A review on giant piezoelectric coefficient, materials and applications

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ABSTRACT

The current work deals with the review of various piezoelectric materials and their piezoelectric coefficient (d33) for probable piezoelectric device applications. In addition, the comprehensive analysis of the data of d33 obtained for distinct compounds is also made. Furthermore, the best suited material compositions are highlighted.

Keywords: Electroceramics; Sensors, Piezoelectrics; PZT; Ferroelectrics.

1. INTRODUCTION

It has been a well known fact that the piezoelectric materials play a major role in electronic devices such as sensors, accelerators, ultrasonic motors, transducers, actuators, filters and resonators, and micro electromechanical systems (MEMS). Piezoelectric ceramics were used for scientific interest (industrial applications) around 1950 [1]. Among all the electroceramics, the piezoelectric ceramics were special due to their characteristics. These materials are actively used because of their environmental friendly substances.

Piezoelectricity is in general the accumulation of charges on the surface of solid material when the material is subjected to the mechanical stress1. Herein, the solid materials such as crystalline solids, ceramic materials, parts of the organic matter viz., bones, DNA, proteins etc., can be considered into the account of piezoelectric materials1. The piezoelectric materials exhibit several potential applications towards the progress of scientific community. In view of this, different applications were noticed in distinct industries like computer, automotive, medical, military, consumer etc [2]. Specifically, these are of high performance multilayer piezoelectric actuators (MPAs), ultrasonic transducers, communication circuit components, sensors, accelerators, filters, resonators, ultrasonic motors, energy harvesters, micro-electromechanical systems (MEMSs) etc [2-5]. These applications are mainly dependent on various piezoelectric characteristics such as electromechanical communication (Kp), relative dielectric permeability (εr/ε0), specific volume electric resistance (ρv), density (ρ), water absorption (W), piezo-modules in a dynamic mode (d31, d33), piezo-modules in a static mode (d31), young’s modulus (Y’), speed of sound (v), good mechanical quality (Qm), relative frequency deviation in the range of working temperatures from the frequency measured at the adjustment temperature (δfδf), corner of dielectric loss tangent in weak electric fields (tg δ), electrical durability (Ei), Curie temperature (Tc), mechanical durability limit with static compression (σcompress.), mechanical durability limit with static bending (σb), mechanical durability limit with static stretching (σs) [6]. As a result of these characteristics, the piezoelectric materials showed the above stated applications. However, it was an observed fact that several scientists put forth on the synthesis of the giant piezoelectric materials containing giant d31 coefficient [7-9]. In this context, the authors intended to review the giant piezoelectric specimen and further to elucidate the piezoelectric parameters of the corresponding samples.

1.1 Theory.

It is a well known fact that the piezoelectric effect can be normally obtained as a result of the interaction between mechanical deformation and the applied input electric field. However, the piezoelectric coefficient is a significant parameter in order to study the piezoelectric properties as well. Let the mechanical stress and the electric charge density be designated by Tjk& D respectively. In case of piezoelectricity, the linear relationship can be found between Tjk& D. This can be referred as direct piezoelectric effect and is mathematically given by Djk = djk · Tjk [10], wherein Tjk refers to the stress tensor of 2nd rank, and djk is called as piezoelectric coefficient (C/N) of 3rd rank. Moreover, the converse of piezoelectric effect also becomes true. According to this converse effect (mechanical deformation of piezoelectric crystal under the external electric field), the mathematical expression can be given by: xjk = djkT · E, where the xjk refers to the strain of 2nd rank, E is associated to the input electric field, and djk is assigned to the parameter of converse piezoelectric coefficient (m/V). In addition, the superscript T indicates the transposition of the matrix. However, as per the thermo-dynamical illustrations [10], the piezoelectric coefficients in direct and converse reactions become identical. It is also indicated that the strain and stress are the symmetrical tensors and therefore the relationship between the two types of piezoelectric coefficients is given by djk=djk. In

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matrix form, the direct and converse piezoelectric effects can be given by

\[
\begin{pmatrix}
D_1 \\
D_2 \\
D_3
\end{pmatrix} =
\begin{pmatrix}
0 & 0 & d_{15} \\
d_{31} & d_{32} & d_{33} \\
0 & 0 & 0
\end{pmatrix}
\begin{pmatrix}
S_1 \\
S_2 \\
S_3
\end{pmatrix}
\]

