Nanogenerators for Human Body Energy Harvesting

Human beings generate huge quantities of energy while performing daily activities, but this energy dissolves in the environment, yet. This article addresses the progress with the development of nanogenerators, i.e. devices able to harvest the biomechanical and the thermal energies by exploiting the piezoelectric, the triboelectric and the thermoelectric physical effects. In designing of nanogenrators, the comfort for the end-user is the main aspect to deal for designers. Therefore, the review focuses on recent materials giving flexibility and stretchability to nanogenerators. In addition, the paper summarizes common fabrics for the nanogenerators design. Finally, the mid-2020s market forecasts, about these promising technologies, highlight the great potentials for the commercialization of nanogenerators because they may help contribute to the route of innovation for developing self-powered systems.

Harvesting the Energy on Human Body – Requirements and Uses

The micro- and nano-power energy harvesting is a research field under rapid development [1, 2]. Since in 1996, T. Starner hypothesized a wearable computing powered by the energy of the body, many researchers improved the techniques for developing energy harvesters (EHs) on the body surfaces [3]. The growing interest in developing self-powered and comfortable devices arises the goal of unlimited monitoring, where the users are untied from the need to change the batteries. In such a way, humans can wear electronic systems without realizing it, especially for elderly and infants [4, 5]. Moreover, the rapid advances in development of miniaturized electronics have led to a gradual reduction of the power consumption for the end-system. Figure 1 lists the power consumption values of commercial off-the-shelf (COTS) components, i.e. datasheet values, and implantable medical devices [X]. These values are in the range 100 nW – 10 mW, which is the typical range of the power harvested on the surfaces of human body [6].

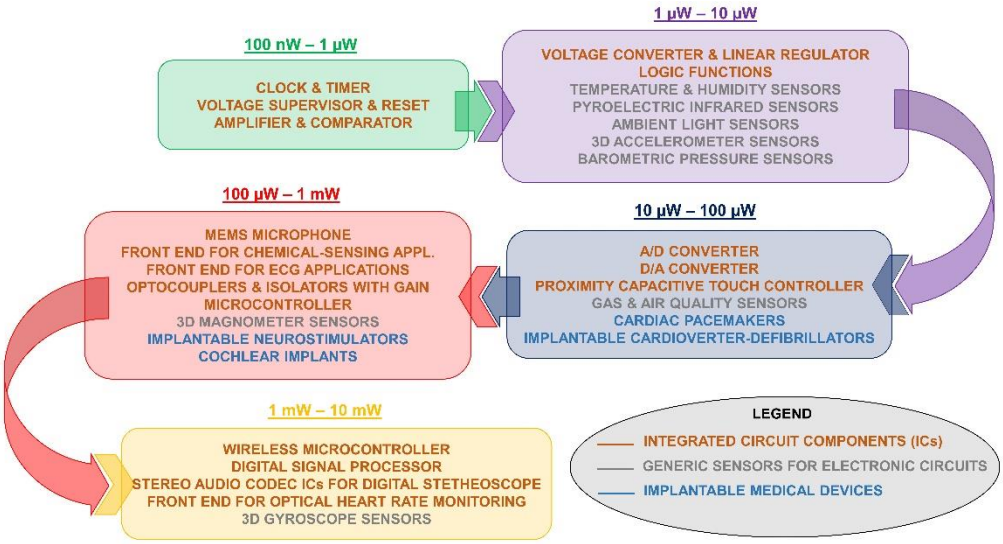

Figure 1. Which devices can be powered by EHs? The average power consumed by generic integrated circuit components and sensors, and by implantable devices, in the range of values: 100 nW – 10 mW. The color codifies the ranges: Green – 100 nW - 1 µW, Violet – 1 µW - 10 µW, Blue – 10 µW – 100 µW, Red – 100 µW - 1 mW, Yellow – 1 mW - 10 mW.

Therefore, the micro- and nano-energy harvesting techniques are receiving more and more interest for designing flexible and stretchable self-powered systems, which ensure well-being for the wearers and allow the development of new technologies for clinical applications [7-9].

From an energy perspective, the human body is a great warehouse of energy: it provides potential energy sources, such as the biomechanical energy through the movements of the body joints and thermal energy by the exchanged heat with the environment. In such a way, the power source on the surface of the body are those mechanical and thermal [10-12].

The newest devices for harvesting the biomechanical and thermal energies on the body surface are nanogenerators (NGs). In 2006, Wang and Song [13] proposed the first NG prototype, and the rapid growth in this decade was unbelievable [X1], because the nano-sizes of NGs enable the design of systems able to maintain their original electronic and structural properties also when are bent, twisted and stretched. These wonderful characteristics of NGs allow their integration into fabrics, thus having proper contact with the surfaces of the body.

In this review, we aim to summarize the latest advances in developing flexible and stretchable NGs for harvesting the energy directly on the body surface. We start the discussion by introducing the three main operating principles for designing NGs, then describing the NGs body applications. In addition, we show NGs based on common fabrics for properly describing the goal of harvesting the energy in each moment of the day. Finally, the review concludes with the market forecasts about new materials for developing foldable NGs, by the mid-2020s.

