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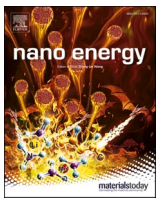

Review

Sustainable electronic biomaterials for body-compliant devices: Challenges and perspectives for wearable bio-mechanical sensors and body energy harvesters

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ABSTRACT

Flexible and body-compliant devices -based on eco-friendly materials represent one of the most challenging needs to monitor human health continuously and seamlessly, while reducing the environmental burden of electronic waste. In this respect, biomaterials are the preferred choice to develop sustainable wearable and implantable systems thanks to their advantages over most synthetic materials: Biocompatibility, biodegradability, tailored and reversible adherence to tissues, and low environmental footprint. In this review, the focus is on flexible bio-mechanical sensors and body energy harvesters based on sustainable active materials and, in particular, on their performance analysis related to different healthcare applications. Thus, the applicability potential of biomaterials extracted from bio-sustainable sources is critically discussed in the framework of developing the next generation of fully sustainable and portable health monitoring systems.

1. Introduction

The development of wearable and mobile technologies, enhancing the functionalities of bio-signals detection in personal healthcare systems, is receiving increasing interest. Indeed, personal health-monitoring technologies are increasingly in demand, especially as the population ages, allowing more effective medical assistance to the elderly and infirm people. Moreover, they enable real-time monitoring, allowing early diagnostics, protecting patients with chronic illnesses from severe harm or regulating professional athletes' activities, thus contributing to the enhancement of the public health system.

Modern personal health-monitoring devices based on portable, flexible, and wearable designs have been developed to detect physiological and vital signs, including heart rate, blood pressure, respiration rate, and local body temperature. They are usually shaped as medical devices, smart eyeglasses and eyewear, wristbands, smart clothing, and watches [1–3]. Biological signals produced by the human body, representing the electrical, chemical, and mechanical activities during

Abbreviations: BC, Bacterial Cellulose; $BaTiO_3$, Barium Titanate; BDO, Butanediol; CMC, Carboxymethyl Cellulose; CNT, Carbon Nanotubes; CNC, Cellulose Nano Crystals; CNF, Cellulose Nano Fiber; CMFs, cellulose microfibers; FF, Diphenylalanine; ECG, Electrocardiogram; FEP, Fluorinated Ethylene Propylene; HZTO, $H_2(Zr0.1Ti_{0.9})_3O_7$; IA, Itaconate; PZT, Lead zirconate titanate; LED, Light Emitting Diode; MWCNT, Multi Walled Carbon Nanotubes; MTP, MXene-coated tissue paper; NGs, Nanogenerators; PEG, Piezoelectric generators; PDLA, Poly(D-Lactic Acid); PLLA, Poly (L-Lactic Acid); PVDF, Poly (Vinylidene Difluoride); PCL, Poly Capro-Lactone; PDMS, Poly Dimethyl Siloxane; PEN, Poly Ethylene Naftalate; PVA, Poly Vinyl Acetate; PEDOTS:PSS, Poly(3,4-ethylenedioxythiophene) polystyrene sulfonate; PHBV, Poly(3hydroxybutyrateco-3-hydroxyvalerate); PLGA, Poly(lactic-co-glycolic acid); PBLG, Poly(γ-benzyl-α,L-glutamate); PAM, Polyacrylamide; PAA, polyacrylic acid; PANI, Polyaniline; PLA, Poly-lactic acid; PVC, Polyvinyl chloride; KNN, Potassium sodium niobate; SA, Sebacate; SCL, Silkworm cocoon layer; SF, Silk fibroin; AgNWs, Silver nanowires; SAlg, Sodium Alginate; TENG, Triboelectric nanogenerators; WSN, Wireless sensor network; ZnO, Zinc Oxide.

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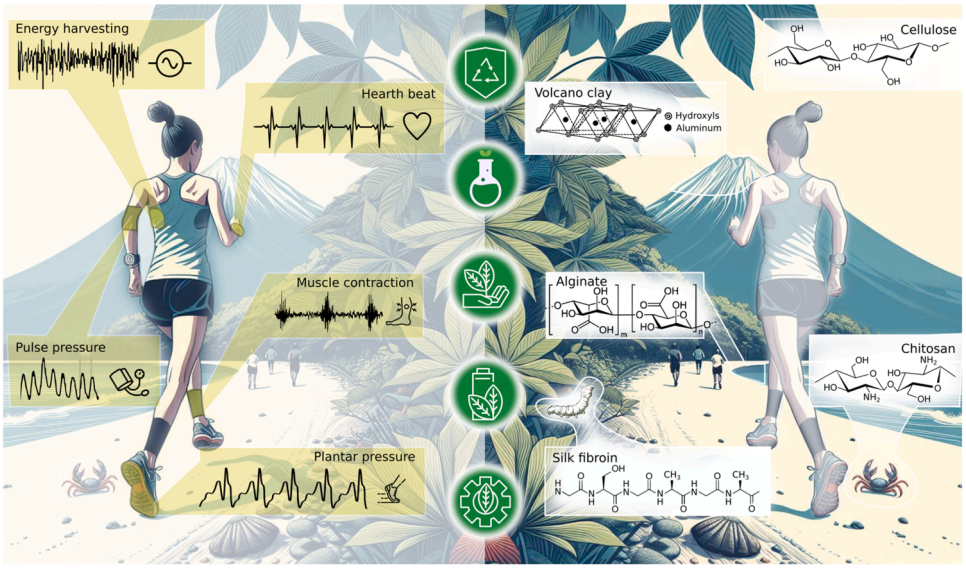


Fig. 1. Sustainable, body-compliant devices made of biocompatible and eco-friendly materials can help monitor physiological parameters or harvest body energy. On the left side, the focus is on the wearable and flexible devices that can conform to the body and monitor physical strains such as hearthbeat, muscle contraction, pulse pressure and plantar pressure, or harvest energy from body movements. On the right side, the nature-derived biomaterials extractable from numerous renewable sources: Cellulose from leaves, clay from volcanos, alginate from algae, silk fibroin from silkworms and chitosan from crustaceans exoskeleton, to cite few, can be adopted to develop a new class of environmentally-sustainable electronic devices.

various local and global biological events contain crucial information about the individual's physiological status and potential clinical cues of upcoming or undiagnosed diseases.

Electronic devices that can adapt to curved surfaces of biological tissues and handle their significant deformations are necessary for physiological parameters assessment. Considerable efforts have been made to design skin-attachable health monitoring sensors featuring high flexibility, lightweight, and ultra-low thickness, as well as enhanced sensing performance and functionality with multiple bio-signals detection (in a single sensing unit rather than in highly complex combined sensors). This has been achieved by developing nano-/micro-scale architectures of organic, inorganic, and hybrid materials [4–9].

Moreover, a paradigm shift toward the choice of nature-derived materials has been imposed by the necessity to promote sustainable materials and material processing. Thus, the advancement of biodegradable alternatives to conventional devices has been the focus of several research studies, mainly driven by the rising demand for degradable electronics that make use of safe, affordable, mass-produced disposable materials to create cutting-edge biodegradable, bioresorbable, or transient devices. This means that the choice of sustainable materials from green sources allows for accomplishing numerous tasks toward the circular economy practice, from the adoption of renewable resources to the shortening of the supply chain and the reduction of the devices' ecological footprint at their life end, see Fig. 1. Moreover, nature-derived materials are typically biocompatible, further supporting their application as compliant and skin-safe devices that can seamlessly interact with the patient or be implanted and degrade without the need for any removal surgery. Although great strides have been made in developing non-invasive mobile and wearable devices with synthetic materials capable of conforming to the human body's soft and curved surfaces, they are somewhat stiff and not fully compatible with the skin. Nature-derived materials are usually intrinsically compliant, however, they typically require non-negligible efforts to let them reach the performances of state-of-the-art synthetic ones [10–13].

The frequency of chronic diseases, the rise in the elderly population, the simplicity of accessing personal health records, the real-time monitoring of health conditions, and the cost-effectiveness of portable medical devices are all factors driving the demand for their use in healthcare. The widespread use of implantable and wearable medical devices has also been made possible by recent developments in front-end electronics and wireless communications for bio-signals post-detection stage [14,15], allowing continuous monitoring of quantitative, high-quality measurements of clinical data [11,16–18]. While big data, artificial intelligence, and flexible electronic technology are being promoted by the development of portable and wearable devices [19,20], on the other side energy availability is a significant barrier to their deployment. For instance, the power consumption of a pacemaker is ≈1 μW, an electrocardiogram (ECG) sensor ≈2.8 μW, a wireless intraocular pressure monitoring device ≈5.3 nW, a wireless sensor network (WSN) ≈71 nW, a chest patch ≈0.96 mW, to cite few [21]. The energy supply of current electronic devices and equipment relies mainly on rechargeable batteries, whose inherent rigidity limits the general flexibility of the entire system. Moreover, batteries' short lifetime reduces and limits the functioning of the wearable systems and the degree to which mobile devices are used, as well as their potential environmental contamination issues are also in conflict with the goals of sustainable development [22]. As a result, several efforts have been made to investigate brand-new and renewable energy sources that are safe for the environment and, at the same time, make wearable systems a self-powered and sustainable technology. In this respect, Body Energy Harvesters (BEH) can extend the lifespan of the battery, offering a long-term power source or even replacing it. Thermoelectric generators (TEG), triboelectric nanogenerators (TENG), and piezoelectric nanogenerators (PENG) are typical examples of environmental energy harvesters that can be potentially applied to wearable devices. Mechanical, thermal, chemical and solar energies are potential sources and, nowadays, different technologies have been developed to exploit and convert them into electrical ones. Also, in this case, the environment can offer numerous highly sustainable paths to develop completely green systems. Furthermore, to ensure portability, it is fundamental to develop flexible BEHs that can seamlessly interact with the biological tissue, matching all the biocompatibility requirements.

The increasing research on sustainable wearable sensors and body energy harvesters has fostered the development of systems that combine both technologies. Indeed, this trend is proven by the necessity to offer seamless, highly portable and energetically autonomous monitoring platforms able to remotely assist patients over long periods, avoiding the presence of bulky instrumentation. In this respect, people suffering from

Table 1
Qualitative analysis of the biomaterials compared to synthetic polymers and ceramics.

Class of active material	List of materials	Fabrication	Scalability	Flexibility	Biocompatibility	Sustainability	Ref.
Biomaterials	Rose petal, leaves, PLLA, glycine, chitosan, cellulose, wood, gelatine, silk, chitine	Drying, coating, freeze-drying, electrospinning, casting, spin coating	Good	Excellent	Excellent	Excellent	[28–35]
Synthetic polymers	PAA, bisacrylamide, polyurethane, PVDF	Electrospinning, casting, melting stretching, thermo-assembly	Good	Excellent	Good	Poor	[36–40]
Synthetic ceramics	Guanidinium perchlorate, ZnO, PZT, PET, PDMS	Coating, embedding	Excellent	Poor	Poor	Poor	[41–46]
Composite biomaterial/ synthetic polymers	PBLG/PDMS, Silk/PVDF, PANI/ cellulose/chitosan, PVDF/chitin, PVA/ alginate/cellulose/CNT, carbon black	Electrospinning, casting, spin-coating, freeze-drying,	Good	Excellent	Excellent	Good	[47–51]
Composite biomaterial /synthetic ceramics	Cellulose/MoS_2/$BatiO_3$, PZT/ cellulose, Strontium/collagen, MXene/ cellulose, silk/MXene	Casting, freeze-drying, electrospinning	Excellent	Good	Good	Poor	[52–56]

cardiovascular diseases, one of the most spread pathologies in the world, would exploit the possibility for a non-invasive, remote, real-time and continuous monitoring of the heart rhythm, triggering a rapid prediction and resolution in case of heart failure [23] [24]. Furthermore, the monitoring of the respiration rate, combined with heart pace detection, gives important information on conditions like asthma, sleep apnea and chronic obstructive pulmonary disease. It also helps checking the health status of infants, providing a fast response in emergency cases [25]. By real-time monitoring, it is also possible to check swallowing disorders in a non-invasive way[26]. Moreover, comfort during the acquisition of physiological parameters gains particular importance during the monitoring of athletes' activity and post-operative rehabilitation. For these cases, the employment of portable systems represents the most efficient solution to check the subject under real conditions [27].

Within this rapidly growing and multifaceted scenario, this work aims to provide a critical and comprehensive overview of nature-derived materials for developing flexible and compliant wearable sensors as well as BEHs, driving the sustainability change toward the next-generation biomedical applications. The first generation of wearable devices, primarily designed to exploit specific physical phenomena for sensing, is now in the throes of scientific maturity. Indeed, many of them are ready to be marketed or are already commercially available. Nevertheless, we emphasize the ongoing development of a second generation of sustainable and biocompatible wearables. The materials most widely used in wearables can be classified as natural materials, biopolymers, synthetic polymers and inorganic materials. In this respect, selecting materials derived from biological resources with specific functional properties is critical to moving toward fully biocompatible and sustainable devices. Table 1 summarizes some of the main properties of the materials used in wearables. Furthermore, technologies and sensing mechanisms will be thoroughly described to achieve a detailed classification of wearables according to the range of applications, the measured parameters and the typical processed material form used to implement the sensing element. This analysis will help to underline the connection between sensors and BEHs for developing the next generation of self-powered sensing wearable devices based on sustainable material.

The review is organized as follows. Section 2 displays the specific features of nature-derived materials. Section 3 describes the main developed sensors based on the mostly-used sensing mechanisms for body-compliant systems that continuously monitor human physiological parameters exploiting nature-derived materials. Piezoelectric, triboelectric, piezoresistive, piezocapacitive and hybrid mechanisms are discussed in the same section. Afterward, Section 4 reports a survey of the BEH devices, mainly based on piezoelectric and triboelectric transduction mechanisms, that can be integrated into the human body. The section discusses the solutions adopted to convert body movements and vibrations into electric energy for the power supply of the sensing units. Section 5 reports on implantable systems made of BEH and sensors and their unique properties when developed with nature-derived materials. Furthermore, Section 6 reports a comprehensive comparative analysis of the reported technologies correlating sensing and energy harvesting mechanisms with the processed material form (e.g. textile, thin film, etc.), including the key performance parameters. Finally, Section 7 reports the conclusions and outlooks.

2. Nature-derived materials

The shift toward the use of nature-derived materials to develop devices for the healthcare field is becoming more and more evident through the last years, driven also by the global environmental crisis. Indeed, the necessity to reduce electronic waste is of paramount importance and biodegradable materials with low carbon footprint represent one of the solutions to this serious issue. Moreover, the exploitation of these innovative materials brings additional value when employed in the biomedical environment due to their characteristics of bioresorbability and biodegradability shown when implanted or used on the skin. These materials display their best potential when adopted in devices for disposable or bioresorbable applications. Indeed, most of them are characterized by a polymeric structure that permits numerous chemical modifications, allowing also to tune their biodegradation based on stimuli such as temperature, pressure, pH, or after a specific time interval [57]. In particular, for implantable systems, their biodegradability helps to avoid further surgery to remove the device, improving the lifestyle of patients and reducing the burden of costs for the healthcare system. On the other side, thanks to structural modifications or by using barrier protective coatings, nature-derived materials can be modified to last for years, enabling their employment for long-term applications. Additionally, they can be easily disposed and replaced by a new device without significant environmental issues. Such versatility in biodegradation is typical of nature-derived molecules, an aspect that is much less likely to be reached with conventional and non-degradable materials [58]. In this section, a description of the unique properties of nature-derived materials, divided into four categories, is presented: *i)* polysaccharides, *ii)* proteins or peptides, *iii)* synthetic polymers and *iv)* combined materials.

2.1. Polysaccharides

Polysaccharides (PSs) are a class of biological macromolecules, consisting of units of monosaccharides such as glucose, linked by glycosidic bonds to form linear or branched chains. They are classified as homopolysaccharides or heteropolysaccharides depending on the nature

of the monomers in the chain of the polymer. Among them, cellulose, a linear homopolysaccharide consisting of β-D-glucose units formed through 1–4 glycosidic bonds, is the most abundant organic polymer on the planet being an important structural component of the primary cell wall of plants and it is the main component of the lignocellulose biomass [59]. This natural polymer possesses interesting properties like biodegradability, biorenewability and low costs of production. Additionally, it offers chemical, physical and mechanical properties tunable from the macroscale to the nanoscale, which find well-assessed applications in paper, food, cosmetic and pharmaceutical industries. Thanks to nanotechnological advances, physical and chemical treatments and bioengineering methods, it is possible to obtain cellulose and cellulose derivatives can be obtained in different forms, including nanocellulose-based materials (e.g. paper, films, membranes, aerogels), nanocrystals and nanofibers, composite fibers and composite films. Recently, plant and bacterial cellulose has been exploited for the development of a new class of devices such as sensors and energy harvesters due to their inherent dielectric properties and the electroactive behavior of nanocrystals, nanofibres and biosynthesized bacterial cellulose. Moreover, cellulose materials exhibit inherent biodegradability (with enzymatic hydrolysis and compostability) depending on crystallinity under both anaerobic and aerobic environmental conditions. Alginate is a linear heteropolysaccharide composed of β-D-mannuronic acid and α-L-guluronic acid linked by 1–4 glycosidic bonds [60]. This natural copolymer is widely distributed in the cell wall of brown algae, including Laminaria and Ascophyllum species and represents the most abundant marine biomass. Chemical and biochemical treatments enabled the synthesis of modified alginic acid derivatives with tailored properties such as solubility, hydrophobicity, affinity for specific proteins and pH sensitivity. The main applications of alginate are in pharmaceuticals, food, biomedical and engineering fields including wound healing, drug delivery and scaffolding due to its biocompatibility, biodegradability, nontoxicity, non-immunogenicity and antimicrobial nature. It possesses gelling and film-forming properties, self-healing ability and key rheological properties, including stretchability, self-healing ability and 3D printability. Additionally, it is a triboelectric material with high transparency, thus allowing it to be a promising candidate for flexible, transparent, and transient wearable electronic devices. While alginate cannot be digested by mammals, due to the lack of specific enzymes for the degradation process, several methods such as physical, chemical and bio-enzymatic can be used to control its degradation in the environment [61]. Pectin is one of the polysaccharides that constitutes the cell walls of higher plants. It has a complex linear heterostructure containing D-galacturonic acid residues substituted by methyl and acetyl groups linked by (1−4) linkages. Pectins are white, amorphous, complex carbohydrates that occur in ripe fruits like peaches, apples, plums, and citrus and certain vegetables, and they are also available as a by-product of fruit juice, sunflower oil and sugar manufacturing. Thanks to their broad availability, pectins are good candidates as raw sources for environmentally friendly materials. Pectin is well-known for its use in the gelling of solutions (jams and jellies) and for its application in film formation, however, it is also characterized by a wider range of uses because of its biodegradability and biocompatibility. Its main applications are in the biomedical field for its pro-biotic behavior in humans or for the controlled delivery of exogenous nutraceuticals, as well as drug delivery, wound healing, cancer therapy and scaffolding. Pectin has been exploited in piezoelectric sensors and energy harvesters being a key component in bio-wasted fruit membranes [62] and composites. The biodegradation of pectin is performed by a set of enzymes generically called pectinase, a group of enzymes that catalyzes the degradation of pectin by hydrolysis, trans-elimination, as well as de-esterification reactions [63] Starch is a natural-derived polysaccharide that can be abundantly harvested from the roots, stems, and seeds of rice, corn, wheat, cassava, potatoes, and other crops [64]. It is a heteropolysaccharide consisting of linear amylose, linked by α-1,4 glycosidic bonds providing good extensibility, and branched amylopectin, linked by α-1,4 glycosidic bonds in the backbone and α-1,6 linkages at the branched points. Wheat starch has numerous physical, chemical, and functional properties of technological interest, including good water solubility, gelatinization, high-temperature adhesive behavior, and easy modification. It has several food applications because it can improve texture and mouth feel or provide adhesion, structure, moisture control and/or thickening. Further, due to its exceptional transparency and flexibility, it can be used in transparent and flexible electric and electronic devices, being also a triboelectric negative material. Starch is degradable in water and soil without toxic residues thus being regarded as an environmentally friendly material [65]. Chitin, a linear homopolysaccharide composed of repeating β(1, 4)-N-acetylglucosamine units, is the second most abundant polysaccharide in the world. Chitin exists in the shells of arthropods such as crabs, shrimps, and insects and is also produced by fungi and bacteria [66]. Therefore, it can be extracted in large amounts from food industrial waste, representing an ideal candidate in a circular economy perspective. Chitin is a rigid and crystalline polymer, thus contributing to its strength and insolubility in water at neutral pH. Owing to its unique biochemical properties such as biocompatibility, biodegradability, non-toxicity, ability to form films, etc., chitin has been adopted in many biomedical applications including polymer scaffolds, drug delivery, wound dressings, tissue engineering, antiaging cosmetics, antimicrobial agents, and vaccine adjuvants. Recently, this biomaterial, being an electroactive biopolymer and a triboelectric material, has been exploited for sensing and energy harvesting. The degradation of chitin in nature is mostly performed by bacteria. Chitosan is the main derivative of chitin, it is obtained by enzymatic or alkaline chemical deacetylation at a minimum of 60% and displays interesting antibacterial and antioxidative properties. The substitution of acetylic groups with amino groups is responsible for the increased hydrophilicity of chitosan compared to chitin. Moreover, the amino group is exploited for the chemical functionalization of the polymer to acquire further properties and to enlarge its application areas, spanning from industrial, to pharmaceutical and medical applications. Chitosan can be physically degraded through different techniques based on ultrasound, plasma, microwaves, etc. [67]. Moreover, it can be enzymatically biodegraded by the human body, mainly thanks to the activity of lysozyme, but also thanks to the bacterial enzymes present in the intestinal tract, such as chitosanase and chitin deacetylase [68]. Furthermore, it was demonstrated that chitosan can degrade in the environment in a period that goes from 7 to a maximum of 10 days, confirming its low carbon footprint [69].

2.2. *Proteins and peptides*

Among natural molecules, proteins and peptides are good candidates for the synthesis of natural polymers whose units are amino acids repeated up to thousands of times. The general molecular structure of an amino acid consists of an amino group (−NH2), a carboxyl group (−COOH) and a side chain (R) typical of each amino acid. Natural proteins contain 20 different types of amino acids and they can be categorized in a variety of ways, such as: *i)* their role as an enzyme or structural element; *ii)* their external form, such as globular or fibrillar protein; *iii)* whether or not they are attached to other biomolecules, such as lipids or sugars or as a host for other ligands. Proteins and peptides can be easily obtained as waste products or by-products from the horticultural and agricultural sectors. Chemical and enzymatic hydrolysis is the most important process for the degradation of proteins and peptides [70]. The most basic (and sole achiral) protein-genic amino acid is glycine, which crystallizes into three distinct forms: α, β, and γ. While α structure is centrosymmetric, β and γ forms are non-centrosymmetric and therefore display piezoelectricity. Glycine is one of the several so-called non-essential amino acids, obtained by the hydrolysis of proteins. It was among the earliest amino acids to be isolated from gelatin (1820), but it can be largely found also in silk fibroin [71]. Since

peptides are short chains of amino acids, they have similar structural and functional characteristics. Their larger size and additional functional groups in peptides introduce more complex intermolecular interactions than between amino acids and thus lead to different self-assembly phenomena in the molecular order which translate into different material performance. Diphenylalanine (FF) is one of the most studied peptides due to its simple structure [72]. FF can often self-assemble into nanostructure in the form of nanowires and nanotubes, propelled by solvent-mediated forces, electrostatic forces, intermolecular hydrogen bonding, and $\pi - \pi$ stacking [73,74]. When an external electric field is applied during the self-assembly process, the polarisation direction of FF nanostructures can be manipulated [75]. Collagen is the primary component protein of skin, tendons, cartilage, bones, and tissues in general and it plays a major role in the structure of vertebrates and invertebrates. It belongs to a superfamily of extracellular matrix proteins which includes 28 known types of collagen, classified as fibrillar or nonfibrillar collagen. Fibril-forming collagens include types I, II, III, V, and IX, however, collagen type I represents about 80% of total collagen and it is found in abundance in the human dermis. The molecular structure of collagen is defined by three polypeptide α-chains (1000 amino acids). This spiral triple helix self-assembles through extensive hydrogen bonding of amine and carbonyl functionalities and packs into a quasi-hexagonal lattice of crystalline fibrils. Under mechanical stress, the displacement of hydrogen bonds redistributes dipole moments toward the longitudinal axis of collagen molecules, thereby inducing permanent polarization [76]. Both chemical and enzymatic hydrolysis can be used to extract collagen [77]. Although biological processes involving enzymes are more promising, chemical hydrolysis is typically largely employed in industry [78]. Gelatin is derived from animal tissue and, in particular, from pig skin (46%), bovine hides (29.4%), and pig and cattle bones (23.1%); less than 1.5% of all gelatin comes from fish [79]. Gelatin is produced by the partial hydrolysis of collagen, which represents 85–92% of this biopolymer. The gelatin-making process gives two types of gelatin, A and B, produced from acid and alkaline pre-treatments, respectively [80]. Under physiological conditions, gelatin degrades quickly in two weeks [81]. Chemical crosslinking is one of the possible ways to increase the stability of gelatin [82]. Glutaraldehyde (GLA) is one of the most popular crosslinkers used to crosslink gelatin because it reacts quickly with the amine groups in gelatin [83]. However, cross-linked gelatine is biodegradable and decomposes within a month. Enzymatic and hydrolytic degradation are the fundamental processes for the degradation of gelatin [84], enabling reduced processing times and waste. Silk fibroin is a naturally occurring fibrous protein secreted by the silkworm Bombyx mori (B. mori) and several species of spiders. Silk fibroin is composed of amino acids such as glycine, alanine, and serine in different percentages [85]. The structure of silk fibroin is composed of a heavy (H) chain polypeptide and a light (L) chain polypeptide joined together by the creation of the H-L complex. The H chain, which can form β-sheet crystallites with a monoclinic unit cell, is responsible for the superior mechanical properties of silk fibroins; the L chain plays a minor role in this process [86]. Since the majority of proteases act outside of the β-sheet regions, in general, an increased β-sheet content reduces silk degradation [87].