Experimentally, the piezoelectric coefficient can be measured and the method is as follows: The desired sample is kept in the sample holder. Further, the direct current voltage is applied with the help of an electrode. Later on, the deformation takes place and meanwhile, it should be measured in the similar direction of the deformation. In next step, one can measure the longitudinal piezoelectric coefficient which is designated by \(d_{zz}\). Herein, the two subscripts sequentially reveal the direction of deformation as well as the applied input electric field. The \(d_{zz}\) parameter can be represented in the tetragonal symmetry group \((0.4\ \text{cm})\) as follows \([10-11]\): \(d_{zz} = (d_{31} + d_{15}) \sin^2 \theta \cos \theta + d_{33} \cos^2 \theta\), where \(\theta\) is the angle between \((001)\) piezoelectric crystal axis & measurement direction. But, in the present mentioned experiment, the direction of \((001)\) crystal axis is perpendicular to the sample. That is, \(\theta\) becomes equal to zero. Hence, the \(d_{zz}\) and \(d_{33}\) become equal.

In general, the trend of produced strain versus the electric field plot shows a butterfly loop model (Fig.1a & b). Herein, the deformation of crystal takes place in linear direction to the applied field. In addition, the formed butterfly shape is also acquired owing to the three effects. These are (i) converse piezoelectric effect, (ii) switching of domain walls, and (iii) movement of domain walls. If the input field tends to zero, the strain also becomes zero as shown in Fig.1a (at point a). Further, the electric field is increased (a-c); the deformation in crystal also increases linearly. As the input field is decreased (c-d), the polarization is altered by 180° at the coercive field (at d). Once the switching is over, the polarization becomes in the parallel direction of the electric field. Therefore, the strain approaches to positive value as shown at e. As a result, further the strain follows a linear manner against the input field. As a whole, it shows a loop. Furthermore, the piezoelectric coefficient can be calculated using the slope of the linear portion of the loop.

1.2 Experimental set up to find piezoelectric coefficient \((d_{33})\).

It is an established fact that the \(d_{33}\) value is a dependent parameter of relative permittivity and remanent polarization \((P)\). According to Landau-Ginsburg-Devonshire (LGD), it is mathematically given by: \(d_{33} = 2Q_{11} \varepsilon_o \varepsilon_{33} P\), herein \(Q_{11}\) shows the electrostrictive constant of the paraelectric phase. Commonly, this value changes from 0.05 - 0.1 \(\text{m}^4/\text{C}^2\) for various samples. The \(\varepsilon_{33}\) & \(\varepsilon_o\) are the permittivity of free space & permittivity of samples respectively and \(P\) reveals the remanent polarization \([2]\). However, the \(d_{33}\) value can be enhanced by optimizing the \(\varepsilon_{33}\& P\). This can be achieved by means of doping element and further controlling the microstructure. In fact, the microstructure as well as transport properties can be well controlled by means of grain orientation even in the case of absence of doping element. This will not affect the phase transition temperatures \((T_c)\) \([3, 4]\).
Specifically, this implies the charge per unit force in the direction of polarization. Usually, this is said to be Berlincourt method. The advantage of this system is to get the $d_{33}$ value with low input frequency and quick in time. At present, this is the easiest system in order to obtain $d_{33}$ value. Besides, many advanced methods provide excellent resolution, consistency etc., to any kind of sample. The schematic representation of various front & back components in the piezometer is shown in Fig.3. Similarly, the schematic experimental set up is shown in Fig.4. It includes the bottom and top probes which contain a gap. In the gap the desired sample is inserted to find $d_{33}$. The adjustment provision is also accommodated.

**2REVIEW ON $d_{33}$ COEFFICIENT AND PIEZOELECTRIC MATERIALS**

### 2.1 Barium Titanate Based Materials (BTX).

It was a reported fact that the piezoelectric materials like barium titanate and its based materials (ceramics & thin films) exhibited extensive applications as actuators, sensors, transducers etc.\[^1\] In general, sensors will have the capacity to obtain mechanical energy from I/P energy. This I/P energy can be either in the form of electrical, electrostatic or thermal energy. In addition, the piezoelectric actuators need the efficiency of piezoelectric parameters such as the maximum strain output, strain, maximum resonance speed, electric charge distribution and the maximum displacement control accuracy [12]. Due to the huge electromechanical response of piezoelectric specimen, they acquired good attention in highly sophisticated equipments like atomic force microscopy and scanning tunneling microscopy. As a whole, it was noticed that the larger is the electromechanical response, the higher is the performance of piezoelectric actuators. This kind of behavior can be attributed to the high numerical values of longitudinal electromechanical coupling factor ($k_{33}$) and longitudinal piezoelectric coefficient ($d_{33}$) [12, 13].