Working Principles of NGs

NGs for harvesting the energy on the human body are in solid matter. The structure of the solid matter can be crystalline or amorphous. In crystalline solid, the constituents of the lattice – ions, atoms and molecules – aggregate each other, to form an ordered and periodic pattern in space. Conversely, the network configuration of the lattice structure is random for the solid matter in amorphous state. The solid matter for developing NGs can be in both lattice structures: amorphous or crystalline, depending on the different nature of the elements – inorganic and/or organic – constituting them.

Up to now, the most known working principles for driving NGs are piezoelectricity and triboelectricity for transducing the biomechanical energy into electricity while the thermoelectric effect, i.e. Seebeck effect, for transducing the thermal energy into measurable electrical one.

The piezoelectric effects relies on the physical and chemical properties of the crystalline phase of the solid matter; if the lattice structure is non-centrosymmetric, the piezoelectric effect may occur under the influence of external perturbations, which produce charge separation across the structure, resulting in electrical polarization of the material.

Instead, the triboelectric NG generates an electric voltage by the frictional contact between solid matters. The contact electrification and the electrostatic induction are the physical effects for the generation of triboelectric charges. When two organic or inorganic materials, with different value of electronegativity, comes in contact each other, generate triboelectric charges on their surfaces, and a

voltage difference occurs from the separation of the electrical charges, due to the spatial displacement of materials.

Finally, thermoelectric NGs generate electrical power when a temperature gradient occurs between two dissimilar conducting materials, generally consisting of multiple couples of p- and n-type elements.

Wearable NGs

Wearable NGs placed on the human body must be comfortable and imperceptible for people; almost like a garment that can conveniently harvest the body energy.

Table 1 lists current piezo-, tribo- and thermos-NGs for harvesting the energy on the body surface. In addition, for each considered working principle, Table 1 highlights common fabrics used as substrate or active material for developing NGs.

Piezoelectric NGs

A single Zno-NWs laterally bonded with silver paste onto a flexible PI substrate, i.e. Kapton®, represents the first piezo-NG for harvesting the biomechanical energy on the body surface [14]. The stretching and releasing movement of the index finger resulted in 0.2% tensile strain onto the structure, generating a piezoelectric potential along the wire [15]. The semiconducting property of the ZnO material with metal electrodes at its ends create a Schottky barrier at least at one end of the NW, thus preserving the piezo-potential in the ZnO-NW and driving the flow of electrons in an external load circuit. Although the NG-structure was flexible enough to follow the movements of the finger, this prototype showed disadvantages, such as the output stability and the mechanical robustness under repeated stress.

In order to increase the robustness of piezo-NG, Lee *et al*. [16, 17] proposed an ultrathin Al-foil as both substrate and electrode for the structure. The prototype allowed the wrinkling detection while slamming the eyelashes. Another robust configuration for harvesting the biomechanical motion of arm stretching relies on the development of ultrathin ZnO film bonded on a PET plastic substrate. The physical properties of the PET allow the mechanical rolling of the whole structure, so that it might be worn at the level of the body joints [19]. The only disadvantage of this NG was the brittleness of the ITO electrode, which limits the rolling curvature of the device to the value of 2 cm. In order to enhance the stretchability of this kind of piezo-NGs, Pradel *et al*. [20] used the silicon rubber, i.e. Ecoflex®, for packaging ZnO ultrathin films. This ZnO-NG generates electrical output by transducing the biomechanical movements of the wrist tendons, detecting even the different movements of each finger. For good sensitivity and high resolution in sensing applications, the best piezoelectric elements are polymers. Particularly, P(VDF-TrFe) nanofibers may be aligned together to form piezo-sheets able to harvest the biomechanical motion and even detect low pressure values with the resolution of about 0.1 Pa [21]. Again, Park *et al*. [22] proposed a flexible and stretchable P(VDF-TrFe) patch able to detect the movement of the skin due to the radial and carotid pulses, with displacement resolution of about 1 µm. Both P(VDF-TrFe)-NGs were packaged into an elastomeric matrix of PDMS, which ensure both stability and flexibility to the devices.

All of the NGs listed above generate measurable quantities of power output in the range of nano-watts. In order to increase these power values, many researchers designed NGs made by materials with better piezoelectric properties than those of ZnO and P(VDF-TrFe).

