

Review on Advanced Piezoelectric Materials (BaTiO₃, PZT)

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Abstract- Piezoelectric materials are some naturally occurring materials which produces mechanical stresses when subjected to some voltages. But natural piezoelectric materials suffers a lot of disadvantages like low strength, sensitivity toward moisture, limited operating temperature range so, to overcome these drawbacks piezoelectric ceramics are made. Two major piezoelectric ceramics used are barium titanate (BaTiO₃) and lead zirconium titanate (PZT). BaTiO₃ is ferroelectric ceramic which exhibit photorefractive effect and piezoelectric properties. BaTiO₃ has a disadvantage of low Curie temperature which can be further overcome by processing. Lead Zirconate titanate is an inorganic compound with chemical formula $Pb[Zr_xTi_{1-x}]O_3$ ($0 < x < 1$). Also called PZT, it is a ceramic perovskite material that shows a marked piezoelectric effect, meaning that the compound changes shape when an electric field is applied. Nowadays, they become the dominant piezoelectric materials in the fields of piezoelectric applications such as actuators, sensors, and transducers in intelligent systems and smart structures, dominating the world market today. Therefore, the aim of this review is giving a state of art in polycrystalline piezoelectric ceramic materials, like BaTiO₃ and PZT and their applications. This review is organized in the following way. In the first part, the history and processing of piezoelectric ceramic materials are briefly introduced, and then the general characteristics of BaTiO₃ and PZT are described with an emphasis on the piezoelectric parameters, compositions and properties, and piezoelectric constitutive relationships. Characterization methods for piezoelectric properties and ferroelectric domain structures of piezoelectric ceramic materials are addressed in the third part. Finally, various applications of piezoelectric ceramic materials in ultrasonic actuators, sensors, transducers, and active vibration controlling, are described.

Keywords- Piezoelectricity, Piezoelectric materials, Poling, BaTiO₃, PZT, Applications.

I. INTRODUCTION

Piezoelectric materials are some materials which produces some mechanical stress when they are subjected to a voltage. From a very long time we are aware of them. Some of the common piezoelectric elements known are quartz, tourmaline and Rochelle salt. These materials have various applications in production, detection of sound, generation of high voltages and frequencies. Natural piezoelectric material have a lot of disadvantages like weak piezoelectric effect, sensitivity to moisture, low mechanical strength and limited operating temperature range [1]. This gave rise to need of more mechanically strong material which can be used on a wide temperature range. Therefore piezoelectric ceramics were developed of which two prime materials are barium titanates (BaTiO₃) and lead zirconium titanate (PZT). These materials gives huge advantages over natural piezoelectric materials like large and ease of piezoelectric effect, high strength and ease of fabrication which makes them two most widely used piezoelectric ceramics in industry now-a-days[2,3]. Piezoelectric ceramics are made with the infliction of piezoelectric effect in polycrystalline ferroelectric ceramics is noticed by poling process, in which an external electric field can alter ferroelectric domains with the grains, thus generating a ceramic material acting very identical to single crystalline with both piezoelectric and ferroelectric properties [1]. Process of manufacturing piezoelectric ceramics includes accurate weighing of high purity oxides, mechanical and chemical mixing, calcinations, milling, powder compaction, burnout, firing of ceramic structure to optimum density at elevated temperature and poling. Later in the 1990s, relaxer type piezoelectric single crystal materials, such as PMN-PT ($Pb(Mg_{1/3}Nb_{2/3})O_3-xPbTiO_3$) and PZN-PT ($Pb(Zn_{1/3}Nb_{2/3})O_3-xPbTiO_3$) were discovered [2,4-8]. All these Pb-based piezoelectric materials govern the actuator applications of piezoelectric materials due to their high longitudinal electromechanical coupling (k_{33}) and large longitudinal piezoelectric coefficient (d_{33}). Table 1 lists the properties of these Pb-based materials [2, 4, 9-12]. The Pb-containing piezoelectrics have been further modified into a number of materials, and the corresponding products have been well commercialized and found mature applications.