Based on these two parameters, one can justify whether the piezoelectric specimen is well suited for the high performance actuator device applications or not [14]. In view of this, many researchers performed extensive investigations on distinct piezoelectric materials. However, in the present review work, we highlighted the piezoelectric materials according to the parent and its based materials. The major role of $d_{33}$ parameter in ensuring the piezoelectric efficiency was done at length and different values were reported in Table.1.

From Table.1, it can be noted that the piezoelectric constant ($d_{33}$) was varying from 4 to 620 pC/N for barium titanate (BT) and BT based materials. These values were achieved as a result of various synthesis approaches to prepare the materials. Besides, the transition temperature ($T_c$) of BT was observed to be changing from 120 to 130°C depending upon the synthesis method. It was also evident from Table.1 that the huge $d_{33}$ value ~ 416 pC/N was found for BT material (prepared through spark plasma sintering) with 100 nm domain size. In the same fashion, the spark plasma sintered (SPS) BT of domain size 500 nm revealed $d_{33}$ ~ 216 pC/N. Likewise, interestingly, the BT sample with domain size of 100 nm which was prepared by hydrothermal synthesis and normal sintering (NS) showed a $d_{33}$ value about 193 pC/N. This established a fact that the kind of sintering process can independently alter the piezoelectric efficiency of samples. During the SPS process the BT attained the tetragonal phases (JCPDS: 05-0626) while the NS method reinforced to obtain the cubic BT phases. This confirmed us that the presence of tetragonal phases in BT allowed getting high $d_{33}$ value. On the other hand, the domain size factor influenced in achieving the high/low $d_{33}$ value. Shao et al.\[^{18}\] and Sharma et al.\[^{13}\] reported that the nano-domain size is the dominant factor in order to enhance the piezoelectric properties of materials. That is, for small sized domains, one can expect high $d_{33}$, while the small value of $d_{33}$ can be observed for the materials possessing large domain size. Using the conventional solid state reaction (SSR) method almost identical $d_{33}$ values were noted around 190-200 pC/N [13, 18]. But the BT single crystal (grown using Bridgeman technique) exhibited $d_{33}$ ~ 86 pC/N which is very small in magnitude when compared with $d_{33}$ of BT prepared by NS & SPS methods. This manifested that the polycrystalline BT material expressed some what high $d_{33}$ while single crystal BT showed moderate value of $d_{33}$. Due to the high $d_{33}$ value of BT, it showed many applications as charge stored capacitors, piezoelectric transducers, and actuators [13, 18].

Further, the BT based ceramics, alloys and composites (as shown in Table.1) [19-35] also expressed the $d_{33}$ value changing from 4 to 620 pC/N. Among these materials, Ba($Zr_{0.2}$-$Ti_{0.8}$)O$_3$-50(Ba$_0$-$Ca_{0.3}$)TiO$_3$ (620 pC/N), BT-x(CT-BS) (570 pC/N) and Ba($Ti_{0.7}$-$Sn_{0.3}$)O$_3$-30(Ba$_0$-$Ca_{0.3}$)TiO$_3$ (530 pC/N) showed the highest value of $d_{33}$ reported till now using BT based materials. In particular, the Ba($Zr_{0.2}$-$Ti_{0.8}$)O$_3$-50(Ba$_0$-$Ca_{0.3}$)TiO$_3$ revealed the highest $d_{33}$ value of 620 pC/N. It was understood that the BT material when mixed with the elements like Zr & Ca performed high piezoelectric coefficient. The reason was due to the presence of three phase structure as reported in the literature\[^7\]. Herein, the three phase structure is related to the rhombohedral-orthorhombic-tetragonal (R-T-O). As a whole, this R-T-O phase structure is responsible for high $d_{33}$. This was occurred in the case of BT-
2.2 Lead Titanate Based Materials (PTX).

The PbTiO$_3$ (PT) prepared via different synthesis methods acquired the $d_{33}$ value ranging from 56 to 79 pC/N [36-38]. This was five times smaller than the $d_{33}$ of BT material. The presence of tetragonal phases may be responsible for this. However, the PT also shows the tetragonal structure. The grain size and synthesis method can also become a reason for this kind of difference. Therefore, several dopants and substitutions were made to the PT material in order to achieve high $d_{33}$. The different $d_{33}$ values of different PT based materials were reported in Table 2.

