The Review of Materials for Energy Harvesting

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***Abstract*—This paper presents a short review of the piezoelectric materials in energy harvesting. Energy harvesting principle, as the method for obtaining energy from environment has been described. Materials and material combinations for creating an energy harvesting composites are discussed, such as ceramic- and polymer-based composites and their mechanical properties. The list of the mostly used piezoelectric materials is presented and elaborated. Possible applications of the energy harvesting materials are discussed, including nanogenerators, biosensors and biomedical applications.**

Keywords—energy harvesting, piezoelectricity, composites, kinetic energy harvesting, bioelectronic medicine

I. INTRODUCTION

Energy harvesting method considers the conversion of environmental energy into electricity, which can be used for powering wireless or remote electronic devices and circuits [1]. While the concept of this conversion is the same as large-scale renewable energy generation, such as wind turbines [2], the quantity of energy generated is significantly smaller [3]–[5]. Therefore, the main difference between the energy harvesting and renewable energy sources is that here harvested energy is collected locally and is also for local use [6]. Energy harvested in this way is generally in the range of tens of microwatts to a few watts [7]. Furthermore, the principle of energy harvesting can be utilized to replace small energy sources, such as batteries. While batteries are inexpensive, they have a limited amount of energy and must be replaced or recharged on a regular basis. Unlike batteries, energy harvesting process is renewable and does not require environmentally harmful disposal.

Piezoelectric materials utilize the inherent material property of piezoelectricity for energy harvesting [8]–[10]. Piezoelectricity represents the appearance of the electric charge accumulated within a material due to the mechanical loading [11]. Piezoelectric materials belong to a group of smart materials [10], [12], meaning that they react to the external stimulus, which is a mechanical loading in their case. Thus, apart from the energy harvesting, piezoelectric materials are also often used for a precise monitoring of the mechanical loading, material motion or process control [13]–[15].

Beside certain ceramics [16], [17], polymers [18] and composites [8], potentially promising piezoelectric materials are natural cellulose and other cellulose-based derivatives as reviewed in [19]. Furthermore, bone exhibits piezoelectricity [20], [21], as well as different proteins and DNA [22], [23] that accumulate electric charge under the stress and release it in the environment at certain conditions. Therefore, biomedical applications of piezoelectric materials are very important, from both aspects of energy harvesting and health monitoring and diagnostics [14], [24]. Also, some novel treatments have been studied based on the actions of piezoelectric materials within a body, such as electrically induced osteogenesis through the use of piezoelectric biocompatible materials discussed in [20]. Furthermore, electrospun piezoelectric nanofibers used as biomaterial scaffolds were shown to deliver targeted micro currents thus regulating cellular fate and controlling the engineered tissue [25]. Energy harvesting by biocompatible piezoelectric devices, has promised to be crucial in the future, especially for the miniature implantable devices, attached to the nerves, which will be able to decipher, record and modulate neural signals [26]. Moreover, other reports have shown that the electric field can guide stem cell differentiation [27], and cell proliferation and tissues regeneration [28].

This paper is a short review of piezoelectric materials that are in use nowadays, mechanical properties of ceramic-based and polymer-based piezoelectric composites. Energy harvesting principle is described, and research directions in this area are elaborated from different application focuses, including biomedical applications.

II. ENERGY HARVESTING PRINCIPLE

The main focus of energy harvesting has been on the development of electronic components capable of generating electrical energy from kinetic energy [29], light sources [30] and thermal gradients [31], [32]. However, since energy harvesting performance is directly related to the quantity of available energy (e.g. kinetic energy, light, thermal gradients), unlike batteries, the individual application requirements must be considered in advance [31]. Final material application determines the design of the device in kinetic energy harvesting and rarely there is a general solution [31].

A. *Thermal energy harvesting*

The interaction and conversion of heat and electricity in solids is described by thermoelectric phenomena, which may be summarized by three thermoelectric effects: (i) Seebeck effect, (ii) the Peltier effect, (iii) and the Thomson effect [1]. Thermoelectric devices have been created and used for power production, cooling, and temperature monitoring based on these phenomena.