2.3. Synthetic polymers

Nature is also a great source of molecules that, in their monomeric form, can be combined with other molecules by synthetic approaches to obtain polymers with diverse and tunable features. This is the case of the lactic acid (2 hydroxy propionic acid) that can be extracted from numerous sources such as corn, sugar beet, starch and soy protein thanks to an eco-friendly fermentation process. It is characterized by two stereoisomeric forms, the dextrorotatory L (+) and the levorotatory D (-) that can be polymerized by ring-opening polymerization or polycondensation in its polymeric form of poly-lactic acid (PLA). By combining the different monomers or further molecules, such as glycolic acid, different polymeric and co-polymeric forms are obtained other than such as poly (D-lactic acid) (PDLA), poly (L-lactic acid) (PLLA) and poly(lactic-co-glycolic acid) (PGLA). PLA has a semicrystalline structure, but by changing the monomeric composition it is possible to tune the properties of the material like crystallinity and biodegradation time. Indeed, PLA is a biodegradable material that can be absorbed into the human body and become an important metabolite for cellular processes in several tissues such as the brain and muscles. It can be absorbed generally in a period that spans between 40 weeks and 6 years, but this can be modified by selecting the appropriate molecular proportions. Therefore, based on the desired activity of the device PLA-based molecules can be implanted for years as well as decompose in a shorter amount of time [58]. Moreover, the industrial degradation of PLA and its derivatives produces carbon dioxide and water, which can be in turn employed for photosynthetic processes to grow further vegetable sources, framing the whole process in the circular economy view [88]. Sebacate is a recurrent monomer in the development of active materials for sensing and energy-harvesting applications. It is coupled to lactic acid, itaconate (IA) and butanediol to obtain elastomeric materials with biodegradation properties. Sebacate is obtained by sebacic acid, a linear fatty acid with ten carbons ending at both sides with carboxylic groups. It is extracted by castor oil which in turn is obtained by a largely diffused castor oil plant, mainly available in India, East Africa, China, Northwest and Southwest Asia, and the Arabian Peninsula. The extraction of sebacic acid from castor oil can follow different routes, including a biotechnological green approach that is more selective than the common chemical methods. However, this route suffers from a scarce yield of products obtained and, therefore, requires further optimization. The polymers obtained with sebacic acid are moisture-proof, with elastic and thoughness properties similar to the plastic, of particular interest for numerous applications such as cellular scaffolds for tissue engineering [89]. Itaconic acid has gained considerable attention during the last decades due to its potential to substitute petroleum-based polyesters. It is formed by two carboxylic acid functionalities and an α,β-unsaturated double bond that makes this molecule attractive as a possible precursor for numerous chemical modifications. It is massively produced by following a green fermentation process starting from polysaccharides, and it easily finds application in various fields from drug delivery to material engineering [90]. Sebacate, lactate and itaconate were selected and coupled to biotechnologically-obtained butanediol to develop a biodegradable and stretchable polyester that demonstrated degradation in vitro in PBS at 37 °C over 30 days. The possibility of obtaining an elastomer that combines the elasticity and strength of synthetic elastomers with biodegradability opens new possibilities in the development of sustainable materials [91]. Poly(γ-benzyl-α, L-glutamate) is an emerging material due to its interesting mechanical properties and its biodegradability. It is approved by the FDA and shows a period of biodegradation in vivo between 8 and 12 weeks. Moreover, a product of its degradation is an important amino acid that regulates numerous functions of the human body [92]. Moreover, plants do represent a rich source of other molecules with interesting properties, such as tannins. They belong to the class of polyphenols and can be extracted from almost every part of plants, such as leaves, wood, roots, seeds and bark due to their role in the protection from fungi and bacteria. Thanks to their ability to easily interact with proteins, tannins can develop strong non-covalent bonds with numerous molecules. This aspect allows to employing tannins as plasticizers or as additives to develop strong adhesive materials. Moreover, this molecule finds application in several fields spanning from the industrial to the medical sector [93].

2.4. Combined materials

Natural biopolymers extracted from eco-friendly natural bio-waste can be used to enhance the sensing abilities of biocompatible polymers and to design biocompatible, flexible, wearable, and piezoelectric

materials for green energy harvesting and biomedical applications [94]. They are characterized by a combination of the aforementioned categories of materials, thus made by polysaccharides, proteins, fats and monomeric molecules. A typical example is cellulose, the most abundant macromolecule available in plants and in the lignocellulosic waste derived from agriculture. Plant-based sources include rose petals [28], root bark [95], wood [32,96,97], cotton [98,99], leaf [28,29], onion skin [100], laver [101] and diatom [102,103]. Such natural materials can be used in two ways to create flexible pressure sensors: *i)* they, or part of them, can be encapsulated or added to a polymer matrix to enhance mechanical strength and structural properties; *ii)* they can be used as a template to reproduce the hierarchical cellular structure of plants. Crystallin cellulose fibrils framed in plant cell walls create a 3D network of interconnected patterns that provides superior structural stability, excellent surface coverage, and efficient thermal and electrical energy distribution. The fibril components, normally cellulose, hemicellulose and lignin in varying amounts, have different decomposition temperatures: Cellulose starts degrading around 315 °C, while hemicellulose at 220 °C and lignin at 160 °C [104].

Wheat [105] and rice [106] are cultivated worldwide, and they contain starch, proteins, and lipids. Starch is the major component and consists of a mixture of amylose and amylopectin, where amylose influences the packing of amylopectin into crystallites and the organization of the crystalline lamella within starch granules. These granules can absorb water and gel, resulting in strong adhesion and providing a good bio-electrochemical environment for efficient ion transport. This allows the production of free-standing thin films that are thermoelectric, flexible, water-repellent, and biodegradable. Starch degradation begins at around 280 °C with the condensation between hydroxyl groups and starch chains, followed by the release of water molecules [107].

Zein [108], gluten, keratin [109] and collagen [110] are proteins derived from animal and plant biomass of the food and agriculture industry. Zein (corn), and gluten (wheat) are currently used in the food packaging industry, and due to their sticky and flexible characteristics, they can be potentially used for health applications and e-skin sensors fabrication. Zein is a byproduct of the bioethanol industry and is widely studied for biomedical applications. All zein fractions are amphiphilic and exhibit piezoelectric properties due to their 3D crystal structures formed by various chemical bonding states. Gluten is a material that is typically obtained from wheat flour. It is made up of glutenin and gliadin, which are intertwined and folded together in a network created through the crosslinking between protein and water. The unique viscoelastic and tensile properties of wheat gluten make it a cost-effective biodegradable polymer that can be used to create flexible films.

Keratin and collagen are two essential proteins that are present in various parts of an animal's anatomy, including feathers and shells. These proteins play a crucial role in maintaining the structural integrity of the animal's body. Keratin is a fibrous protein that forms the primary structural component of hair, nails, and feathers. Chicken feathers [109] comprise 90% keratin characterized by an elastic α-helix and β-sheet structure interconnected to each other by intra- and inter-molecular hydrogen bonding, which causes the development of electrical dipoles and remarkable mechanical strength also due to the presence of disulfide bonds. Feather keratin is biodegradable, biocompatible and has potential for use in biomedical applications. Collagen is a protein that provides strength and elasticity to the skin, tendons, and bones. The inner eggshell membrane is made up of several types of fibrils and is composed of proteins such as collagen (types I, V, and X), osteopontin, and sialoprotein. Both the outer and inner membranes consist of interwoven protein fibers, with the inner membrane being thicker and more compact. The hydrogen bonding between various micro-fibrils and proteins induces polarization under external pressure, making it an excellent candidate for capacitive pressure sensing applications. Understanding the distribution and function of these proteins in different animal tissues is essential for developing new materials and therapies for various biomedical and industrial applications.

Another interesting class of materials is biocomposites which can be created from the combination of nanometric inorganic solids, such as clay, silica, and biopolymer. These materials have attracted great interest due to their properties, such as high strength, flexibility and biodegradability making them suitable for a variety of biomedical applications [111]. Nanoclays are a type of tiny particles that are composed of layered mineral silicates. These particles are characterized by unique anisotropic plate-like structures, which are approximately 1 nm thick and 100 nm in diameter. Their combination with biopolymer showed an increment of melting temperature and crystalline rate [112]. This makes them useful in a wide range of applications, such as in the manufacturing of composites, coatings, and other materials. Bioinspired piezoelectric and triboelectric materials can be created using organic piezoelectric nanomaterials. Phages, which are protein structures containing DNA or RNA genomes, can be simple or complex in structure [113]. When phages are aligned in a single direction, they can produce decent piezoelectricity. By vertically aligning the rod-shaped M13 bacteriophage, nanostructures with significantly improved piezoelectricity were achieved through unidirectional polarization.

3. Sensors

3.1. Piezoelectric sensors

The direct piezoelectric effect consists of generating a charge separation upon applying a mechanical stimulus for materials with intrinsic non-centrosymmetry. This effect was extensively exploited in the past years to develop sensors able to convert mechanical stimuli into electrical signals. Moreover, such devices do not require a power supply, boosting their portability and sustainability. Nature provides several materials exhibiting piezoelectricity [114]. On the other hand, thanks to tailored modification of the molecules, it is possible to add further functionalities according to the needs of each specific type of sensor. In this context, an example was presented by the group of Sun [32] who developed an innovative piezoelectric material obtained by the delignification of balsa wood. Thanks to a green chemical approach, they removed lignin and hemicellulose, obtaining an ordered and porous structure made of crystalline cellulose. Such material is completely biodegradable in a few weeks and shows a 130-fold compressibility compared to native wood. The developed flexible device exhibited high stability under repeated cycles of compressions at a pressure of ≈13 kPa and was adapted to body parts to sense finger bending. The authors also proposed it for applications in energy harvesting by placing an array of sensors onto the floor or tables to capture energy from steps or finger tapping.

PLA has gained the attention of numerous research groups due to its natural origin. Derived from starch, it shows biodegradability, flexibility and good piezoelectric characteristics, therefore enabling its adoption in numerous devices. As a first example, Zhukov's group[115] developed a ferroelectric material based on a PLA foam that, after compression and poling, showed excellent piezoelectric coefficients with a d_{33} of 600 pC/N and a d_{31} of 44 pC/N in longitudinal and transversal directions, respectively. Their fabrication approach allows to exploit not only the non-centrosymmetry of the molecules but also its microstructure and porosity to enhance the sensitivity of the material. Due to the material flexibility, the authors applied it to different devices, such as biosensors embedded in tissues, artificial muscles, and energy micro-harvesters. Due to its high availability in nature, PLA is an interesting material to be employed at scalable levels in industry. For this reason, Tuukkanen's group [116] studied the piezoelectric behavior of both commercial PLA films and in-house films fabricated by a roll-to-roll approach. They investigated the piezoelectric response under different types of stimuli, and they found that the bending sensitivity of the film was much higher than the compressive one. This difference in sensitivity increased for commercial films compared to in-house films, probably due to

additional processes performed on the commercial PLA. Therefore, PLA-based devices were tested for bending sensing as well as strain sensing in force sensors.

Among the PLA-based materials, the chiral PLLA was investigated for the development of force sensors by Oh's group [117]. The authors applied a melt-spinning process to obtain flexible PLLA fibers with high piezoelectric properties. The percentage of the beta-form of PLLA was increased by the following two approaches: *i)* by controlling the drawing ratio and annealing temperature and *ii)* by introducing barium titanate ($BaTiO_3$) nanoparticles to induce the PLLA fibers orientation. By adjusting these parameters, the piezoelectric behavior of the fibers was optimized to produce flexible textiles for sensing. The developed sensitive patches were tested with palm tapping, blade of hand tapping, twisting and bending, indicating the suitability of the materials for sensing different types of physiological strains.

Moreover, another form of PLA-based material is represented by the lactate monomeric form, which shows a piezoelectric behavior. Wang et al. [118] developed a highly stretchable elastomer based on lactate crosslinked with itaconate and coupled to flexible chains of sebacate (SA) and butanediol (BDO). The high stretchability of the obtained material allowed to reach an elongation of ≈ 4.4 times the initial length. This feature helps in orienting the C═O groups on the lactate molecules, thus enhancing the piezoelectric output. Moreover, its high flexibility and self-healing ability were exploited to develop a device capable of continuous contact with the skin during different activities, resulting in more reliable monitoring.

One of the main opportunities for working with nature-derived materials is the possibility of developing implantable devices and avoiding a second operation for their removal. It is also possible to reduce the burden of electronic waste by producing biodegradable devices. These two goals were reached by Ali et al. [119] who employed PLA and glycine to develop a completely biodegradable piezoelectric material with a straightforward fabrication approach. The presence of glycine helped the crystal orientation of PLA, thus improving its piezoelectric performance. The resulting material is a flexible and lightweight film with a layer thickness of 15 μm. The developed thin sensor was attached to the skin and employed to monitor cough, carotid, and wrist artery pulse, as well as wrist and elbow bending.

Xu et al. [120] employed the chiral PDLA in the form of a stereo-complex with PLLA and quantum dots to obtain electrospinned piezoelectric fibers with luminescent properties. The stereo-complex formed by PDLA and PLLA showed a higher mechanical and thermal resistance, but its piezoelectricity was reduced due to the opposite charges that the two isomers bring. Therefore, applying an electric field during the spinning process was necessary to orient the fibers and restore the piezoelectric properties. The resulting device was applied as a force sensor as well as an energy harvester to light up a bulb.

Focusing on developing bioresorbable and flexible material, Curry et al. [121] developed a PLLA-based pressure sensor with an easy fabrication approach. The aim was to obtain a sensor able to monitor gastrointestinal pressures and diaphragmatic contractions. For gastro-related applications, employing safe and biodegradable materials is particularly important. Another advantage of the developed sensor is that once implanted, there is no need for a second operation to remove it. The devices were tested in terms of sensitivity to compression and vibration to sense mouse intrabdominal pressures.

In another work, Curry et al. [30] employed PLLA to develop a piezoelectric sensor for monitoring intrabdominal pressure. The device also operated as an ultrasound transducer for blood-brain barrier disruption to deliver drugs and viral vectors. The authors employed electrospinning to obtain flat films with enhanced crystallinity and orientation of the C═O groups, improving the piezoelectric behavior of the device. The device can be implanted, and then its natural degradation avoids a second intervention for removing it.

PLLA demonstrates high performance also when coupled to other molecules to develop composite materials. An example is the fabrication of piezoelectric, biocompatible, biodegradable and poling-free PLLA blended with Vitamin B2, which significantly enhances the crystallinity and molecular orientation of PLLA [122]. The developed PLLA/Vitamin B2 BEH exhibited a good electrical response under both mechanical and acoustic stimulations. By fixing a sound source at 3 mm from the PLLA/Vitamin B2 BEH, the output voltage of the BEH increases by increasing the frequency of the sound from 1 to 27 kHz. This device has great application potential in environmental mechanical and acoustic energy harvesting, wearable devices, and human–machine control systems.

Some classes of biomaterials can be coupled to synthetic piezoelectric materials to enhance their performances. This is the case of silk fibroin (SF), a protein obtained by silkworm cocoons that shows interesting mechanical and piezoelectric properties thanks to its hierarchical and chemical structure. Wang et al. [34] found that the amount of beta-form of polyvinylidene difluoride (PVDF) increased when it was electrospinned with SF in a core-shell structure due to interfacial interactions between the two materials. The authors realized a nanofibers mat sandwiched between two aluminum foils and attached to the metacarpophalangeal joint to monitor palm movements successfully. They also employed it to control the movements of a toy car by applying different types of strains by hand on the sensor, demonstrating its potential as a remote-sensing controller.

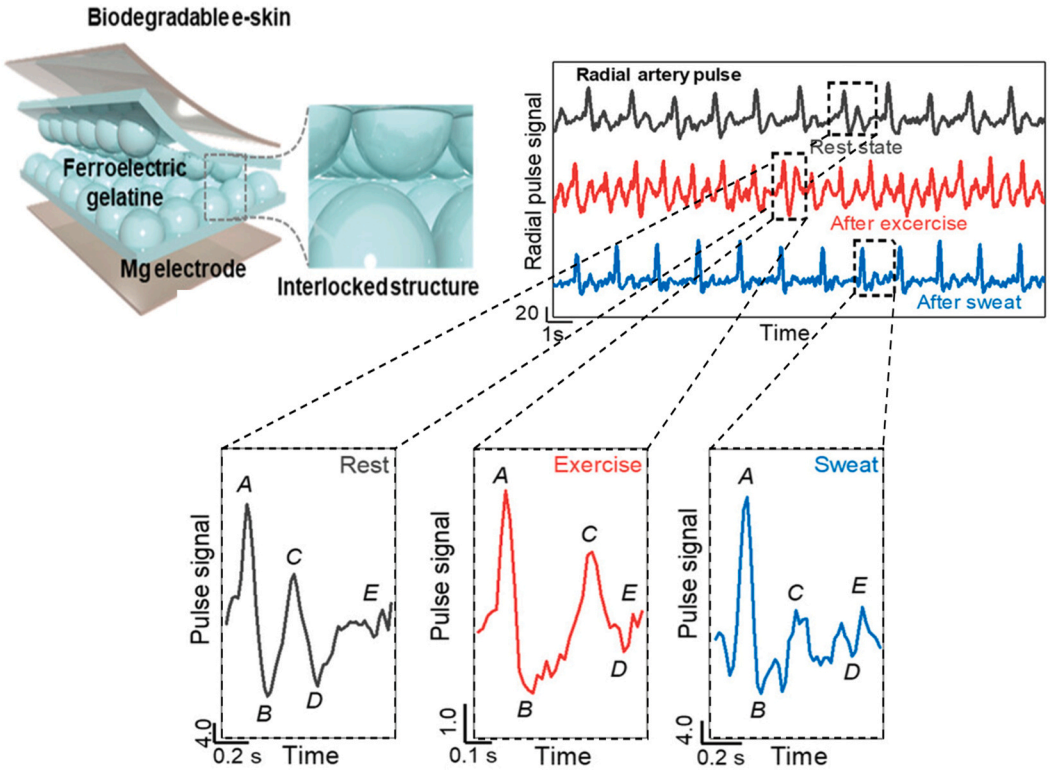
The combination of regenerated silk and gold nanorods allows the development of a piezoelectric material with adhesive properties. Thanks to the application of mild temperature and pressure conditions, it was possible to attach the previously described materials on poly (3hydroxybutyrateco-3-hydroxyvalerate) (PHBV) substrate to obtain a flexible sensor, as described in the work of Bittolo Bon et al. [35], thus demonstrating that the features of natural materials can be adapted to obtain the desired functionalities. The resulting sensor was able to respond to different forces, making it helpful in producing smart and innovative electronics in regenerative medicine.

SF can also be employed as nanofibers to develop biodegradable and eco-friendly pressure sensors for oral medical diagnosis, as demonstrated by the group of Liu [34]. In particular, the sensor was employed to continuously monitor the status of the collar system and chewing muscles to prevent oral diseases. The alignment of silk nanofibers was controlled by electrospinning parameters to enhance the piezoelectric output of the material. This, combined with micro-structured electrodes, allowed it to reach a high sensitivity of 30.6 mV/N and stability at different humidity changes, resulting in an ideal solution for the humid oral environment.

SF, tannin, and graphite can be combined to obtain a piezoelectric material with interesting bioresorbable and adhesive behavior to be exploited on soft tissues such as the gastrointestinal tract [125]. Moreover, it was also 3D printed via extrusion technique. The material, also fabricated by simple solvent casting, was positioned on the intestine of rats to monitor gastrointestinal movements. In Minhyun et al. [126], silk was coupled to PVDF-trifluoroethylen to obtain a transparent and flexible material for sensing. They applied an innovative chemical process to obtain flexible and transparent silk fibers. Device performances were measured both in compression and bending and, after placing it on a human wrist, it was possible to measure the artery pulse.

Collagen is the protein involved in the central structural systems of mammalians and is usually organized in bundles to offer mechanical resistance. This protein has piezoelectric properties that, combined with its strength and flexibility, result in a material with a high potential for sensing applications. Moreover, with the purpose of mimicking the structure of the bones that shows piezoelectricity, Fang et al. [54] combined oriented strontium carbonate nanocrystals and collagen fibrils to obtain a piezoelectric material that can be employed to develop flexible sensors. The obtained device was tested under compression and bending, and thanks to its piezoelectric response and flexibility, it was proposed as energy-harvesting device and electronic skin sensor.

Piezoelectric devices can also be developed with gelatine, an animal-



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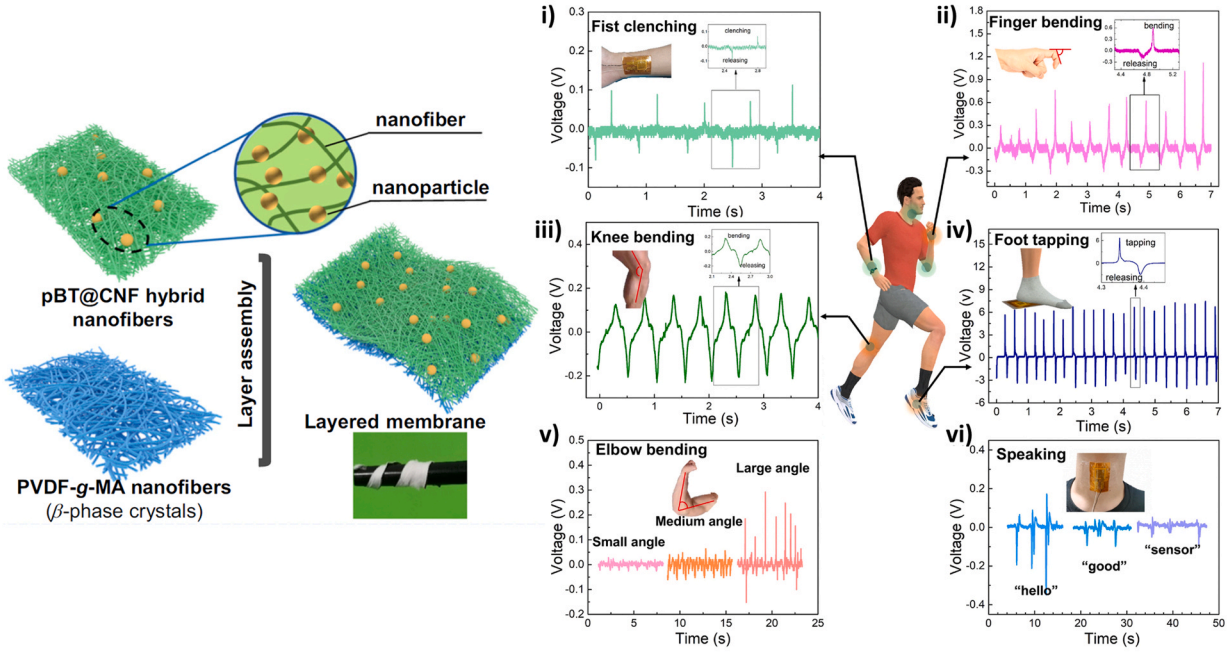


(b)

Fig. 2. (a) Schematic 3D view of biodegradable ferroelectric gelatine e-skin and pulse signals acquired on the wrist under different physiological conditions, corresponding to rest, exercise, and sweating conditions, respectively [33]; (b) Schematic illustration of the fabricated cellulose/PVDF nanofiber membrane wrapped on a Chinese chopstick and sensing performance during the detection of human motions as (i) fist clenching, (ii) finger bending, (iii) keen bending, (iv) foot tapping during a walking movement, (v) elbow bending at different angles, and (vi) speaking different words [98].

derived molecule originating from a degraded form of gelatine. The group of Ghosh [33] employed it to develop an edible and flexible detector for pressure and strains. The gelatine was crosslinked with glutaraldehyde and shaped in a microstructured form. This allowed the development of a flexible device able to sense a very high number of strains correlated to tiny human movements such as arterial pulse from the wrist, breathing pace, swallowing, coughing, drinking, vasoconstriction, and vasodilatation. Also, it was possible to map pressure exerted at different points by fingers (Fig. 2a).

