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Development of Piezoelectric property materials for the Nanogenerator production

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ABSTRACT

The new trend of materials science and mechanical energy harvesting facilitates the understanding of high performance piezoelectric material systems. In this system is soft, flexible or stretchable formats, with special opportunities for use in bio-integrated applications, from mechanical energy harvesting into sensing devices. This paper reviews the piezoelectric materials and its types of recent developments. It converts mechanical energy into electrical energy for applying small signal like stress, strain, and vibrational movement of human body surface. It can be able to harvest such body-twisted biomechanical and thermal energies by exploiting piezoelectric, triboelectric, and thermoelectric physical property.

Key-words: Piezoelectric material, flexible, mechanical energy, electrical energy, nanogenerators

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INTRODUCTION

Piezoelectricity is the capability of certain materials to generate an alternating current (AC) voltage when subjected to mechanical stress or vibration, or to vibrate when subjected to an AC voltage, or both. The most common piezoelectric material is quartz. The some other materials of ceramics, Rochelle salts, and various other solids also exhibit this effect. The word "Piezoelectric" is derived from the Greek "piezein", which means to squeeze or press, and "piezo", which is Greek word for "push"[1]. The unique characteristic of piezoelectric effect is reversible, materials exhibits, first one is direct piezoelectric effect (generation of electricity while stress is applied) as shown in Fig. 1 and second one is the converse piezoelectric effect (generation of stress while an electric field is applied) as shown in Fig. 2. A shifting of positive and negative charge centers in piezoelectric material takes place during the mechanical stress, which followed by results in an external electrical field. An outer electrical field either stretches or compresses the piezoelectric material, when reversed.

The piezoelectricity is measured by piezoelectric coefficient or piezoelectric modulus (D). It can be defined as the change in volume that it undergoes when subjected to an electric field or as the polarization that it undergoes when mechanical stress is applied. This is mathematically represented as

$$D = P/\sigma \quad (1)$$

where, P: polarization and σ : stress².

WORKING PRINCIPLE

The working principle of piezoelectricity is illustrated in Fig. 3. Normally, charges in a piezoelectric crystal are exactly balanced, even if they are not symmetrically arranged. The effects of charges exactly cancel out, leaving no net charge on crystal faces. More specifically, electric dipole moments-vector lines separating opposite charges exactly cancel one another out. If crystals are squeezed (massively exaggerated in this picture), it forces the charges out of balance. Now, the effects of charges (dipole moments) no longer cancel one another out and net positive and negative charges appear on opposite crystal faces. Thus, squeezing of crystal, produces voltage across its opposite faces.

Piezoelectric materials

Piezoelectric materials are broadly into two category (i) natural and (ii) man-made materials [3, 4]. Natural piezoelectric materials are Berlinite (structurally identical to quartz), cane sugar, quartz, Rochelle

salt, topaz, tourmaline, sodium potassium tartarate and bone. Man-made piezoelectric materials are Ceramics, sintered form of finely ground powdered mixture made of ferroelectrics of oxygen-octahedral types are lead zirconate titanate [Pb(Zr, Ti)O₃], Lead titanate (PbTiO₃), Barium titanate (BaTiO₃), Zinc oxide (ZnO) and polymer is polyvinylidene fluoride (PVDF).

Developments in piezoelectric materials

The selection of piezoelectric material for a power harvesting application is the major influence creation on its functionality and performance. Although number of different piezoelectric materials have been developed, lead zirconate titanate (PZT) as a power harvesting material. Despite of its extreme brittle nature causes limitations in strain. Lee [5] highlights that these piezoceramics are susceptible to fatigue crack growth when subjected to high frequency cyclic loading.

Another common piezoelectric material is poly vinylidene fluoride (PVDF). It exhibits considerable flexibility when compared to PZT. Lee [5, 6] developed a PVDF film coated with poly(3,4-ethylenedioxythiophene)/ poly(4-styrenesulfonate) [PEDOT:PSS] electrodes. They compared the PEDOT:PSS coated films to inorganic electrode materials, indium tin oxide (ITO) and platinum (Pt) coated films. It was found that Pt electrodes and ITO electrodes showed fatigue crack on surface at vibration frequency of 33 kHz and 213 Hz, respectively. However, PEDOT:PSS film does not damage even at vibration for 10 h at 1 MHz. Mohammadi [7] developed a PZT fiber-based flexible composite piezoelectric material of various diameters (15, 45, 120, 250 μm) and were aligned, laminated, molded in an epoxy [8]. The voltage output of sample was tested by dropping a 33.5 g (20 mm diameter) stainless steel ball on piezoelectric material from a height of 10 cm. The peak power was also calculated considering a 1 M Ω load resistance. A maximum voltage and power output of 350 V and 120 mW was obtained for the thickest transducer, 5.85 mm thick, with the smallest fiber diameter, 15 μm . Upon studying the relationship between voltage output of the harvester and its physical geometry, it was determined that thicker plates have the capability of larger fiber displacements, and that samples with smaller diameter fibers have the highest piezoelectric coefficient, d_{33} and lowest dielectric constant defined in this study as K_3 , both of which contribute towards higher power outputs and more efficient systems.