B. *Kinetic energy harvesting*

Kinetic energy may be obtained from a variety of sources. These have been classified as human-, industrial-, transportation-, and structural-based, and the sections that follow offer examples of data that describe the shape that kinetic energy can take in each scenario. It is clear that the features connected with each instance differ greatly. This illustrates a unique difficulty with kinetic energy harvesting in those generators for different purposes that are frequently extremely different. It is critical that generators are built from the start with a thorough understanding of the application and the properties of the kinetic energy being targeted.

To transform a kinetic energy to electrical energy, some sort of transduction mechanism is clearly necessary [1]. This mechanism must be integrated into the mechanical system intended to optimize the energy linked from the application environment to the transducer.

Depending on the kind of transducer, the transducer can create energy via mechanical strain or relative displacement inside the system. The application of active materials, such as piezoelectric materials, is an obvious example of how strain can be turned directly into electrical energy. However, piezoelectric material characteristics essentially restrict the efficiency of piezoelectric generators [1]. The energy harvesting potential can vary depending on the volume of the transducers and piezoelectric materials used. Therefore, Liang et al. [9] introduced Normalized Power Density (NPD) metric for comparing the energy generation performance of piezoelectric materials which takes into account the devices volume, excitation frequencies and stimulating accelerations.

Additional kinetic energy transducers include electromagnetic and electrostatic transductors, where electromagnetic transductors have the highest efficiency if size constraints are disregarded.

Piezoelectric materials have been used for many years to convert mechanical energy into electrical energy. Electrical polarization in linear correlation to the strain occurs in material under the stress, due to the action of the electric dipoles. An applied electric field, on the other hand, causes the dipoles to spin, causing the material to deform. As a result, piezoelectric materials are utilized in a wide range of commercial sensors and actuators, and they are also a viable choice for kinetic energy harvesting applications. Single crystal materials such as quartz, piezoceramics (e.g. lead zirconate titanate (PZT)), screen printable thick films based on piezoceramic powders and some polymer materials (e.g. polyvinylidane fluoride (PVDF)) are capable of exhibiting the piezoelectric effect [1]. These materials, and others listed in the Table I, have anisotropic piezoelectric behavior, meaning that material properties differ depending on the direction of the strain and also on the orientation of the polarization.

A number of constants describe the piezoelectric characteristics of a material. Since piezoelectric materials are polarized along their thickness [1], when the direction of the applied mechanical strain is the same as the direction of polarization of the material, the constraints are indicated by the subscript 33 and if the strain is applied perpendicular to the direction of the polarization, the constraints are indicated by the subscript 31, shown on Figure 1, where k – electromechanical coupling coefficient, V – voltage of generated electrical energy and d – piezoelectric constant.

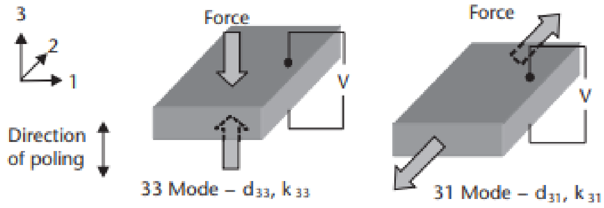

Fig. 1. Piezoelectric constraints [1]

The majority of piezoelectric harvesters operate in mode 31 indicated on Figure 1 [1], hence the surface of a piezoelectric element is frequently correlated to the surface of a mechanical spring element, which transforms vertical displacements into lateral strain across the piezoelectric element [1].

III. MATERIALS FOR ENERGY HARVESTING

As energy harvest composites require piezoelectric properties, they are most commonly consisted of piezoelectric fibers (ceramic or polymer) and matrices (mainly polymer) in order to achieve mechanical stability, as well as piezoelectric properties.