TABLE 1. Piezoelectric properties of Pb-based systems and BaTiO₃ based systems.[13]

Materials	Poly/Single Crystal	d ₃₃ (pC/N)	k ₃₃	Reference
PZT-5A (soft)	Polycrystalline	374	0.71	[2]
PZT-8 (hard)	Polycrystalline	225	0.64	[9]
PMN-70PT	Single crystal	1500	>0.9	[4]
92%PZN-8%PT	Single crystal	2200	>0.9	[10]
0.5BZT-0.5BCT	Polycrystalline	620	0.65	[11]
0.7BTS-0.3BCT	Polycrystalline	530	0.57	[12]

The fabrication of majority piezoelectric ceramics starts with powder preparation. The powders are then pressed to the necessitate shapes and sizes, and the green shapes are in turn processed to mechanically strong and dense ceramics. The more important processes that impact the product features and properties are powder preparation, calcining and sintering. The next steps are machining, electroding and poling (application of a dc field to orient the ferroelectric domains and induce piezoelectricity). The electromechanical properties of piezoelectric ceramics are largely impacted by their processing conditions. First, high purity raw oxide materials are correctly weighed according to their required ratio and then are mechanically or chemically mixed. During the calcination step, the solid phases behave to form the piezoelectric phase. After calcination, the solid mixture is milled to thin particles. Shaping is accomplished by a variation of ceramic processing techniques, counting powder compaction, tape casting, slip casting, and extrusion. During the shaping operation, organic materials are habitually added to the ceramic powder to improve its flow and binding characteristics. The organic is then separated at a low temperature (500-600°C) burnout step. After organic removal, the ceramic structure is launched to an optimum density at high temperature. Lead containing ceramics such as PbTiO₃, PZT, Pb (Mg_{1/3}Nb_{2/3}) O₃ are fired in sealed crucibles in an optimized PbO atmosphere to prevent lead loss above 800°C.

II. BARIUM TITANATE (BaTiO₃)

The BaTiO₃ was found during World War II as a high capacitance material and was the first found ferroelectric compound with a perovskite structure. Later on, the electrostriction effect for the unpoled polycrystalline ceramics of BaTiO₃ as well as the piezoelectricity for the electrically poled samples was found, leading to many applications earlier than the Pb-containing material, PZT [13]. Having piezoelectric properties BaTiO₃ comes with some disadvantages like low Curie temperature. It is discovered earlier which made its demand in industry high but further treatments can be done to enhance its properties.

Barium titanate is an inorganic compound which seems white as a powder and is clear when made as huge crystals. It is ferroelectric ceramic which exhibits the photorefractive effect and piezoelectric properties. The solid exists in one of the five polymorphs depending on temperature. From high to low temperature, these crystals symmetries of the five polymorphs are hexagonal, cubic, tetragonal, orthorhombic, and rhombohedral crystal structure. All of these phases exhibit the ferroelectric effect apart from cubic phase. Lower symmetry phases are stabilized at lower temperatures and involve movement of the Ba²⁺ to off-centre position. The remarkable properties of this material arise from the cooperative behaviour of the Ba²⁺ ions.

Since the piezoelectric effect is objectively low for a BaTiO₃ single crystal poled along the polar axis, a so-called domain wall engineering technique has been employed, making use of the piezoelectric anisotropy of single crystals [13]. It has been executed by poling the single crystal with the electric field along the direction which has an intersection angle with respect to the crystallographic polar axis. For example, Figure 1 shows the schematic model of the engineered domain configurations for the tetragonal 4 mm ferroelectric crystals [13]. If the poling electric field is along the [110] direction, it will generate two equally-preferred ferroelectric domains with spontaneous polarizations (Ps) along [100] and [010], respectively. In the case of [111] poled crystal, [100], [010], and [001] are three complimentary PS alignments for ferroelectric domains. Complex domain configuration has been formed, which enables enlarged piezoelectric response of the domain-engineered crystal. [13]