Chitosan is an emerging material representing the second most abundant polysaccharide after cellulose. It can be extracted in large amounts from seafood waste as chitin and then easily converted into chitosan thanks to a deacetylation process, representing a highly sustainable and green source. Due to its non-centrosymmetric structure, chitosan displays an interesting piezoelectric behavior, and different research groups have started to focus their attention on this high-potential material for the development of piezoelectric sensors. As an example, in the work of Hosseini et al. [31] a chitosan layer was blended with glycine to develop a pressure-sensitive detector. The amount of glycine added to chitosan was tested to find the best ratio to have the best electrical features. Moreover, the developed sensor, which is fully biodegradable, was tested with hand tapping.

The potential of chitosan was addressed by de Marzo et al. [127] who applied a chemical treatment to enhance the piezoelectric efficiency of chitosan, improving it from 6 pC/N to a value of 15.56 pC/N. This allowed to exploit this material in the form of thin, flexible, and transparent film and to develop numerous devices able to sense ultrasound. The authors also developed the first type of miniaturized ultrasonic transducer based on this piezoelectric natural material.

Kim et al. [35] presented a bottom-up regeneration strategy to create a chitin nanofibers film with silver electrodes. The piezoelectric properties of freestanding chitin film were adequately evaluated by measuring the current and voltage outputs. In the low-pressure region (below 10 kPa), the output current density increased following the applied normal pressure. At 100 kPa, it increased monotonically to 177 nA/cm^2. The piezoelectric response of the freestanding chitin film was used in a wide vibration frequency range for the fabrication of chitin-based speakers and microphones.

Also, cellulose has demonstrated its potential in the sensing field thanks to its piezoelectric properties. These properties, together with the wide availability in nature as waste material, make cellulose a good candidate for developing innovative sensing devices. In this context, Wang et al. [98] developed a composite material consisting of cellulose nanofibers (CNF), $BaTiO_3$ nanoparticles embedded in polydopamine, and PVDF grafted to maleic acid. This composite material was fabricated by assembling the cellulose layer into the PVDF layer in an interconnected structure. The localized stress produced by the nanoparticles and the electrospinning technique, which promotes PVDF beta orientation, is responsible for enhancing the piezoelectric properties. The developed device was attached to different body parts to detect physiological movements, such as fist clenching, finger and knee bending, footsteps, elbow bending, and, if attached to the throat, voice recognition, thus demonstrating its application as a self-powered sensor (Fig. 2b).

Another natural material, the pectin extracted from the plant cell wall, was employed by Bahnu et al. [128] to develop degradable and flexible pressure sensors. However, its high hydrophilicity and brittleness make it necessary to blend it into other materials, such as graphite and PEG, to obtain composites with improved mechanical and electrical features. The developed sensor was tested with finger touch, by touch with conductive and non-conductive objects, and load charge.

The Lu group [129] developed a sensor based on biodegradable and recyclable materials to avoid any e-waste linked to device fabrication. In particular, they employed Bacterial Cellulose (BC) and imidazolium perchlorate to develop a piezoelectric and ferroelectric material able to respond to pressure to sense stretching and twisting. To demonstrate its potential, the material was employed to develop a self-powered keyboard.

Through the development of plantar sensors, it is possible to monitor human body movements, which play a central role in the case of diabetics and physical rehabilitation therapies. Guan et al. [53] mixed piezoelectric Lead zirconate titanate (PZT) with microfibrillated cellulose to overcome the ceramic material's brittleness and retain flexibility. The device was placed below the plantar of the foot to monitor different activities, such as walking, jumping, and squatting, with good pressure sensitivity.

The most common approach for the development of devices based on nature-derived materials is to extract and use them with minimum chemical modifications; however, another strategy is represented by the synthesis of new molecules and materials starting from natural sources. The group of Xu [52] presented a flexible Tempo Oxidized Cellulose Nanofibrils/MoS_2/BaTiO3 composite film to fabricate a piezoelectric BEH for physiological monitoring systems. The incorporation of tetragonal BaTiO3 nanoparticles allowed to obtain a freestanding film with an excellent piezoelectric coefficient of 45 pC/N. The obtained PENG showed high sensitivity to tiny environmental forces with a real-time response of 0.3 V peak voltage under acoustic vibration provided by hand clapping or by falling leaf. The device was used to monitor real-time human movements and physiological health parameters, such as blood pressure and heart rate. Additionally, the device exhibited pyroelectric properties and was designed as an electronic skin to monitor temperature.

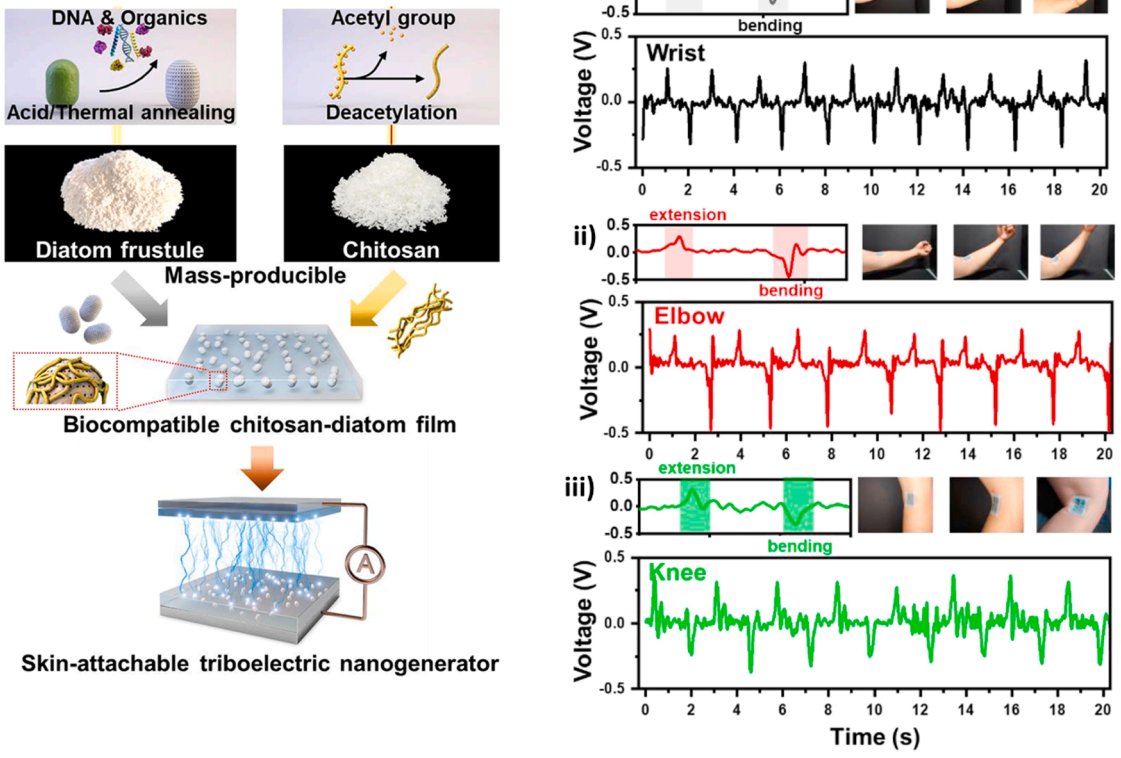
Kumar et al. [130] reported on the fabrication of an entirely organic biocompatible PENG having multifunctional energy harvesting capabilities at a time (mechanical/acoustic/wind) by integrating Vitamin B2 with PVDF to create a composite having a d33 piezoelectric coefficient of 50.3 pC/N. The device showed ultra-sensitive performances generating an output voltage at the human audible sound at different sound pressure levels of acoustic energy. The BEH also acted as an acoustic energy harvester when attached to a sound speaker, reproducing three different instruments: solo piano, guitar, and violin, playing the music with a sound pressure level (SPL) of ~84 dB, generating maximum output voltages of ~0.30, ~0.42 and ~0.53 V, respectively.

Nguyen et al. [47] combined an aminoacid-derived molecule, the poly(γ-benzyl-α, L-glutamate) (PBLG), with the transparent and biocompatible Poly Dimethyl Siloxane (PDMS). PBLG was electrospinned as a membrane and then soaked in PDMS to obtain a flexible film protected from humidity for biomechanical and underwater acoustic sensing. The electrospinning technique allowed obtaining aligned fibers showing a high sensitivity and high output signals for energy harvesting purposes.

Another interesting work that exploits amino acid is presented by the group of Li [131], which developed an omnidirectional stretchable piezoelectric sensor made of self-aligned DL Alanine. The highly ordered structure allows not only to match the stretchability of the skin but also to reach a satisfactory piezoelectric behavior to monitor hand, finger and swine muscles movements.

3.2. Triboelectric sensors

Triboelectric sensing is based on the generation of charges between two mating surfaces upon dynamic contact. Sensors for human movements exploiting triboelectric mechanisms are characterized by lightweight and high sensitivity. Moreover, many materials derived from natural sources show the ability to generate charges when coupled with the electronically complementary countersurface. Therefore, efforts were focused on the development of triboelectric sensing devices based on nature-derived materials. An interesting example of triboelectric sensors was reported by Kim et al. [102]. They employed two natural materials, chitosan and diatoms derived from ocean wastes. The presence of diatoms in chitosan increased the triboelectric performances of the film thanks to the hydrogen-bond interactions and the geometrical



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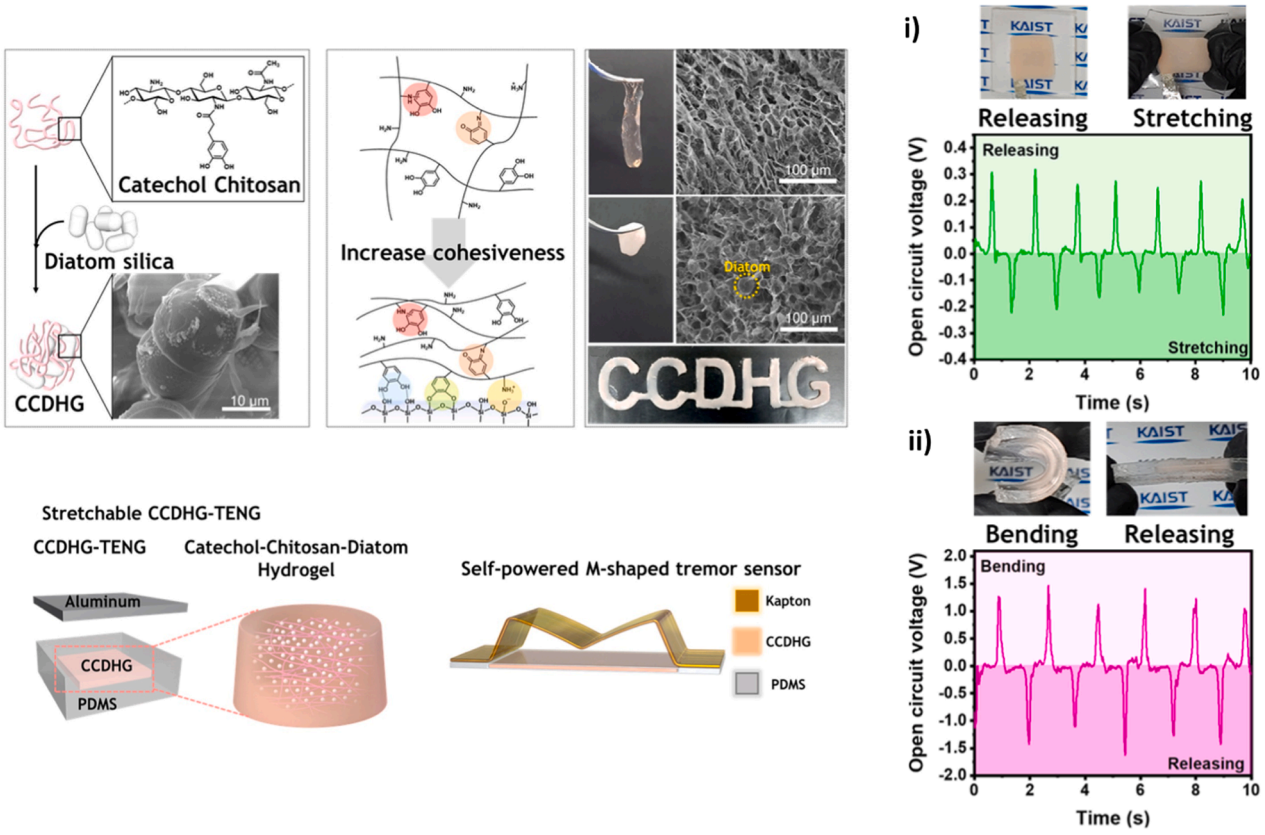

(b)

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Fig. 3. (a) Schematic illustration of the biocompatible chitosan-diatom triboelectric BEH and photograph of the motion sensor and the output voltage acquired during the movement of i) wrist, ii) elbow and iii) knee [102]. (b) Fabrication method of catechol-chitosan-diatom hydrogel from catechol-chitosan and corresponding optical images of both catechol-chitosan and catechol-chitosan-diatom hydrogel (CCDHG); a schematic image of the CCDHG-TENG and the tremor sensor are also reported. Moreover, i) and ii) report the open circuit voltage of e-skin based on CCDHG-TENG working under stretching and bending, respectively[103].

structure of the diatoms. Such a sensor was employed to characterize the movements of joints like wrist, elbow, knee, and ankle (Fig. 3a).

The marine environment inspires several research groups as it represents the source of numerous natural compounds that can be employed in the sensing field. The group of Kim [103] developed a hydrogel-fabricated sensor based on chitosan, diatoms, and catechol that enables the device to be easily attached and keep it conformal to the skin. The triboelectric device was employed for energy harvesting but also like an e-skin for the detection of movements such as stretching, tapping and bending. Moreover, the authors developed a 3D structure able to sense tremors with different intensities for Parkinson's disease sensing (Fig. 3b).

Paper is a very interesting material for the development of sensors due to its availability and easy waste removal. The group of Wu [132] employed a polyethylenimine-coated paper to develop a triboelectric, low-cost, water-resistant, antibacterial and flame-retardant electronic skin. The developed device, able to detect the sliding/bending movement of the fingers, proved to be disposable since the polyethylenimine is water soluble. Moreover, the authors realized sensors array to distinguish the spatial recognition of the pressure distribution.

A water-soluble and biodegradable triboelectric sensor based on two types of cellulose, CNC and methylcellulose, with graphite as a conductive electrode, was developed by Wang et al. [133]. The paper can also be recovered from office waste to obtain a material that meets the circular economy requirements. The sensor was then employed to sense the respiration rate, to test the breathing status of bedridden patients, and to monitor the movements of joints such as the elbow and wrist.

The self-healing ability is one of the most desired properties in sensing materials as it allows to place them on the skin and avoid any damage during the usage of the device. As an example, the group of Han [134] developed a highly stretchable hydrogel based on gelatine and polyacrylic acid (PAA) to conformally apply onto the skin surface as a flexible and stretchable electronic skin. The triboelectric device was able to respond to both stretching and compression stimuli and also to light a light-emitting diode (LED).

The group of Li [135] employed sodium alginate (SAlg) mixed with glycerol to obtain a triboelectric sensor. The electrodes consisted of SAlg and silver nanowires (AgNWs). The thickness and the size influenced the device performance in terms of generated charges, and the best results were obtained for the thinnest and largest devices. The transparent sensor was able to detect different movements, such as finger bending, and forehead and throat movements, to map finger pressure and was employed as a game controller.

PVDF-based sensors also exploit the triboelectric effect, and in particular, different strategies can be adopted to enhance the crystal orientation of PVDF fibers when coupled to specific molecules. As an example, Eom's group [136] exploited the annealing of PVDF on a chitin substrate to enhance the performance of the material. The device, transparent and stable in water, was tested for pressure sensing, and its performances were demonstrated by employing it to monitor artery pulses on the neck as well as to sense music.

3.3. Piezoresistive sensors

Among the mechanisms exploited to develop flexible sensors, the piezoresistive effect is the most employed when nature-derived materials are involved. It is based on flexible materials that change resistance or current due to the applied strain or pressure. Piezoresistive sensors have several advantages: Easy fabrication and manufacturing, efficient response, simple read-out, low cost, and low power consumption. On the other hand, this type of sensor requires bulky reading systems and a power supply.

Among the employed materials, cellulose and its related products, such as paper and nanocellulose (cellulose nanocrystals and nanofibers, to cite a few), are widely used due to their availability and easy processing. As an example, Koval's group [137] developed a piezoresistive strain sensor based on nanocellulose obtained from non-wood cellulose, with a relatively easy fabrication approach. The device was made of a thin nanocellulose film with a sputtered layer of conductive nickel on the top. The overall thin sensing layer was able to detect bending and straightening movements upon changing resistance.

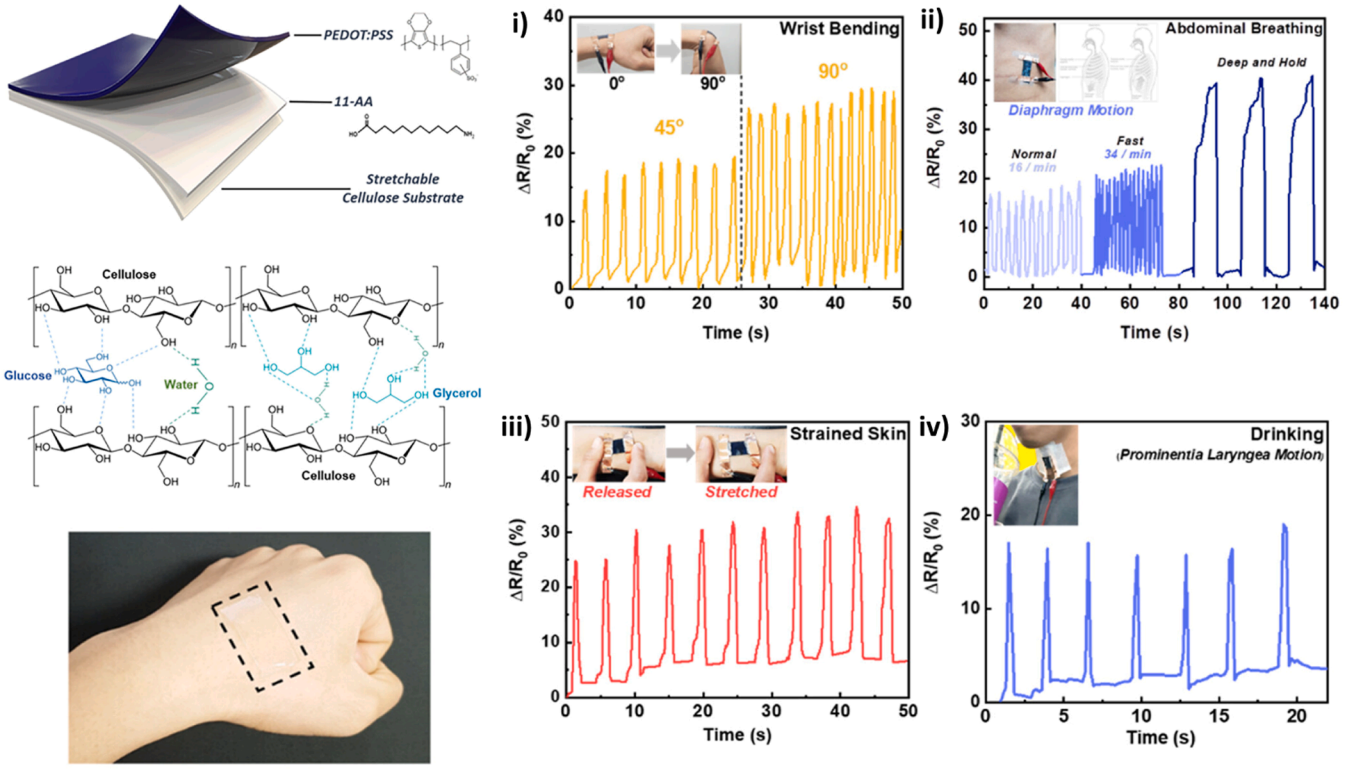
Due to its biodegradability, renewability, and low cost, a paper substrate was exploited by Liu's group [138] to fabricate a sensor able to detect very small strains. The paper was covered with a conductive layer of silver nanoparticles and a super-hydrophobic coating for underwater sensing. The latter was inspired by nature for both material choice and its surface micro-structuring. Indeed, they fabricated scorpion-inspired "V"-shaped microgrooves to reach a high sensitivity with a strain resolution of 0.089%. This was due to the deformation of the grooves, whose resistance in the conductive layer changes upon small compressive and tensile strains. This allowed them to employ the device for detecting strains generated by movements of fingers, elbow, swallowing, and for voice recognition. Papillae-like microstructures, like in Lotus leaves, conferred super hydrophilic behavior for underwater sensing to detect drops falling on the water surface. These characteristics pave the way for application in environmental sensing.

Hydrogels can also be designed with self-healing ability, thus when implemented in sensors, can improve their durability. This is the case of the CNF-polyvinyl acetate (PVA)-borax-graphite hydrogel, developed by Zheng's group [139]. Thanks to the dynamic interaction between the borax and PVA that ensured self-healing ability, as well as to the mechanical strength and conductivity of the CNF-graphite composite, they were able to develop a soft, stretchable, and durable strain sensor. The sensor was tested to monitor finger bending and, when attached to a puppet, was able to distinguish different positions of the arm. Also, it was employed to discriminate between different handwritten forms, which could be beneficial for sign language recognition and deaf-mute subjects.

The development of a highly stretchable, self-healing and conductive hydrogel was reported by Chen's group [140] that fabricated a hierarchical triple-network structure based on biodegradable cellulose nanofibrils, polyacrylic acid and polypyrrole crosslinked by ferric ions. The gel exhibited mechanical strength, viscoelasticity and stretchability mimicking the human skin's behavior. The developed device was attached to human fingers, wrists and cheeks to detect small/large bending and stretching movements.

Wibowo's group [141] focused on the development of a stretchable and flexible sensor by combining a cellulose film with layers of poly(3, 4-ethylenedioxythiophene) polystyrene sulfonate (PEDOTS: PSS) thanks to surface modifiers. The piezoresistive effect allowed to employ it as a wearable sensor to detect body motions and for voice recognition (Fig. 4a).