The two main types of piezoelectric composites are composites with ceramic and polymer matrix, mainly reinforced by fibers, and including both the synthetic and natural materials, with outstanding piezoelectric properties [33]. The list of the mostly used piezoelectric materials is given in Table I.

A. *Ceramics*

Ceramics with the highest piezoelectric properties are those with perovskite structure, such as lead magnesium niobate (PMN), lead titanate (PT), lead zirconate (PZT) and lead zirconate titanate (PLZT). A new parallel connected freeze-cast PZT composite, proposed by Zhang et al. [17], has advantages over conventional monolithic PZT composite mainly due to a larger piezoelectric sensitivity, as well as piezoelectric anisotropy, which is crucial for energy harvesting applications.

However, lead based ceramics have some issues related to lead oxide toxicity, so the researches took turn towards other, lead–free piezo materials for biomedical uses [33]. As focus was turned away from these composites the quest was to find composites with similar properties. Again, lead-free composites with properties comparable to lead-based ceramics still have the same crystalline perovskite structure, which include: alkaline niobates (K, Na, Li) NbO3, barium titanate ($BaTiO_3$), alkaline bismuth titanate $(K, Na)_{0.5}$ $Bi_{0.5}$ TiO_3, and barium zirconate titanate-barium calcium titanate (BZT-BCT) [34]–[36]. The main drawbacks of these ceramics are their poor mechanical properties, brittleness, and fragility at low tensile strains. These are the reasons why piezoelectric polymers found their applications among energy harvesting composites for biomedical purposes.

Established method to produce piezoelectric materials is electrospinning, but there are only several studies reporting electrospun ceramics without addition of polymers into solvent. It has been reported that high temperature postprocessing, like calcination, can also improve the final ceramic fibers and piezoelectric properties.

TABLE I. *A BRIEF OVERVIEW OF COMMONLY USED PIEZOELECTRIC MATERIALS AND MATERIAL COMBINATIONS FOR ENERGY HARVESTING APPLICATIONS*

Materials	Common applications	Ref
PLLA	Piezoelectric fabric, nanogenerators, nanofibers, sensors, biomedical sensors, wearables	[7]
Polyurethanes	Health monitoring	[8]
Cellulose and derivatives	Energy harvesting	[19]
PVDF and its copolymers	Human movement energy harvest, Tissue engineering scaffolds, Biosensors, Health monitoring	[18], [24], [33], [37]–[45]
PVDF	Nanogenerators, Capacitors, Sensors	[41]
CNT and PVDF	Nanofiber actuators	[46]
Ag and PVDF	Sensors	[47]
GK Graphite powder and PVDF	Use in biology, chemistry, and electronics	[48]
PVDF with DMF-acetone solvent	Powering microelectronic devices, LEDs, digital Displays, micro-photodetectors, self-cooling textiles	[49]
Coated zinc oxide and PVDF	Motion sensors, flexible microsensors, biosensors	[16], [50]
BNT-ST combined with PVDF-TrFE	Powering wearable sensors and devices for monitoring exercise and health monitoring	[51]
BaTiO3 and PVDF	Wearable electronics for human motion monitoring	[36]
Polycarbone acrylate di-epoxide (PCADE) and PVDF	Enzyme membrane reactors, filtration, sensors, catalytic systems, in energy harvesting / conversion /storage, in structural composites and in biomedical applications	[52]
PVDF and PZT	Energy scavenging applications, nanofibers nanogenerators	[9], [14], [17], [53]
PZT	Energy harvesting, Transducers, Sensors, Medical diagnostics, Neurological treatments, Tissue regeneration, Cancer treatments, Antifouling treatment, Bionic and Smart devices	[54]–[56]
PZT, PMN-PT, ZNO, BaTiO3	Cell and tissue characterization, Storing energy from natural organ motion	[20], [57]
TiO_2	Fuel and solar cells, supercapacitors, Li-ion batteries	[58]
NFC, NFCh, CNC, MFS	Nanogenerators, wearables	[59], [60]
Barium titanate (BTO) - based materials	Dental regeneration	[61]
PCMB and P3HT	Solar cells	[62]