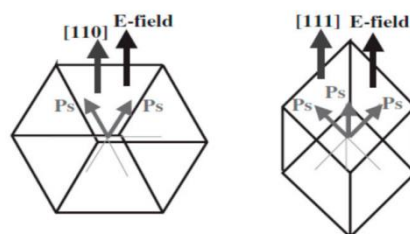


Figure1. Schematic model of the engineered domain configurations for the tetragonal 4mm ferroelectric Crystals (Ps: spontaneous polar vector along the <001>c directions) [14].

Piezoelectric anisotropy is the reason due to which the piezoelectric effect in batio₃ single crystal is enhanced [15]. It should be noted that the piezoelectric coefficients (e.g., d₃₃) usually show orientation-dependent phenomenon related with their Gibbs free energy, considering the crystal symmetry.

III. LEAD ZIRCONATE TITANATE (PZT)

Lead Zirconate titanate is an inorganic compound with chemical formula $\text{Pb}[\text{Zr}_x\text{Ti}_{1-x}]\text{O}_3$ ($0 < x < 1$). Also called PZT, it is a ceramic perovskite material that shows a marked piezoelectric effect, meaning that the compound changes shape when an electric field is applied. One of the commonly studied chemical composition is $\text{PbZr}_{0.52}\text{Ti}_{0.48}\text{O}_3$. The increased piezoelectric response and poling efficiency near to $x=0.52$ is due to the increased number of allowable domain states at the MPB. At this boundary, the six possible domain states from the tetragonal phase $\langle 100 \rangle$ and the eight possible domain states from the rhombohedral phase $\langle 111 \rangle$ are equally favourable energetically, thereby allowing a maximum 14 possible domain states.

Like structurally similar Lead scandium tantalate and Barium Strontium titanate, PZT can be used for manufacture of uncooled staring array infrared imaging sensors for thermo graphic cameras. Both thin film (usually obtain by chemical vapour deposition) and bulk structures are used. The formula of the material used usually approaches $\text{Pb}_{1.1}(\text{Zr}_{0.3}\text{Ti}_{0.7})\text{O}_3$ (called PZT30/70). Its properties may be modified by doping it with lanthanum, resulting in lanthanum-doped lead zirconium titanate (PLZT, also called lead lanthanum zirconium titanate), with formula $\text{Pb}_{0.83}\text{La}_{0.17}(\text{Zr}_{0.3}\text{Ti}_{0.7})_{0.9575}\text{O}_3$ (PLZT17/30/70).

In the 1950s, Jaffe and his co-workers found that the PZT system could exhibit strong piezoelectric effects. The maximum piezoelectric response was found in the PZT compositions near the morphotropic phase boundary (MPB) between the rhombohedral and tetragonal phases. Since then, the PZT system containing various additives has become the dominant piezoelectric ceramic for a variety of applications. The phase diagram of PZT pseudo-binary is shown in Figure 2(a), where T_c line is the boundary between the cubic paraelectric phase and the ferroelectric phases [16]. A significant feature of the PZT solid solution is the MPB, which divides the region of ferroelectric phase into two parts: a tetragonal phase region (on the Ti-rich side) and a rhombohedral phase region (on the Zr-rich side). In the PZT system, at room temperature, the MPB occurs close to $\text{Zr}/\text{Ti}=53/47$. The MPB represents an abrupt structural change within a solid solution with variation in composition but nearly independent of temperature. Usually it occurs because of the instability of one phase (such as ferroelectric tetragonal phase) against the other (ferroelectric rhombohedral phase) at a critical composition where two phases are energetically very similar but elastically different. The MPB compositions have mixed symmetries and therefore are easily poled in the polycrystalline form. Moreover, the phase boundary reduces anisotropy energy, lowers the domain wall energy, and thus, increases the domain wall mobility. In turn, this provides a high extrinsic domain wall contribution to the electromechanical properties [9]. It is noticed that the dielectric constant, piezoelectric and electromechanical behaviour attain maximum in the vicinity of the MPB composition, as shown in Figure 3 [16].