An interesting approach to enhance the piezoresistive performances of materials is to improve the space between molecular planes. One example is given by Yang's group [142] which used film made of bacterial nanofiber cellulose intercalated into an MXene layer. This nature-derived filler helps to enhance the flexibility and compressibility of the MXene structure and improves its mechanical resistance thanks to the presence of hydrogen bonds. This allowed obtaining high sensitivity



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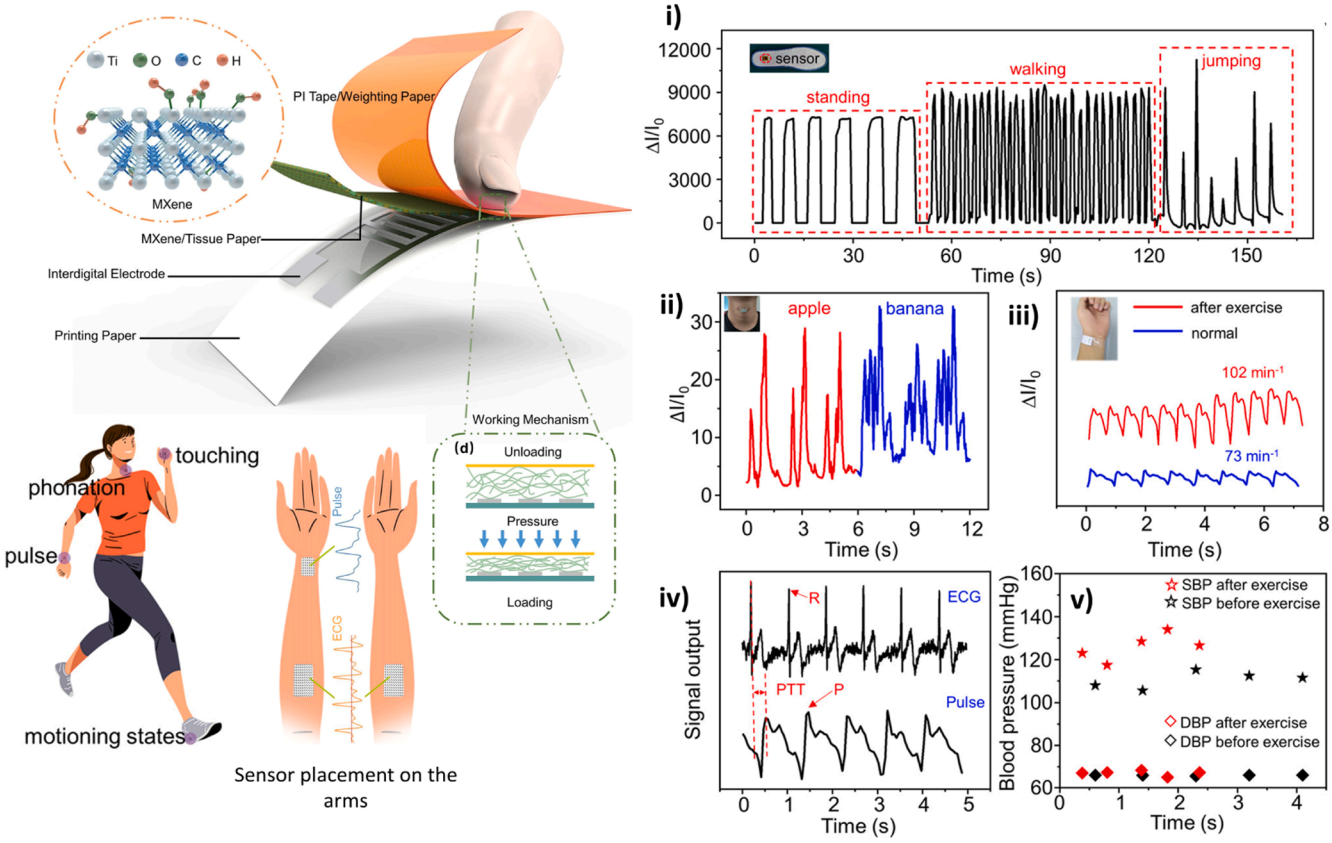

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Fig. 4. (a) Schematic of the cellulose/PEDOT: PSS hybrid film with its chemical structures, including the added glycerol and glucose. A picture of the cellulose film attached to the skin (back of the hand) showing the stretching capabilities (up to 300%). Real-time relative resistance responses of on-skin sensors during i) wrist bending, ii) abdominal breathing, iii) skin stretching and movement of the throat (motion of the prominent laryngeal while drinking water) [141]. (b) Schematic of the paper-based wearable sensor and various scenarios of physiological monitoring. The sensor consists of interdigital electrodes and MXene/tissue paper (the working mechanism is also shown). Moreover, three different motion states - standing, walking, and jumping - are also displayed in i), whereas throat movements while reading the words "apple" and "banana" are shown in ii). Wrist pulse signal before and after exercise is shown in iii), whereas the concurrent recording of ECG and arterial pulse signals with the pulse transit time (PTT) for blood pressure monitoring are in iv). Finally, v) shows the calculated beat-to-beat systolic blood pressures (SBPs, stars) and diastolic blood pressures (DBPs, rhombus) before (black) and after (red) exercise using the PTT method [168].

in a very low-pressure range for sensing both tiny and large movements, such as finger gestures, thumb, knee, and elbow bending, swallowing, heartbeat and voice recognition. An example of a sensor developed only with cellulose was proposed by Chen's group [143] that employed two kinds of paper to realize a soft and flexible resistive sensor. In particular, they employed a smooth printing paper to obtain interdigitated electrodes, whereas the sensing part was obtained with a porous crepe paper, the latter made conductive with a thermal approach. The sensor was tested by compression and bending and was able to record respiration, phonation, music, wrist pulse, and falling water droplets, as well as sense the pressure distribution with sensors arranged in an array.

The combination of BC and chitosan was exploited in the form of a very light aerogel by Huang's group [144]. The aerogel was made conductive by employing polyaniline (PANI), a conductive polymer, and the developed structure was embedded into a PDMS shell, to ensure mechanical resistance to repeated cycles as well as to achieve a stable signal. The obtained sensor was employed to detect fingerbending swallowing during drinking and to distinguish words by attaching it to the throat, being able to detect very light pressures down to 32 Pa.

Cellulose and its derivatives can also be employed to improve the performances of already existing materials. For example, deep eutectic conductive solvents represent a groundbreaking solution to develop transparent, stretchable and conductive materials, even if they exhibit poor mechanical resistance. Wang's group [145] solved the latter issue by adding a green and sustainable mechanical reinforcement based on BC, thus exploiting its ultrapure crystalline fibers. By exploiting a mild fabrication approach, they developed a piezoresistive sensor able to detect very light pressure, strain stimuli, and temperature to monitor human gestures and limb movements. When coupled to an emissive layer, it also worked as an electroluminescent device thanks to the ionic conductivity of the material.

The group of Liu [146] exploited a fabric-based material to develop wearable and flexible sensors. In particular, the authors covered some cotton fibers with carbon nanotubes (CNT) and polyester fibers, employing Ni-based coatings shaped as interdigitated electrodes. Finally, they assembled the two materials to obtain either a single element or sensor arrays sensitive to different stimuli, thanks to the hierarchical organization of the fibers. Indeed, these devices were able to sense pressures as low as 2 Pa. The device was employed for sensing finger bending and hand gestures, but also to recognize sound and heart pulse, as well as to develop pressure maps when arranged in an array. The strength of this approach is the employment of a common cotton fabric as a base to develop flexible and sensitive e-textile materials.

The MXene was coupled to BC fibers to obtain a strong, super compressive, and sensitive carbon aerogel in the work of Chen [147]. It was fabricated by freeze drying, allowing the alignment of the lamellae of MXene, while the BC helped to keep them in the same position, thus strengthening the structure. The developed device was able to sense a large range of pressures. It was applied to sense small movements like jugular vein pulse, voice pressures, wrist and elbow bending, and facial expressions.

Piezoresistive materials can also be fabricated superhydrophobic to achieve self-cleaning and to allow operability in humid conditions. In particular, Yun's group [148] employed carboxymethyl cellulose (CMC) as a linker to combine carbon black, multilayer graphene and SiO_2 nanoparticles to develop superhydrophobic piezoresistive flexible and wearable sensors. The device was used to sense different stimuli, from finger, elbow, wrist, check and throat bending strain to pressure field exerted on a sensor array. Microcracks also decorated the sensor to enhance its sensitivity.

An example of paper-based electronics was given by Tao's group [149]. The authors developed a multi-layered structure to tune its sensitivity. Indeed, a higher number of layers makes the sensitivity rise in the small pressure range, whereas it falls in the larger range. This behavior is ascribed to the presence of holes in the paper that reduce the conductivity of the material. When stress is applied, the layers get more in contact between them, the holes are reduced, and the resistance falls rapidly, ensuring high sensitivity at small pressures. This behavior is due to the presence of small air gaps between the layers, as validated both by simulations and experiments. The high sensitivity of the sensors was demonstrated by recording voice, wrist pulse, and respiration rate. They also demonstrated that it is possible to distinguish between different exercises by placing the sensors on the soles, like jump, push-up, squat, walk and run.

The piezoresistive effect of graphene-coated cellulose paper was employed for applications in sensing pressure and humidity changes. Khalifa's group [150] employed two different methods of dip-coating and vacuum filtration to absorb graphene on the surface of the papers. In both cases, they were able to obtain a device sensitive to pressure and apply it to sense different loading and body movements like finger tapping. They found that the vacuum filtering method allowed obtaining a more sensitive device.

The compressibility of soft structures and its change in resistance was exploited by Zhai's group [151], which adopted a mixture of CNT, cellulose nanocrystals (CNC), and polyurethane to obtain a soft aerogel, responding to compression at different pressures. The obtained device allowed to reach good responsiveness and relatively high sensitivity, and it was employed to monitor different movements such as finger, wrist, and elbow bending, squatting and rising, walking and running.

In the work of Gao et al. [152], biodegradable and nature-derived materials have been used as sensing materials and homogeneous substrates for electrode printing. In particular, a porous and flexible structure of tissue paper was coated with AgNWs to obtain the sensing layer. Instead, a very smooth nanocellulose paper was printed with silver interdigitated electrodes and integrated with the sensing part. In this way, it was possible to obtain a flexible and easily disposable sensor, able to respond to different pressures and to sense bending, pressing movements, wrist pulse, voice records and to recognize pressure locations.

Pyo's [153] developed a highly sensitive sensor that, at the same time, can work under a broad range of pressures. In particular, the authors fabricated a structure consisting of CNT and Ni-coated fabric based on cotton. The hierarchical structure, given by different fabric layers, allowed to broaden the sensing pressure range to detect heart pulses on the wrist for developing a wearable musical keyboard and a flexible and foldable fabric computer keyboard. The sensors can be positioned on a glove to develop a haptic device able to recognize the pressure in handling objects.

Hao et al. [96] produced a wood-based pressure sensor characterized by high conductivity through a coating treatment with reduced graphene oxide (rGO). The obtained wood-based pressure sensor exhibited a high sensitivity of 1.85 kPa^{-1} over a broad range of pressure (0 −60 kPa), high stability over 10k cycles, a fast response time (150 ms), and a low detection limit (60 Pa). The high sensitivity allows to discriminate

different pressures and to monitor subtle body movements, for example, muscle movement during speech, and can be used for real-time monitoring of physiological signals such as heartbeat, able to distinguish three distinct waves of the arterial pulse.

Long et al. [154] presented a porous and coral-shaped molybdenum carbide-graphene (MCG) composite as piezoresistive sensing material fabricated by a direct laser writing method on paper substrates to make paper-based strain/pressure sensors. The strain sensor exhibited the gauge factors for the tensile and compressive strain of 73 and 43, respectively, with fast responding/recovering time and good stability. The prototype sensor was able to clearly distinguish weak pressure signals, such as breathing patterns at the chest for application in human health monitoring, and small mechanical vibration sources, such as for acoustic sensing from sounds.

Besides cellulose, many biomaterials can be exploited for piezoresistive sensing, such as sea-derived chitosan and alginate. Moreover, the piezoresistive effect can be significantly exploited when combined with wrinkled structures, typically observed on soft and elastic surfaces. Lei's group [49] demonstrated the possibility of combining physical crosslinking and pre-stretching of a chitosan-based gel to form a 3D self-wrinkling structure. The structure is then functionalized with a conductive layer of PANI to develop a strain sensor to detect finger bending based on the piezoresistive effect. The obtained sensor showed a high sensitivity in a small pressure range between 15 and 50 Pa, paving the way for applications such as electronic skin.

Material stretchability is another important feature for the development of sensors, as reported by Liu et al. [155] who proposed a hydrogel based on PVA, chitosan, and phytic acid acting as cross-linker. This hydrogel, due to its thermoplastic property, can be extruded and employed as an ink to develop arbitrary forms. The developed sensors exhibited a linear response in a wide working range. The device generated stable output signals after being stored even for 35 days, and at the same time, it can be easily degraded. The sensor was tested by monitoring finger and elbow movements and during running.

It was demonstrated that SAlg and tannic acid, both biodegradable molecules from natural sources, can be combined in the presence of crosslinked polyacrylamide (PAM) to develop a highly stretchable, elastic, and self-healing gel with piezoresistive behavior to be employed as strain sensors for large limb movements and subtle muscle movements [156]. The presence of tannic acid conferred a very good self-adhesiveness, thus making it suitable to stick to different materials such as plastic, glass, stone and biological tissues, both in air and underwater. This property greatly enhanced the adhesion to the human skin, thus promoting the body movements sensing with high precision. Large movements, such as knee bending, and small movements, such as smiling, wrist movements, finger bending, swallowing, and respiration before and after exercise, were detected.

Also, the protein-based SF can be used to fabricate strain-sensitive piezoresistive devices. Yang et al. [157] developed a piezo sensor that combines conductivity and conformal attachment to the skin, even in the case of sweat conditions. For this purpose, the authors combined a conductive polymer (polypirrolidone) and the SF, then coupled this layer with a silk-based gel, interlocked with the sensing part. The strong interlocking between the two polymers allowed to obtain elastic and stretchable sensors able to sense the heartbeat with high sensitivity.

An ultralight aerogel was developed by Abadi's group [158] taking inspiration from the nacre. They employed a combination of SF, graphene oxide, and MXene nanosheets to develop a highly porous and conductive material with intrinsic flexibility for piezoresistive pressure and strain sensing. The hierarchical organization of the sheets, combined with the lightweight and high compressibility of the involved materials, allowed to develop a flexible device able to sense very tiny stimuli, such as movements due to facial expressions.

Natural proteins can be combined with synthetic materials to increase the performance of the sensors. In Chao's group [56], an SF nanofiber membrane was coated with conductive MXene to develop a sensing layer. Then, it was placed on an SF nanofiber membrane printed with MXene ink interdigitated electrode as electrode layer to obtain a flexible piezoresistive sensor to record pressures. The sensor endows a combination of flexibility, lightweight, breathability, and environmental sustainability due to its biodegradation.

Optical transparency, mechanical toughness and thermal stability are shown by a material developed by Reizabal's group [159]. They were able to reach these characteristics by combining SF with AgNWs. The developed piezoresistive sensor was highly sensitive to pressure and was also able to light up LEDs.

An excellent example of piezoresistive materials involving different classes of biomaterials is presented by Zicong's group [160], where electrospun fibers consisting of poly-lactic acid (PLA), SF, and collagen (CL) were adopted to obtain flexible, sensitive, and skin-compatible sandwich-structured piezoresistive pressure sensors. The piezoresistive sensor showed a sensitivity of 24.13 KPa^{-1}. The human finger-tapping tests confirmed that the sensor was more sensitive to relatively small pressures. Real-time measurements of human wrist pulse, neck pulse, and respiration rate were conducted. Since the sensor might record the throat vibration during speech, it could be used to distinguish between human voices and transform auditory data into electronic signals.

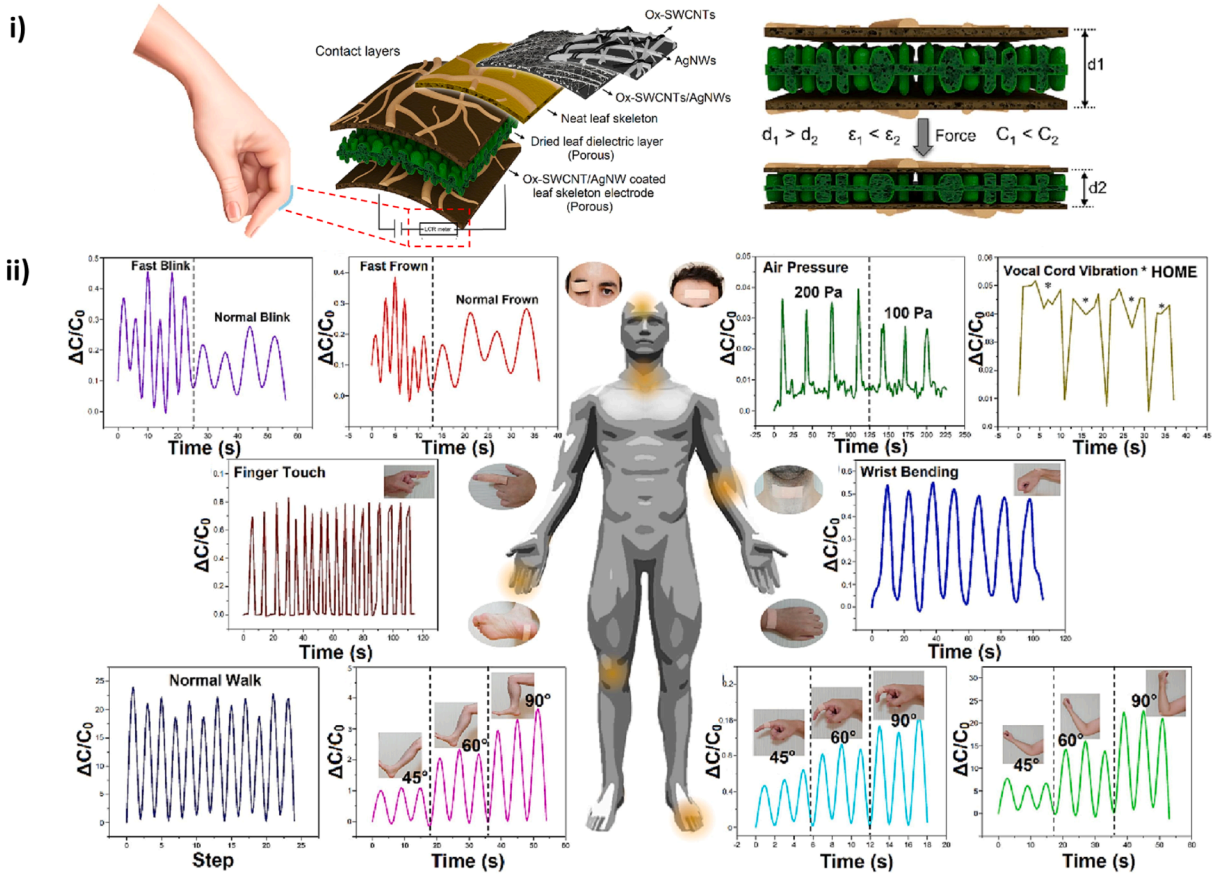
Liu's group [161] had the idea to develop a biocompatible, biodegradable, and conductive ink based on gelatine, PVA, and multi-walled carbon nanotubes (MWCNT). The ink was easily screen-printed onto fabrics, maintaining a simple and scalable fabrication approach. The authors, to enhance the performance of the material, instead of performing the nanopatterning technique, applied a pre-stretch to the device, creating micro-cracks to realize a random patterning. The fabricated device applied onto a glove, was able to distinguish between nine different finger gestures with the help of an artificial intelligence algorithm.

Collagen was also explored by Zhang et al. [162], which employed a 3D-based approach to obtain a flexible material that changes its conductivity properties under pressure stimulus. In particular, they adopted a porous aerogel consisting of crosslinked collagen fibers and MXene to obtain a biodegradable structure with high mechanical resistance and thermal insulation. It was tested in terms of response to pressure, showing a stable signal even at different temperatures and demonstrating high thermal stability. It was then employed to sense different kinds of human motions, such as finger tapping, elbow bending, finger bending, and different movements, such as walking, running, and jumping in a low or fast way.

A gelatin-based ionic conductive hydrogel was fabricated by Zhang's group [163]. The developed material has an interesting temperature-dependent adhesion which enhances the contact with the skin at body temperature, whilst displaying an easy detachment at lower temperature. Moreover, when combined with metallic electrodes, it can be employed as a strain-sensitive resistive device to monitor body movements. Thanks to its high adhesion, the device can closely and sensitively monitor movements of elbow, arm and finger bending and water wallowing on the throat.

An interesting work was reported by Li's group [105], which mixed carbon black and wheat flour to obtain a carbon-black dough to monitor body movements such as elbow bending and sweat monitoring. The authors' idea was to exploit the mix of molecules naturally present in the flour, such as sugar, starch, lipids, and gluten, to obtain a mechanically resistant dough that can also interact with body fluids. The presence of carbon black makes the dough conductive enough to be exploited as a biodegradable piezoresistive sensor.

Another example of the combination of biodegradable polymers with conductive materials was presented by Scaffaro's group [164], which combined biodegradable PLA and PEG with GO as conductive material. The amphiphilic nature of GO allowed to improve the interactions between all the materials and to obtain a homogeneous system without the adoption of complex and environmentally unsustainable protocols. This material can detect both pressure and strain stimuli.



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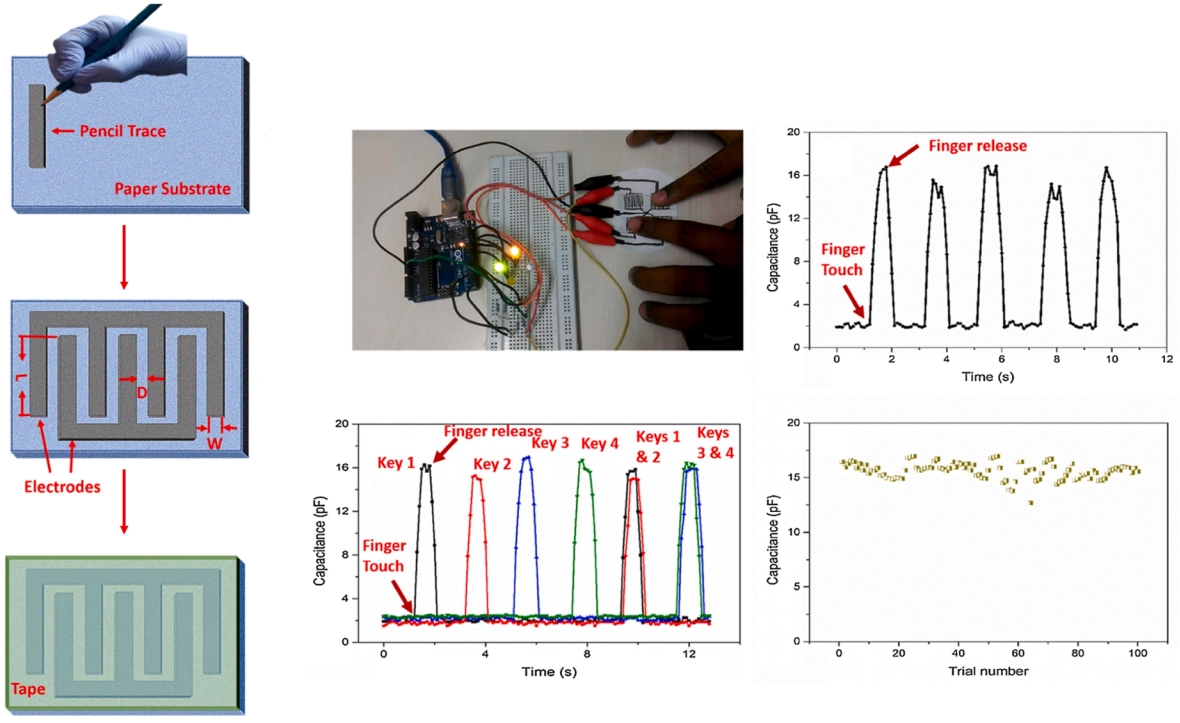

(b)

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Fig. 5. (a) Composed layers and working principle of developed e-skins i). Human whole-body physiological monitoring by Ox-SWCNT/AgNW all-leaf biodegradable e-skin is also shown in ii). The detected parameters include blinking, normal walking and joint bending, finger touch and vocal cord vibrations [29]. (b) Interdigitated capacitive touch sensor based on graphite on paper. The picture also shows the touchpad connected to LEDs and interfaced to Arduino UNO during the measurement of the capacitance variation of a single key sensor and touchpad itself with touch; the capacitance of a sensor after several touching cycles - namely, as a function of trial number - is also shown [170].