Piezofiber power harvesting materials have also been investigated by Churchill *et al* [9] who tested a composite consisting of unidirectionally aligned PZT fibers of 250 μm diameter embedded in a resin matrix. It was found that when a 0.38 mm thick sample of 130 mm length and 13 mm width was subjected to a 180 Hz vibration that caused a strain of 300 μm in the sample, the composite was able to harvest about 7.5 mW of power. The results of this study show that a relatively small fiber-based piezoelectric power harvester can supply usable amounts of power from cyclic strain vibration in the local environment.

Sodano¹⁰ presented a comparison of several piezoelectric composite devices for power harvesting that are normally used for sensing and actuation. The power harvesting ability of macro-fiber composite (MFC), quick pack IDE (model QP10ni), and the quick pack model (QP10n) actuators was tested. The MFC contains piezofibers embedded in an epoxy matrix which affords it extreme flexibility, and it utilizes interdigitated electrodes, which allow the electric field to be applied along the length of the fiber and act in the higher d_{33} coupling mode, as shown in Fig. 4. Thus interdigitated electrode pattern of the MFC and the quick pack IDE results in low-capacitance devices which limits the amount of power that can be harvested.

Flexible piezoelectric materials are attractive for power harvesting applications because of their ability to withstand large amounts of strain. Larger strains provide more mechanical energy available for conversion into electrical energy. A second method of increasing the amount of energy harvested from a piezoelectric is to utilize a more efficient coupling mode. Two practical coupling modes exist; -31 mode and -33 mode. In -31 mode, a force is applied in the direction perpendicular to the poling direction, an example of which is a bending beam that is poled on its top and bottom surfaces. A force is applied in the same direction as the poling direction, such as the compression of a piezoelectric block that is poled on its top and bottom surfaces in case of -33 mode. An illustration of each mode is presented in Fig. 5. Conventionally, -31 mode has been the most commonly used coupling mode, however -31 mode yields a lower coupling coefficient k , than -33 mode.

Sodano [12] compared the efficiencies of piezoelectric materials viz., traditional PZT, quick pack (QP) actuator, and macro-fiber composite (MFC), subjected to 0–500 Hz chirp, and exposed to random vibrations recorded from an air compressor of a passenger vehicle. It was found that the efficiency of PZT for each vibration scheme was fairly consistent (4.5% at resonance, 3.0% for a chirp, and 6.8% for random vibrations) and was higher than the other two devices. A summary of various piezoelectric materials discussed above are depicted in Table 1.

Ng and Liao [15, 16] presented two types of bimorphs (Fig. 6) along with a unimorph piezoelectric harvester. The first bimorph (series triple layer) consisted of two piezoelectrics with a metallic layer

sandwiched between them. The second bimorph (parallel triple layer) similar to series triple layer and piezoelectrics were connected electrically in parallel mode. The finding is showed that under low load resistances and excitation frequencies the unimorph generated the highest power, under medium load resistances and frequencies. The parallel triple layer had the highest power output, and under high load resistances and frequencies the series triple layer produced the greatest power. Thus, a series connection should a increased device impedance leading to more efficient operation at higher loads.

Properties of Piezoelectric Materials

Modulus and Density

Piezoceramics have modulus values are in the range of 10 to 100 GPa with a density typically in the range of 7000 to 8000 kg/m³. Piezoelectric polymers are softer materials with elastic modulus in the range of 1 to 3 GPa with a density in the range of 1000 to 2000 kg/m³.

Strain and Stress

The piezoelectric materials typically produce 0.1 % strains. The stresses that can be produced by hard piezoceramics are on the order of tens of megapascal. Piezoelectric polymers can produce stresses that are 1/100 of the stresses that can be produced by piezoelectric ceramics.