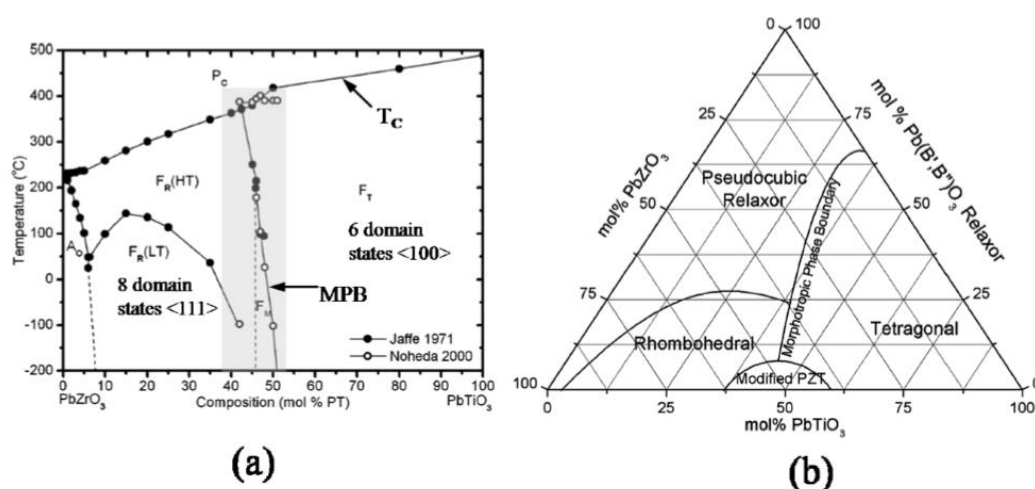


Figure 2. (a) Phase diagram of PbZrO_3 - PbTiO_3 system. P_c : paraelectric cubic, F_T : ferroelectric tetragonal, $F_R(\text{HT})$: ferroelectric rhombohedral (high temperature form), $F_R(\text{LT})$: ferroelectric rhombohedral (low temperature form), A_0 : antiferroelectric orthorhombic, T_c : cubic temperature, and MPB : morphotropic phase boundary. Close to the MPB , a stable monoclinic phase is discovered. Reproduced with permission from [16, 17]

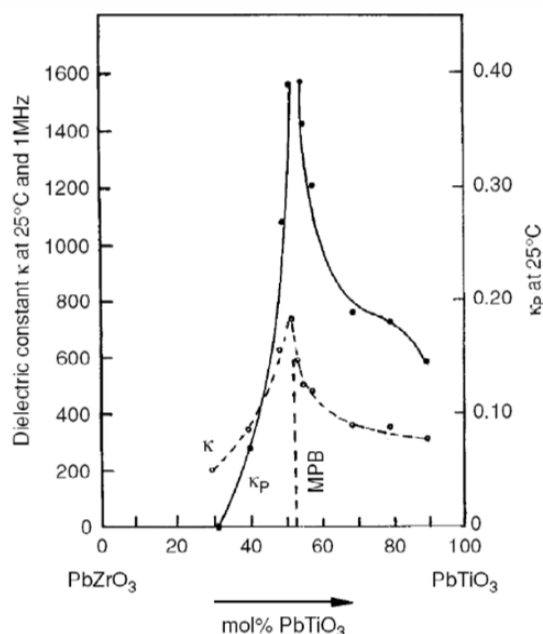


Figure3. Dielectric constant (κ) and electromechanical coupling factor (κ_P) for the PbZrO_3 - PbTiO_3 piezoelectric ceramic system. [16]

IV. APPLICATIONS OF ADVANCED PIEZOELECTRIC MATERIALS (BaTiO_3 , PZT)

Applications of piezoelectric materials have now expanded into many fields since the discovery of the effect by the Curie brothers in 1880. Significant progress in applications was made possible after the discovery of PZT ceramic materials. Piezoelectric devices can be splitted into four common categories: generators, sensors, actuators, and transducers depending of what type of physical effect used. For all of these basic functionalities, different designs are available. In the subsequent sections, some typical examples are chosen for detailed discussion, not only because of its technical importance but also its providing the opportunity to illustrate important ideas.

