

Short Review

Perovskites based Lead Free Piezo Ceramics

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Abstract

Lead based piezoceramics such as Lead Zirconate Titanate (PZT) and Lead Titanate (PT) have been widely used for applications for their superior piezoelectric properties. They have large value of piezoelectric coefficients (d_{33}), electromechanical coupling factors (k) and large strain with respect to the applied electric field which are useful parameters for sensors and actuators. But their applications are often limited due to the presence of lead (Pb) in their compound. Lead is toxic in nature and pollute the environment. For the past few years search for lead free materials especially ceramics have been taken and found many materials matches with the properties of lead-based compounds. This article compiles on the collections of lead-free ceramics especially perovskite-based compounds which are having promising properties, that could replace the applications based on lead-based compounds. This article listed studies on compounds based on (K,Na) NbO₃, (Bi,Na) TiO₃, BaTiO₃ and BiFeO₃.

Keywords: Lead-free piezoelectrics, Perovskite structure, Ultrasonic transducer

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Introduction

Piezoelectrics are the materials which converts electrical energy to mechanical energy and vice versa. The structure of materials at the unit cell level lacks centro-symmetry. These materials also have non-vanishing value of polarization even without the application of external electric field below a certain critical temperature. This type of behavior is exhibited by the materials called ferroelectrics which exists in single crystals, polymers and ceramics. The single crystals like Quartz, Rochelle salt and Tourmaline are widely studied [1]. In polymers PVDF, PDMS, PLLA etc., are widely studied. The ceramic materials are broadly divided into two groups as lead based and lead-free compounds. The lead-based compounds are based on Lead Zirconate titanate (PZT) and lead titanate. The lead-free compounds include Barium titanate, Bismuth sodium titanate (BNT), Lithium Niobate, Zinc Oxide, potassium sodium niobate (KNN) etc. have been widely investigated for this purpose. The parameters like piezoelectric coefficients (d_{33} , d_{31} etc.), electromechanical coupling factors (k_{33} , k_{31} , k_p etc.), and voltage coefficients (g) are important for specific applications. Most of the ceramics are lead (Pb) based compounds which are having superior piezoelectric properties. But it has been observed that the lead is toxic and poses threat to environment and health issues. Their applications in the field of medical are very much limited. Often the lead-based compounds are coated with TiO₂ whenever their applications aimed at human body [2]. In order to find the alternate materials various compounds have been investigated in the past which are free from lead such as potassium sodium niobates, Bismuth sodium titanate, Barium titanate etc. This article discusses about lead free ceramics especially perovskite-based materials which are being actively studied presently.

Potassium sodium niobate ceramics ($\text{K}_{0.5}\text{Na}_{0.5}\text{NbO}_3$ – (KNN))

Among lead-free piezoelectric materials potassium sodium niobate attracting investigators for its superior properties. The KNN material exhibits perovskite structure of type ABO_3 . K and Na goes to A-site, Nb goes to B-site and O goes to face centered position of Perovskite cubic cell. The Cubic to tetragonal transition (ferroelectric) occurs at 420°C , tetragonal to orthorhombic transition occurs at 200°C and orthorhombic to rhombohedral transition occurs at -123°C to -163°C . Very high ferroelectric phase transition temperature (420°C) and also low density (around 4.5 g/cm^3) make it as a suitable material for applications. Enhanced property of piezoelectricity in this compound is due to the presence of morpho-trophic boundary separating two orthorhombic phases. Low density is important for acoustic impedance matching with surrounding medium when the mechanical energy of the materials is transferred to surrounding medium. These materials show excellent piezoelectric properties in which d_{33} varies between $80\text{--}720\text{ pC/N}$. The lower side of the d_{33} values ($< 200\text{ pC/N}$) exhibited by normal polycrystalline samples and the high side of the d_{33} ($> 200\text{ pC/N}$) exhibited by textured ceramics. The KNN in polycrystalline ceramics form shows d_{33} lies near the value $70\text{--}100\text{ pC/N}$ [3]. It has also reported to be 95 pm/V with the converse method (d_{33}^*) of measurement [4]. With the doping Sr and Ba at the A site d_{33} reaches as high as $100\text{--}110\text{ pC/N}$ [1, 5]. With lithium and tantalum doping the value of d_{33} reaches as high as $190\text{--}230\text{ pC/N}$ [6]. The hot-pressed sample shows d_{33} value 170 pC/N [3]. With the development of processing methods especially template grain growth method in which the grains of the materials oriented in a particular crystallographic direction which can enhance the piezoelectric properties. The materials which are prepared in this manner are called textured ceramics. The textured ceramics of KNN shows as high as 416 pC/N [7]. However, with the modification of phase boundaries, the d_{33} values were improved between $400\text{--}700\text{ pC/N}$ [8] [Table 1].

