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Self-Powered Wearable Biosensors

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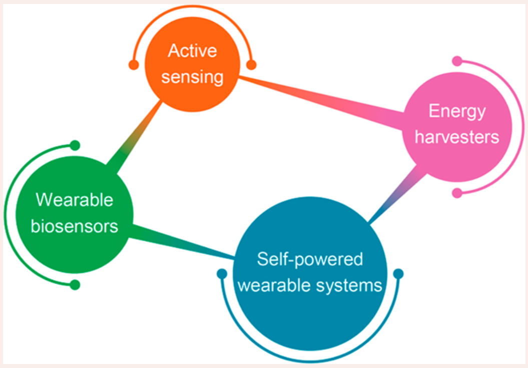

CONSPECTUS: Wearable biosensors hold the potential of revolutionizing personalized healthcare and telemedicine. Advances in chemical sensing, flexible materials, and scalable manufacturing techniques now allow wearables to detect key physiological indicators such as temperature, vital signs, body motion, and molecular biomarkers. With these systems operating on the skin, they enable continuous and noninvasive disease diagnosis and health monitoring. Such complex devices, however, require suitable power sources in order to realize their full capacity. Emerging wearable energy harvesters are attractive for addressing the challenges of a wearable power supply. These harvesters convert various types of ambient energy sources (e.g., biomechanical energy, biochemical energy, and solar energy) into electricity. In some circumstances, the harvested electrical signals can directly be used for active sensing of physiological parameters. On the other hand, single or hybrid wearable energy harvesters, when integrated with power management circuits and energy storage devices, could power additional biosensors as well as signal processing and data transmission electronics. Self-powered sensor systems operate continuously and sustainably without an external power supply are promising candidates in the next generation of wearable electronics and the Internet of Things.

This Account highlights recent progress in self-powered wearable sensors toward personalized healthcare, covering biosensors, energy harvesters, energy storage, and power supply strategies. The Account begins with an introduction of our wearable biosensors toward an epidermal detection of physiological information. Advances in structural and material innovations enable wearable systems to measure both biophysical and biochemical indicators conformably, accurately, and continuously. We then discuss emerging technologies in wearable energy harvesting, classified according to their capability to scavenge energy from various sources. These include examples of using energy harvesters themselves as active biosensors. Through seamless integration and efficient power management, self-powered wireless wearable sensor systems allow real-time data acquisition, processing, and transmission for health monitoring. The final section of the Account covers the existing challenges and new opportunities for self-powered wearable sensors in health monitoring and human−machine interfaces toward personalized and precision medicine.

1. INTRODUCTION

In recent years, wearable electronics have greatly improved the quality of daily life and have become indispensable tools.[1,2] Wearable devices targeted for detecting diversified biophysical and biochemical signals offer a noninvasive means for extracting physiological indicators.[3,4] The real-time monitoring of these indicators can provide valuable information for the early diagnosis and prevention of a number of health conditions such as cardiovascular diseases, gout, diabetes, and coronavirus disease 2019 (COVID-19).[5−8] Emerging nanotechnology, materials science, and flexible electronics have led to wearable biophysical sensors that are capable of monitoring human activities, body motion, and electrophysiological signals (e.g., electroencephalogram (EEG) and electrocardiogram (ECG)).[1,3] In addition, wearable biochemical sensors are emerging for noninvasive detection of molecular-level indicators (e.g., electrolytes and metabolites) from biofluids.[9,10]

To ensure wearable biosensors can achieve continuous operation and make accurate measurements, it is crucial to develop renewable and sustainable power supplies.[11] The recent materials and nanotechnology advances have led to new wearable devices that can harvest energy directly from the human body and the surrounding environment. These wearable energy harvesters are capable of converting different energy sources such as biomechanical energy, biochemical energy, thermal energy, and solar energy into electricity.[12] In some cases, they can work directly as active sensors, as their generated outputs correspond to the external stimuli such as

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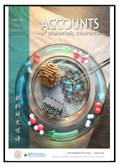