107 Table 1. Current NGs for harvesting the energy on human body surface*

NG type	NG active material	Electrodes	Substrate	NG dimension	Mechanical load / tensile strain	NG flexibility	Durability test	Human body application	Voltage	Current	Ref.
Piezo	Single ZnO-NW[a]	Ag	PI film	Area: - Thickness: 50 µm	Tensile strain measured: 0.05 – 0.2 %	Bending radius: 2 cm	Stretching and releasing at 22 cycles/min for 120 min	Finger bending	0.25 mV	150 pA	[14, 15]
Piezo	ZnO film[a]	Al	Al foil & PMMA	Area: 13 x 5 mm^2 Thickness: 18 µm	-	Bending radius: 3 mm	Pressing and releasing at 2 Hz for 20 h	Blinking motion	0.2 V	2 nA	[16, 17]
Piezo	ZnO & PVDF composite[a]	Au	PDMS	Area: 30 x 30 mm^2 Thickness: 30 µm	-	-	-	Finger bending	0.33 V	62 nA	[18]
Piezo	ZnO film[a]	ITO & Ag	PET	-	-	Rolling curv. radius: 2 cm	666 cycles of rolling movement (2000 s)	Rolling and muscle stretching	0.28 V	-	[19]
Piezo	ZnO film[a]	AZO	Ecoflex	Area: 50 x 50 mm^2 Thickness: 305 µm	Compressive strain measured: −0.22%	-	-	Movement of the tendons of the wrist	1 V	100 pA	[20]
Piezo	P(VDF-TrFe) sheet[a]	Ag	PI	Area: 9 x 18 mm^2 Thickness: 245 µm	Ultra-high sensitivity for measuring pressure: 0.1 Pa	-	1000 cycles of bending movement at 2 Hz frequency	Skin vibration	1.5 V	40 nA	[21]
Piezo	P(VDF-TrFe) sheet[a]	Au	PDMS	Area: 9 x 18 mm^2 Thickness: 100 µm	Precise sensor for pressures of 1 kPa and deformations below 1 µm	Bending radius: 2.5 mm	30% strain of uniaxial stretching over 1000 cycles	Radial and carotid blood vessels pulse	0.6 V	-	[22]
Piezo	PVDF sheet[a]	Cu	PVC & PDMS	Area: 4 x 20 mm^2 Thickness: 100 µm	Tensile strain measured: 0.5%	-	Five days of bending movement at 5 Hz frequency	Blinking motion	0.2 V	2 nA	[23]
Piezo	PVDF film[a]	Au	PI	Area: 40 x 70 mm^2 Thickness: 350 µm	Tensile strain measured: 0.001613%	-	-	Wrist bending, elbow folding, cardiac impulse, human breathing, heel strike	25 V	20 µA	[24]
Piezo	AlN film[a]	Mo & Cr/Al	PI & Parylene	Area: 4 × 6 mm^2 Thickness: 100 µm	-	Bending radius: 5.5 mm	-	Follows the skin deformation	0.7 V	0.3 µA	[25]

NG type	NG active material	Electrodes	Substrate	NG dimension	Mechanical load / tensile strain	NG flexibility	Durability test	Human body application	Voltage	Current	Ref.
Piezo	$BaTiO_3$ & P(VDF-HFP) composite[a]	Al	PDMS & PI	Area: 2 × 2 mm^2 Thickness: 350 µm	Applied pressure of 0.23 MPa	-	5400 cycles of bending movement	Finger tapping	75 V	15 µA	[26]
Piezo	PZT film[a]	Cr/Au & Ti/Au	PET	Area: 20 × 50 mm^2 Thickness: 225 µm	Tensile strain measured: 0.3%	Bending radius: 21 mm	115000 cycles of bending movement	Wrist movement	120 V	2 µA	[27-29]
Piezo	PMN-PT particles[a]	Ag-NWs	Ecoflex	-	Tensile strain measured: 200 %	-	15000 stretching cycles	Knee bending	0.7 V	50 nA	[30]
Piezo fabric	PVDF-$NaNbO_3$ nonwoven fabric[b]	PU yarns & Ag-coated PA yarns	PDMS	Area: 25 × 25 mm^2 Thickness: 2000 µm	Compressive pressure of 0.2 MPa	-	1000000 cycles of compression test at 1 Hz	Heel strike	3.4 V	4.4 µA	[31]
Piezo fabric	Single ZnO & PVDF fiber[b]	Au	PS & PDMS	Fiber length: 20 mm Thickness: 6.2 mm	Tensile strain measured: 0.1%	-	-	Elbow folding	0.1 V	-	[32]
Piezo fabric	PVDF filaments as spacer yarns[b]	Ag-coated PA66 yarns	PU	Area: 53 × 150 mm^2 Thickness: 3.5 mm	Compressive pressure of 0.10 MPa	-	150 cycles of compression	Fabric to wear	14 V	30 µA	[33]
Piezo fabric	$BaTiO_3$/PVC fibers[b]	Cu	PET	-	-	-	Maximum tensile strength of 54 MPa	Elbow folding, fabric to wear	1.9 V	24 nA	[34]
Tribo	PTFE & Al (rhombic grid structure)[c]	Al & Cu	PET	Area: 50 × 50 mm^2 Thickness: 0.6 mm	-	-	100000 cycles of external contact force	Natural walking with back load of 2 kg	400 V	230 µA	[35]
Tribo	PTFE & Al (zig-zag structure)[c]	Al	PI	Area: 38 × 38 mm^2 Thickness: 0.95 mm	Contact force of 500 N	-	100000 cycles of external contact force	Hand palm tapping	215 V	656 µA	[36]
Tribo	PTFE & Al (zig-zag structure)[c]	Al	PET	Area: 30 × 270 mm^2 Thickness: -	Deformation amplitudes during the contact force: 2.5 cm	-	-	Foot plantar pressure	840 V	55 µA	[37]