Energy Production Capacity

The Volumetric energy density is the Product of stress and strain the capacity to do work per unit volume. Use of energy densities hard and soft piezoelectric polymers are in the range of 10 to 100 kJ/m³. The highest stresses may be obtained by limiting strain of piezoceramics by 0.1% but will be on the order of 10 to 100 % for soft piezoelectric polymers. Speed of response for a material that is used as an actuator is also important. Since piezoelectricity is associated with molecular changes, the response time for piezoelectric materials is very small (on the order of few microseconds)¹⁷.

Uses of piezoelectric materials

There are situations where in mechanical energy is converted (pressure or movement) into electrical signals or vice-versa. Piezoelectricity is found in useful applications, such as the production and detection of sound, generation of high voltages, electronic frequency generation, microbalances, to drive an ultrasonic nozzle, and ultrafine focusing of optical assemblies. It is also the basis of a number of scientific instrumental techniques with atomic resolution, the scanning probe microscopies, such as STM, AFM, etc., and everyday uses, such as ignition source for cigarette lighters, and push-start propane barbecues, as well as the time reference source in quartz watches¹⁸.

(i) Piezoelectric transducer: A transducer is simply a device that converts small amounts of energy from one kind into another (for example, converting light, sound, or mechanical pressure into electrical signals).

(ii) Ultrasound equipment: In generally piezoelectric transducer is converts electrical energy into extremely rapid mechanical vibrations. In fact, that it makes sounds, but ones too high-pitched for our ears to hear. These ultrasound vibrations can be used for scanning, cleaning and etc.

(iii) Microphone: Conversion of sound energy (waves of pressure traveling through air) into electrical energy using piezoelectric crystals. The attachment of crystal with vibrating part of microphone, as pressure waves from voice arrive will make crystal to move back and forth, thus generates corresponding electrical signals and stereo converts back into audible sounds.

(iv) Quartz clock or watch: The reverse-piezoelectric effect is used to keep time (period) very precisely. Electrical energy (battery) is fed into a crystal to make it oscillate thousands of times a second. The electronic circuit, slower, further once-per-second beats a tiny motor and some precision gears to drive second, minute, and hour hands around the clock-face.

(v) Spark lighters: The squeezing a piezoelectric crystal, while press the switch generating a voltage and making a spark fly across a small gap.

(vi) Inkjet printer: Generally, inkjets squirt their syringes using electronically controlled piezoelectric crystals, which squeeze their "plungers" in and out; Canon bubble jets fire their ink by heating it instead.

Piezoelectric nanogenerator

Nanogenerator converts mechanical or thermal energy into electricity as produced by small-scale physical change.

It is an energy harvesting device. It converts external kinetic energy into electrical energy by a nano-structured piezoelectric material. Although its definition may include any types of energy harvesting devices using nano-structures to convert various types of ambient energy (e.g. solar power and thermal energy) [19], is typical approaches are piezoelectric, triboelectric, and pyroelectric nanogenerators.

Principle

The working principle of nanogenerator can be explained in two ways with respect to force exerted (i) perpendicular and (ii) parallel to the axis of nanowire (Fig. 8).

When a piezoelectric structure is subjected to external force by moving tip, deformation occurs throughout the structure. This will create electrical field inside the nanostructure; stretched part with positive strain will exhibit the positive electrical potential, whereas the compressed part with the negative strain will show the negative electrical potential. This is due to the relative displacement of cations with respect to anions in its crystalline structure. The nanowire will have an electrical potential distribution on its surface in tip, while the bottom of the nanowire is neutralized, since it is grounded. The nanowire will generate maximum voltage. This maximum voltage can be calculated by the following equation²⁰

$$V_{max} = \pm \frac{3}{4(k_0 + k)} [e_{33} - 2(1 + \nu)e_{15} - 2\nu e_{31}] \frac{a^3}{l^3} v_{max}$$

where k_0 is the permittivity in vacuum, k is the dielectric constant, e_{33}, e_{31}, e_{15} are the piezoelectric coefficients, ν is the Poisson ratio, a is the radius of the nanowire, l is the length of the nanowire and v_{max} is the maximum deflection of the nanowire's tip. The formation of schottky contact will generate direct current output signal consequently.

In second case, a model with a vertically grown nanowire stacked between the ohmic contact at its bottom and the schottky contact at its top is considered (Fig. 8). When the force is applied toward the tip of nanowire, uniaxial compression will be generated in the nanowire. Due to piezoelectric effect, tip of nanowire will have a negative piezoelectric potential, increasing the fermi level at the tip. Since the electrons starts flowing from tip to bottom through the external circuit, as a result, the positive electrical potential will be generated at the tip. The potential at the tip is maintaining because of the schottky contact will barricade the electrons being transported through the interface. As the force is removed, piezoelectric effect diminishes, and the electrons will be flowing back to the top in order to neutralize the positive potential at the tip. The second case will generate alternate current output signal.