It was noticed that the PT based materials in the form of composites, thin films etc., attained the $d_{33}$ values altering from 10 to 3500 pC/N [36-61]. The sol-gel processed PZT thin film showed a small piezoelectric coefficient about 10 pC/N. Specifically, the single crystal PT based materials attributed to the high $d_{33}$ value. This was obtained owing to the existed high strains achieved for $<001>$ oriented rhombohedral crystals [45]. In view of this, the highest value of $d_{33} \sim 3500$ pC/N for (1-$x$)[Pb(Mg$_{1/3}$Bi$_{2/3}$)$_{0.5}$]O$_3$-$x$[PbTiO$_3$] (PMN-PT) prepared via Bridgeman technique. In addition, some single crystals were found with high piezoelectric response like Pb(Mg$_{1/3}$Bi$_{2/3}$)$_{0.5}$O$_3$-$x$[PbFe$_{0.5}$Co$_{0.5}$]O$_3$-PbZrO$_3$-PbTiO$_3$ (PMN–PZT)-S-Crystal, PZN-8%PT crystal orientation $<001>$ [Pb(Zn$_{1/3}$Nb$_{2/3}$)$_{0.5}$]O$_3$-PbTiO$_3$Pb(Zn$_{1/3}$Nb$_{2/3}$)$_{0.5}$O$_3$-PbTiO$_3$], PZN, crystal orientation $<001>$ etc. Therefore, it was confirmed that the crystal orientation plays a vital role in acquiring the high $d_{33}$ value. The corresponding transition temperatures were also noted in Table 2. Thus, these materials were extensively used for variety of applications such as data memory, storage, energy harvesting, solar energy conversion, and high power transducers. The $T_c$ values of various samples were also noted in Table 2.

| BT based materials | $d_{33}$ (pC/N) | $T_c$ (°C) | Synthesis Method | Applications | Ref. |
|-------------------|----------------|---------|-----------------|--------------|-----|
| BaTiO$_3$         | 200            | 120     | conventional method (SSR) | capacitors, piezoelectric transducers | [19] |
| BaTiO$_3$ (BT-100-SPS) | 416          | 120     | Spark plasma sintering (SPS) | capacitors, piezoelectric transducers | [20] |
| BaTiO$_3$ (BT-100-NS) | 193           | 120     | hydrothermal synthesis | capacitors, piezoelectric transducers | [20] |
| BaTiO$_3$ (BT-500-SPS) | 216           | 120     | spark plasma sintering (SPS) | capacitors, piezoelectric transducers | [20] |
| BaTiO$_3$         | 191            | 130     | conventional method (SSR) | Actuator | [21] |
| BaTiO$_3$, single crystal | 86            | 130     | Bridgeman Technique | Actuator | [21] |
| Barium Titanate and nano-akermanite, (BT/nAK) BT90/nAK10 | 4 | 130 | freeze-casting technique | | [22] |
| 0.95BaTiO$_3$-0.05CaTiO$_3$-Co | 150 | 105 | | | [23] |
| 0.9BaTiO$_3$.0(1-x)CaTiO$_3$.xBaSnO$_3$ [BCT-xBS] | 469 - 1335 | | SSR | electromechanical actuator applications | [24] |
| Ba(Ti$_{0.8}$Sn$_{0.2}$)$_3$O$_3$-30(Ba$_{0.5}$Ca$_{0.5}$)$_3$TiO$_3$ | 530 | | SSR | electromechanical actuator applications | [25] |
| Ba(Zr$_{0.7}$Ti$_{0.3}$)O$_3$-50(Ba$_{0.5}$Ca$_{0.5}$)$_3$TiO$_3$ | 620 | | SSR | electromechanical actuator applications | [25] |
| (Ba$_{0.98}$Ca$_{0.02}$)$_3$(Ti$_{0.96}$Sn$_{0.04}$)O$_3$ | 510 | | SSR | electromechanical actuator applications | [25] |
| (Ba$_{0.97}$Ca$_{0.03}$)$_3$(Ti$_{0.94}$Sn$_{0.06}$)O$_3$ | 440 | | SSR | electromechanical actuator applications | [25] |
| BT-x (CT-BS)     | 570            |         | conventional method (SSR) | electromechanical actuator applications | [25] |
| (Ba$_{0.93}$Ca$_{0.07}$)$_3$(Ba$_{0.5}$Sn$_{0.5}$)O$_3$ | 387 | 108 | SSR | | [26] |
| (Ba$_{0.5}$,Ca$_{0.5}$)$_3$(Ti$_{0.86}$Zr$_{0.14}$)$_3$O$_3$ | 375 | 115 | SSR | | [27] |
| $\chi$(Na$_{0.8}$Bi$_{0.2}$)$_3$TiO$_{3-y}$ -$(K_x Bi_{1-x})$TiO$_3$ | 145 | 302 | | | [23] |
| Ba$_{2}$TiO$_3$ (x y = z; 1; y:z = 2:1) | 281 | | | | |
| BCS (Ba$_{0.6}$Sr$_{0.4}$Zr$_{0.6}$Ti$_{0.4}$O$_3$-CoFe$_2$O$_4$) | | | | | |
| 0.94Na$_{0.5}$Bi$_{0.5}$TiO$_3$-0.06BaTiO$_3$ | 102.6 | | sol-gel method | | [29] |
| Ba$_{0.8}$Ca$_{0.2}$TiO$_3$ | 290 | | SSR | sensors, actuators, fuel injectors & transducers | [30] |
| (Ba$_{0.85}$Ca$_{0.15}$)$_3$(Zr$_{0.8}$Ti$_{0.2}$)O$_3$(BCZT) | 486 | 90 | SSR | actuators, ultrasonic transducers, | [31] |