To develop lightweight sensors, Yan's group [165] proposed an air-rich polyurethane sponge dip-coated with silver nanoparticles, CNT, CNC and tannic acid to obtain a self-healing and pressure-sensitive piezoresistive sensor. The idea was to endow the sponge with electrically conductive silver that forms nanoparticles in the presence of tannic acid. The brittle coating given by CNT and CNC made the sponge very sensitive to pressure thanks to the formation of nanocracks that change the material resistivity. Furthermore, the sponge had self-healing properties at high temperatures. The obtained sensor was highly compressible and sensitive to different loads. It was tested to sense the finger and wrist bending, the stamp done by feet, and also the falling of a drop onto it, showing in all cases the possibility of having a highly affordable operatibility.

The employment of brittle materials to develop soft sensors was also pursued by Zhang's group [166], which adopted highly compressive polyurethane and CNT foam coated with a mix of CNT and CNC, fabricated by a freeze-drying approach. The brittleness of the coating made the sensor very sensitive to small compression ranges and enhanced its sensitivity. The sponge was studied in compression and employed to sense the throat during drinking, cheeks, finger, and wrist movements and steps. Also, a sensor array was developed to distinguish between different loads in different positions.

Zaho et al. [167] presented a flexible piezoresistive pressure sensor based on a hybrid porous microstructure, fabricated using sugar as a template, to form a porous PDMS structure replicating the surface of Epipremnum Aureum leaves. The obtained flexible pressure sensor exhibited a high sensitivity of 83.9 kPa^{-1} at an applied pressure lower than 140 Pa. For this reason, it was used to detect physiological parameters, such as wrist pulses for heartbeat monitoring, as well as for speech recognition.

Based on the use of composite materials, Su et al. [55] presented the fabrication of a pressurized film of MXene (Ti3C2Tx)/BC used as a device for the detection of sound signals, able to distinguish different voices and display sound images. The device exhibited high sensitivity in the low-pressure range (0 −0.82 kPa, with sensitivity 51.14 kPa^{-1}), wide linear range (response/recovery time of 99/93 ms), and high stability (5k cycles). Traditional human motions, including finger pressing/knocking, exhaling/blowing, and nodding/swallowing/coughing, were monitored. The device showed resistance variations at different acoustic signs caused by different throat muscle motions; it could recognize the smallest unit of different languages (Chinese/English), variation of voice attributes (volume tonality), and similar signals in vocal/non-vocal states, thus providing application in dysphonia problems. Additionally, the sensor could distinguish between distinct natural sounds by sensing air-pressure waves created by sound transmission.

A flexible pressure sensor using MXene-coated tissue paper (MTP) sandwiched between a polyimide encapsulating layer and a printing paper with interdigital electrodes was demonstrated by Yang et al. [168]. The MTP pressure sensor showed an ultrahigh sensitivity of 509.5 kPa^{-1}, a low limit (1 Pa), and a broad range (0.5 to 100 kPa) of detection, with outstanding stability over 10k loading/unloading cycles. The MTP sensor can be used in human health monitoring and for diagnosis/prevention of diseases, such as early-stage Parkinson's disease. If positioned on the throat, the device distinguished different acoustic signals and different types of words. It can be used to prevent cardiovascular disease thanks to pulse rate monitoring combined with an arterial pulse. The flexible pressure sensor can also be utilized to create a remote respiration monitoring system that can wirelessly detect different respiratory conditions and abnormalities (Fig. 4b).

3.4. Piezocapacitive sensors

Capacitive sensors can detect the change in the distance between the two electrodes upon application of a squeezing pressure, thanks to the capacitance variation. These sensors are characterized by the ability to sense also static pressures, differently from piezoelectric and triboelectric sensors; however, they necessitate an external power source. Numerous are examples of natural materials useful to develop sensors able to sense strains and pressure thanks to a capacitive mechanism [169].

The development of capacitive e-skin pressure sensors is of particular interest as they are characterized by a facile fabrication approach, low detection limit and high sensitivity. Elsayes' group [28] employed a water-soluble material obtained from freeze-dried rose petals to obtain a sustainable and biodegradable sensor for monitoring pressure. The sensor was employed to detect finger gestures when positioned on a glove.

An innovative solution to exploit nature-derived material is to use vegetables as a scaffold to develop the sensors. This approach was employed by Zarei and coworkers [29] as they used a dried leaf as a substrate to deposit single-walled CNT and silver wires for electrodes. Then, they sandwiched a dried leaf in the middle of two electrodes obtaining the capacitive sensor. It is flexible and thin and can be attached conformally to the skin to measure pressures. Its hierarchical structure allows to sense a wide range of pressures, namely 0.01–97 kPa. It was employed to sense both subtle muscular movements and also larger movements, spanning from blink, frown and finger movement up to walking and knee bending. The sensor also showed biodegradability and breathability on the skin (Fig. 5a).

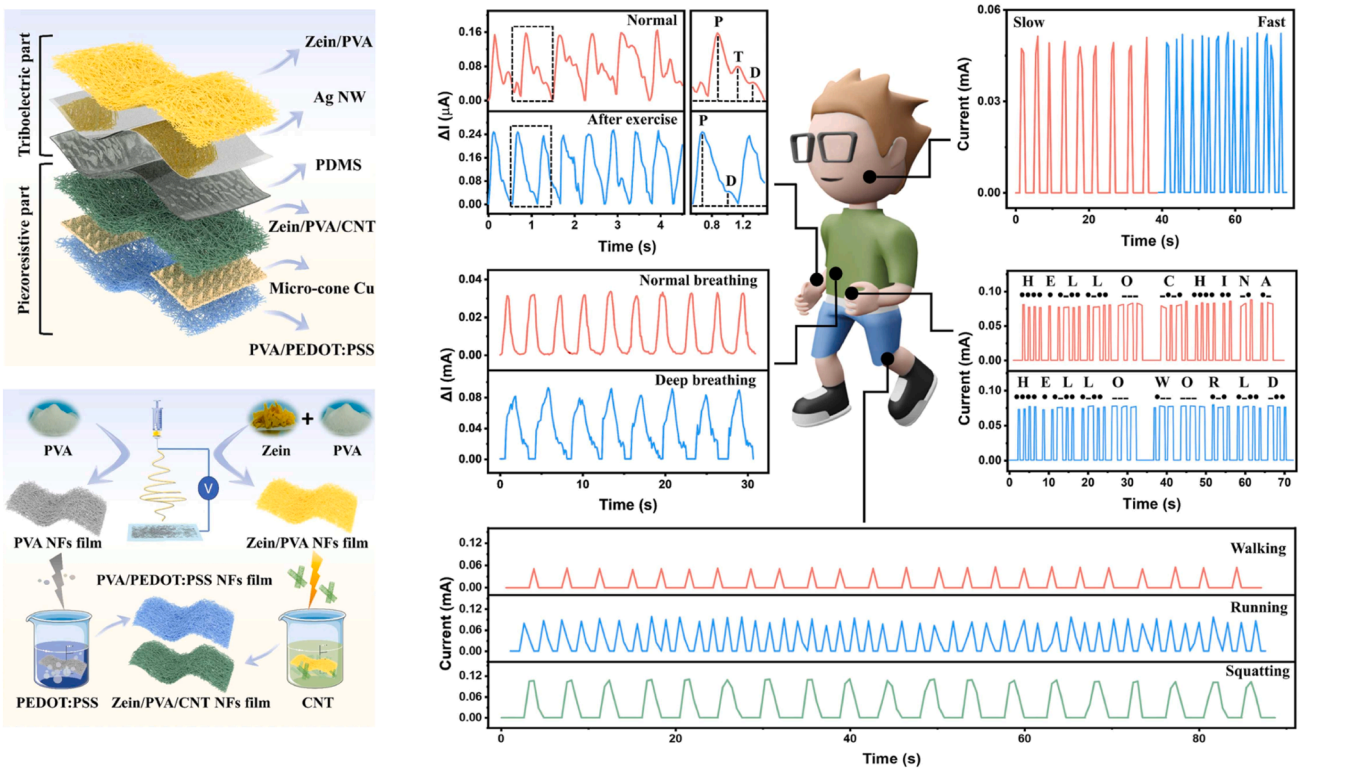
Another example of a piezocapacitive sensor was developed by Khalid's group [171] by employing degradable PLGA and polycaprolactone (PCL) nanofibrous membranes. Thanks to their high sensitivity at low-pressure ranges, they can be employed to record the arterial pulse on the wrist as well as a touch sensor to map different pressure points.

Gelatine, an animal-derived source, can be thermally modulated in a coil-helix structure to obtain a strong structure. Qin's group [172] employed this material and this crosslinking system together with choline chloride and ethylene glycol to obtain a flexible, stable and transparent hydrogel whose capacitance can be employed to sense pressure and strain stimuli. Indeed, the soft hydrogel can be applied conformally on the skin and follow curved surfaces to detect finger joint movements and to localize pressure touches on an array.

The sensitivity to capacitance that pig gelatine-derived materials showed was also exploited by Peng's group [173] to develop a soft sensor based on an ionic gelatin membrane. The changes in capacitance were tested by applying different pressure on the developed device and its sensitivity was tested by placing the same on different body joints such as finger, wrist, elbow, and knee.

To follow the principles of sustainability, the fabrication process applied has the same importance as the material chosen. Kanaparthi's group [170] developed also this concept when designing and developing an innovative capacitive pressure sensor based on graphite and cellulose. They designed a cleanroom-free and solvent-free approach by simply drawing with a pencil an interdigitated structure on paper. The sensor responded to different applied pressures and also to different strains by changing its capacitance (Fig. 5b).

Fangyi's group [174] reported a facile method to fabricate a high sensitivity, large detection range bimodal capacitive fiber sensor



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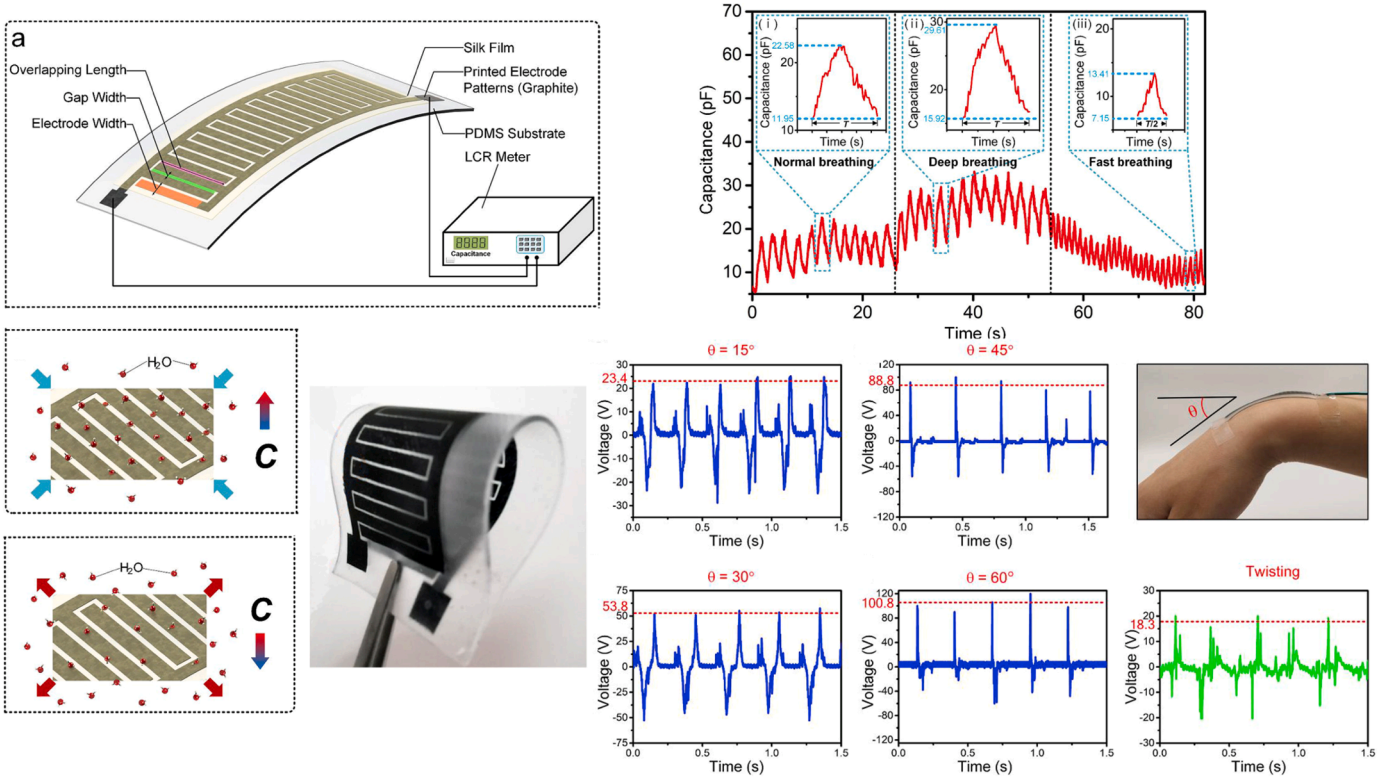

(b)

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Fig. 6. (a) Schematic diagram of the piezoresistive-triboelectric hybrid electronic device (PTHD) showing its structure (below is also reported the fabrication process of the PTHD). The picture shows also the detection of some human activities when the PTHD is attached to the skin in different body positions, such as wrist pulse before and after exercise, belly breathing during normal or deep breathing, and further movements including walking, running, and squatting [108]. (b) Measurement setup of the proposed wearable PS-TENG used for detecting water molecules (below is also reported the absorption and desorption mechanism of water molecules respectively and a picture of the fabricated sensors showing its flexible structure). On the left, the plots show the ability of the sensor to monitor human breathing, including normal, deep, and fast states, and to recognize the motion of human joints, such as bending and twisting of wrist joints [177].

assembled by integrating AgNWs/BC electrodes with coaxial PDMS dielectric. The fiber-based sensor, operating in touch mode, displayed a monotonic capacitance increase in the pressure range between 0 to 460 kPa. Moreover, in a lower pressure region until 0.5 kPa, a linear response with a high sensitivity of 5.49 kPa^{-1} was found. The capacitive fiber was embedded into a choker sensor and then attached to a necklace. The chocker sensor was used to detect human voice and recognize the radial artery pulse waveform and heartbeat rate, confirming the excellent sensitivity and resolution of the capacitive sensor. In touchless mode, the sensor is highly sensitive to objects up to 30 cm distance and, as proof of this, they developed a touchless piano played without touching the keys.

3.5. Hybrid sensors

Great performances can be reached by sensors developed with nature-derived materials when different sensing mechanisms are combined. This is the case of the work from Wei's group [175], who fabricated a strain-sensitive material based on a mix of SAlg, derived from algae, and PAA into a chelate of calcium ions with CNT. The obtained hydrogel is able to detect strains by a collaborative piezoresistive and piezocapacitive effect, which helps in better discriminating between different movements. In particular, thanks to the resistive sensitivity of 6.29 gauge factor and the capacitive sensitivity of 1.25 kPa^{-1}, the developed device detected finger and knee bending and chest movements during breathing. It was also employed as a touch keyboard to discriminate between signatures of different letters. Moreover, thanks to the presence of calcium ions, which can fastly react with SAlg and PAA, the obtained hydrogel showed an interesting self-healing behavior. Another feature that characterizes this material is its shear-thinning behavior, which allows to employ it for the 3D printing of the sensor.

The necessity to develop materials able to perfectly conform to human skin and avoid breakage, pushed researchers toward the development of hydrogels with self-healing abilities, like the material developed by Jing's group [176]. They employed nanochitin with PAA coordinated by metal ions to develop a self-healing, transparent and stretchable hydrogel with mechanical flexibility and stability in a very large temperature range, as well as at sub-zero temperatures. The material was employed to develop a multifunctional sensing platform since it was able to detect different stimuli by exploiting different sensing mechanisms. The piezoresistive effect was used to determine the strain ratio and thus recording voice, finger bending and temperature changes. Capacitance measurements were employed to record pressure and droplet fall, while the triboelectric effect was employed to power small LED and as BEH.

Wang's group [51] developed a soft and stretchable 3D hydrogel characterized by a mix of polyvinyl alcohol, SAlg, BC, modified CNT and carbon black, overall fabricated by a facile and scalable freeze-thawing process. The developed device was able to sense strains thanks to its piezoresistive behavior, as well as to sense pressure thanks to a capacitance mechanism. The sensor can therefore recognize different types of stimuli and act as a multifunctional platform. The potentiality of the device was demonstrated by sensing finger, elbow and step movements, as well as by recognizing the different weights of objects statically placed on top of it.

By combining more materials together, it is possible not only to employ different sensing mechanisms but also to sense different types of parameters. This is the case of Wen's group [177] who employed PDMS and graphite to develop a triboelectric strain sensor and a layer of SF to obtain a humidity sensor based on the change in capacitance. In this way, the contribution of the different parameters could be easily distinguished. Moreover, the fabrication approach based on screen-printing is highly scalable and allows to mass produce flexible sensors. The humidity-sensitive part was employed to measure human breathing, an important parameter to define health status, while the triboelectric mechanism was employed to monitor the movements of human joints such as wrist bending and twisting (Fig. 6b).

Another example of a nanodevice based on a mixed sensing mechanism was presented by Sun's group [178], They developed a composite material by mixing gelatin with PAM and PEDOT: PSS to obtain a hydrogel that can be stretched up to 300% and that possesses conductivity features, with high sensitivity at both small and large strains. It worked with piezoresistivity when used to sense stretching; its performances were proved by sensing facial expressions, finger movements, wrist, elbow and knee bending during walking, running, jumping and squatting. On the other side, triboelectricity was employed to harvest energy to power capacitors and LEDs.

Gelatin, a molecule originating from animals, can be crosslinked with NaCl in a glycerol/water solvent to obtain an organo-hydrogel able to sense strains and human movements. Wu's group [179] developed this sensing material with numerous features like stretching resistance up to 300% of its initial length, transparency, high conductivity, freezing resistance, self-healing and long-term stability. The sensor, in the form of gel, was placed on different body parts and it was able to exploit a double mechanism: Piezoresistivity was used to detect movements like finger, elbow and wrist bending, as well as subtle movements like swallowing, smiling and pouting and monitoring physical exercise, while the triboelectric effect allowed to employ the device as a BEH for a self-powered calculator.

Cellulose showed numerous advantages when compared to other molecules of natural origin and allows to develop multifunctional platform. As an example, Qian's group [180] developed a transparent organo-hydrogel based on cellulose crosslinked in a glycerol/NaCl/-water with dehydration and cooling tolerance. The material was employed to develop triboelectric BEH. Moreover, the resistance changes were employed to sense the strains generated by finger, wrist, and elbow movements. Also, it was possible to exploit its piezoelectric behavior to develop pressure-sensitive sensors able to respond to finger tapping.

Sustainable development was also one of the main objectives of Zheng's group [108], which developed a smart material using zein and PVA. The developed device showed the ability to sense a very large range of pressures and to monitor movements from tiny to large scales thanks to a combined triboelectric and resistive effect. In this way, it was possible to sense small movements such as wrist pulse, belly breathing, cheek muscle during chewing and finger-tapping and likewise strong movements like walking, running and squatting. Furthermore, it was used as a smart switch to activate different mechanisms in a smart house before being easily degraded (Fig. 6a).

This study [176] discusses the fabrication of a hydrogel composed of carboxyl-functionalized chitin nanofibers bound by hydrogen bonds with polyacrylic acid (PAA/NCT). The hydrogel showed a high sensitivity (gauge factor GF=2.69) and good stability. The strain sensor developed was attached to the throat of a volunteer to detect acoustic sensing capabilities and the results showed that the sensor displayed two distinct peaks when the volunteer said "hello". Next, various human

movements such as knee flexion, hand gestures and finger bending were detected and all generated clear signals. Pressure sensing performance in the presence of an external force was also evaluated, and the developed capacitive sensor showed good sensitivity (0.063 N^{-1}) to the subtle pressure of a drop of water. In addition, the PAA/NCT hydrogel was assembled into a TENG, which can act as an energy accumulator with a power density of 1.06 W/m^2.

Huang's group [48] demonstrated that, by matching appropriate materials, it was possible to obtain an enhanced tribo-piezoelectric effect. Silk serves exactly for this scope when coupled to PVDF and allowed to obtain a more efficient charge harvesting, considering that PVDF needs to be polarized in a specific direction. In this way, they developed a flexible device able to monitor different movements like finger bending and the heartbeat of a mouse and, at the same time, harvest energy from these movements.

Silk is an attractive piezoelectric natural material due to its softness and piezoelectric efficiency. In the work reported in [181], engineered silk was used to produce a hydrogel enriched with zinc oxide (ZnO) nanorods and encapsulated within two thin silk membranes, in order to enhance the overall piezo-triboelectric efficiency and to obtain soft, stretchable, flexible, biocompatible and biodegradable devices for sensing human movements. The device can be attached to different body parts and, thanks to its stretchability can be employed during bending, pressing, stretching and twisting. To demonstrate its performance, it was employed to monitor elbow movements, muscle movements and respiration rate.

Ocean-derived materials represent an attractive and sustainable solution to develop functional materials. This is the case of alginate which originates from algae. Alluri's group [182] employed this material, combined with $BaTiO_3$ nanoparticles, to obtain beads by ionotropic gelation which are then blended into a PDMS slab. This composite material showed a mixed tribo- and piezo-electric effect. It was then attached to the forearm and employed to test the forearm twisting, bending and the movements of each singular finger.

Shi's group [183] employed a 3D cellulose structure to embed $BaTiO_3$ piezoelectric nanoparticles that, together with PDMS, share one electrode to realize a mixed piezo- and tribo-electric sensor and BEH. The sensor was tested with different applied pressure ranges and with finger tapping.

To reach the goal of a device entirely made of nature-derived materials, Pongampai's group [184] improved the performance of chitosan by mixing it with $BaTiO_3$ nanorod and coupling it with cellulose and CNT electrode to develop a piezo-triboelectric device. Furthermore, it was employed to measure pressure and to charge LED.

The fabric employed as a substrate to develop a wearable sensor was used by Guo's group [185]. They covered a common fabric with electrospinned SF and PVDF to obtain a piezo-enhanced triboelectric sensor. Its potential is represented by the possibility to scale this fabrication approach with different sizes of fabric to develop a smart textile. The device was self-powered and was employed to sense body movements such as elbow bending or arm friction and swing. They proposed to employ the device as a fall alert sensor for elderly people and persons working in highly-risk areas.

It is interesting to find sensing materials in unexpected natural systems. An example is represented by the inner shell membrane of eggs since Saquib's group [186] demonstrated its sensing ability according to three different mechanisms: Capacitive, piezoelectric and triboelectric. The triboelectricity allowed to harvest energy, the piezoelectricity operated in dynamic sensing, while the capacitance was adopted in both static and dynamic sensing. The sensor obtained is foldable, flexible and disposable and was tested with a moving door for piezoelectric sensing and with wind and mouth blowing for capacitive measurements.

4. Body energy harvesters

4.1. Piezoelectric body energy harvesters

PLGA, derived from PLA, mixed with $BaTiO_3$ nanoparticles was employed by Selvarajan'group [187] to develop a biodegradable material and thus a flexible device able to recover energy from low frequencies underwater vibrations. The device was safely implanted and naturally biodegraded in the body, enabling the possibility to safely power implants.

Hanani's group [188] developed a biocompatible material with lead-free molecules able to show piezoelectric performances without the need for a poling process. In particular, the authors developed $H_2(Zr_{0.1}Ti_{0.9})_3O_7$ (HZTO) nanoparticles functionalized with polydopamine and embedded in a PLA matrix, obtaining a flexible piezoelectric material able to generate current and to light up LED from gentle finger tapping and hand slapping.

An interesting work involving another PLA-based material was presented by Wu's group [189] who employed a biodegradable piezoelectric material composed of potassium sodium niobate (KNN) nanowires, (PLLA) and PHBV. They fabricated a completely biodegradable device for the electrical stimulation of cells for nerve tissue repair that was implanted without the need for a second removal operation. The device was remotely powered by ultrasound as a safe energy-transfer method.

Wang's group [190] developed an elastomeric material totally based on biodegradable biomolecules, in particular Lactate, 1,4-BDO, and SA monomers, that undergoes a green product cycle for its whole life. The interchain penetration of the chosen molecules allowed to obtain a flexible and elastic structure that enabled an efficient displacement and therefore a high energy generation. The authors employed this material to develop a device that was tested under repeated pressure and proposed as a next-generation degradable wearable BEH device.