DEVELOPMENTS IN PIEZOELECTRIC NANOGENERATOR

The piezoelectric energy conversion are proposed and investigated by many research groups^{21, 22}. In particular, flexible energy harvester called a nanogenerator which can generate electrical energy from not only mechanical energy but also tiny biomechanical energy (e.g., heartbeat, muscle motions, and eye blinking) have attracted attention in response to the demands of infinite self-powered sources for operating flexible and wearable electronic systems [23].

ZnO nanowires: The first nanogenerator is the piezoelectric ZnO nanowires (NW) based energy device (Wang and co-workers), (Fig. 8a) [24–27]. A single ZnO NW on a flexible polyimide (PI) substrate successfully convert from the human finger motions (Fig. 8a-i) and the running/scratching motions of a hamster to electricity [24, 25]. To enhance the output performance of nanogenerator have used the transferred lateral ZnO nanowire arrays on a flexible substrate [26], that have provided the technical advancement to operate commercial electronic devices using the nanogenerator technology (Fig. 8a-ii). Choi [27] reported that the transparent and flexible nanogenerator which involves the ZnO nanorod (NR) arrays and indium tin oxide (ITO) electrodes coated polyether sulfone (PES) flexible substrates (Fig. 8a-iii).

polyvinylidene fluoride (PVDF): Since polymer materials have the naturally flexible properties and mechanical stabilities, the piezoelectric polymers as polyvinylidene fluoride (PVDF) have been used to fabricate polymeric flexible nanogenerators (Fig. 8b) [28–30]. Fig. 8b-i presents the piezoelectric PVDF nanofibers placed on a working substrate using a unique direct write technique. The PVDF based nanogenerator generates electrical outputs of about 5–30 mV and 0.5–3 nA when the substrate is deformed by stretching and releasing motions [28, 29]. Cha [30] proposed the polymeric nanogenerator made of porous PVDF by employing a novel template-assisted method (Fig. 8b-ii). This sonic wave driven energy harvester produces higher output performance compared to dense PVDF based nanogenerators due to the effective nanoporous structure.

Perovskite-structured ceramic [PbZr_xTi_{1-x}O₃ (PZT) and BaTiO₃]: In 2010, there have been new approaches to use perovskite-structured ceramic [PbZr_xTi_{1-x}O₃ (PZT) and BaTiO₃] thin films with inherently high piezoelectricity for higher energy conversion efficiency [31–34] (Fig. 8c). The PZT and BaTiO₃ thin films on bulk substrate are transferred onto flexible PI substrates by adopting unique transferring techniques after high temperature crystallization process (Fig. 8c-i, ii). The perovskite thin film-based nanogenerators show higher power density compared with other flexible piezoelectric devices with the similar device structure. Qi [33] presented the energy harvester of wavy piezoelectric PZT ribbons, that can scavenge stretching motions to generate.

Bio-ecocompatible piezoelectric ceramics: Recently, the research on large-area, low-cost, mechanically-stable, and high-output nanocomposite generator has been invented by casting piezoelectric

nanocomposites onto flexible plastic substrates at low temperature [35-39]] (Fig. 8d). In particular, several research groups have developed lead-free and high performance flexible energy harvesters using bio-ecocompatible piezoelectric ceramics such as BaTiO₃ [35], (K, Na) NbO₃ [36, 38, 37], and Li-doped (K, Na) NbO₃ [39]. Subsequently, a new concept of ultra-stretchable elastic composite generator was also developed by employing the Ecoflex silicone rubber-based piezoelectric composites and long silver nanowire-based stretchable electrodes [40]. A summary of the various piezoelectric materials used in Nanogenerators for harvesting energy from the surface of the human body as shown in Table 2.

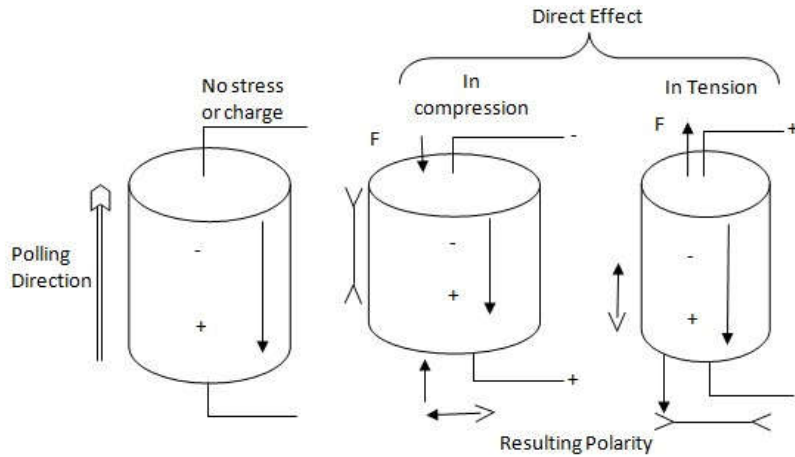


Fig. 1 Direct piezoelectric effect (redrawn)