A. Piezoelectric Actuators

Actuators are devices that can convert input energy into mechanical energy [19]. Among the varieties of actuators with different input energy (considering electromagnetic, electrostatic, and thermal energies [20–22], piezoelectric actuators feature high strain output, high response speed, and high displacement control accuracy. As a result of those advantages, piezoelectric actuators have found wide applications ranging from high-tech equipment such as scanning tunnel microscopy (STM) and atomic force microscopy (AFM), to our daily life devices such as digital cameras and cellular phone terminals [13]. Most piezoelectric actuators depend on a material with sufficiently large electromechanical response known as the piezoelectric material. When applying an electric field in a particular direction, the piezoelectric material can produce a series of strain components satisfying various specific application requirements for actuators. The degree of electromechanical response for piezoelectric material, which describes the strain level with respect to external electric field, plays a important role on the performance of actuators.

Actuators are used in areas, including optics, astronomy, fluid control, and precision machinery. Piezoelectric strains induced by an electric field are used for actuator implementations. Figure 4 shows the design classification of ceramic actuators [23]. Simple devices composed of a disk or a multilayer structure use the strain induced in a ceramic by the applied electric field directly. Compound devices do not use the induced strain directly but use the louden displacement through a special magnification mechanism such as a unimorph, bimorph or moonie. The most popularly used multilayer and bimorph structures have the following characteristics: The multilayer structure does not have a large displacement (10 μm) but has advantages in generation force (1 kN), response speed (10 μs), lifetime (1011 cycles), and electromechanical coupling factor k_{33} (0.70). Unimorph and bimorph structures are defined by the number of piezoelectric ceramic plates: only one ceramic plate is bonded onto an elastic shim, or two ceramic plates are bonded together.

A composite actuator structure called a “moonie” is developed to amplify the small displacements induced in piezoelectric ceramics. The moonie consists of a thin multilayer element and two metal plates that have narrow moon-shaped cavities bonded together, as shown in Figure 4(c) [23]. This device has characteristics intermediate between the conventional multilayer and bimorph actuators; it has an order of magnitude larger displacement (100 μm) than the multilayer, much larger generative force (100 N), and faster response (100 μs) than the bimorph.

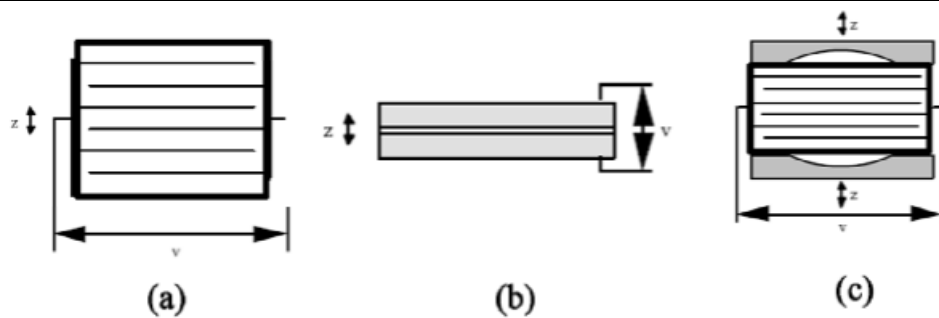


Figure 4. Structures of piezoelectric ceramic actuators. (a) Multilayer, (b) bimorph, and (c) moonie structures. [23]