Table 1. Data on BT and its based materials.
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Table 2. Data on PT and its based materials.

| PT based materials | d33 (pC/N) | Tc (°C) | Synthesis Method | Applications | Ref. |
|--------------------|------------|--------|------------------|--------------|-----|
| PbTiO3             | 56         | 475    | tartrate precursor method | energy harvesters, smart sensors | [36] |
| PbTiO3 (at 750°C calcined temp) | 79         |        | tartrate precursor method | | [37] |
| (1-x)[Pb(Mg1/3Nb2/3)O3]-x[PbTiO3] (PMN-PT) | ~3500     |        | Bridgman technique | | [39] |
| 1-x)[Pb(Mg1/3Nb2/3)O3]-x[PbTiO3] (PMN-38PT) | 300-1200   |        | high temperature flux technique | solid state actuators | [45] |
| PZT thin films     | 10         |        | sol-gel processing | | [41] |
| Cellulose paper derived ceramics (CPDC) | 50         |        | | | [42] |
| PZT60/40 films, Pb(Mg1/3Nb2/3)O3–PbZrO3–PbTiO3 (PMN–PZT)-R – Ceramic | 100        |        | | | [43] |
| Pb(Mg1/3Nb2/3)O3–PbZrO3–PbTiO3 (PMN–PZT)-T–Ceramic | 230        |        | | | [44] |
| Pbn0.5–Y0.5TiO3     | 1500       |        | Bridgman technique | | [44] |
| PZN-8% PT crystal orientation <001> | 2500       |        | high temperature flux technique | | [45] |
| Pbn0.5–Y0.5TiO3     | 1100       |        | Bridgman technique | | [44] |
| PZN, crystal orientation <001> | 1530       |        | high temperature flux technique | | [45] |
| PZN, crystal orientation <111> | 1100       |        | high temperature flux technique | | [45] |
| PZN-8% PT, crystal orientation <111> | 83         |        | high temperature flux technique | | [45] |
| PZT                | 750        |        | high temperature flux technique | | [45] |
| Pb1/3B1/3O2-PT     | 1500       |        | high temperature flux technique | | [45] |
| PZN-PT             | 1600       |        | high temperature flux technique | | [45] |
| PZN direction <111> | 83         |        | high temperature flux technique | | [45] |
| PZN-8% PT          | 84         |        | high temperature flux technique | | [45] |
| PZN along <001>    | 1100       |        | high temperature flux technique | | [45] |
| PZN-8% PT along <001> | 2500     |        | high temperature flux technique | | [45] |
| 0.955Pb(Zn1/3Nb2/3)O3–0.045PbTiO3 along <001> | 2280      |        | grown using the high temperature flux technique | | [46] |
| 0.955Pb(Zn1/3Nb2/3)O3–0.045PbTiO3 <111> | 92         |        | | | [46] |
| PZT ceramics       | 219.4      |        | Bridgman technique | | [47] |
| Lead zirconatetitanate (PZT) thinfilms | 12         |        | sol-gel technique | | [48] |
| PZT Fe             | 230        |        | SSR | | [49] |
| PZT Nb             | 470        |        | SSR | | [49] |
| PZT6              | 290        | 330    | | | [50] |
| PZT7              | 425        | 350    | | | [50] |
| PZT5A4            | 460        | 360    | | | [50] |
| PZT507            | 820        | 165    | | | [50] |
| PLZT             | 108        |        | | | [51] |
| PZT-5A           | 3.74       |        | | | [52] |
| PZT-5H           | 5.93       |        | | | [52] |
| PZT-7A           | 1.53       |        | | | [52] |
| 0.67Pb(Mg1/3Nb2/3)O3–0.33PbTiO3>1900 | |        | | | [53] |
| PZT-4D          | 246        |        | | | [54] |
| PZT-5H           | 677        |        | | | [54] |
| PZT(52/48)        | 135        |        | tartrate precursor method | | [36] |
| 0.3 PZT          | 87         |        | | | [36] |
| PZT(57/43)       | 200        |        | | | [36] |
2.3. Bismuth Titanate Based Materials (BITX).