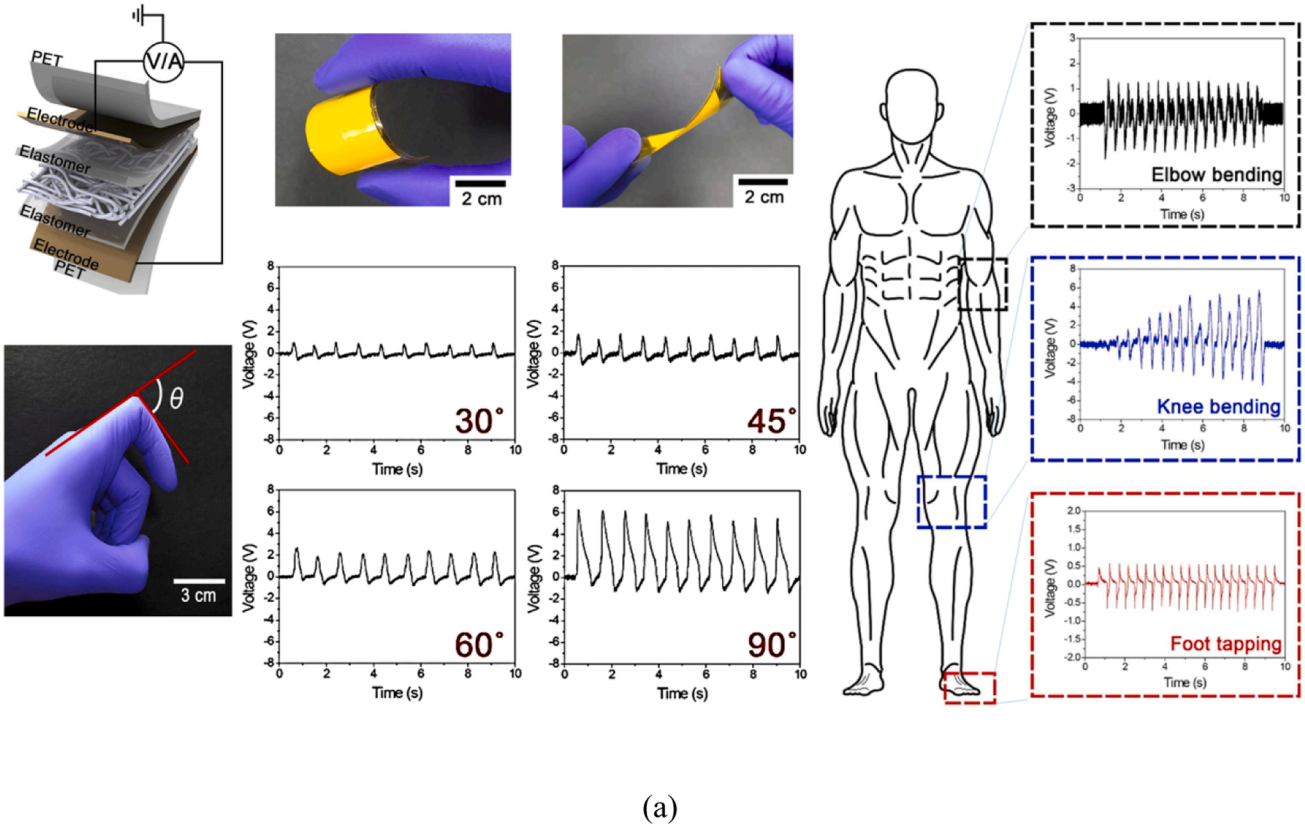
Another molecular-building approach was employed by Wang's group [118] to develop a flexible piezoelectric material. They combined the piezoelectricity of the lactate molecules with stretchable chains of elastomer, in particular BDO and SA as flexible chains and IA as a cross-linking agent. This allowed to obtain a device that showed enhanced piezoelectric behavior as a consequence of its large deformation and self-healing that increases the life of the device and enables its use in strong stress conditions.

An interesting example of direct nature-derived material was presented by Karan'sgroup [110] where the eggshell membrane is extracted from eggs and simply washed, dried and used as a piezoelectric BEH. The flexibility and the performance of the material allow the development of a device to harvest energy from tiny movements like throat coughing or swallowing, voice and arterial pulse, and to power up multiple LEDs.

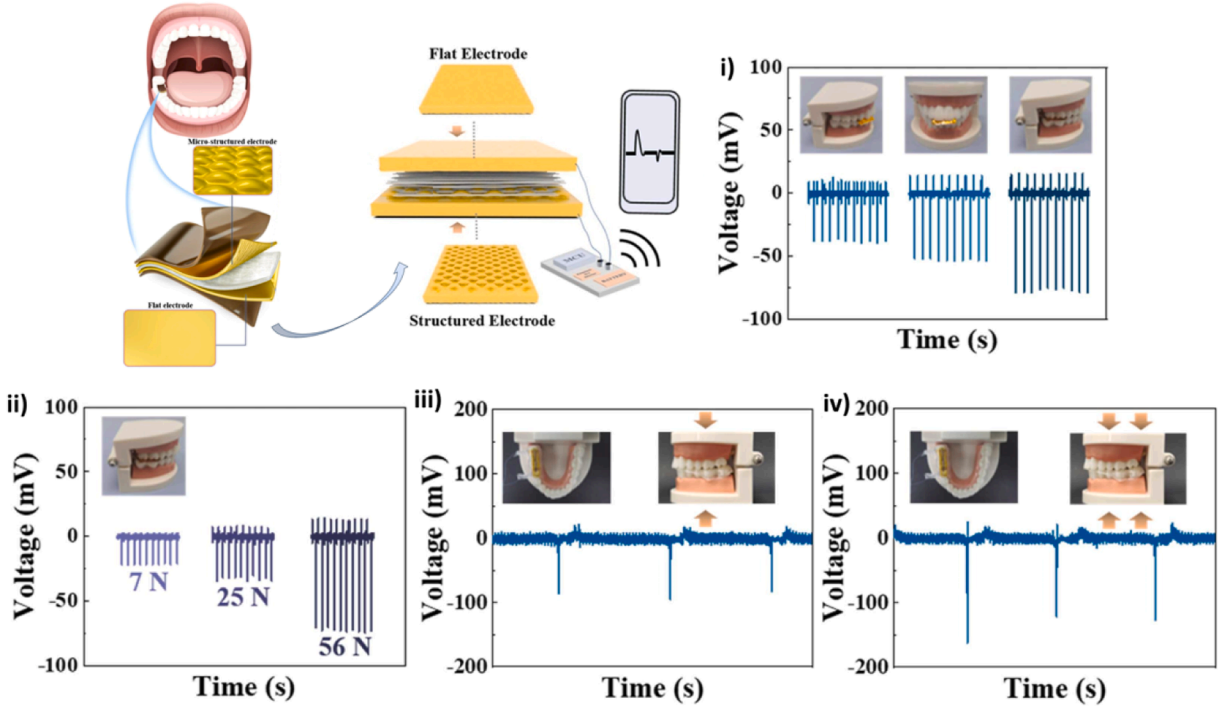
Eggshell membranes are known to be intrinsically piezoelectric, but they can also be employed in combination with another synthetic material, like PVDF. The surface interactions that occur between these two materials are responsible for an enhancement of the piezoelectric output, as it was demonstrated by Badatya's group [191]. The authors realized a flexible device by sandwiching these two piezoelectric materials between two ITO-coated poly(ethylene terephthalate) (PET) layers to harvest energy from body movements.

Interestingly, another common biowaste was demonstrated to show interesting performances as BEH, and in particular onion skin was directly extracted from the vegetables by Maiti's group [100] and combined with gold electrodes, polypropylene foil and PDMS to develop a very sensitive piezoelectric BEH, able to sense a broad range of stimuli. As an example, they exploited footsteps to power up LED.

A further example of as-used nature biowaste was reported by Kar's group [109] who presented a piezoelectric BEH based on chicken feathers. This material indeed shows a piezoelectric behavior due to the presence of keratine, while disulfide bonds are responsible for mechanical strength, resulting in a durable and efficient material. It was proposed as a lightweight and flexible wearable piezoelectric generator



(a)



(b)

Fig. 7. (a) Schematic illustration and fabricated flexible electrospun silk piezoelectric generator device packaged with Kapton film. On the left, the detection of some bending angle of a finger (from 30° to 90°) and the generated voltage signals corresponding to the detection of the movements of three different body parts (elbow, knee, and foot, respectively) [198]; (b) Schematic illustration of the OSFM-SE pressure sensor with its structure showing the flat and the structured electrodes. The picture shows also the output voltage of OSFM-SE pressure sensor when attached to incisors and molar at different forces (small and large occlusal forces, respectively) [34].

able to harvest energy from body movements, therefore it was tested with gentle finger impacts, wrist pulses and to light up LEDs.

The idea to develop a smart and soft material led Vivekananthan's group [192] to the development of a combination of cotton fabric and smart collagen nanofibril in a film that showed an interesting piezoelectric behavior. The obtained piezoelectric device guaranteed a comfortable wearability measured under different pressures.

The work of Sakar's group [99] presents one of the stronger existing cellulose fibers coming from Sonchus asper, also famous for its medical properties like diuretic, refrigerant, sedative, and antiseptic properties. These fibers become piezoelectric after stress application and thus were employed together with PDMS to develop piezoelectric BEHs.

Another example of as-used natural materials is presented by Shao's group [193]. The authors found that the Ginkgo biloba, one of the most ancient trees, can grow leaves whose cellulose crystals are already aligned and oriented displaying a marked piezoelectric behavior. Therefore, they extracted and dried yellow leaves and employed them to develop a piezoelectric energy harvester. They measured the ability of the device to sense both strong and subtle body movements to harvest energy from different actions and proposed to apply the device on different body areas during sports to recover energy from several movements to power up implantable devices.

Ba et al. [194] combined cellulose nanofibrils and $BaTiO_3$ nanoparticles to develop a composite material by a scalable and easy fabrication approach that involves roll coating and screen printing to obtain a flexible piezoelectric film that was used to harvest energy from body movements such as footsteps and hand bending.

To avoid any environmental toxic compound, Sun's group [32] developed a biodegradable BEH based on a wood sponge obtained by an innovative treatment. Due to the performance of the device and its mechanical durability, it was employed to harvest energy from walks when incorporated into the floor as a large-area device. It can also be positioned in different places of the home, such as the desk, to recover energy from finger tapping and hand movements.

Crab shells, a common biowaste material in the food industry, were employed by Hoque and coworkers [50] to extract chitin nanofibers that, combined with PVDF, result in a piezoelectric and biocompatible material with interesting features like flexibility, low costs, and lightweight. The device fabricated was tested with finger tapping and during sonication to demonstrate its ability to harvest energy from different frequencies. The device was also tested to lighten up LEDs.

Similarly, chitosan is one of the most abundant polysaccharides with piezoelectric properties on Earth and different approaches were applied to improve its properties. Li's group [195] electrospinned chitosan nanofibers and red-emission quantum dots to obtain a piezoelectric nanospun membrane. The presence of the quantum dots increased the organization of chitosan fibers both at macroscopic and microscopic levels and allowed to increase its piezoelectric performance while retaining the flexibility of the material. Moreover, the presence of quantum dots enlarged the potential to employ such material to develop optoelectronic devices. The BEH fabricated was employed to harvest energy from everyday movements, such as chewing, finger and knee bending and heel steps.

Another approach is followed by Lee's group [73] who exploited a bottom-up approach to develop a microscopically finely organized device by self-assembling FF nanotubes. The material obtained is a highly aligned piezoelectric membrane employed to develop a device that harvests energy from finger pressing. The fabrication approach is scalable and was adapted to develop also large-scale harvester recovering energy from body movements.

Another solution to employ FF in the form of microrods was presented by Tao and coworkers [196] who grew the rods in strong conditions on a hard surface and then transferred them on a flexible piezoelectric PLA surface obtaining a free-standing and flexible material to develop wearable piezoelectric BEHs. The device can be employed to scavenge energy from pressures.

Furthermore, FF showed interesting piezoelectric properties when developed in nanowires, overcoming the performance of most common ceramic piezoelectric materials. Jenkins's group [197] demonstrated both by simulation and by device fabrication the possibility of employing this biodegradable peptide-based material to fabricate a flexible piezoelectric BEH. It was tested with periodic force exerted onto the device.

Electrospun nanofiber membranes were produced by Sohn's group [198] employing silk as animal-derived biomaterial. This protein-based material showed improved piezoelectric properties when molecular chains are aligned by electrospinning. Moreover, the authors better clarify the mechanisms behind the piezoelectricity of silk by demonstrating an improvement in performance after soaking the material in alcohol. The obtained device is employed to harvest energy from knee, elbow and finger bending and from footsteps (Fig. 7a).

Liu's group [34] developed a device based on SF specifically for oral health applications. It can sense and record mechanical movements originating from the mouth and it was also employed as a BEH to power oral implants (Fig. 7b).

Maity and coworkers [199] demonstrated that natural components can be employed to enhance the properties of synthetic molecules. In particular, they used natural sugar or sucrose to coat piezoelectric PVDF fibers. This approach allowed not only to stabilize the structure by preserving flexibility, but also the surface interactions with the sugar molecules helped to align PVDF in its β-phase and thus enhance the piezoelectric output. The device developed harvested energy from vibrations, such as air blowing, vibrating belt and several musical instruments playing.

Das et al. [112] proposed another example of the enhancement of the properties of synthetic materials by using nature-derived substances. In particular, they employed high crystallinity volcano clay nanoparticles and ball mill incorporated into PVDF thin film to develop two types of portable, cost-effective, biocompatible, eco-friendly piezoelectric BEHs. The devices were tested during jogging and by applying them on a vortex machine to demonstrate the ability of the devices to harvest energy from body movements.

Lee's group [200] employed a new paradigm to align biological molecules. They employed bacteriophage M13 as a rod-shaped piezoelectric entity and aligned them vertically by using a mixed genetic engineering and template-assisted method. The authors aligned the nanorod on a soft polyethylene naphthalate (PEN) substrate to develop a flexible BEH that can be worn and can recover enough energy by finger pressing to power up a small screen.

4.2. Triboelectric body energy harvesters

TENG provides an innovative solution by supplying energy from high entropy sources. This new class of flexible devices is characterized by unlimited applications not only in harvesting energy from human body movements but also when employed as a sensing element, exponentially increasing the potential of their applications [201–204]. Moreover, fabricating triboelectric BEHs with nature-derived material is a new trend able to boost their sustainability by reducing the environmental footprint [97,205–207]. As a further example, Zheng et al. [208] reported an environmentally friendly and multi-functional wheat starch TENG (S-TENG) for wearable electronic devices. S-TENG was used not only to drive and intelligently control electronic equipment, but also to effectively harvest energy from body movements and wind. In addition, the output of S-TENG was not negatively affected by the increase in environmental humidity but increased abnormally. In the range of 20% relative humidity (RH) to 80% RH, S-TENG can be potentially used as a sensitive self-powered humidity sensor.

Silk-based materials find applications also for triboelectric BEHs. Wang et al. [209] designed an ultra-robust and natural silkworm cocoon layer (SCL)/ PDMS–TENG as a BEH to scavenge waste energy from human motions. The electrical output of the SCL/PDMS–TENG is 126 V

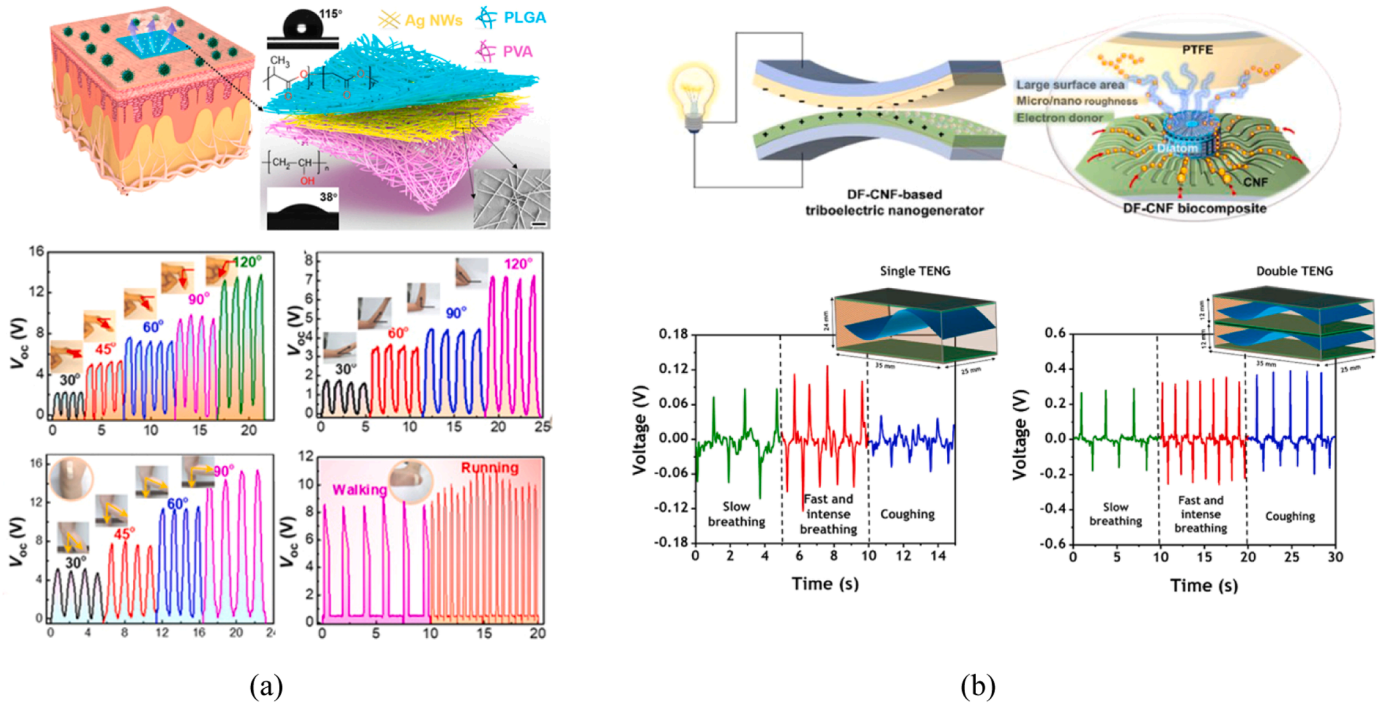


Fig. 8. (a) Breathable, biodegradable, and antibacterial e-skin conformally attached to the epidermis and schematic illustration of its three-dimensional structure [221]. (b) Schematic illustration of DF–CNF composite and the signal generated by the (on left) single and (on right) double DF–CNF biocompatible TENG during human respiration at different rates and coughing [217].

as open-circuit voltage, 3 μA as short-circuit current and 216 mW/m^2 as power density. The integrated self-charging TENG was demonstrated to power small electronics and monitor human motions.

Dudem et al. [210] developed a soft fully packaged TENG device based on crystalline silk microparticles (SMPs) embedded into a polyvinyl alcohol film and PTFE plastic cups. The TENG device exhibited maximum open-circuit voltage V_{OC}, short-circuit current I_{SC}, and short-circuit charges Q_{SC} values of ~280 V, 17.3 μA and 32.5 nC, respectively, as compared to the TENG with pure PVA (i.e., ~76 V, 3 μA, and 8.3 nC). Such a device revealed a maximum effective electrical output power density of ~14.4 W·m^{-2} at a load resistance of 40 MΩ and stability even after 10k cycles over several weeks. The packaging with silicone rubber allowed the protection of the device from humidity and to attain durable electrical performance in even harsher environments to obtain a soft, comfortable-to-wear, and skin-friendly device.

Xu et al. [211] reported a flexible, stretchable and fully bioabsorbable TENG based on doped silk film SF to harvest biomechanical energy in vitro or in vivo for intelligent wireless communication. The SF-TENG, which employed PTFE as paired material, offers significant advantages over conventional generators in terms of bending resistance, skin affinity and self-powered efficiency.

A substrate-free self-powered e-tattoo sensor that provided on-skin power harvesting characteristics and tactile information was proposed by Gogurla et al. [212]. The ultrathin device consists of electro-spun SNF layers incorporating CNTs. The device can activate electronic devices to monitor whole-body joint movements.

Further, Dudem et al. [213] reported a natural eco-friendly TENG based on micro-architected silkworm fibroin films characterized by high surface roughness and resistance to aqueous solubility obtained by means of soft imprinting lithography and alcohol annealing treatment, respectively. The micro-architected silkworm fibroin-based device exhibited very high surface charge density, I_{SC}, V_{OC}, and power density values of 101μC/m^2, 89 mA/m^2, 395 V, and 22.05 W/m^2, respectively, as compared to the flat TENG (i.e., 78 μC/m^2, 40 mA/m^2, 230 V, and 15.13 W/m^2). The TENG device was employed for real-time human body sensing applications.

Kim et al. [103] reported a highly stretchable and self-healable hydrogel conductor mixed with ocean biomaterials including catechol, chitosan and diatom to realize a TENG for energy harvesting from human motion and self-powered skin-attachable tremor sensors for monitoring the health condition of Parkinson's disease patients. The device showed an instantaneous power density of 29.8 mW/m^2.

The group of Kim [102] developed a chitosan-diatom TENG for skin-attachable wearable devices. The porous diatom bio-silica can be used as a biocompatible additive to greatly change the electro-positivity and surface properties of chitosan films, showing bio-affinity and much higher output power paired with fluorinated ethylene propylene (FEP) film for vertical contact-separation. The time-averaged power density of the chitosan-diatom TENG is measured as 15.7 mW/m^2, which is 3.7 times higher than that of the pure chitosan TENG. For practical applications, the electrode-encapsulated chitosan-diatom film was successfully applied for a self-powered watch and a skin-attachable motion sensor.

Zheng et al. [214] prepared a transparent TENG by pairing an eco-friendly, flexible, and transparent chitosan/starch composite films by drop casting process, with high transmittance exceeding 88% in the visible light range, with the FEP film, and transparent PET/ITO electrodes. The optimized chitosan/starch-FEP-based TENG exhibited a maximum power density of ~5.07 Wm^{-2} and an ultrahigh sensitivity of 46.03 VkPa^{-1} in the pressure range of 1.25–6.25 kPa achieved by the device as a self-powered force sensor.

Bai et al. [215] proposed a TENG based on a biocomposite consisting of porous cellulose acetate and polyethyleneimine (PEI) paired with flexible low-temperature vulcanized silicone rubber. The developed TENG presented outstanding electrical output stability and durability, and it is effectively capable of responding to multiple mechanical stimuli originating from the human hand, including holding, twisting, jabbing, flapping, and bending. Furthermore, the sensing pocket was applied in different positions on the human body to monitor various movements that come from arms and feet in real-time.

Another type of porous nanocomposite was developed by Bai's group [216] who proposed a wearable TENG based on a material that incorporates nano-Al_2O_3 fillers into cellulose acetate networks. Several concentrations of casting solution and content of nano-Al_2O_3 fillers were tested to improve triboelectric charge yield, paired with low-temperature vulcanized silicon rubber. The optimized TENG can deliver an electrical performance of ~2.5 mW/cm^2 on a 0.8 MΩ external resistor and it was capable of charging various capacitors, lighting LED arrays, and driving commercial wristwatches. Furthermore, a TENG-based elbow supporter and a grip ball, as self-powered sensors, were proposed to realize real-time detection of human actions during sports exercise.

Rajabi-Abhari et al. [217] reported a work in which diatom bio-silica was used as a biomaterial to enhance the output performance of CNF-based TENGs. The device employed tribopositive bio-silica having hierarchically porous three-dimensional structures and high surface area into CNF layer to obtain a composite film mechanically strong, electron-rich, low-cost, and frictionally rough. Moreover, a practical application of the BEH was examined with a self-powered smart mask for human breathing monitoring (Fig. 8b).