NG type	NG active material	Electrodes	Substrate	NG dimension	Mechanical load / tensile strain	NG flexibility	Durability test	Human body application	Voltage	Current	Ref.
Tribo	FEP & Al[c]	Al & Cu	PI	Area: 52 × 57 mm^2 Thickness: 24 mm	-	-	180000 cycles of external contact force	Heel strike	700 V	-	[38]
Tribo	Cu & CPP[c]	Al & Cu	PA	Circular contact area: 1800 mm^2 Thickness: 100 µm	-	-	10000 cycles of external contact force	Body movements during sleeping	55 V	0.8 µA	[39]
Tribo	Cu & PI[c]	Cu	PDMS	Area: 75 × 50 mm^2 Thickness: -	Tensile strain measured: 22%	Curved surface up to 36 cm^{-1}	3600 stretching cycles	Elbow and knee bending, bicep muscle tension, neck tilting, swallowing motion	600 V	45 µA	[40]
Tribo	Au & epoxy (zig-zag structure)[c]	Cu	PI	Area: 15 × 30 mm^2 Thickness: 1.64 mm	-	-	-	Finger tapping	160 V	8 µA	[41]
Tribo	Al & PDMS (double arch-shape structure)[c]	ITO	PET	Area: 20 × 40 mm^2 Thickness: -	-	-	-	Slam the biceps with forearm	120 V	88 µA	[42]
Tribo	PET & PDMS (with sponge divider)[c]	ITO & Cu	-	Area: 90 × 270 mm^2 Thickness: 3.5 mm	-	-	-	Foot plantar pressure	220 V	40 µA	[43]
Tribo	PES & PDMS (arch-shape structure)[c]	Ag-NWs	-	-	Tensile strain measured: 60%	-	10000 stretching cycles	Breathing, coughing, swallowing, drinking, eating	35 V	4 µA	[44]
Tribo	Human skin & PDMS[c]	ITO	PET	Area: 10 ×10 mm^2 Thickness: 1 mm	The detection limit of the pressure is about 0.4 kPa	-	-	Hand palm tapping	60 V	3.5 µA	[45]
Tribo	Human skin & PDMS[c]	Cu	-	Circle diameter: 15.5 mm Thickness: 1 mm	-	-	5000 cycles of external contact force	Monitoring the motions of elbow, knee, heel, finger joints	40 V	2 µA	[46]
Tribo	Human skin & PDMS[c]	ITO	PET	Area: 12.5 × 25 mm^2 Thickness: -	-	-	-	Finger tapping	200 V	12 µA	[47]

NG type	NG active material	Electrodes	Substrate	NG dimension	Mechanical load / tensile strain	NG flexibility	Durability test	Human body application	Voltage	Current	Ref.
Tribo	Human skin & PDMS[c]	Ti/Cu	Ecoflex	Area: 18 × 22 mm^2 Thickness: -	-	-	-	Finger tapping	21.9 V	0.55 µA	[48]
Tribo	Human skin & PDMS[c]	Au	PI	-	The range of movement of middle joint of finger: 60°-180°	-	-	Monitoring the finger angle displacements	70 V	-	[49, 50]
Tribo	Human skin & PDMS[c]	Ag-NWs	PET	Area: 50 × 50 mm^2 Thickness: -	-	-	30000 cycles of external contact force	Detecting the velocity of touch	5 V	-	[51]
Tribo	Human skin & PDMS[c]	Ag-NWs	-	Area: 20 × 30 mm^2 Thickness: -	-	-	-	Finger tapping, elbow and wrist bending	56 V	3.1 µA	[X2]
Tribo	Human skin & Ecoflex[c]	Ag-NWs	-	Area: 40 × 70 mm^2 Thickness: 1.5 mm	Tensile strain measured: 300%	Bending radius: 20 mm	3600 cycles of external contact force	Foot plantar pressure and arm swinging	250 V	-	[52, 53]
Tribo	Al & Rubber[c]	Al	PTFE	Area: 30 × 88 mm^2 Thickness: 250 µm	Stretching displacement of 100 mm	-	5000 cycles of stretching and releasing movement	Breathing, knee motion	65 V	-	[54]
Tribo fabric	Latex fabric & PDMS[d]	Graphene	PET	Area: 30 × 30 mm^2 Thickness: 2.4 µm	Contact pressure of 30 – 40 kPa	-	2 weeks of external contact force cycles	Fabric to wear	30.9 V	3.9 µA	[55]
Tribo fabric	Nylon fabric (PA 6) & PTFE[d]	ITO	PET	Major axis: 25 mm Minor axis: 20 mm Thickness: 100 µm	High sensitivity for measuring pressure in the range: 2.5 – 1200 Pa	-	40000 cycles of external contact force	Monitoring of the arterial pulse onto the carotid, wrist, and chest	5 V	0.3 µA	[56]
Tribo fabric	Latex fabric & FEP[d]	ITO	PP	Area: 65 × 100 mm^2 Thickness: 90 µm	Tensile strain measured: 5.4%. Contact force of 140 N	Bending radius: 0.84 mm	-	Hand palm tapping	340 V	78 µA	[57]
Tribo fabric	Denim fabric & PI[d]	Al	PET & sponge	Area: 50 × 50 mm^2 Thickness: 90 µm	-	-	2000 cycles of external contact force	Walking, running and sitting activities	10 V	0.75 µA	[58]
Tribo fabric	Al & PDMS[d]	Al & Cu	Fabric	Area: 20 × 70 mm^2 Thickness: 800 µm	-	-	-	Fabric to wear for walking activity	17 V	0.28 µA	[59]