It was also observed that the rhombohedral bismuth titanate and its based materials revealed the considerable piezoelectric response. Besides, the undoped BIT expressed the ferroelectric transition temperature at 670 °C [62]. In connection with d33, it showed value of 40 pC/N. Comparatively, this was smaller than the d33 of BT & PT which suggested moderate piezoelectric efficiency. Therefore, it was expected for ferroelectric and piezoelectric devices application [63]. For further increase of d33 value, several additives were mixed to the BIT. As a result, few elements such as Sr, Na, Nd, K etc., were doped to BIT. The obtained d33 data were listed in Table.3 [62-71]. It was clear from Table.3 that the d33 of BIT based samples was observed to be changing from 17 to 650 pC/N. In this case also, the maximum value of d33 of 650 pC/N was achieved for BNBT-5.5 single crystal [64]. For BT, and PT based materials also the similar observations were noted. Moreover, it was noticed that the single crystal piezoelectric materials depending upon the type of dopant/substituent, the piezoelectric coefficient was much improved. The Tc values of various samples were also noted in Table.3.

2.4. Sodium Niobate Based Materials (NNX).

The sodium niobate (NaNbO3 (NN)) and its based materials revealed the piezoelectric coefficient ranging from 28 to 410 pC/N. It was observed that the undoped NN showed d33 ~ 28 pC/N. It is of small value in comparison with the BT, PT, & BIT. However, the potassium elements improved three times its d33 value. Latter, the different combinations were prepared using the solid state reaction method. The d33 values of those materials were listed in Table.4. It was found that the d33 was changing from 80 to 410 pC/N [72-78]. These materials showed data memory and storage, energy harvesting, solar energy conversion applications. The Tc values of various samples were also noted in Table.4.

| Table 3. Data on BIT and its based materials. |
|---------------------------------------------|
| BIT based materials | d33 (pC/N) | Tc (°C) | Synthesis Method | Applications | Ref. |
|----------------------|------------|---------|-------------------|--------------|------|
| (Bi27Ti3O51)3, BIT | 40         | 670     | polymeric precursor method | ferroelectric and piezoelectric devices | [62] |
| SrBi2Ta2O9 films   | 17         |         | polymeric precursor method | ferroelectric & piezoelectric devices | [62] |
Table 4. Data on NN and its based materials.