An example of the employment of cellulose nanocrystals (CNC) was presented by Wang [133] who developed a TENG based on cellulose nanocrystal CNC nanofibers loaded into methylcellulose (MC) films, extracted from recycled wasted papers. The obtained composite was a flexible, transparent and water-soluble CNC/MC film as the positive-triboelectric materials that can be completely degraded in the natural environment. The CNC/MC films showed better positive triboelectricity than metal films when in contact with PTFE film as a negative-triboelectric material. Moreover, a completely water-soluble device with open-circuit voltage and short-circuit current of 2 V and 30 nA was prepared by replacing PTFE film with MC film as a negative triboelectric layer obtaining a biodegradable wearable bandage sensor able to precisely distinguish the various breathing states (slight, normal, deep, rapid breathing) and completely washed away in 2 min

Fachechi et al. [218] reported on triboelectric energy harvesters based on CMC porous aerogel films, filled by PDMS polymer. The fabricated devices with a 1 cm^2 area, tested by a novel approach for measuring key parameters in triboelectric nanogenerators, exhibited a peak-to-peak voltage of 27.8 V, a peak-to-peak current of 2.2 μA and a maximum generated power of 0.9 μW under pressing-release (5 N at 1 Hz).

Park and his group [95] proposed a TENG based on nonwoven nanofiber (NF) material comprising *Ulmus davidiana* var. *japonica* and PCL for enhancing triboelectricity paired with a Teflon layer. Moreover, this wood-derived material showed an antifungal activity against athletes' feet, which holds great promise for wearable and body-attachable human kinetic-energy harvesters. The wood-based TENGs during the energy harvesting possessed a high energy harvesting efficiency of 1600 V/N·m^2.

Jiang et al. [106] proposed TENGs based on several fully bioabsorbable natural materials for in vivo implantable devices. The polymeric components of TENGs originated from nature, include cellulose, chitin, rice paper, SF, and egg white. The device architecture consisted of two different bioabsorbable polymers acting as friction layers and ultrathin Mg films serving as the top and back electrodes with spacers for vertical contact-separation mode. Various triboelectric outputs of these natural materials were achieved by a single material and their pairwise combinations. The maximum voltage is 55 V, current 0.6 μA, and power density 21.6 mW/m^2. The modification of SF encapsulation film made the operation time of the TENG tuneable from days to weeks. Further, the authors demonstrated the full degradability and resorbability of this device in Sprague–Dawley rats, avoiding a second operation and other side effects. Using the proposed TENG as a voltage source, the beating rates of dysfunctional cardiomyocyte clusters were accelerated and the consistency of cell contraction was improved. This provided a new and valid solution to treat some heart diseases such as bradycardia and arrhythmia.

Wang et al. [219] developed BEHs based on chitosan films and natural materials such as starch, lignin, glycerol, and acetic acid. The outputs can be optimized as 13.5 V and 42 nA from chitosan-10% acetic acid composite film due to the sticky surface by adding a layer of Ecoflex silicone rubber.

The group of Khandelwal [101] proposed a TENG biodegradable and edible device based on laver coated with an edible silver leaf which served as the active layer and a rice sheet as substrate. The device output performance was tested using paper, tissue paper, polyvinyl chloride (PVC), and FEP. The best electrical performance was achieved by exploiting the FEP-laver as the opposite layer.

Zhang et al. [220] reported TENG devices as mechanical BEH and self-powered sensor devices based on cellulose II, fabricated by treating cellulose via a dissolution–regeneration process. The device architecture consists of a layer of cellulose aerogel paired with a PTFE layer. For an applied force of 40 N, V_{OC}, I_{SC}, and Q_{SC} of the cellulose II aerogel-based TENG resulted to be ≈ 65 V, 1.86 μA and ≈ 23 nC, respectively.

Peng et al. [221] designed a TENG exploiting single electrode mode by sandwiching Ag NW electrode between the top PLGA triboelectric layer and the bottom PVA substrate. The e-skin device based on the proposed material had a maximum matching peak power density of 130 mW/m^2 and a voltage response pressure sensitivity of 0.011 kPa^{-1}, which enables the whole-body physiological signal monitoring such as blinking, pulsing, speaking, and respiring, and major joint motion detections, including knuckle, elbow, knee, and ankle (Fig. 8a).

Sun's group [222] reported a fully sustainable TENG based on fish gelatin layers as friction layers that can degrade completely within 30 days in the soil. By modifying the two friction layers with dopamine and fluorinated silane respectively to serve as a promising triboelectric pair, the obtained TENG showed remarkable output performances with a power density of up to 100 μW cm^{-2}. The device was used in self-powered wearable sensing of human movements and in human-machine interaction.

An all-cellulose BEH was proposed by Zhang [223] who developed an eco-friendly, recyclable and interactive device TENG. The TENG in contact-separation mode was composed of pure BC as one friction layer and BC-CNT-polipyrrolidone membranes as an electrode and the other friction layer. The all-cellulose TENG was readily utilized as a wearable sewing interface to control an electronic piano.

Chen et al. [224] developed a TENG to replace a traditional power supply for synthesizing different metallic nanoparticles using an electrochemical approach. The TENG device consists of two polyethylene terephthalate (PET) sheets used as the substrates and covered with thin aluminum films. PDMS and gelatin, properly nanostructured covered the top of two PET/Al sheets separately. The conductive substrate is represented by carbon fibers and metallic nanoparticles. Additionally, the authors integrated the as-prepared carbon modified with the TENG to construct a wearable self-powered sensing system that exhibited significant selectivity and sensitivity toward lactate detection.

One of the most important aspects which need to be considered when developing flexible devices is that it is necessary to match the mechanical features of the skin in order to have high compliance and reliable measurements. For this reason, in the work of Leng [225] a hydrogel based on bovine serum albumin and glycerol is presented, which displays mechanical matching with the skin and is able to retain moisture and conductivity. The hydrogel can be used as well to register electrophysiological signals and can be coupled to PDMS to obtain a flexible and self-powered TENG. The device capabilities were demonstrated by developing a robot and a smartphone Bluetooth control systems.

Jao et al. [226] proposed a TENG to harvest biomechanical energy from human motions, in which a chitosan-glycerol film was explored. The TENG design consisted of an as-prepared chitosan-glycerol film, containing nanostructures on the surface, regarded as positive triboelectric material and attached to a metal layer paired with

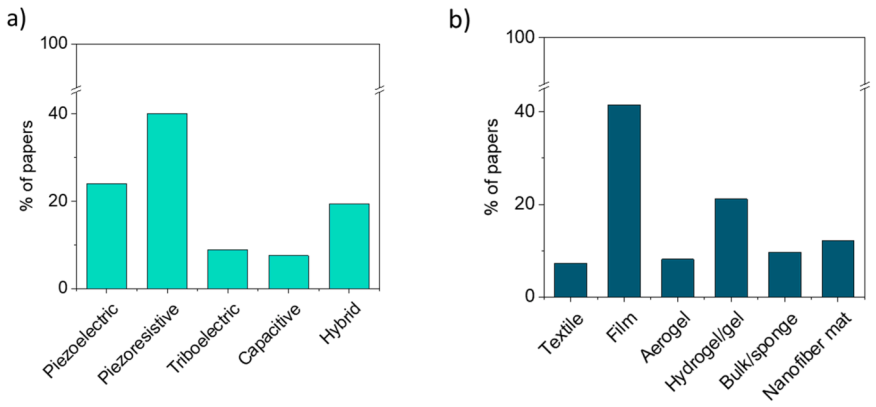


Fig. 9. (a) Distribution of sensing mechanisms. (b) Distribution of material form for sensing.

polytetrafluoroethylene (PTFE), attached to another metal layer, acted as negative triboelectric material. The output characteristics revealed stability under various humidity conditions of the developed devices, indeed TENG can reach 130 V with a contact area of 5 × 3 cm^2, peak currents of 14 μA and a maximum power above 2.5 W/m^2. Moreover, the TENG was further developed into various kinds of self-powered healthcare sensors for humidity, sweat, and gait phase detection. More importantly, the designed humidity sensor based on the TENG exhibits a promising advancement in sensitivity compared with conventional TENG-based humidity sensors.

He et al. [227] proposed a self-powered cellulose fiber-based TENG system constructing 1D CNFs on CMFs (cellulose microfibers) skeleton to form a 2D hierarchical nanostructure with high efficiency for $PM_{2.5}$ removal. The TENG design consists of a CMFs/CNFs/Ag nanostructured film paired with FEP/Ag layer. The so-obtained TENG system was capable of monitoring breathing status without using an external power supply.

Huang et al. [228] proposed an all-fiber-structured TENG fabricated via a one-pot electrospinning method that integrated ethyl cellulose/-polyamide 6 co-electrospun nanofiber-incorporated MXene ($Ti_3C_2T_X$) sheets into the PVDF nanofiber skeleton. The ethyl cellulose and polyamide 6 act as the triboelectric positive material whereas MXene sheet incorporated into poly(vinylidene fluoride) are strongly electronegative conductive nanofibers and act as a triboelectric negative material. The assembled all-fiber TENG exhibited excellent durability, stability and output performance that reached a peak power density of 290 mW/m^2 at 100 MΩ of load resistance. The TENG was capable of monitoring human movements as a self-powered sensor.

5. Implantable sensing and energy harvesting systems

The possibility of implanting self-powered devices would help to avoid the implantation of batteries, which bring intrinsic dangers inside the body. To avoid this problem while retaining a high power recovery efficiency, the group of Huang [48] developed a self-matched piezo and tribo implantable energy harvester in which the piezoelectric effect enhanced the triboelectric effect. The device is fabricated thanks to the combination of PET and PVDF with a type of recombinant spider silk. The flexibility that the device display allows is to be positioned in many locations of the body to monitor movements. One of the most important characteristics of devices developed with nature-derived material is that they are usually biodegradable at different and sometimes also controllable rates. This feature is extremely useful when applied for implantable systems as it eliminates the need for a second surgery to remove the implant. Moreover, the employment of devices able to collect energy from body movements promotes the use of self-powered autonomous systems able to work without conventional electronics. Therefore, the combination of biodegradable sensors and biodegradable BEHs paves the way for implantable systems that can closely monitor physiological parameters and increase comfort for patients. An example is given by the group of Zheng [229], who developed an implantable and biodegradable triboelectric energy harvester based on PLGA, PHBV, PVA and PCL able to power electrodes for tissue repairment in 9 weeks. The system can then degrade and be absorbed by the body in only 90 days. Another approach for system biodegradation is presented by the group of Li. [230] who developed a triboelectric nanogenerator based on PLGA, PCL and PLA for wound healing applications. The biodegradation of the device is controlled by the presence of AuNRs and triggered by NIR light. In this way, the device is able to work properly for 28 days and then, after the degradation trigger starts, is completely photo-biodegraded in 14 days. In the work of Jiang [106], a combination of Chitin, Cellulose, silk fibroin, rice paper and egg white is used to develop an implantable triboelectric nanogenerator able to help dysfunctional cardiomyocytes to recover their contraction efficiency and then degraded in maximum 84 days. This device does not require a second operation to be discarded. The importance of flexibility for implantable systems is underlined by Gogurla and coworkers [181] who presented a flexible, enzymatically degradable and implantable piezo-tribo harvester based on silk and incorporated ZnO nanorods. The flexibility of the device, comparable to that of skin, improves the comfort of the device when placed on the skin or implanted and allows to correctly and autonomously measure body movements without further power sources. It is also employed to activate low-power electric devices such as LEDs, an oximeter and stopwatches. The work of Selvarajan [187] presents a transient biodegradable piezoelectric system based on PLGA and $BatiO_3$ particles which can be activated by both low-frequency and high-frequency ultrasonic stimuli, generating up to 10 mW/cm^2. It was tested by powering LEDs and can completely biodegrade in physiological conditions after 100 days. Implantable energy-harvester systems find interesting applications also in the context of nervous tissue regeneration, as underlined in the work of Wu [189]. They developed a piezoelectric device powered by ultrasound and fabricated with KNN nanowires, PLLA and PHBV which can send electrical pulses to nerves to promote their regeneration. Moreover, they were able to tune the biodegradation time of the system by changing the encapsulation layer. Triboelectric nanogenerators can be employed as multifunctional platforms for in vivo monitoring and therapeutic delivery, as it was demonstrated by the Zhang's group [231]. They presented a biodegradable silk-based triboelectric nanogenerator to monitor epileptic seizures. The strength of this work lies in the possibility of tuning the operating life and the mechanical sensitivity by changing the molecular size of silk, the surface and the structural organization of the device.

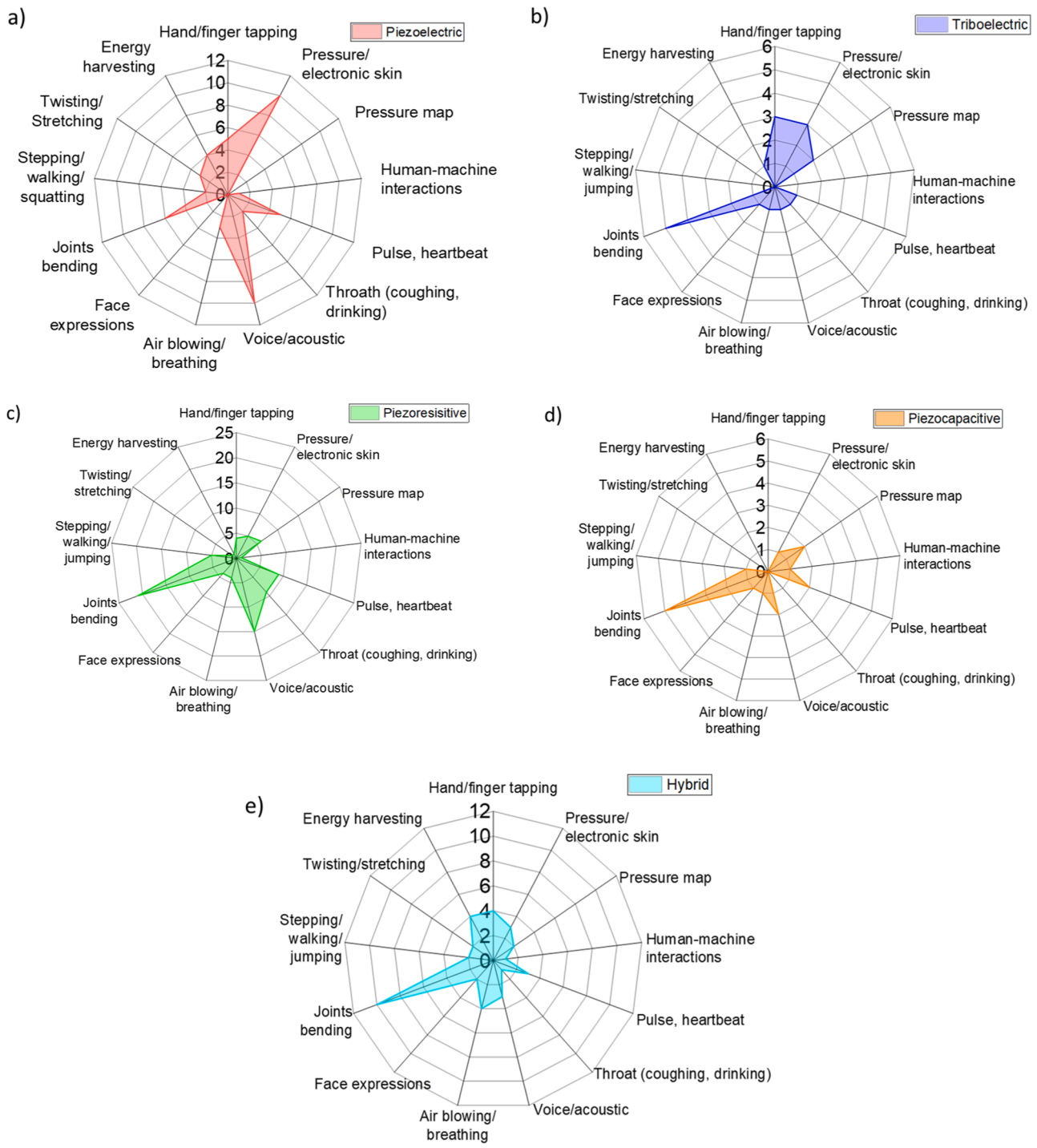


Fig. 10. Distribution of applications for each reviewed transduction mechanism. (a) Number of papers regarding piezoelectric sensors. (b) Number of papers regarding triboelectric sensors. (c) Number of papers regarding piezoresistive sensors. (d) Number of papers regarding piezocapacitive sensors. (e) Number of papers regarding hybrid sensors.

6. Comparative analysis

The employment of biomaterials derived from nature to develop wearable devices is a relatively young trend in the fabrication of innovative sensors and it is estimated that their number will grow in the future due to the necessity to supply sustainable electronic solutions and reduce the e-waste amount. Most reviews in the field of wearable sensing and power supply devices fabricated with both conventional and biological materials mostly focus on material fabrication techniques or merely list the type of devices that can be developed [232–236]. However, as far as we know, the literature misses an analysis in which the type of nature-derived material and its material form are correlated to the existing wearable applications. Therefore, in this Section, the applications of wearable devices and power supply devices are analyzed from the perspective of the material kind, the sensing mechanism and the processed form of the active material. This study will help to obtain some guidelines on how to boost the employment of nature-derived materials thanks to their unique features to develop next-generation wearable devices.

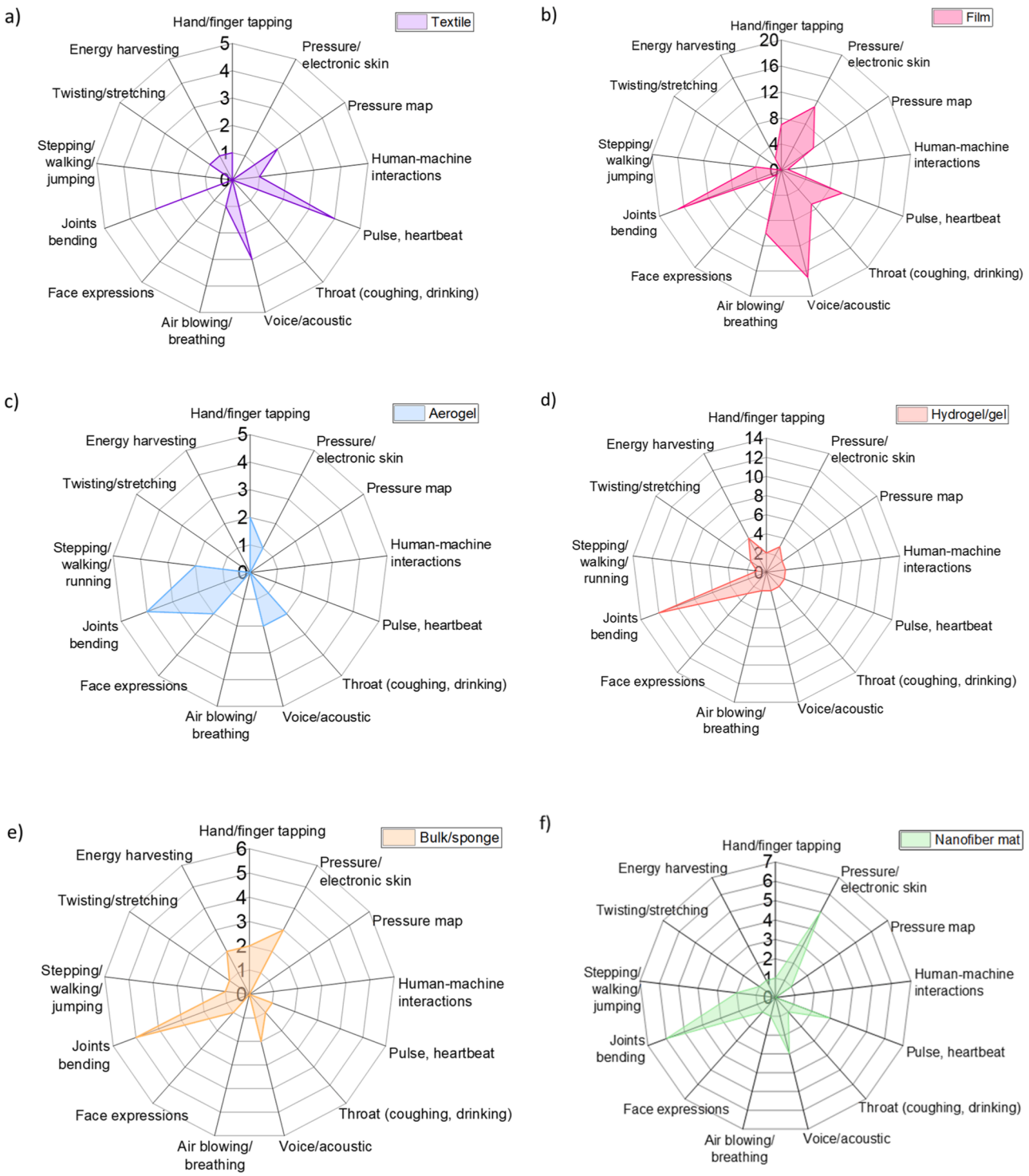


Fig. 11. Distribution of applications for each reviewed material form. (a) Number of papers regarding textile materials. (b) Number of papers regarding film materials. (c) Number of papers regarding aerogel materials. (d) Number of papers regarding hydrogel materials. (e) Number of papers regarding bulk and sponge materials. (e) Number of papers regarding nanofiber materials.

6.1. Sensors

The analysis about the number and type of sensors described in this review identifies the physical mechanisms mostly adopted to develop sensitive sensors. In particular, among the mechanisms involved (piezoelectric, triboelectric, piezoresistive, piezocapacitive and hybrid mechanisms), it emerges that piezoelectric and piezoresistive devices are the most employed to develop pressure and strain sensors based on nature-derived materials. The reason behind this is represented by the stability of the signal and the fact that piezoelectric and piezoresistive materials are less influenced by environmental contributions which can make the measurement less affordable. However, piezoresistive sensors always require a power-consuming reading platform, while piezoelectrics are characterized by energy-independent signal generation, improving the portability of the system. Also, the hybrid mechanism is relatively broadly employed due to the possibility of developing

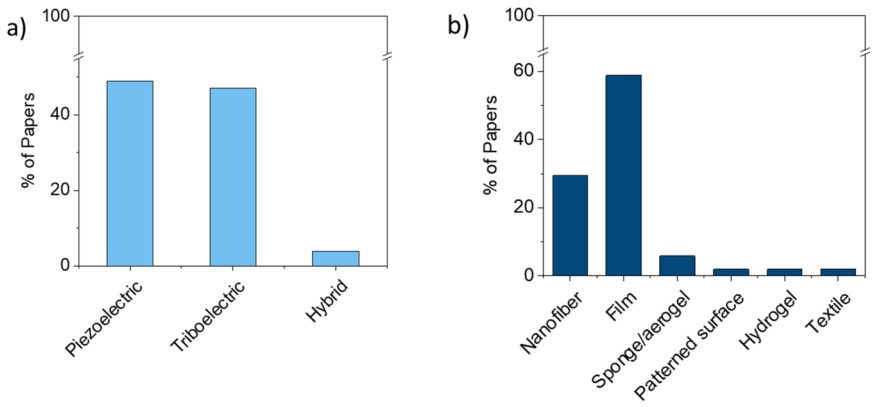


Fig. 12. (a) Distribution of BEH mechanisms. (b) Distribution of material form for BEH devices.

platforms able to monitor multiple parameters at once, thus increasing the portability and the comfort for the patient (Fig. 9a). Another important point when planning the development of an innovative device is the choice of the processed form that the material will assume to exploit as much as possible its properties. Among the papers analyzed, the most diffuse forms are textile, film, aerogel, hydrogel/gel, bulk materials/sponge and nanofiber mat. It emerges that films, produced mainly by solvent casting or spin coating techniques, are the most employed due to their low fabrication costs and very low thickness that improve conformability. Also, hydrogels and soft gels are largely applied because of their stretchability. Thirdly, we find nanofiber mats that show a hierarchical structure allowing to greatly enlarge the pressure range of stimuli recorded (Fig. 9b).

When developing an instrument for monitoring human parameters or for specific applications, it is of paramount importance to understand how to exploit the sustainable active material to understand how to improve it and to reach the best possible performance. Therefore, it is useful to have an overview of which type of sensing mechanism is mostly employed for a specific application to better drive our choice. The overview of the analysis is represented in Fig. 10. By analyzing the type of sensing mechanism for each paper, it emerges that the monitoring of joints and muscle strains is the most studied to collect data on health during sports activities or to monitor the conditions of elderly and bedridden. There is not a specific preferred mechanism, as generally triboelectric, piezoresistive, piezocapacitive and hybrid mechanisms are chosen. Instead, the analysis of the applications of piezoelectric devices underlines that they are mainly chosen for applications related to pressure monitoring and sound or voice recognition. This aspect can be influenced by the higher sensitivity that piezoelectric materials demonstrate for lower pressures and lighter stimuli like the one involved in sound emission or gentle finger pressures.