B. Ultrasonic Motor

Piezoelectric materials play a major role in ultrasonic motors and other piezoelectric actuators because of their purpose to transform electrical energy into mechanical energy. Various parameters of some important piezoelectric materials are displayed in Table 1. Since the characteristics of Piezoelectric can be widely adjusted by substituting or doping additives, the data show only a rough range. In the table, T_c is the Curie temperature and E_c is the coercive field. The necessity in properties of piezoelectric materials has to be identified according to their specific purposes of the devices. These used in ultra-high frequency (UHF) and high-frequency devices need the material to have low permittivity and low high frequency dielectric loss. For energy transducer implementation, the coupling coefficient and acoustic impedance of the material are often stressed. Materials with excellent frequency stability and high Q_m values can be used as standard frequency oscillators [24, 25].

There are different types of ultrasonic motors available such as –

- Standing wave piezoelectric ultrasonic motor
- Travelling wave piezoelectric ultrasonic motor
- Linear piezoelectric ultrasonic motor
- Bar type travelling wave rotary piezoelectric motor
- Double rotor piezoelectric ultrasound motor

Concentrated research on piezoelectric ultrasonic motors began after the discovery of Sashida's first practical ultrasonic motor in 1982. The elliptical movement on the stator surface is created by a proper superposition of two orthogonal flexural waves. The piezoelectric ring has a segmented electrodes and the piezoelectric ceramic under each segment is polarized in such a way that one group of segments excites the sine mode and the other group excites the cosine mode. As a type of ultrasonic motor, a linear ultrasonic motor (LUSM) also uses the converse piezoelectric effect of piezoelectric ceramics and the ultrasonic vibration of an elastic body. It transmits the micro amplitude movement of a stator into the macro linear motion of a slider by the friction force between the stator and slider. The bar-type travelling wave rotary piezoelectric ultrasonic motor consist of two types: single degree of freedom (SDOF) and multi degrees of freedom (MDOF) ultrasonic motors. Both of them use the vibration of the bar type round stator to achieve conversion from electrical energy to mechanical energy.[26]

C. Piezoelectric Ceramics Based Sensors

A piezoelectric ceramic sensor is a gadget that utilizes the piezoelectric effect to count pressure, acceleration, strain or force by modifying them to an electrical signal. The principle of performance of a piezoelectric ceramic sensor is that a physical dimension, transformed into a force, acts on two combating faces of the sensing element. Depending on the design of a sensor, different "modes" to load the piezoelectric element can be used: longitudinal, transversal and shear [27]. The way a piezoelectric material is cut produces three main operational methods for sensors which are:

- Transverse
- Longitudinal
- Shear

In transverse type, force is applied along neutral axis(y) displaces charges along (x) direction, perpendicular to line of force whereas, in longitudinal type the amount of charge displaced is strictly proportional to the applied force and self-sufficient of the piezoelectric element size and shape. Putting various elements mechanically in series and electrically in parallel is the only way to improve the charge output. The charge produced in shear effect is strictly proportional to the applied and independent of element size and shape. In opposing to the longitudinal and shear effects, the transverse effect makes it possible to fine –tune sensitivity on the applied force and element dimension.

Two prime groups of materials are utilized for piezoelectric sensors: piezoelectric ceramics and single crystal materials. The ceramic materials (such as PZT) have a piezoelectric constant/sensitivity that is nearly two orders of magnitude higher than those of the natural single crystal materials and can be produced by cheap sintering processes. The piezoeffect in piezo-ceramics is maintained, so their high sensitivity degrades over time. This degradation is highly correlated with increased temperature. The less-sensitive, natural, single-crystal materials (gallium phosphate, quartz, tourmaline) have higher when carefully handled, almost enormous long term stability. There are also new single crystal materials fruitfully available such as Lead Magnesium Niobate-Lead Titanate (PMN-PT). These materials offer improved sensitivity over PZT but have a lower maximum operating temperature and are currently more expensive to manufacture.