Materials	Dielectric permittivity (ϵ') and loss ($\tan\delta$)	d_{33} (pC/V)	k_p
KNN	400-440 and 0.02-0.09	70-100 85-95 pm/V (d_{33}^*)	0.25-0.35
KNN- Sr (1%)	420 and 0.05	110	0.39
KNN-Li 3%; Ta 20%	920-1256 and 0.02	190-230	0.46-0.54
KNN-hot press	330 and 0.014	160	0.45
KNN-textured	1570	416	0.61
KNN-Textured	~ 5000 and 0.14	650	

Table 1: Dielectric and piezoelectric properties of KNN-based materials.

The temperature-dependent variations in the real and imaginary parts of the dielectric constant for a typical KNN ceramic are shown in Figures 1 and 2.

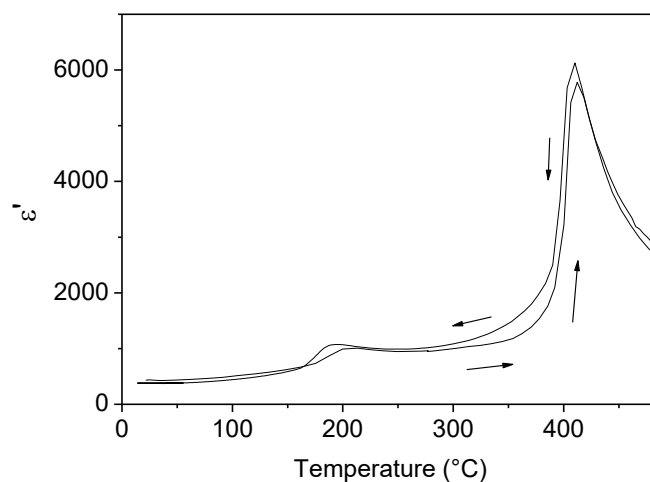


Figure 1: The Variation of real part of dielectric constant (ϵ_r) with temperature for KNN at 1KHz.

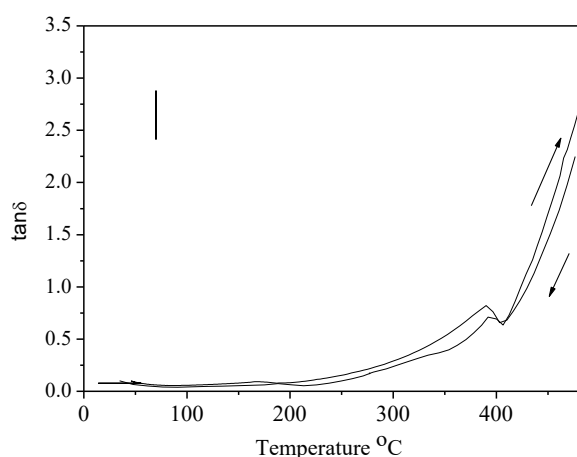


Figure 2: The variation of loss tangent ($\tan \delta$) with temperature at 1KHz.

Bismuth sodium titanate - ($\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$ (BNT))

Bismuth Sodium Titanate popularly known as BNT exhibits perovskite structure. It has rhombohedral symmetry at room temperature [9]. Large piezoelectricity is due to presence of MPB exist between ferroelectric Rhombohedral and tetragonal phases. Ordering of A-site ions (Bi and Na) in 1:1 ratio is present in the compound.