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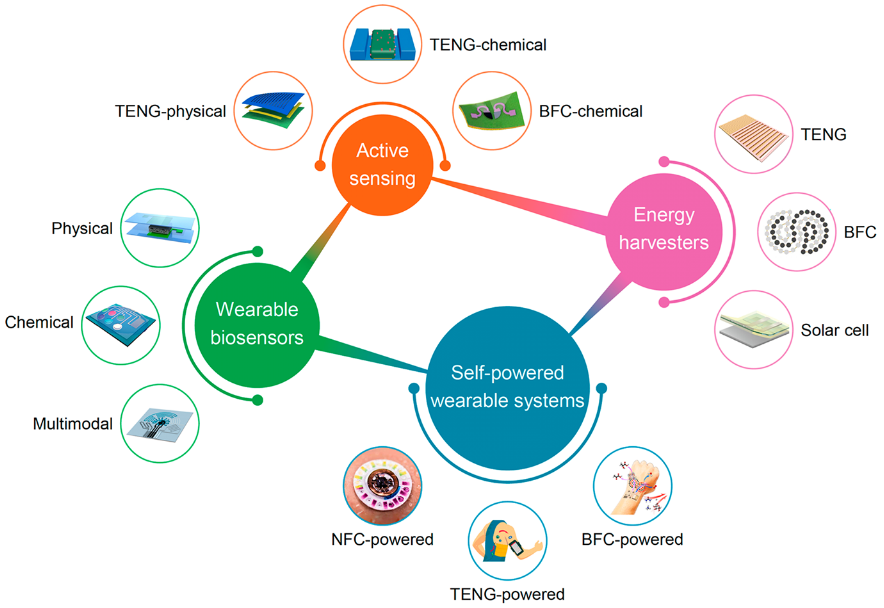

Figure 1. An overview of the self-powered wearable sensors. Reproduced with permission from ref 14. Copyright 2016 Springer Nature. Reproduced with permission from ref 16. Copyright 2017 Springer Nature. Reproduced with permission from ref 15. Copyright 2020 Springer Nature. Reproduced with permission from ref 19. Copyright 2019 American Academy for the Advancement of Science. Reproduced with permission from refs 17 and 18. Copyright 2020 American Academy for the Advancement of Science. Reproduced with permission from ref 21. Copyright 2016 American Chemical Society. Reproduced with permission from ref 20. Copyright 2018 Elsevier. Reproduced with permission from ref 22. Copyright 2016 Royal Society of Chemistry. Reproduced with permission from ref 23. Copyright 2018 Royal Society of Chemistry.