B. *Polymers*

Polymers that exhibit piezoelectricity include some natural materials as chitin, collagen, silk, cellulose [19], [59], and elastin [63], as well as composites [8]. Nanofibers have been investigated for energy harvesting and process control [62]. Also, many of these polymers are biocompatible, hence usable in contact with human body. Polymers which have these characteristics are polyvinylidene fluoride (PVDF) [8], [36], [37], [64]–[66], polyvinyl chloride (PVC), liquid crystal polymers (LCPs), polyamides, polyhydroxyalkanoates, such as poly-b-hydroxybutyrate (PHB), and poly-3-hydroxybutyrate-3-hydroxy valerate (PHBV) [67]. Less studied piezoelectric polymers include PHB, PHBHV, parylene-C, nylon and P4HB.

Among these composites, PVDF and its copolymers are the most studied one and with the highest piezoelectric effect, due to their oriented molecular dipoles. PVDF, and PVDF-based composites have remarkable mechanical properties (high chemical resistance, good mechanical strength, transparency, flexibility etc.), but they also provide extraordinary advantages regarding healthcare applications because of its stability under gamma radiation (used in sterilizing medical devices) and is FDA approved [33], and above all, PVDF has shown superior piezoelectric properties [68]. Furthermore, ZnO flexible polymers [16] have shown a good potential for biocompatible nanogenerators, by achieving the open circuit voltages up to 470 V.

Many research papers reported the use of nearfield and far-field electrospinning manufacturing to produce single polymer piezoelectric PVDF fibers, [69], [70] or nanofibrous structure of PVDF meshes [65].

IV. APPLICATIONS OF ENERGY HARVESTING MATERIALS

Piezoelectric materials have been utilized in a variety of applications due to their unique properties, including ultrasonics, robotics, energy harvesters, energy conversion, aircraft, household industries, damage detection, automobile engines, sensors, actuators, transducers, sensors, medical diagnostics, neurological treatments, tissue regeneration, cancer treatments, antifouling treatment, bionic and smart devices. They are an important class of materials in the field of bioengineering. Further, we will briefly list various uses of piezoelectric composites.

A. *Nanogenerators – energy harvesters*

Harvesting energy from inherent human body movements has received a lot of attention in recent years: Furthermore, implantable self-powered devices capable of scavenging energy from various natural sources have been created to transform biomechanical energy into other useful types of energies [71].

Kato et al. [72] developed a new vibration energy harvester by dispersing lead-free piezoelectric (Na0.5K0.5) NbO3 (NKN) ceramic particles in PVDF nanofibers. They demonstrated that their energy harvester may be utilized to power transducers in ubiquitous networks. The harvester's piezoelectric coefficient d_{33} increased by employing the high quantity of NKN particles (50 percent) and after the corona poling, it was shown that the particles were suitably polarized.

Dye-sensitized solar cells (DSCs) have garnered a significant deal of interest since its invention as one of the low-cost photovoltaic technologies due to the lower manufacturing costs and less expensive raw ingredients [8]. DSCs are typically composed of a TiO_2 photoanode, Ru-based dyes, an electrolyte containing a redox couple, and a Pt photocathode. TiO_2 nanoparticles may absorb a large number of dye molecules and create photogenerated electrons in traditional DSCs. Additionally, it has been reported [62] that by using the [6,6]-phenyl-C61-butyricacid methyl ester (PCBM) nanofibers as internal material and combining it with various external materials (such as poly(3-pentylthiophene), or P3HT) can be also used as solar cells. Dye-sensitized solar cells based on PEDOT nanofiber auxiliary electrode showed the highest potential in terms of conversion efficiency [62].