The polarization in the compound is due to off center displacement of Ti^{4+} ion and also due to the displacement of A-site ions. These compounds possess high remanent polarization ($37 \text{ } \mu\text{C}/\text{cm}^2$) and high coercivity ($E_c=73 \text{ kV}/\text{cm}$). It is difficult to pole the sample because of high coercivity and often it exhibits d_{33} values vary in the range $73\text{-}95 \text{ pC}/\text{N}$ [10-13]. In order to improve the piezoelectric properties additives and substitutions have been tried. BNT- BaTiO_3 solid solution showed enhanced piezoelectric properties ($d_{33} = 125 \text{ pC}/\text{N}$) which is due to the presence of Rhombohedral-Tetragonal phase boundary [14]. These compounds also exhibit relaxor behavior. The presence of depolarization temperature (T_d) due to relaxor behaviour restricts the use of the material high temperatures range. The value of T_d for pure BNT is 190°C . There is an inverse relationship exists in this compound in BNT- BaTiO_3 solid solution with d_{33} and T_d . With increase in d_{33} the T_d value decreases [14]. With the doping of K, Li at A-site the value of d_{33} reaches in the range $232\text{-}281 \text{ pC}/\text{N}$ with varying concentrations [15]. The ion (Hf, Zr, Sn, Nb, and Ta) substituted in BNKT can decrease the depolarization temperature (T_d) and induce a phase transition from a polar (ferroelectric R-T) phase to a non-polar (pseudocubic) phase, finally resulting in improved strain (0.34-0.47%)

[16, 17]. The texture ceramics also show d_{33} above 300 pC/N [18].

Barium titanate (BaTiO₃) (BT)

BT is widely studied compound and is first reported lead free piezoelectrics. The structure of BT is perovskite structure ABO₃ with Ba²⁺ goes to A-site, Ti⁴⁺ goes to B-site and O²⁻ goes to face center position of perovskite structure. The cubic to tetragonal ferroelectric phase transition temperature often called Curie temperature occurs at 120 °C [19]. Application of the material is limited by this temperature. The compound is widely used as Sonar. The room temperature value of electromechanical coupling factor (k_p) and d_{33} are in the range 0.3 to 0.4 and 190-200 pC/N respectively [20]. By improving processing condition and doping the piezoelectric properties are improved by large extent. Often BT are combined with KNN and BNT to shift the Curie temperature to higher value since the T_c of KNN and BNT are 420 °C and 383 °C respectively. The B-site substitution like Ca and Zr doping a large value of d_{33} around 637 pC/N and the k_p value around 0.59 have been reported [21]. The texture ceramics of above composition shows excellent piezoelectric properties with d_{33} as high as 755 pC/N have been reported [22]. Doping of BiFeO₃ and Bi (Mg_{0.5}Ti_{0.5}) O₃ leads to large value of d_{33} around 154 pC/N and high T_c of 482 °C have been reported [23]. Zhu et al, also reported that high value of d_{33} in this solid solution is due to the presence of Morphotrophic phase boundary exist between Pseudocubic and Rhombohedral structure [23].

Bismuth iron oxide (BiFeO₃)

Bismuth Iron Oxide (BFO) is a multiferroic material exhibits both ferroelectric and magnetic ordering at room temperature. By modifying composition, piezoelectric properties can be induced in the material. The substitution of Sm at the Bi site exhibits high piezoelectric coefficient around 37 pC/N [24]. The observation of this high piezoelectric coefficient is due to two polar phase boundaries (R3C/ Pna21). The substitution of Ba at A-site also exhibits large value d_{33} around 134 pC/N [25]. A large value of d_{33} around 405 pC/N and high T_c of around 450 °C was obtained Ga modified in BFO which is due to the presence of Rhombohedral - Tetragonal phase boundary [26]. By modifying macroscopic ferroelectric domains, phase boundaries and also processing parameters the piezoelectric properties can be improved in this compound. The P-E loop of the samples usually shows linear behavior. It can be modified into saturated P-E loop by air quenching the samples at different temperature [27]. The quenched samples also show high strain values compared to the slowly cooled samples. The strain value as high as 0.4% was reported on Bi (Mg_{1/3}Nb_{2/3}) O₃ or (Ba, Ca) ZrO₃ doped samples [28, 29].