NG type	NG active material	Electrodes	Substrate	NG dimension	Mechanical load / tensile strain	NG flexibility	Durability test	Human body application	Voltage	Current	Ref.
Tribo fabric	Al & PDMS[d]	Au	Au-coated fabric	Area: 70 × 70 mm^2 Thickness: -	Bending length of 3 cm	-	-	Arm bending	139 V	39 µA	[60]
Tribo fabric	Ag & ZnO/PDMS[d]	Ag	Ag-coated fabric	-	Compressive force of 10 kgf	-	12000 cycles of external contact force	Fabric to wear	120 V	65 µA	[61]
Tribo fabric	PET & FAS/PDMS[d]	Al foil & Ag-NWs	PA 6	Area: 30 × 30 mm^2 Thickness: -	-	-	12000 cycles of external contact force	Fabric to wear	575 V	12.1 µA	[62]
Tribo fabric	CNT & PTFE[d]	CNTs	Cotton threads	Fiber length: 90 mm and 8 helix turns	Tensile strain measured: 2.15%, with max. tensile stress of 210 MPa	-	90000 cycles of strain	Finger bending	9.1 V	0.1 µA	[63]
Tribo fabric	Ni & Parylene[d]	Ni	PES	Area: 100 × 100 mm^2 Thickness: -	Bending movement of 180°	-	1000 cycles of bending movement	Fabric to wear	50 V	4 µA	[64, 65]
Tribo fabric	Cu coils & Silicone rubber fiber[d]	Cu & CNT	-	Fiber diameter: 3.5 mm Fiber length: 100 mm	Tensile strain measured: 100%	-	10800 stretching cycles	Hand motion, fabric to wear	142 V	-	[66]
Tribo fabric	Al/PDMS & PU/PI[d]	Carbon thread	Fabric	Area: 15 × 60 mm^2 Thickness: -	-	-	4000 cycles of sliding motion	Fabric to wear under armpit for walking activity	15 V	0.15 µA	[67]
Tribo fabric	Nylon fabric (PA 6) & FEP[d]	Ag	Fabric	Area: 100 × 100 mm^2 Thickness: 300 µm	-	-	24000 cycles of bending movement	Fabric to wear	12.6 V	3.7 µA	[68]
Tribo fabric	Nylon fabric (PA 6) & PES[d]	Ag	Fabric	Area: 50 × 110 mm^2 Thickness: -	-	-	-	Bending of arm and leg joints, shaking of clothes	90 V	1 µA	[69]
Thermo	n: Bi_2Te_3 p: Sb_2Te_3[e]	Ag	PDMS	Thermocouples: 2778 Area: 50 × 50 mm^2 Thickness: 4 mm	Temperature gradient of 5 °C	-	-	-	250 mV	2 µA	[70-72]
Thermo	n: Bi_2Te_3 p: Sb_2Te_3 & PEDOT:PSS[e]	Ag film	PI	Thermocouples: 15 Area: 30 × 50 mm^2 Thickness: -	Temperature gradient of 5 °C	Bending radius: 30 mm	1000 cycles of bending movement	Worn on the wrist	12.1 mV	-	[73]

NG type	NG active material	Electrodes	Substrate	NG dimension	Mechanical load / tensile strain	NG flexibility	Durability test	Human body application	Voltage	Current	Ref.
Thermo	p-n: Bi_2Te_3[e]	Cu	PDMS	Thermocouples: 50 Area: - Thickness: -	Environmental temp.: 18.3 °C	-	-	Worn on the wrist, on the upper arm and on T-shirt, walking activity	7 mV	30 µA	[74]
Thermo fabric	n: Bi_2Te_3 p: Sb_2Te_3[f]	Ag foil	Silk fabric & PVA	Thermocouples: 12 Area: 40 × 80 mm^2 Thickness: 2.5 mm	Environmental temp.: 20 °C	-	100 cycles of bending and twisting movement	Arm-attached, walking activity	6.02 mV	-	[75]
Thermo fabric	p: $Bi_{0.5}Sb_{1.5}Te_3$ n: $Bi_2Se_{0.3}Te_{2.7}$[f]	Ag	PI	Thermocouples: 12 Area: 6 × 25 mm^2 Thickness: 4.7 mm	Environmental temp.: 5 °C	-	-	Placed on T-shirt, resting activity	11.5 mV	12.7 µA	[76]
Thermo fabric	n:$(0.98Bi,0.02Sb)_2(0.9Te,0.1Se)_3$ p:$(0.25Bi,0.75Sb)_2(0.95Te,0.05Se)_3$	Ag wire	PES fiber cloth	Thermocouples: 12 Area: 50 × 65 mm^2 Thickness: -	Temperature gradient of 2.9 °C	-	-	Arm-attached, running activity	2.6 mV	-	[77]