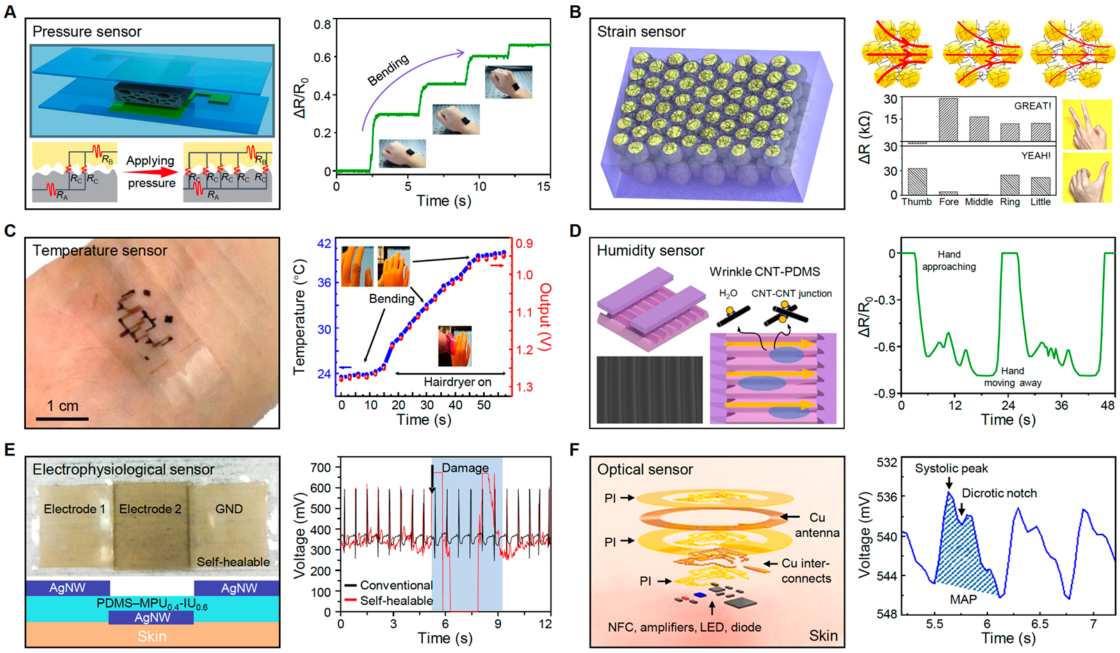

Figure 2. Wearable biophysical sensors. (A) Porous CNT-PDMS based piezoresistive pressure sensor for body motion monitoring. Reproduced with permission from ref 20. Copyright 2018 Elsevier. (B) Strain sensor with immobilized MWCNT networks for finger gesture recognition. Reproduced with permission from ref 28. Copyright 2017 Wiley-VCH. (C) Stretchable temperature sensor for strain-independent temperature sensing. Reproduced with permission from ref 30. Copyright 2018 Springer Nature. (D) Wrinkled CNT-PDMS as a humidity sensor for hand approaching detection. Reproduced with permission from ref 32. Copyright 2019 American Chemical Society. (E) Self-healable electrocardiogram sensor for measurement of cardiac signals. Reproduced with permission from ref 34. Copyright 2018 Springer Nature. (F) Epidermal optical sensor for heart rate and mean arterial pressure recording. Reproduced with permission from ref 37. Copyright 2016 American Academy for the Advancement of Science.

motion, bending, strain, and molecular concentration.[13] Meanwhile, the integration of energy harvesters with wearable biosensors and signal processing circuits enables the development of fully self-powered sensor systems.

This Account provides a systematic introduction and highlights recent advances of self-powered wearable biosensors in the field of personalized healthcare (with a focus on our own works) as shown in Figure 1.[14−23] First, a brief introduction of wearable biosensors with engineered materials and novel layouts is given. Subsequently, wearable energy harvesters with different mechanisms are discussed in detail. Following these sections, the latest strategies of energy harvesters as active sensors and as key components in self-powered sensor systems are illustrated. Finally, a perspective on the future development and challenges of self-powered wearable sensors is provided.

2. WEARABLE BIOSENSORS FOR HEALTH MONITORING

Wearable biosensors provide feasible approaches to monitor epidermally physiological signals from both physical motions and biofluids.[3] This section summarizes various types of sensing modalities and working principles that serve as the foundations for wearable biosensors, along with their clinical implementations. Section 2.1 introduces wearable biophysical sensors that are available for noninvasively measuring biopotentials, physical motions, and optical signals associated with human activities. With the conformal attachment on the skin, they can detect various physical indicators. Section 2.2 discusses sensing technologies and preparation methods of wearable biochemical sensors. By exploiting different essential sensing elements, wearable biochemical sensors realize continuous tracking of chemical biomarkers from biofluids that indicate health status and allow for an early disease diagnosis.

2.1. Wearable Biophysical Sensors

Pressure and Strain Sensors. Epidermal pressure and strains caused by the arterial pulse, human motion, and breathing rate are closely related to human physiological activity.[3] Soft wearable tactile sensors are also of great importance for applications in personalized healthcare, electronic skin, and prosthesis control.[24] Wearable pressure and strain sensors operate by detecting stimuli via changes in sensor material properties including piezoresistive, capacitive, piezoelectric, and triboelectric effects.