| NN based materials | $d_{33}$ (pC/N) | $T_r$ (°C) | Synthesis Method | Applications | Ref. |
|--------------------|----------------|-----------|------------------|--------------|------|
| NaNbO$_3$          | 28             |           | Molen salt method | Storage memories, energy harvesting, solar energy | [72] |
| (K$_{0.465}$Na$_{0.465}$Li$_{0.07}$)NbO$_3$ | 80           | 420       |                 |              | [73] |
| (K$_{0.46}$Na$_{0.54}$Li$_{0.06}$)NbO$_3$ | 240          | 460       |                 |              | [73] |
| (K$_{0.46}$Na$_{0.54}$Li$_{0.06}$)Nb$_{0.78}$Ta$_{0.22}$Sb$_{0.06}$O | 410          | 253       |                 |              | [73] |
| 0.95(K$_{0.95}$Na$_{0.05}$)NbO$_3$+0.05SrTiO$_3$ | 200          | 277       |                 |              | [73] |
| 0.95(K$_{0.95}$Na$_{0.05}$)NbO$_3$+0.05SrTiO$_3$ | 200          | 430       |                 |              | [73] |
| 0.94(K$_{0.95}$Na$_{0.05}$)NbO$_3$+0.06LiNbO$_3$ | 235          | 460       |                 |              | [73] |
| 0.95(K$_{0.95}$Na$_{0.05}$)NbO$_3$+0.05SrTiO$_3$ | 283          | 392       |                 |              | [73] |
| 0.7Bi$_2$Na$_3$TiO$_7$-0.2Bi$_2$K$_{2.5}$TiO$_4$-0.1Bi$_2$Li$_4$Ti$_2$O$_9$ | 216          | 350       |                 |              | [73] |
| 0.5Na$_2$Bi$_2$Ti$_3$O$_7$-0.5K$_{2.5}$Bi$_2$Ti$_3$O$_7$ | 150          | 320       |                 |              | [73] |
| (Li$_{0.95}$Na$_{0.05}$)Ti$_2$O$_3$-xCeO$_3$ | 272          |           | SSR              |              | [74] |
| 0.963(K$_{0.48}$Na$_{0.52}$)(Nb$_{0.95}$Sb$_{0.05}$)O$_3$-0.037(Bi$_{0.86}$Na$_{0.14}$)HIO$_3$ | 512          | 238       | ultrasonic transducers, actuators, & sensors | [75] |
| (K$_{0.42}$Na$_{0.58}$Li$_{0.00}$)Nb$_{0.87}$Ta$_{0.13}$Sb$_{0.06}$O$_3$ | 255          |           | SSR              | [76] |
| Li$_{0.95}$Na$_{0.05}$K$_{0.05}$Li$_{0.05}$Ta$_{0.05}$Sb$_{0.03}$ | 306          |           | SSR              | [77] |
| Na$_{0.32}$K$_{0.4}$Li$_{0.28}$La$_{0.06}$Nb$_{0.95}$Ti$_{1.65}$O$_{10}$ (KNN:LNT-La) | 215          | 420       | SSR              | [78] |

2.5. Polymer Based Materials (POLX). In general, the piezoelectric polymers will exhibit the piezoelectric nature owing to the molecular structure of the polymers [79]. The $d_{33}$ data of various polymer materials were shown in Table 5. The results were noted to be of order 1-2000 pC/N. From the Table 5, it was clear that the pure organic polymers revealed the moderate piezoelectric coefficient. On the other hand, the same polymers were doped with inorganic piezoelectric elements such as Pb, Na, K, Nb etc., and the obtained $d_{33}$ values were of greater than 1000 pC/N [79]. For instance, the PMN–0.29PT/epoxy (1–3) composite, and PMN0.29PT expressed the huge piezoelectric response by providing the high $d_{33}$ [79]. Therefore, these were used as electromechanical applications.

2.6. Multiferroic Materials (MUFX). In general, the multiferroic materials can exhibit the electrical, piezoelectric, ferroelectric and magnetic behavior. The multiferroic materials as shown in Table 6 showed inverse piezoelectric coefficient ranging from 36 to 400 pm/V [80, 81]. This confirmed us the range of produced strain of different materials for the applied electric field. In the literature, the highest value of inverse $d_{33}$ about 400 pm/V was noted for (0.67-x)BiFeO$_3$-0.33BaTiO$_3$SrZrO$_3$ material [81]. It was achieved owing to the presence of tetragonal peaks. Hence, these materials were used for actuators, sensors, information storage and some micro motor systems.

2.7. Glass based materials (GLX).
The Sr-fresnoite \((\text{Sr}_2\text{Ti}_2\text{Si}_2\text{O}_{8})\) + added \(\text{SiO}_2\) which is a glass based piezoelectric materials revealed the \(d_{33}\) of 10 pC/N \([82]\) (Table.7). This implied that the weak piezoelectric nature was observed in the case of present glass material. This kind of manner may be acquired as a result of the existed less intense polycrystalline phases although it is mixed with silicon dioxide. However, it was suited for high temperature applications.