Another important part of the development of innovative devices based on nature-derived material is the form in which the material needs to be processed. In this case, both performance and compliance with different body parts play an important role. The distribution of the material forms related to the applications is displayed in Fig. 11. Textiles, thanks to their conformability and adaptability to smart dresses, are largely employed in most applications, including pressure maps, wrist pulse, heartbeat, voice or sound recognition, joint bending monitoring and tapping. Films are the most employed for developing pressure sensors, voice and acoustic recognition and joint movements, probably due to their sensitivity in recording lighter stimuli. Aerogel, hydrogel, gel and sponges, thanks to their high compressibility, are instead most employed for developing devices that can monitor large bending from finger, elbow and knee. Nanofiber mats are instead the most employed to sense a large range of stimuli due to the hierarchical structure of their form.

6.2. *Body energy harvesters*

The devices applied as BEHs analyzed in this review require a separate analysis from sensors since they are evaluated on the basis of different qualitative and quantitative parameters. In particular, the most important feature of BEHs, is the ability to generate high electric signals both for short and open-circuit measurements. Moreover, by definition, they work as power supply and need to be energetically independent. Indeed, the reviewed papers present only piezoelectric and triboelectric materials in a similar amount (Fig. 12a). Regarding the mainly employed material forms to develop efficient BEHs based on natural materials, films and textiles are the most developed. This is due to the high compliance and flexibility that these two material forms usually display. Moreover, while films are characterized by low costs of fabrication and high sensitivity, textiles can sense a wider pressure stimuli range thanks to their hierarchical structure (Fig. 12b).

7. Conclusions: challenges and perspectives

The need to develop sustainable devices based on green-chemistry materials became of paramount importance during the last decades, especially considering the growing number of wearable devices, necessary to continuously monitor human health, and the consequent amount of electronic waste produced. This class of materials brings intrinsic advantages in the development of wearable sensors and energy harvesters and it is not always expected to find the same features in conventional synthetic polymers or ceramics, such as sustainability, low costs, flexibility, biocompatibility and controlled biodegradability. Moreover, when implanted they can be absorbed by the body and be replaced by living tissues reducing rejection complications. Additionally, their large availability makes them perfect candidates for industrial scalability. Although nature-derived and sustainable biomaterials research is paving the way for a new generation of wearable devices, the road toward their optimization is still in its infancy due to some challenges that need to be faced. The first aspect to be considered is the intrinsic variability of materials extracted from raw sources that could be responsible for differences from one batch to another. Indeed, most of these molecules are complex polymers with high molecular weights and, therefore, multiple efforts are required to standardize the extraction and purification to obtain homogeneous substances suitable to be further processed for the fabrication of reproducible devices [237]. Moreover, most nature-derived materials cannot be processed with standard fabrication techniques but require mild protocols optimized specifically for their processing. In particular, these materials may be damaged when treated with high-temperature or rough solvents. The combination of low homogeneity and the necessity for mild processing conditions makes the miniaturization of the final devices challenging. It is emblematic that most of the references in this review report devices in

the size of cm or mm and only a few of them report micron-sized sensors. Despite these challenges, it is also true that these materials reveal a large potential thanks to their intrinsic characteristics. As an example, biological-assisted sorting or production could be a possible solution for improving their purification, enabled by the feature of bio-recognition or bio-production that mostly nature-derived molecules possess. Moreover, their high potential for chemical modification will help to improve their processability or, vice versa, will force us to use mild and more sustainable fabrication protocols. Furthermore, the possibility to use the same material in different forms (thin film, nanofibers, textile, hydrogel, etc.) will give a boost to the development of micron or even nano-sized sensors and energy harvesters.

Therefore, the efforts required by nature-derived materials to obtain devices with the required characteristics are counterbalanced by the advantages that would come from their use in everyday practice.

In this work, an overview of wearable pressure sensing devices and BEHs based on bio-derived active materials is presented. The devices are classified based on their mechanism (piezoelectric, triboelectric, piezoresistive, piezocapacitive and hybrid). Moreover, a comprehensive analysis of their working mechanism and their processed material form by healthcare applications is given. This review provides broad guidelines to help in defining the best material for specific wearable applications, underlining the potential of nature-derived materials and considering their important role in our near future.

CRediT authorship contribution statement

Mastronardi Vincenzo: Conceptualization, Formal analysis, Investigation, Methodology, Visualization, Writing – original draft. **de Marzo Gaia:** Conceptualization, Formal analysis, Investigation, Methodology, Visualization, Writing – original draft. **De Vittorio Massimo:** Supervision, Validation, Writing – review & editing. **Algieri Luciana:** Formal analysis, Investigation, Methodology. **Antonaci Valentina:** Formal analysis, Investigation, Writing – original draft. **Blasi Laura:** Formal analysis, Investigation, Writing – original draft. **Todaro Maria Teresa:** Formal analysis, Investigation, Writing – original draft. **Scaraggi Michele:** Validation, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Table sensors

Material	Sensing mechanism	Sensitivity	Pressure/Force range	Stretchability	Ref.
CNT into a chelate of calcium ions with PAA and SAlg	Capacitive	-	-	-	[175]
Paper-based	Capacitive	-	-	-	[138]
Gelatine, choline chloride (ChCl) and ethylene glycol (EG)	Capacitive	-	lower than 1 kPa	-	[172]
Rose petals	Capacitive	-	-	-	[28]
Ionic collagen fiber	Capacitive	-	-	-	[173]
Leaf, SWCNT and AgNWs	Capacitive	-	0.86 ± 0.16 kPa	-	[29]
PLLA Nanofibers	Piezoelectric	25 mV kPa^{-1}	-	-	[30]
Glycine/chitosan	Piezoelectric	2.82 mV kPa^{-1}	-	-	[31]
Pectine/PEG/Graphite	Piezoelectric	-	0.1–1 kPa	-	[128]
Silk/PHB/Gold Nanorods	Piezoelectric	-	recorded up to 1 N		[124]
PBGL/PDMS	Piezoelectric	10.2 (d_{33}) 54 pC N^{-1} (d_{31}), 615 mV N^{-1}	-	-	[47]
BC hydrogel/Imidazolium Perchlorate	Piezoelectric	4 mV kPa^{-1}	0.2-31.25 kPa	-	[129]
Wood Sponge	Piezoelectric	-	tested at 13.3 kPa	-	[32]
Porcine skin gelatine	Piezoelectric	41 mV Pa^{-1}, 24 pC N^{-1}	-	-	[33]
PLA	Piezoelectric	600 pC N^{-1} (d_{33}), 41 pC N^{-1} (d_{31})	-	-	[115]
PLLA-$BaTiO_3$	Piezoelectric	-	-	-	[117]
PLA	Piezoelectric	10 pC N^{-1}	-		[116]
Cellulose/PVDF/Maleic Acid Polydopamine/$BaTiO_3$	Piezoelectric	27.2 pC N^{-1}	-	-	[98]
Cellulose/MoS_2/$BaTiO_3$	Piezoelectric	45 pC N^{-1}	-	-	[52]
Inner eggshell	Piezoelectric	16.93 V MPa^{-1}	0 -0.098 MPa	-	[186]
SF-/PVDF	Piezoelectric	-	-	-	[123]
Silk/Tannin/Graphite	Piezoelectric	-	-	-	[125]
Silk/PVDF	Piezoelectric	6.24 pm V^{-1}, 73 μV Pa^{-1}	-	-	[126]
Chitosan	Piezoelectric	15.56 pCN^{-1} 80 mV kPa^{-1}	0 – 0.5 kPa		[127]
PZT/Cellulose	Piezoelectric	31 pC N^{-1}	lower than 31 kPa	-	[53]
Lactate elastomer	Piezoelectric	-	-		[118]
Strontium collagen	Piezoelectric	3.45 pm V^{-1}	-	-	[54]
SF	Piezoelectric	30.6 mV N^{-1}		-	[34]
Chitin	Piezoelectric	-	lower than 10kPa	-	[35]
PLLA	Piezoelectric	-	0-18 kPa	-	[121]
PDLA/PLLA/CQD	Piezoelectric	-	-	-	[120]

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Material	Sensing mechanism	Sensitivity	Pressure/Force range	Stretchability	Ref.
PLGA/PCL	Piezoelectric	-	0-4.6 kPa	-	[171]
PLA/Glycine	Piezoelectric	13.2 mV kPa^{-1}, 8 pC N^{-1}	2-69.5 kPa	-	[119]
DL Alanine	Piezoelectric	$d_{13} = 5.72 \pm 1.64$ pC N^{-1}, $d_{33} = \sim 5.5$ pm V^{-1}	150 mV at 6 N	40% tensile strain	[131]
Cellulose paper coated with Graphene	Piezoresistive	5.9 N^{-1}	-		[150]
Nikel on nanocellulose substrate	Piezoresistive	-	-	0.1958 %	[137]
X-SF-MXeneY-Graphene Oxide	Piezoresistive	14.23 kPa^{-1}; 12.53 kPa^{-1}	lower than 1.4 kPa	-	[158]
Chitosan	Piezoresistive	-	-	-	[49]
PANI into BC/CH composites	Piezoresistive	1.41 kPa^{-1}	-	-	[144]
BC and polymerizable deep eutectic solvents (PDESs)	Piezoresistive	-	-	GF (Gauge Factor) 1.29	[145]
CNFs and graphene co-incorporated PVA-borax hydrogel	Piezoresistive	-	-	GF 3.8	[139]
TEMPO-oxidized cellulose nanofibrils were homogeneously dispersed into PAA hydrogel	Piezoresistive	-	-	GF 7.3	[140]
Ionic hydrogel strain sensor comprises SAlg, tannic acid, and chemically cross-linked PAM	Piezoresistive	-	-	GF 2.0	[156]
Stacked CNT and Ni-fabrics	Piezoresistive	26.13 kPa^{-1}	-	-	[153]
Graphene Oxide papers	Piezoresistive	larger than 17.2 kPa^{-1}	-	-	[149]
Cotton fabric, Polyester textile - Ni electrodes - CNTs	Piezoresistive	14.4 kPa^{-1}	-	-	[146]
Graphene oxide as a compatibilizer for PLA - PEG blends	Piezoresistive	-	-	GF 70	[164]
CNTs/graphene/Waterborne polyurethane/CNC	Piezoresistive	-	-	-	[151]
MXene/BC	Piezoresistive	51.14 kPa^{-1}	0 - 0.82 kPa	-	[55]
Natural-wood/rGraphene Oxide	Piezoresistive	1.85 kPa^{-1}	-	-	[96]
SF/PolyPyrrole	Piezoresistive	-	-	-	[157]
Tannic acic/SAlg/PAA	Piezoresistive	-	-	-	[156]
Nanocellulose-MXene	Piezoresistive	95.2 kPa^{-1}	lower than 50 Pa	-	[142]
Paper-MXene	Piezoresistive	509.5 kPa^{-1}; 344.0 kPa^{-1}	-	-	[168]
All-paper	Piezoresistive	1.5 kPa^{-1}	-	-	[152]
CNT-BC	Piezoresistive	12.5 kPa^{-1}	-	-	[147]
SF/MXene	Piezoresistive	298.4 kPa^{-1}; 171.9 kPa^{-1}	1.4–15.7 kPa; 15.7–39.3 kPa	-	[56]
Collagen/Mxene	Piezoresistive	61.99 kPa^{-1}	-	-	[162]
Carbonized creep paper	Piezoresistive	2.56 kPa^{-1}; 5.67 kPa^{-1}	0.42-2.53 kPa; 0-0.42 kPa	-	[143]
SF/AgNWs	Piezoresistive	12-26 GPa^{-1}	-	-	[159]
Cellulose/PEDOT:PSS	Piezoresistive	83.9 kPa^{-1}	-	-	[141]
Gelatine/PVA/MWCNT	Piezoresistive	-	-	GF ~ 1296 under 12–20% strain	[161]
PVA/CS, phytic acid	Piezoresistive	-	-	GF 1.77	[155]
CMC/carbon black/multilayer graphene and SiO_2 nanoparticles	Piezoresistive	-	-	GF 70.2	[148]
Gelatin/Polyacrylamide	Piezoresistive	-	-	GF 0.560 and 0.835	[163]
Carbon black / wheat flour	Piezoresistive	-	-	-	[105]
Chitosan/diatom	Triboelectric	-	-	-	[102]
Gelatin/PAM/PEDOT:PSS	Triboelectric	-	-	GF 1.58	[178]
Chitosan/diatom/catechol	Triboelectric	-	-	-	[103]
Paper and polyethilenimmine	Triboelectric	-	60 N	-	[132]
CNC/methyl cellulose/graphite	Triboelectric	-	-	-	[133]
PVDF-TrFE/chitin nanofiber film	Triboelectric	12.4 nA kPa^{-1}; 2.6 nA kPa^{-1}	98 Pa - 9.8 kPa; 9.8 - 98 kPa	-	[136]
SAlg/glycerol/AgNWs	Triboelectric	0.237 V kPa^{-1}	-	-	[135]
Silk hydrogel/ZnO nanorods	Piezo-Triboelectric	320 pC N^{-1}	-	-	[181]
Silk fibroin/PVDF	Piezo-Triboelectric	-	-	-	[185]
Silk/PVDF/Poly Ethylele Tereftalate	Piezo-Triboelectric	-	-	-	[48]
Cellulose/$BaTiO_3$ aerogel papers based PDMS nanocomposites.	Piezo-Triboelectric	-	-	-	[183]
$BaTiO_3$-Nanorods/Chitosan	Piezo-Triboelectric	-	-	-	[184]
$BaTiO_3$/SAlg	Piezo-Triboelectric	-	-	-	[182]
PVA/PEDOT:PSS and Zein/PVA/CNT	Piezoresistive-Triboelectric	281.59 kPa^{-1}	0–60 kPa	-	[108]
Cellulose/glycerol/NaCl/H2O	Piezo-capacitive	0.0103 kPa^{-1}	-	-	[180]
AgNWs/BC	Piezo-capacitive	5.49 kPa^{-1}	-	-	[174]
PVA/SAlg/BC/modified CNT and carbon black	Piezo-capacitive	0.033 kPa^{-1}	-	-	[51]
Graphite/cellulose paper	Piezo-capacitive	-	-	-	[170]
PLGA-PCL	Piezo-capacitive	0.863 ± 0.025 kPa^{-1}	-	-	[171]

Appendix B. Table nanogeneratos

Material	Mechanism	Sensitivity	Power/Power density	Force	Frequency	Max Output	Active area	Ref.
SF	Piezoelectric	3.5 pC N^{-1}	1.2 μW cm^{-3}	10 N	0.5 Hz	7 V and 150 nA	3 × 4 cm^2	[198]

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Material	Mechanism	Sensitivity	Power/Power density	Force	Frequency	Max Output	Active area	Ref.
Potassium Sodium Niobate/ PHBV/PLLA	Piezoelectric	150 pC N^{-1}	0.7 W cm^{-2}	0.5 N	10 Hz	0.6 μA and 6 V	1.5 × 1.5 cm^2	[189]
PVDF/Clay nanoparticles	Piezoelectric	39.75 pC N^{-1}; 50.10 pC N^{-1},	4115 μW cm^{-3}; 7187 μW cm^{-3}	5 N	10 Hz	1.6 μA and 85 V; 1.9 μA and 125 V	4.9 × 3.0 × 0.2 cm^3	[112]
HZTO/polydopamine/PLA	Piezoelectric	26 pm V^{-1}	463.5 mW cm^{-3}	2 N	23 Hz	5.41 V and 0.26 mA	3.0 × 1.5 cm^2	[188]
Cotton fibers/PDMS	Piezoelectric	-	182.06 μW cm^{-3}	5 N	10 Hz	~1.0 μA and 81.2 V	3.6 × 2.1 cm^2	[99]
PLLA/Dyphenilalanine rods	Piezoelectric	12.4 pm V^{-1}	1.56 W m^{-3}	60 N	0.13 Hz	1.78 V and 70 nA	1.2 × 1.2 cm^2	[196]
PVDF/eggshell membrane	Piezoelectric	-	-	10 kgf	6 Hz	15 V and 150 nA	-	[191]
$Ba_{0.85}Ca_{0.15}Zr_{0.10}Ti_{0.90}O_3$ / Polydopamine/ PLA	Piezoelectric	-	7.54 mW cm^{-3}	8.2 - 18.5 kPa	4 Hz	14.4 V and 0.55 μA	1.5 × 1.3 cm^2	[238]
$BaTiO_3$/PLGA	Piezoelectric	-	10 mW cm^{-2}	10 N	-	8 V	1 × 1 cm^2	[187]
Chitin/PVDF	Piezoelectric	35.56 pC N^{-1}	6600 mW cm^{-3}	25 kPa	6 Hz	49 V and 1.9 mA	2 × 1.5 cm^2	[50]
Cellulose fibrous untreated onion skin	Piezoelectric	2.8pC N^{-1}	1.7μW cm^{-2}	34 kPa	3 Hz	18 V and ~ 166 nA	-	[100]
Lactate, 1,4-BDO, and SA	Piezoelectric	-	-	4 gf	0.3 Hz	303.75 mV cm^{-2} and 1.92 10^{-2} mA cm^{-2}	4 × 4 cm^2	[190]
Eggshell membrane	Piezoelectric	23.7 pC N^{-1}	238.17 mW cm^{-3}	81.6 kPa	1-5 Hz	26.4 V and 1.45 μA	2.0 × 1.5 cm^2	[110]
Cellulose nanofibril/MoS_2/ $BaTiO_3$	Piezoelectric	45 pC N^{-1}	-	-	-	8.2 V and 0.48 mA	-	[52]
Collagen / cotton	Piezoelectric	-	-	5 N	-	45 V and 250 nA	3 × 3 cm^2	[192]
Chicken feathers fibers	Piezoelectric	1.6–2.1 pC N^{-1}	6 mW cm^{-2}	0.13–0.31 MPa	4 Hz	10 V and 1.8 mA	2.0 × 6.0 cm^2	[109]
Diphenilalanine nanotubes	Piezoelectric	46.6 pm V^{-1}	8.2 nW	42 N	-	2.8 V and 37.4 nA	-	[73]
lactate-BDO, SA and IA	Piezoelectric	-	-	4 gf	-	0.24 V cm^{-2} and 19.75 μA cm^{-2}	4 × 4 cm^2	[118]
Yellow ginkgo leaves	Piezoelectric	6.44 pm V^{-1}	25.7 mW cm^{-3},	16 N	3 Hz	131 V cm^{-3} and 2.5 mA cm^{-3}	2 × 2 cm^2	[193]
Sugar encapsulated PVDF fibers	Piezoelectric	33 pC N^{-1}	33 mW m^{-2}	10 kPa	5 Hz	100 V	10.5 × 8 cm^2	[199]
FF nanorods	Piezoelectric	-	0.1 nW	-	1 Hz	0.6 V and 7 nA	30 × 12 × 0.25 mm^3	[197]
Cellulose nanofibril/$BaTiO_3$	Piezoelectric	-	-	-	-	0.25 V	3 × 1.5 cm^2	[194]
Chitosan fibers and red quantum dot	Piezoelectric	-	12 μW cm^{-3}	4 N	3 Hz	125 V cm^{-3} and 1.5 μA cm^{-3}	1.0 × 1.0 cm^2	[195]
SF	Piezoelectric	30.6 mV N^{-1}	5.9 mW m^{-2}	31 N	1 Hz	155.2 mV and 22.9 nA	-	[34]
Wood sponge	Piezoelectric	-	0.6 nW cm^{-2}	3 N	-	0.69 V and 7.1 nA	15 × 15 × 14 mm^3	[32]
M13 bacteriophage	Piezoelectric	13.2 pm V^{-1}	236 nW	17 N	0.5 Hz	12 V and 300 nA	-	[200]
Chitosan/$BaTiO_3$	Piezo/tribo	-	431.8 μW	-	-	110.8 V and 10 μA	-	[239]
Silk/ZnO	Piezo/tribo	-	1 mW cm^{-2}	15 N	1 Hz	12.5 V and 0.04 μA	5 × 5 mm^2	[181]
Wheat starch	Triboelectric	-	113.2 μW cm^{-2}	-	4 Hz	151.4 V and 47.1 μA	25 cm^2	[208]
Crystalline silk microparticles/PVA	Triboelectric	-	14.4 W m^{-2}	5 N	5 Hz	280 V and 17.3 μA	-	[210]
Chitosan/starch	Triboelectric	46.03 V kPa^{-1} in the range 1.25–6.25 kPa	5.07 W m^{-2}	2-12 N	3 Hz	1080 V and 16.9 mA m^{-2}	1 × 1 cm^2	[214]
Cellulose acetate /polyethyleneimine	Triboelectric	-	2.21 mW cm^{-2}	16 N	1.5 hz	478 V 6.3 μA cm^{-2}	3 × 3 cm^2	[215]
Al_2O_3 fillers into cellulose acetate	Triboelectric	-	2.5 mW cm^{-2}	18 N	1.5 Hz	448 V and 45 μA	4 × 4 cm^2	[216]
Bio-Silica and Cellulose Nanofibril	Triboelectric	-	85.5 mW m^{-2}	8 N	5 Hz	388 V and 18.6 μA	4.9 cm^2	[217]
CNC/ methylcellulose	Triboelectric	-	180 μC m^{-2}		0.5 Hz	205 V and 18 μA	2 × 2 cm^2	[133]
CMC/PDMS	Triboelectric		0.9 μW	5 N	1 Hz	27.8 V and 2.2 μA	1 cm^2	[218]
SCL/ PDMS	Triboelectric	-	216 mW m^{-2}	5 N	3 Hz	126 V and 3 μA	2 × 3 cm^2	[209]
Root bark and PCL	Triboelectric	-	9 μW cm^{-2}	20 N	8 Hz	80 V	5 × 5 cm^2	[95]
Chitosan/diatom	Triboelectric	-	15.7 mW m^{-2}	8 N	5 Hz	150 V and 1.02 μA	3 × 4 cm^2	[102]
Silk/rice paper/chitin/egg white/cellulose	Triboelectric	-	21.6 mW m^{-2}	-	1 Hz	55 V and 0.6 μA	1 × 2 cm^2	[106]
Chitosan	Triboelectric	-	17.5 μW m^{-2}	-	-	13.5 V and 42 nA	2 × 1 cm^2	[219]
Laver/rise sheet	Triboelectric	-	0.02 mW m^{-2}	-	-	23 V and 315 nA	2 × 2 cm	[101]
Silk fibroin	Triboelectric	-	-	20 N	-	3 μA and 50 V	2 × 2 cm^2	[211]
CNT/silk nanofibers	Triboelectric	0.069 kPa^{-1}	6 mW m^{-2}	13.3 kPa	0,5 Hz	150 V and 1.24 μA	2 × 2 cm^2	[212]
Cellulose	Triboelectric	1.66 V N^{-1}	127 mW m^{-2}	40 N	4 Hz	65 V and 1.86 μA	6.15 cm^2	[220]
PLGA/PVA	Triboelectric	0.011 kPa^{-1}	130 mW m^{-2}	40 KPa	5 Hz	90 V and 1.5 μA	2 × 2 cm^2	[221]
Fish gelatine	Triboelectric	-	100 μW cm^{-2}	5 N	5 Hz	500 V and 4 μA	3 × 3 cm^2	[222]
Bovine Serum Albumine/ Glycerol	Triboelectric	-	0.15 W m^{-2}	30	5 Hz	474 V and 0.7 μA	3 × 3 cm	[225]
BC/CNT	Triboelectric	-	3 μW	6 N	5 Hz	29 V and 0.6 μA	6 × 6 cm^2	[223]
Chitosan	Triboelectric	-	-	-	1 Hz	130 V	5 × 3 cm^2	[226]
CNF/microfibers	Triboelectric	-	7.68 μW cm^{-2}	-	1 Hz	21.9 V and 0.73 μA	-	[227]
Ethyl cellulose/polyamide/ MXene/PVDF	Triboelectric	-	290 mW m^{-2}	43 N	5 Hz	45 V and 1.8 μA	2 × 2 cm^2	[228]
Silk	Triboelectric	-	22.05 W m^{-2}	5 N	5 Hz	395 V	2 × 2 cm^2	[213]

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