain size. This is completely different from the dielectric behavior BaTiO$_3$ and 35/65 PZT. The dielectric properties of 65/35 PZT, modified by manganese, can be found elsewhere [28].

2.3. Dielectric Properties of Other Ferroelectrics

The room temperature dielectric constant of different ferroelectrics is shown in table 8.

Table 8. Summary of Dielectric Constant (at room temperature) of Different Ferroelectrics

| S.No. | Material                                | Value | Ref.     |
|-------|-----------------------------------------|-------|----------|
| 1.    | Barium zirconate (BaZrO$_3$)            | 430   |          |
| 2.    | Bismuth ferrite (BiFeO$_3$)             | 40    |          |
| 3.    | Calcium titanate (CaTiO$_3$)            | 165   |          |
| 4.    | Potassium niobate (KNbO$_3$)            | 700   |          |
| 5.    | Potassium tantalite (KTaO$_3$)          | 242   |          |
| 6.    | Lead titanate (PbTiO$_3$)               | 200   | 29-39    |
| 7.    | Lead zirconate (PbZrO$_3$)              | 200   |          |
| 8.    | Strontium titanate (SrTiO$_3$)          | 332   |          |
| 9.    | Lead zinc niobate (PbZn$_{0.33}$Nb$_{0.66}$O$_3$) | 7    |          |
| 10.   | Neodymium aluminate (NdAlO$_3$)         | 17.5  |          |
| 11.   | Calcium ceriate (CaCeO$_3$)             | 21    |          |
2.4. **Correlation between Grain Size and Ferroelectric Properties**

Since the dielectric properties depend on grain size, the ferroelectric properties also would depend on grain size. Tetyana Malysh, in his thesis work [40], tried to study the dependence of ferroelectric properties on grain size. The results are summarized in the table 9. The compound investigated in this work is the lanthanum substituted PZT (PLZT).

| S.No. | Grain Size (µm) | Dielectric constant | Remanant Polarization (µC/cm²) | Ref. |
|------|----------------|-------------------|-------------------------------|------|
| 1.   | 0.16           | 3000              | 9                             |      |
| 2.   | 0.43           | 4500              | 11                            | 40   |
| 3.   | 0.54           | 4600              | 25                            |      |
| 4.   | 1.84           | 4800              | 30                            |      |

It can be inferred from the figure that the magnitude of polarization and dielectric constant is increasing continuously with increase in grain size of PLZT.

2.5. **Correlation between Grain Size and Piezoelectric Properties**

Since the ferroelectric properties depend on grain size, the piezoelectric properties (piezoelectric charge coefficient i.e. \(d_{33}\) also would depend on grain size. In 2014, an attempt has been made to verify this [41]. In that attempt, BaTiO₃ ferroelectrics of different grain sizes, ranging between 0.29 µm and 8.61 µm, have been synthesized through hydrothermal technique. The values of electrical properties of BaTiO₃, with respect to grain size are summarized in table 10.

| S.No. | Grain Size (µm) | Dielectric constant | \(d_{33}\) (µC/cm²) | Ref. |
|------|----------------|-------------------|------------------|------|
| 1.   | 1.19           | 6000              | 510              |      |
| 2.   | 1.28           | 5400              | 500              | 41   |
| 3.   | 1.97           | 5000              | 450              |      |
| 4.   | 2.21           | 4000              | 350              |      |
| 5.   | 4.32           | 2000              | 200              |      |
| 6.   | 8.61           | 1500              | 160              |      |

One can observe that, from table 9, the compounds of larger grain size exhibit smaller dielectric constant values. The same trend has been for the \(d_{33}\) values also. Compounds of lower grain size exhibit high dielectric constant and \(d_{33}\) values. Therefore, for BaTiO₃ ferroelectric, grain size \(\propto\) \(\frac{1}{\text{dielectric constant}}\) and grain size \(\propto\) \(\frac{1}{d_{33}}\).

3. **Conclusions**

Based on the work that has been reviewed, the following conclusions were made:

- The relation i.e. grain size \(\propto\) \(\frac{1}{\text{dielectric constant}}\) and grain size \(\propto\) \(\frac{1}{d_{33}}\) is valid for barium titanate, and may not be true/applicable to all ferroelectrics.
• BaTiO$_3$ nanocomposite shows significant electrical properties i.e., open circuit voltage and short circuit current is 5.5 V and 350 nA respectively.
• Pb (Zr$_{0.20}$Ce$_{0.15}$Ti$_{0.65}$) O$_3$ and KNbO$_3$ exhibit large values of dielectric constant at room temperature i.e. 380 and 700 respectively.
• Lanthanum substituted PZT of grain size 1.84 $\mu$m, exhibit large values of dielectric constant (4800) and remanant polarization (30 $\mu$C/cm$^2$)

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