Fig.5 shows the overall, review analysis of giant \(d_{33}\) values of piezoelectric materials. It was also noted that the Pb-based single crystal: \((1-x)[\text{Pb}(\text{Mg}_{1/3}\text{Nb}_{2/3})\text{O}_3]-x[\text{PbTiO}_3]\) (PMN-PT) showed the highest value of \(d_{33}\) \(\sim 3500\) pC/N. The polymer composite: PMN0.29PT revealed the maximum value of \(d_{33}\) \(\sim 2000\) pC/N. In case of BIT based materials, the existed high strains achieved for \(<001>\) oriented rhombohedral BNBT-5.5 single crystal exhibited the maximum value of \(d_{33}\) of 650 pC/N. The BT based composite material: \(\text{Ba}(\text{Zr}_{0.2}\text{Ti}_{0.8})\text{TO}_3-50(\text{Ba}_{0.7}\text{Ca}_{0.3})\text{TiO}_3\) performed the high piezoelectric coefficient of 620 pC/N. The rest of the materials performed considerable \(d_{33}\) values useful for various sensors, transducers, actuators and storage applications.

### Table 5. Data on various polymer based materials.

| Polymer materials | \(d_{33}\) (pC/N) | \(T_c\) (°C) | Synthesis Method | Applications | Ref. |
|-------------------|-------------------|--------------|------------------|--------------|------|
| PVDF              | 13-28             |              |                  |              | [79] |
| PVDF-TrFE         | 24-38             |              |                  |              | [79] |
| ParyleneC         | 2.0               |              |                  |              | [79] |
| PI (α-CN)         | 5.3-16.5          |              |                  |              | [79] |
| APB/ODPA          | 16.5              |              |                  |              | [79] |
| Polymide P150 (β-CN) | 5.3           |              |                  |              | [79] |
| PMN–0.29PT/epoxy (1–3) composite | 1200 | |                  |              | [79] |
| Cellular polypropylene | 200              |              |                  |              | [79] |
| Fluorinated and Post treated cellular PP | 270            |              |                  |              | [79] |
| COC based cellular electrets | 13              |              |                  |              | [79] |
| Cellular Polyethylene enaphthalate (PEN) | 45            |              |                  |              | [79] |
| PTFE/FEP multilayer VCP | 225           |              |                  |              | [79] |
| FEP multilayer    | 1000              |              |                  |              | [79] |
| Cellular PDMS     | 1148              |              |                  |              | [79] |
| Micromachined integrated cellular Parylene | 1200  | |                  |              | [79] |
| PMN0.29PT         | 500               |              |                  |              | [79] |

### Table 6. Data on multiferroic materials.

| Multiferroic materials | \(\text{Inverse } d_{33}\) (pm/V) | \(T_c\) (°C) | Synthesis Method | Applications | Ref. |
|------------------------|----------------------------------|--------------|------------------|--------------|------|
| \(\text{Li}_{0.55}\text{Bi}_{0.55}\text{Fe}_{0.55}\text{O}_3\) film | 107.5 | | sol-gel methods | actuators and sensors | [80] |
3. CONCLUSIONS

A review of piezoelectric materials with their $d_{33}$ values was performed. As a result of this review, it was concluded that (i) the Pb-based single crystal: (1-x)[Pb(Mg_{1/3}Nb_{2/3})O_x]-x[PtTiO_3] (PMN-PT) showed the highest value of $d_{33} \approx 3500$ pC/N. (ii) The polymer composite: PMN0.29PT revealed the maximum value of $d_{33} \sim 2000$ pC/N. (iii) In case of BIT based materials, the highest high strains achieved for <001> oriented rhombohedral BNBT-5.5 single crystal exhibited the maximum value of $d_{33}$ of 650 pC/N. (iv) The BT based composite material: Ba(Zr_0.2Ti_0.8)O_3-50(Ba_0.7Ca_0.3)TiO_3 performed the high piezoelectric coefficient of 620 pC/N. (v) The sodium niobate based material: (K_0.44Na_0.56)L_0.06(Nb_0.84Ta_0.16)(SiO_2) expressed the huge value of $d_{33} \sim 410$ pC/N. Further the high inverse $d_{33}$ of multiferroic materials was observed for Li_0.06Bi_0.95Nb_0.05Fe_2O_5 film and (0.67-x) BiFeO_3-0.33BaTiO_3-SrZrO_3. In addition, the $d_{33}$ value of glass material was found and noted to be very small in magnitude (Sr-fresnoite (Sr_2Ti_3O_7) + added SiO_2) (10 pC/N).

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