*ZnO-NW, zinc oxide nanowire; Ag, silver; PI, polyimide (e.g. Kapton®); Al, aluminum; PMMA, poly(methyl methacrylate); PVDF, polyvinylidene fluoride; Au, gold; PDMS, polydimethylsiloxane; ITO, indium tin oxide; PET, polyethylene terephthalate; AZO, aluminum-doped zinc oxide; P(VDF-TrFe), poly(vinylidene fluoride-co-trifluoroethylene); Cu, copper; PVC, polyvinyl chloride; AlN, aluminum nitride; Mo, molybdenum; Cr, chromium; $BaTiO_3$, barium titanate; P(VDF-HFP), poly(vinylidene fluoride-co-hexafluoropropylene); PZT, lead zirconate titanate; Ti, titanium; PMN-PT, lead magnesium niobate-lead titanate; $NaNbO_3$, niobium sodium oxide; PU, polyurethane; PA, polyamide; PS, polystyrene; PA6 & PA66, nylon; PTFE, polytetrafluoroethylene; FEP, fluorinated ethylene propylene; CPP, cast polypropylene; FAS, fluoroalkylsilanes; CNT, carbon nanotube; PES, polyester; Ni, nickel; PVA, poly(vinyl acetate); PEDOT:PSS, poly(3,4-ethylenedioxythiophene)-poly(styrenesulfonate); Bi_2Te_3, bismuth telluride; Sb_2Te_3, antimony telluride; BiSbTe, bismuth antimony telluride; BiSeTe, bismuth selenium telluride.

Such proposed piezo-materials are inorganic PZT and $BaTiO_3$ materials. Thus, a flexible composite thin film made by P(VDF-HFP) and hemispherical $BaTiO_3$ nanoparticles can generate voltage output up to 75 V and current output of about 15 μA under finger pressing. The hemispherical surface of each nanoparticle enhanced the piezo-power generation thanks to the improvements of the surface contact between the active area and the electrode [26]. The entire device, designed onto a PI film, even guaranteed robustness under 2h-long repeated bending cycles. Instead, the flexible piezoelectric PZT thin film developed onto a PET substrate, with interdigitated metal electrodes, harvested the energy from the biomechanical movements of the wrist, since its bending radius values was about 1.6 cm [27-29]. The motion of the wrist produced up to 120 V of voltage output and 2 μA of current output. Finally, Jeong *et al*. [30] proposed the most flexible element for the field of biomechanical energy harvesting. A piezo-elastic-composite made by particles of PMN-PT and multiwalled carbon nanotubes (MWCNTs) packaged in Ecoflex® silicon rubber allowed very large stretchability up to 200% of the initial value, generating small quantities of electrical power under the folding movements of the human joints. In order to maintain the robustness of the device, very long Ag-NWs are stretchable electrodes, which ensure high-conductivity and electrical output stability.

Triboelectric NGs

At the early stage of the development of tribo-NGs, the researchers focused on the design of tribo-NGs composed by a metal plate, positively charged, and a polymeric plate, negatively charged.

Concerning the biomechanical energy harvesting, Yang *et al.* [35] proposed a backpack tribo-NG able to harvest the energy generated by walking with 2.0 kg load. Multiple PTFE and Al plates, placed on a flexible PET substrate, operate as tribo-layers and form an integrated rhombic structure. During the contact and releasing process of the tribo-layers, the NG generates huge quantities of power output about milliwatt. Again, tribo-NGs composed by zigzag structures of multiple PTFE and Al plates, embedded in shoe insole, generate energy while walking [36, 37]. Moreover, a patch-NG made by a thin Cu cantilever spring sandwiched into a folded CPP film detects the body movements during sleep, without needing to external power source [39]. Concerning these kind of NGs, the power output strongly depends on the good adhesion between the two active layers. Modifying the tribo-layers by adding surface porosity or by implementing micro-cubic or micro-pyramid array structures [41, 42], the power output greatly increases, even reaching values of hundreds of volts and hundreds of microamperes.

The polymeric physical state of tribo-layers allows greater flexibility than the metal-based ones, although at the expense of a lower power output. The stretchability of NGs even depends from the nature of the electrodes. ITO electrodes are the most widely used as transparent conducting film in tribo-NGs, which mainly operate under compressive stress, but their brittle nature does not allow withstanding high tensile strain. Therefore, conducting polymers and metal or carbon nanostructures are the alternatives to the ITO electrodes for developing stretchable devices [78]. Particularly, Yang *et al*. [40] developed an imperceptible and stretchable tribo-NG for harvesting the biomechanical energy of human joint movements. The tribo-NG consisting of a wavy Kapton® film sandwiched between two Cu serpentine electrodes, onto a PDMS substrate, supports a tensile strain up to 22%, making the NG-structure suitable to be attached on the skin. Again, to increase the stretchability of tribo-NGs, Hwang *et al*. [44] proposed a device composed by two polymeric tribo-layers and very long Ag-NW electrodes. In

such a way, the NG withstands to tensile strains of about 40%, while breathing, coughing and swallowing, among many others human physiological activities.