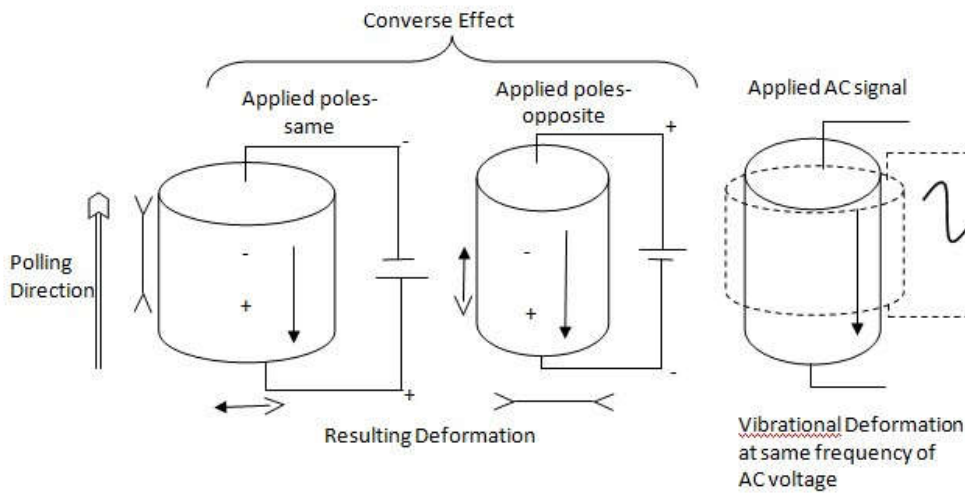


Fig. 2 Converse piezoelectric effect (redrawn)

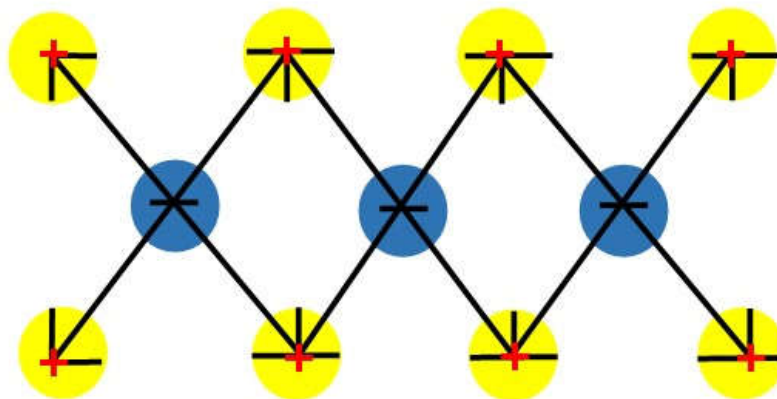


Fig. 3 Working principle of piezoelectricity (redrawn)