Lead-free piezoelectrics in medical and clinical applications

Lead-free piezoelectric materials are rapidly replacing toxic, lead-based ceramics like PZT in clinical medicine. By utilizing safer alternatives such as potassium sodium niobate (KNN), barium titanate (BaTiO₃) and bismuth sodium titanate (BNT), the medical field is achieving safer diagnostic, surgical and therapeutic breakthroughs. The properties of lead-free piezoelectrics are suitable for medical applications. These lead-free ceramics especially KNN possesses inherently lower acoustic impedance. The PZT materials have acoustic impedance in the range (30-34 M Rayl). The KNN and BNT have acoustic impedance in the range (24-27) and (28-30) respectively. This allows for much easier acoustic impedance matching to human tissue, vastly improving bandwidth and pulse-echo sensitivity. These materials are also have high depolarization temperature (T_d). The value of T_d varies from 200 °C to 300 °C for KNN and 100 °C to 200 °C for BNT compounds. This flexibility makes non-vanishing piezoelectric properties for wide range of temperature. These materials are also having high electromechanical coupling factors (>0.4). This is suitable for higher efficiency in conversion of electrical energy to mechanical energy. Some of the applications are mentioned below.

Diagnostic medical imaging

The lead-free piezoelectric materials serve as the core vibrational elements inside the diagnostic ultrasound transducers. making them highly effective for sharp, high-resolution intravascular ultrasound (IVUS) and pediatric needle transducers. One such application is HIFU High- Intensity Focused Ultrasound (HIFU) for Tumor Ablation. The mechanical deformation in by external oscillating applied voltage specifically produced pressure waves in the surrounding medium as ultrasonic waves. These waves can be focused on to specific tissue. Because of focusing a large intensity of ultrasonic waves is concentrated at particular area, which specifically destroys the tumor cells in the affected bodies. HIFU is a non-invasive therapeutic technique, it targets and destroys deep-tissue tumors [30].

Minimal invasive and precision surgery

Handpieces built with lead-free Langevin-type transducers deliver high-frequency micro-vibrations tailored for selective bone cutting (such as in oral, maxillofacial, and dental surgeries). These devices safely cut hard bone tissues while leaving adjacent soft nerves and blood vessels completely unharmed. Lead-free materials also exhibit lower mechanical losses during continuous high-power actuation. This creates upto a 30% reduction in device self-heating compared to PZT, protecting sensitive biological tissues from thermal agitation during surgery [31].

Smart therapeutics and tissue engineering

Tissues like skin, muscle, ligament, tendon, bone, cartilage, dentin, cementum, and hair are all reportedly piezoelectric given their non-centrosymmetric spiral structures. These cells react directly to localized electric stimuli, leadfree piezoelectrics can also be used as an important role as stimuli.

Wound Healing Scaffolds: Flexible lead-free sheets made from composites of polymers and piezoelectric materials harvest subtle biomechanical movements from the patient's body like breathing and stretching. They convert this motion into localized micro-electrical fields that actively accelerate cell proliferation and wound closure.

Bone Tissue Regeneration: Implantation of biocompatible, polarized KNN or ZnO structures mimics the natural piezoelectricity found in real human bone. Under physiological loading, the implant stimulates osteogenesis (bone growth) and accelerates deep structural integration with the skeleton [32].

Precision drug delivery systems

Piezoelectric materials are localized inside the body and activated remotely via ultrasound which activates the release of drug on specific location. This functioning of this phenomenon bypasses the need for tethered wires or internal batteries. The piezoelectric materials also act as piezodynamic agents. When vibrated by ultrasound, they generate Reactive Oxygen Species. This mechanism directly destroys drug-resistant tumor cells, and triggers localized drug release simultaneously [33, 34].

Piezoelectric materials for energy harvesting

The leadfree piezoelectrics can also be used as energy harvesters when certain instruments like pace makers integrated with our human organ. The pacemakers are usually powered by batteries. These batteries can be replaced with conventional piezoelectric materials which can produce voltage with stress coming from the movement of body or heartbeats.

Only few of the applications are mentioned above. Many more are possible with lead-free materials. The applications depend on the specific piezoelectric coefficients of the materials.

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