D. Smart structures

There is an increasing consciousness of the upper hand to be obtained from the enlargement and misuse of smart materials and structures in implementation ranging from hydrospace to aerospace. With the potentiality to react autonomously to changes in their environment, smart systems can offer a simplified prospective to the control of various material and system characteristics such as noise, shape and vibration, etc., depending on the smart materials used. Use of piezoelectric materials in smart structure includes:

I. Active Vibration Damping

Smart structures that use different piezoelectric patches to control the vibration of thin plates have been of countable interest in recent years. The development of finite element codes, such as ANSYS, makes it possible to fully model coupled thermo mechanical-electrical systems of one or more dimensions and obtain reciprocal relations between piezoelectric actuator voltages and system response. By combining such models into a closed-loop control system, very effective active vibration suppression can be reached.

Piezoelectric actuators and sensors are also required for active vibration control. Researchers at TU Darmstadt in Germany have been investigating ways to lower vibrations by attaching piezoelements. When the material is bent by a vibration in one direction, the system responds to the bend and sends electric power to the piezoelement to bend in the other direction. The idea is to detect the changes in the vibration behaviour of the entire wheel caused by the surface changes on the rolling contact area, as schematically shown in Figure 5 [28]. Piezoelectric sensors are kept on different areas of the wheel, changing the displacements on the surface into electrical signals.

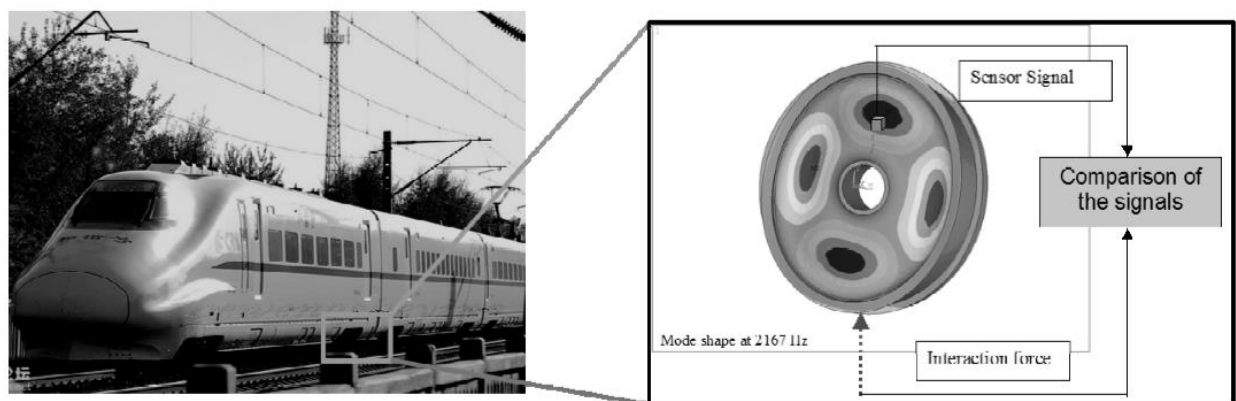


Figure 5. The method used for the assessment of the roughness of the wheel. A piezoelectric sensor detects the vibrations of the wheel, leading to an assessment of its wear status (source: TU Darmstadt). Reproduced with permission from [28]

II. Active Noise Control

The sources for noise are numerous. For example, noise can be emitted from engines, exhaust systems, fans, and blowers. Active noise control (ANC) was developed as a way to reduce if not eliminate some of these different types of noise. Active control of sound is very identical in nature to ANC and is often narrated in the same terms. ANC works on the simple principle of destructive interference, where the undesirable sound wave is tackled with a sound wave of equal amplitude, but 180 degrees out of phase. The result is that the sound waves eliminate each other, and the undesirable sound is reduced or eliminated. This principle is implemented in smart structures, including noise cancellation headsets, transformer quieting systems, and interior noise reduction in automobiles and aircraft [30, 31]. Piezoelectrics are used more often to monitor the noise associated with panel vibrations such as those present in transformers. In underwater acoustics, piezoelectrics simply far outperform other types of sensors and actuators for the creation and reception of sound. It is generally very distinct to produce broadband sound sources for underwater due to difficulties related with the design and manufacture of acoustic transducers. Active control of sound principles can be utilized to generate such a source.