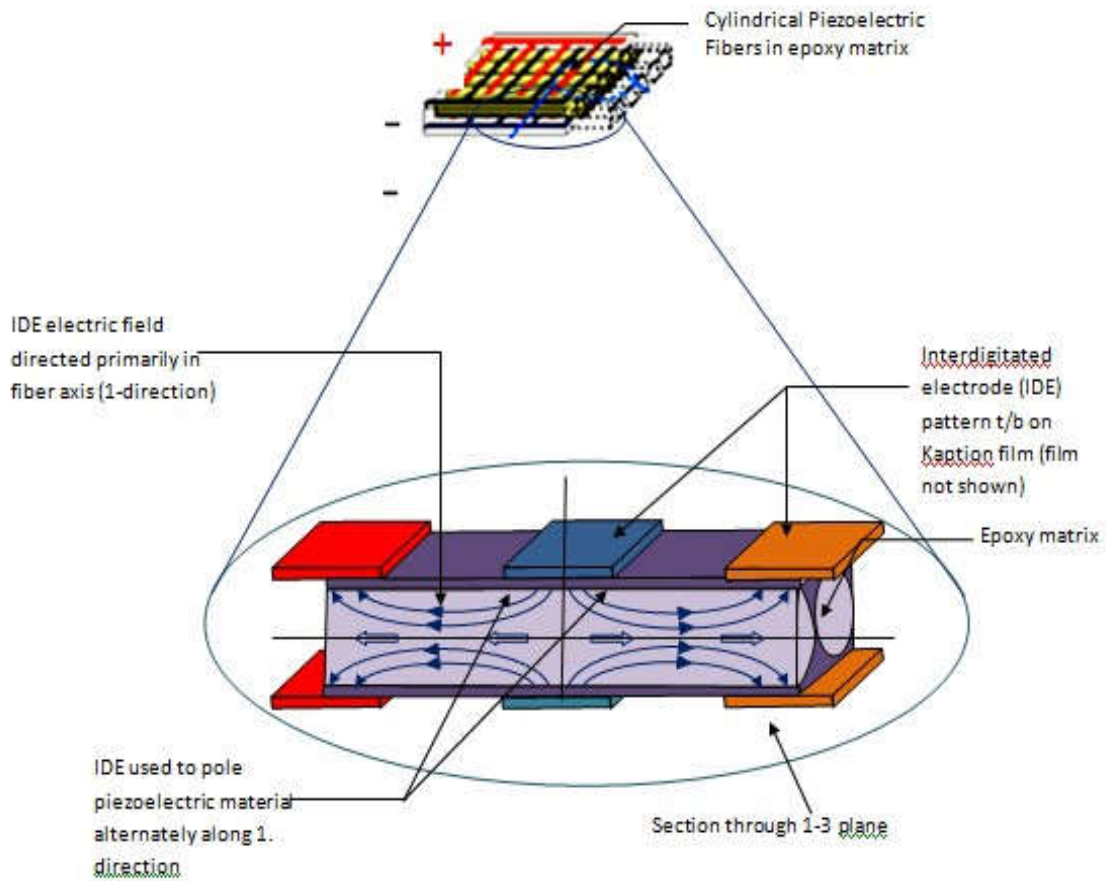


Fig. 4 Schematic cross sectional view of an active fiber composite (AFC) actuator [31]
Flexible piezoelectric materials are attractive for power harvesting applications (redrawn)

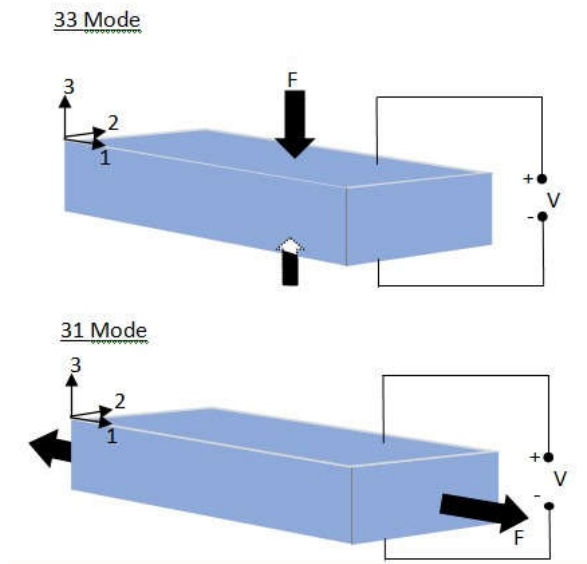


Fig. 5 Illustration of -33 mode and -31 mode operation for piezoelectric materials [33] (redrawn)

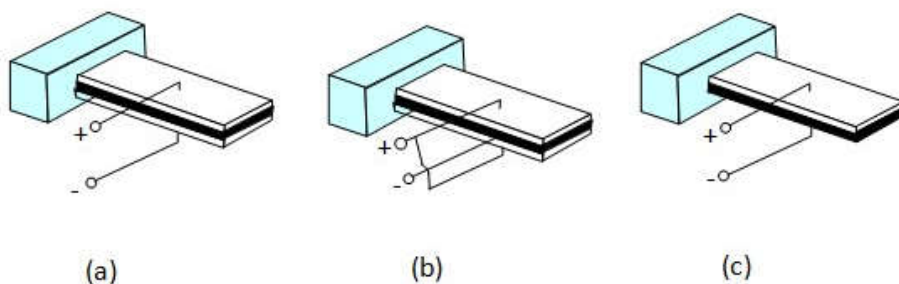


Fig. 6 (a) A series triple layer type piezoelectric sensor, (b) A parallel triple layer type piezoelectric sensor, (c) A unimorph piezoelectric sensor (redrawn)