As a basic structure of the all tribo-NGs listed above, two electrodes are mandatory for the proper device operating, but in some case may limit the application fields and increase the development costs. The fundamental single-electrode mode can overcome these limitations, and greatly expands the applicability of tribo-NGs in the research field of biomechanical energy harvesting and human-machine interfacing. According to the tribo-series [79], human skin is a triboelectric layer, which presents electrostatic charges on their surface once it is in contact with dielectric or ferroelectric materials [80]. The PDMS polymer film is widely used as friction layer for developing tribo-NGs, which involve the skin layer in the generation of tribo-charges. In the scientific literature, these NGs works as self-powered tactile sensors for tracking location, velocity and force of the human touch [45, 51] [X2]. In addition, tribo-NGs based on the friction contact between epidermis and PDMS polymer can monitor the human motion, by measuring the angle displacements of the main body joints [46, 49, 50]. However, the PDMS polymer may be not suitable to follow all movements of the limbs, due to its elongation limit. Thus, Lai *et al*. [53] developed a tribo-layer made of Ecoflex® silicon rubber, in which Ag-NWs were dispersed within, to form a stretchable electrode. This tribo-NG sustains tensile strains of over 300%, making the device highly conformable for electronic-skin applications.

Thermoelectric NGs

Bi_2Te_3 and Sb_2Te_3 are the conventional n- and p- type elements for developing the multiple thermocouples constituting the thermo-NGs, thanks to their high thermoelectric efficiency at room temperature. However, the brittle nature of these inorganic compounds does not allow flexibility for the NG, so it makes necessary the use of polymeric materials for supporting and packaging the thermoelectric elements to impart the desirable flexibility to the NG-structure.

A flexible thermo-NG adheres to the body surface better than a rigid one, optimizing the thermal conductance between the skin and the generator, so raising up the temperature difference between the hot and cold sides of the NG. For example, Hyland *et al*. [74] wrapped a rigid thermo-NG into a PDMS polymer, increasing the device area but conferring a minimum of flexibility for the entire structure. In this way, the NG was easily placed on the chest and the researchers tested the performance of the device under different walking speeds. In order to enhance the device flexibility, Jo *et al*. [81] designed a PDMS polymer substrate with several holes, which were filled by columns of thermoelectric elements. The entire structure comprises eight p-n junctions in 250 mm^2, with a thickness of more than 4 mm that does not allow large bending movements. To enhance the thermo-NG flexibility, We *et al*. [73] proposed an hybrid composite thermo-NG, by infiltrating an organic conducting polymer, the PEDOT:PSS, into the pores of an inorganic thermoelectric film. The minimum value of the NG bending radius was about 3 cm, so that it could be worn on the wrist. In addition, this NG supports over 1000 cycles of bending movements without showing any degradation in output performance. Another example of flexible thermo-NG is the wavy-shape module presented by Francioso *et al*. [70, 72]. The researchers assembled the p- and n-thermolegs on a wavy substrate made of two PDMS layers covered with a thin layer of Kapton®. The former PDMS layer was Ag-filled to enhance the contact between the skin and the hot thermo-junctions, while the latter one insulates the cold junctions from the hot ones. This wavy-shape NG counts up to 2778 p-n thermocouples and generates 250 mV when the temperature gradient is

about 5 K. This value of voltage output is very high for the field of energy harvesting by the body heat; however, the authors performed just simulation tests.

Fabric NGs

NGs based on fabrics can be a great innovation for harvesting energy directly on the body surface because humans wear clothes in each moment of the day. Particularly, the fabrics made by polymeric materials are very common in textile market thanks to their low cost large-scale manufacturing. Therefore, adding active polymer into common fabrics could represent a new trend in developing smart-clothes for harvesting the energy. Among all the working principles for designing NGs, the triboelectric one is the most used because relies on the contact/release process of any materials with different electronegativity values; as opposed instead to the piezoelectric and thermoelectric ones, which depend on the physical and chemical properties of the chose active elements for harvesting the energy. For example, NGs based on piezoelectric fabrics should be flexible, almost stretchable and just the PVDF polymer ensures these features. Zeng *et al*. [31] proposed a wearable NG made by PVDF-$NaNbO_3$ nonwoven fabric as active material, and conductive yarns as electrodes. This NG works under compressive stress and shows great stability also over 1000000 compression–release cycles, which is a value more high than a NG made by the only $NaNbO_3$-NWs elements [82]. PVDF monofilaments can be also “3D spacers” between two knitted layers made of PA66 yarns, thus having an all-fiber NG [33]. The technologies for knitting "3D spacer" fabrics are highly developed in the textile field, therefore this example of piezo-NG can open the way for easily design of clothes for harvesting the energy.