Fuel cells represent devices where electrochemical oxidation of hydrogen or hydrogen rich fuel is converted in the presence of a metal catalyst into electric current. Among different kinds of fuel cells, the direct methanol fuel cells (DMFCs) have been widely studied because they possess optional high energy density and can be operated at room temperature. To achieve the uniform Pt dispersion, porous nanomaterials with larger surface areas are used as electrode materials. Many continuous and porous nanofibers have been produced by electrospinning and used as catalyst supports.

Supercapacitors, also known as electrochemical double layer capacitors, which store energy electrostatically through the reversible adsorption of ions in an electrolyte onto electrodes, have received a lot of attention as an energy storage device due to their high energy density compared to common electrolytic capacitors [58].

B. Biosensors

The biosensor device utilizes physico-chemical cues that can sense and translate physical, chemical or biological reactions to the electrical signals [73]. Novel biosensors based on biocompatible piezoelectric materials have recently attracted a lot of interest. Because polymer-based piezoelectric materials are flexible and may deform at lower applied stresses, they are a good option for detecting numerous mechanical-like signals in the human body, such as pressure sensing applications [57]

Human physiological signals such as pulse or blood pressure monitoring [14] and respiration may be detected using biosensors. Furthermore, piezoelectric materials can be used as strain sensors for smart skin or knee prosthesis [14]. Such signals will provide data that will allow essential function monitoring and lifestyle awareness. Because of their flexibility, piezoelectric polymers are excellent candidates for mechanical (pressure) sensors whether applied to a skin or embedded in wearable/portable electronics [74].

Park et al. [45] suggested a bendable and stretchable P(VDF-TrFE) sensor with a resolution of around 1 mm that can detect movement of the skin on the neck caused by a carotid pulsed behavior. P(VDF-TrFE) nanofiber meshes were immersed in a polydimethylsiloxane (PDMS) elastomeric matrix, which assured bendability and stability of the device.

C. Biomedical applications

PVDF has emerged as a promising material for stimuli-responsive scaffolds in tissue engineering due to its piezoelectric characteristics and biocompatibility. Tissue engineering seeks to repair damaged organs and tissues that cannot regenerate on their own by producing biomaterial-assisted biological replacements. Biocompatible scaffolds are essential components for tissue engineering because they can direct tissue development and regeneration in three dimensions. Surmenev et al. [24] reported that various implantable PVDF-based scaffolds were used for collecting energy from the in-vivo movements. Therefore, PVDF and its copolymers (for example, PVDF-HFP and PVDF-TrFE) in the form of 3D scaffolds can give electrical stimulation to cells, promoting tissue regeneration [20], [28], [33], osteogenesis [20], [24], [25], nerve regeneration [25], stem cell differentiation [27], and cell proliferation [24], [28].

V. Conclusion

The review of piezoelectric materials that can be exploited for energy harvesting applications was presented in this paper. In recent years, piezoelectric nanofibers, such as PVDF and PZT, have received extensive attention in the construction of piezoelectric energy harvest devices due to their voltage output, high sensitivity, mechanical flexibility, biocompatibility, stability, and low cost. Further improvements in PVDF nanofibers piezoelectric properties and voltage output have been achieved by adding BT nanoparticles due to their high voltage piezoelectric coefficient or by adding small amounts of graphene. This led to the effective use of the electrospun piezoelectric nanofibers in rechargeable portable electronic devices and self-powered sensors in many fields. For example, piezoelectric nanogenerators have been shown to effectively power implanted and wearable biomedical devices, or as the self-powered sensors for health monitoring, and even as a micro-current stimulator for neuro and bone tissue regeneration.

Moreover, the rapid technology progress and constant thrive towards device minimization, power consumption reduction and flexibility of devices, along with discovering new sources of vibration means that the interest in piezoelectric energy harvesting devices is expecting to continue to grow. With further exploration of new piezoelectric materials and their improvements through combinations of composite constituents, and especially considering certain nanoparticles as reinforcements, it can be expected that piezoelectric nanogenerators will be capable of powering the majority of wireless electronic devices in the near future.

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