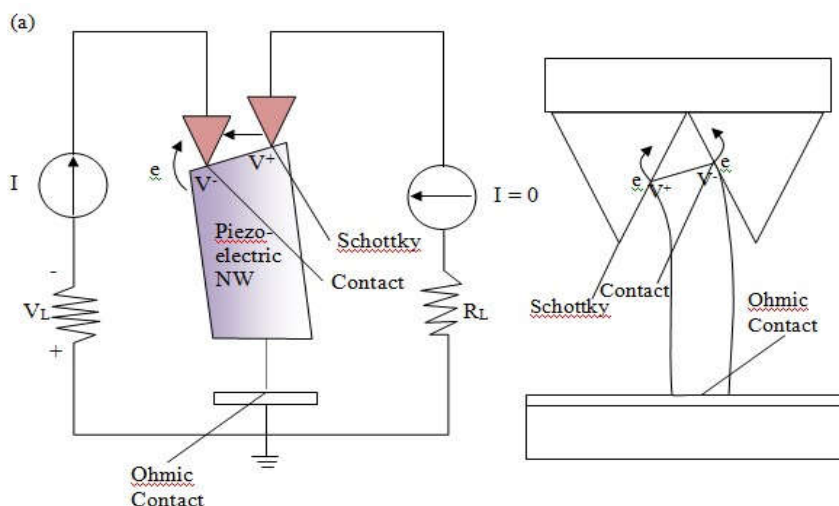


Fig. 7 (a) An active fiber composite (AFT) tip is swept through the tip of nanowire. Only negatively charged portion will allow the current to flow through the interface. (b) The nanowire is integrated with the counter electrode with AFT tip-like grating. As of (a), electrons are transported from the compressed portion of nanowire to the counter electrode because of Schottky contact (redrawn)

(a) ZnO Nanowire-based Nanogenerators

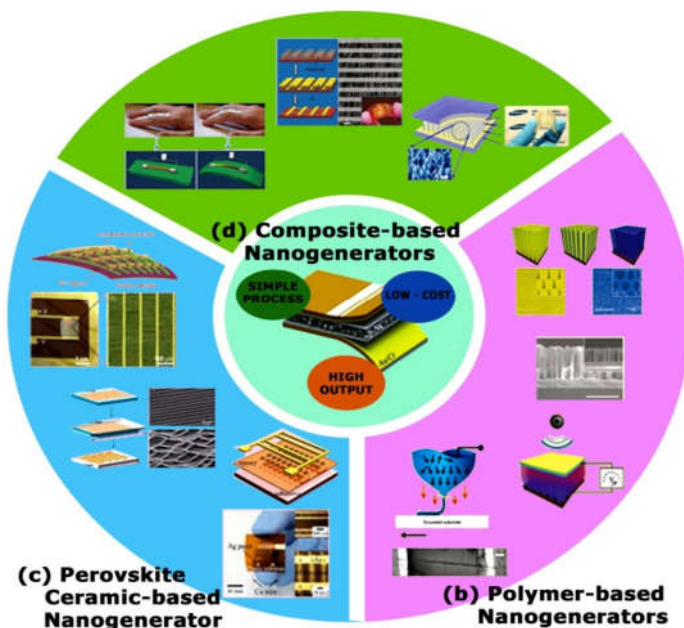


Fig. 8 Some flexible energy harvesting devices. Nanogenerators based piezoelectric materials such as [60] (a) ZnO NW [12, 14, 15], (b) PVDF polymer [16, 18] (c) Perovskite ceramic thin film [19, 21,22] (redrawn)

Table. 1 Summary of several piezoelectric materials investigated

Author	Type of material	Advantages/disadvantages	Power harvesting capabilities
Lee [6]	Monolithic PZT	Most common type of device. Not flexible. Susceptible to fatigue crack growth during cyclic loading	N/A
Lee [5, 6]	PVDF film coated with PEDOT/PSS electrodes	Resistance to fatigue crack damage to electrodes	N/A
Mohammadi [7]	Piezofiber composite	Increased flexibility	120 mW from 34 × 11 mm plate of 5.85 mm thickness
Churchill [9]	Piezofiber composite	Increased flexibility	7.5 mW from 130 × 13 mm patch of 0.38 mm thickness
Sodano [10]	MFC composite, quick pack IDE, quick pack	MFC—flexibility MFC and quick pack IDE—low-capacitance devices quick pack—energy harvesting capability	quick pack proved to harvest the most energy
Schonecker [11]	Monolithic PZT, quick pack, MFC	MFC—flexibility quick pack and monolithic PZT—energy harvesting capability	PZT proved to be most efficient (6.8% for random vibration excitation)