As regards the design of textile-based NGs, which exploit the triboelectric effect, nylon fabric represents a good choice as contact layer for the generation of energy when it rubs with polymeric materials, such as PDMS [55], PTFE [56], FEP [57, 68] and PES [69], among many others. Again, denim fabric in contact/release process with PI represents a tribo-NG well suited for harvesting the biomechanical energy [58]. However, these fabric NGs do not rely on knitting techniques, but rather they seem as single or multiple patches assembled on fabrics. Thus, Liu *et al*. [83] proposed a textile tribo-NG without a multilayer structure, where PET fibers interlace at the top surface of the fabric and penetrate across its thickness on the bottom side, so the final structure looks like a three-dimensionally penetrated fabric. Again, Zhao *et al*. [84] fabricated a textile tribo-NG by weaving of Cu-coated PET (Cu-PET) warp yarns and PI-coated Cu-PET weft yarns. This NG show washability capability, resulting suitable for the common daily using. However, to have greater stretchability, a conductive polymer should replace the Cu metallic material.

Developing thermo-NGs based on fabric structures is very difficult since the brittle nature of the inorganic thermoelectric elements do not allow easy integration into textiles. However, Lu *et al*. [75] deposited thermoelectric p-n columns on both sides of a silk fabric. In this way, the NG supports multiple bending and twisting movements, thus opening the possibility to integrate the thermo-NGs into clothes. Again, by using a sewing process, Kim *et al*. [76] utilized conductive fabric threads for electrically connecting in series the thermo-columns into a PI substrate. Then, the entire structure was sewn into a t-shirt for harvesting the thermal energy during walking. In addition, Siddique *et al*. [77] proposed a similar structure, composed by PES fabric as substrate, Ag-threads as conductive element and p- and n-types Bi_2Te_3 as thermoelectric elements. The textile and flexible thermo-NG can easily wrap the forearm for harvesting the thermal energy also in harsh environment. Finally, by coating commercial fabrics with

PEDOT:PSS polymer is possible developing flexible and foldable thermos-NGs. Although such polymer properly works in the temperature range of about 300 - 390 K, which are the proper temperatures for the human life, the voltage output is too low for considering this NG able to harvest the human thermal energy [85].

Concluding Remarks and Future Perspectives

The current promising progress in the field of nanotechnology allows the development of new devices – nanogenerators (NGs) – for harvesting mechanical and thermal energy directly on the body surface. NGs will bring many exciting opportunities for continuous powering wearable systems, since the scientific research on wearables open up new handy prospects for the non-invasive monitoring of people's health in the medicine and the fitness fields [86, 87]. Therefore, the overall impact of NGs may expand with the progress on new foldable and stretchable materials, which make devices comfortable for the end-users. *IDTechEx*, a company for the business analysis, provides the market forecast for the mid-2020s about recent materials for developing NGs. Figure 2 groups these technologies. Thus, companies around the world will invest many billions of dollars, with particular regards for the field of functional materials.

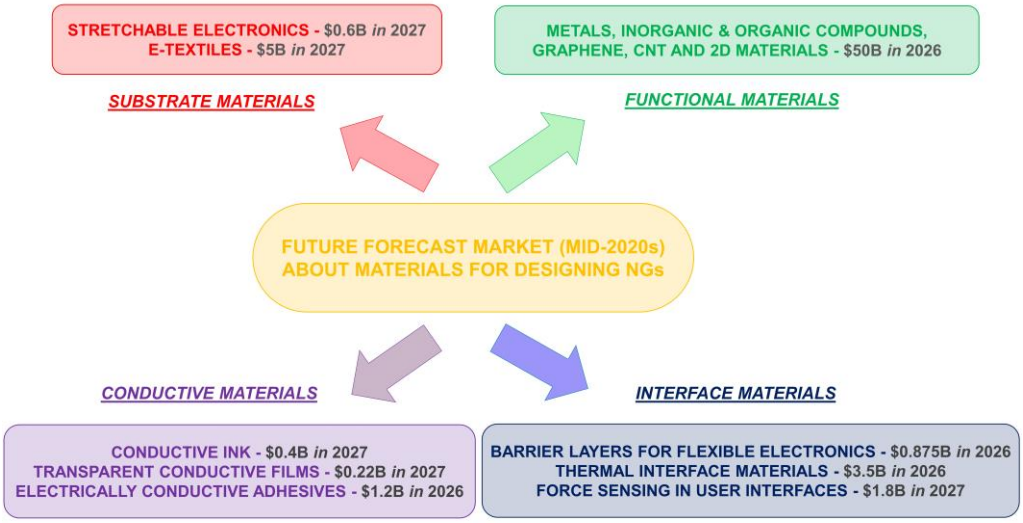

Figure 2. Market forecast for the mid-2020s The graph shows the money that will be spent in the mid-2020s for developing useful materials to design wearable NGs. [Data from IDTechEX Company, http://www.idtechex.com].

In addition, the improvements in the design of extensible electrodes and malleable electronic components could transform rigid devices into pliable ones, so achieving smart systems for a broad range of applications, such as biomedical devices, components for soft robotics and human-machine interfacing, among many others [88, 89].

In summary, with the maturation of the technologies proposed in this review, in the near future we could have electronic devices powered by NGs, which harvest the energy directly on the body surfaces.

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