III. Active Shape Control

Active shape control is of substantial attraction for reflectors and antennas that must maintain accurate dimensions for optimal performance. This is of specific importance for structures in space that are made of lightweight materials and are exposed to thermal distortion. Instruments such as millimetre-wave and sub-millimetre-wave passive instruments and those operating up to the infrared spectrum for space radio astronomy need to maintain micrometer accuracy, with overall dimensions of up to a few meters [29]. Yet thermal distortions of space structures of this size even those using low co-efficient of thermal expansion materials such as carbon fibre reinforced plastic can result in RMS surface errors on the order of 0.8mm [29]. For these applications, smart structures involving attached piezoelectric actuators have been proposed [29].

IV. Active Health Monitoring

Comprehensive methodologies [32] for locating and determining the extent of linear crack in beams, plates and pipes have been developed based on the time-of-flight analysis of Lamb wave propagation via appropriate data analysis methods such as wavelet transform started by Wang et al. [32] and Hilbert-Huang transform (HHT) [32]. These methodologies and applications are reviewed hereinafter. Nowadays, an important application field of IDT is in the health monitoring of structures. IDT is a thin piezoelectric film surface bonded on either piezoelectric or unpiezoelectric substrate for the use of wave excitation or reception of structures. On the surface of the wafer, a type of electrodes is designed, which consist two alternating sets of fingers connected to external electric power sources for the energy supply. Since IDT is bonded on a piezoelectric layer which is surface bonded on the metal substrate, the piezoelectric effects must be modelled in the dispersion characteristics of the piezoelectric coupled structure. Jin et al. [32] derived the lamb wave propagation in a metallic semi-infinite medium covered with piezoelectric layer.

Owing to their exceptional mechanical and electric coupling properties, piezoelectric materials hold many potential applications in the fields of structural health monitoring and repair. It is found from research that piezoelectric materials have exceptional sensing and exciting properties. It can be concluded that plain piezoelectric sensors and actuators and IDT have the capability to detect the cracks in beam, plate and pipe with reasonable accuracy. In addition, as an option to conventional materials for structural restore, the piezoelectric materials consist promising features (e.g., light weight) to overcome the problems caused by conventional repair methods. The delamination and notch crack in beam and plate structures can be successfully repaired via removing the singularity at the crack tip. Breakthroughs are supposed to get rid of the obstacles existing in coupling coefficients and price for the implementation in industry scale.

V. CONCLUSIONS

In this review, we present an overview of the state of art in piezoelectric ceramic materials, which includes their processing, properties, characterization, and the potential applications. More than one century after the discovery of piezoelectricity, piezoelectric ceramics have become commercially viable. Among the fabrication process of piezoelectric ceramics, poling process is the most critical step, which is necessary to induce the piezoelectricity in the polycrystalline ferroelectric ceramics. To meet with stringent requirements for specific applications, compositional modifications of piezoelectric ceramics with different doping conditions are possible to adjust the properties to a remarkably wide range of requirements. Characterization of the piezoelectric properties of piezoelectric ceramics is crucial for establishing the relationship between the manufacturing process and ceramic performance, which enables ones to adjust the manufacturing process of the piezoelectric ceramics to produce tailored materials. Insights obtained through specifications have led to many new devices and uses. Significant progress has been made in the applications of piezoelectric ceramics since the discovery of PZT ceramics. Various potential applications of piezoelectric ceramic materials in ultrasonic actuators, sensors, transducers, and active vibration controlling, are presented in this review, and the personal perspectives towards future trends of piezoelectric ceramic materials are also presented.

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