Table. 2 Various piezoelectric materials used in Nanogenerators for harvesting energy from human body surface

Nanogenera tor active material	Electrode	Substrate	Nanogenera tor dimension	Mechanical load/tensile strain	Nanogenera tor flexibility	Human body application	Voltage	Current
Single ZnO NW [24, 25]	Ag	PI film	Area: _ ; Thickness: 50 µm	Tensile strain measured: 0.05-0.2%	Bending radius : 2 cm	Finger bending	0.25 mV	150 pA
ZnO film [41]	ITO and Ag	PET	-	-	Rolling curv. Radius: 2 cm	Rolling and muscle stretching	0.28 V	-
Single ZnO and PVDF fiber [42]	Au	PS and PDMS	Fiber length: 20 mm; Thickness: 6.2 mm	Tensile strain measured: 0.1%	-	Elbow folding	0.1 V	-
P(VDF-TrFe) sheet [43]	Ag	PI	Area: 9× 18 mm ² ; Thickness: 245 µm	Ultra-high sensitivity for measuring: 0.1 Pa	-	Skin vibration	1.5 V	40 nA
PVDF-NaNbO ₃ nonwoven fabric ⁴⁴ , 2013	PU yams and Ag coated PA yams	PDMS	Area: 25 × 25 mm ² ; Thickness: 2000 µm	Compressive pressure: 0.2 MPa	-	Heet strike	3.4 V	4.4 µA
ZnO film [45, 46]	Al	Al foil and PMMA	Area: 13× 5 mm ² ; Thickness: 18 µm	-	Bending radius: 3 mm	Bending motion	0.2 V	2 nA
ZnO and PVDF composite [47]	Au	PDMS	Area: 30× 30 mm ² ; Thickness: 30 µm	-	-	Finger bending	0.33 V	62 nA
ZnO film [48]	AZO	Ecoflex	Area: 50× 50 mm ² ; Thickness: 305 µm	Compression strain: -0.22%	-	Movement of wrist tendons	1 V	100 pA

BaTiO ₃ and P(VDF-HFP) composite [49]	Al	PDMS and PI	Area: 2 × 2 mm ² : Thickness: 350 μm	Applied pressure: 0.23 MPa	-	Finger tapping	75 V	15 μA
PVDF filaments as spacer yarns [50]	Ag-coated PA66 yarns	Pu	Area: 53×150 mm ² : Thickness: 3.5 mm	Compressive pressure: 0.10 MPa	-	Fabric to wear	14 V	30 μA
PZT film [51-53]	Cr/Au and Ti/Au	PET	Area: 20 × 50 mm ² : Thickness: 225 μm	Tensile strain measured: 0.3%	Bending radius: 21 mm	Wrist movement	120V	2 μA
BaTiO ₃ / PVC fibers ⁵⁴ , 2015	Cu	PET	-	-	-	Elbow folding, fabric to wear	1.9 V	24 nA
PVDF sheet [55]	Cu	PVC and PDMS	Area: 4 × 20 mm ² : Thickness: 100 μm	Tensile strain measured: 0.5%	-	Blinking motion	0.2 V	2 nA
PVDF film [56]	Au	PI	Area: 40 × 70 mm ² : Thickness: 350 μm	Tensile strain measured: 0.001613%	-	Wrist bending, elbow folding, cardiac impulse, human breathing, heel strike	25V	20 μA
PMN-PT particles [57]	Ag-NWs	Ecoflex	-	Tensile strain measured: 200%	-	Knee bending	0.7 V	50 nA
P(VDF-TrFe) sheet [58]	Au	PDMS	Area: 9 × 18 mm ² : Thickness: 100 μm	Precise sensor for pressure of 1 kPa and deformations below 1 μm	Bending radius: 25 mm	Radial and carotid blood vessel pulsing	0.6 V	-
AN film [59]	Mo and Cr/Al	PI and Parylene	Area: 4 × 6 mm ² : Thickness: 100 μm	-	Bending radius: 5.5 mm	Follows deformations of skin	0.7 V	0.3 μA

CONCLUSION

This paper summarizes recent progress in piezoelectric technologies that rely on mechanics and piezoelectric materials concepts for functions that lie beyond those available with conventional approaches. Here, we are discussed about piezoelectric principle, types, piezoelectric materials and properties. Then, the developments of piezoelectric materials were discussed in various method of preparation and that can be used as a piezoelectric nanogenerator in modern technique. This nanogenerator produces are voltage, current and etc., it can be used in daily human routine work.

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