Piezoresistive pressure sensors are prevalent due to their cost-efficient fabrication, simple designs, and easy acquisition of both static and dynamic responses. Active materials including conductive polymers, carbon nanotubes, graphene, and nanowires are promising candidates with mechanical robustness and electrical reliability.[25] Microstructure-engineered designs such as wrinkles, cracks, woven materials, and porous materials can greatly enhance the sensitivity by accommodating geometrical deformations.[25] The synergistic effect of actively conductive materials with intrinsic structure provides a feasible approach to obtain a satisfying performance. Capacitive pressure sensors perform with both high sensitivity and low hysteresis. The microstructured dielectric layer allows for the detection of subtle changes in pressure.[26] Piezoelectric and triboelectric pressure sensors provide suitable approaches to dynamic measurements with fast responses and high signal-to-noise ratios. Upon deformation, mechanical to electrical energy conversion occurs allowing for a quantification of applied pressure. Thus, wearable pressure sensors that exploit different sensing modalities have a broad utility in health monitoring, human−machine interfaces, and soft robotics applications.[24] Figure 2A shows a porous piezoresistive pressure sensor based on a carbon nanotube−poly(dimethylsiloxane) (CNT-PDMS) conductive elastomer through a solution-evaporation fabrication method.[20] With an optimized dimension and content ratio, this pressure sensor achieves 0.51 kPa^{-1} in a 2 kPa linear region and induces stable resistance responses in accordance to the joint bending states, as well as other muscle movements.

The conventional strain sensors, mainly based on brittle materials, typically suffer from low stretchability and are inappropriate for the detection of human motion. Two mainstream strategies are available to construct stretchable conductive materials. The first involves introducing stretchability into intrinsically brittle materials to develop different geometric patterns, such as cracks and buckled structures.[27] The second strategy adopts percolating conductive nanomaterial networks including nanoparticles, nanowires, and nanotubes.[25] On the basis of these strategies, Figure 2B presents a highly robust and stretchable strain sensor by a three-dimensional (3D) self-assembly of carbon nanotubes and microsphere composites.[28] When the strain sensor is stretched, an applied stress induces the disconnection of overlapped carbon nanotubes due to the weak interfacial binding and large stiffness mismatch between the stretchable elastomer matrix and nanomaterials, resulting in an increasing electrical resistance.

Temperature Sensor. Body temperature is a critical indicator to monitor human activities and determine health conditions. It maintains an extraordinarily narrow range between 36 and 37 °C through thermoregulation, and abnormal changes provide insightful information related to cardiovascular health, cognitive condition, wound healing, and many other syndromes.[29] Traditional methods rely on simple thermometers and are not applicable for a continuous point-of-care use. Temperature-sensitive materials embedded in a flexible or stretchable substrate can exhibit a high sensitivity, fast response, long-term reliability, and skin compatibility. Zhu et al. present circuit design strategies to improve the accuracy and robustness of a wearable temperature sensor based on stretchable carbon nanotube transistors (Figure 2C).[30] The stretchable temperature sensor circuit can trace sensor output as a function of temperature, and negligible change in the temperature output occurs under a repeatedly uniaxial strain of 15% in the device. To further minimize the strain-induced errors, smaller feature sizes will enable the integration of instrumentation electronics closer to the sensor elements and thereby allow an accurate temperature monitoring under different stretching strains.

Humidity Sensor. Besides the measurement of the typically physical parameters mentioned above, the analysis of skin humidity can yield insights into various aspects of physiological health. A real-time measurement of the hydration levels of human skin can be used to monitor respiration and water evaporation,[31] which are important in monitoring disease states and in assessing factors related to an abnormal skin response. Figure 2D shows a wearable humidity sensor with a wrinkled CNT-PDMS, the resistance of which changes due to the presence of water molecules.[32] The wrinkled structure supports a more hydrophilic and anisotropic wetting surface with an enlarged surface area, thus enhancing the humidity sensing performance. Through the modulation of the CNT ratio, the wearable humidity sensor shows great sensitivity and reliable repeatability, especially in human motion or breathing monitoring.

Electrophysiological Sensor. Biopotential signals are effective indicators for medical diagnosis and health monitoring. Wearable electrophysiological sensors are available to measure biopotentials including ECG, EEG, electromyography

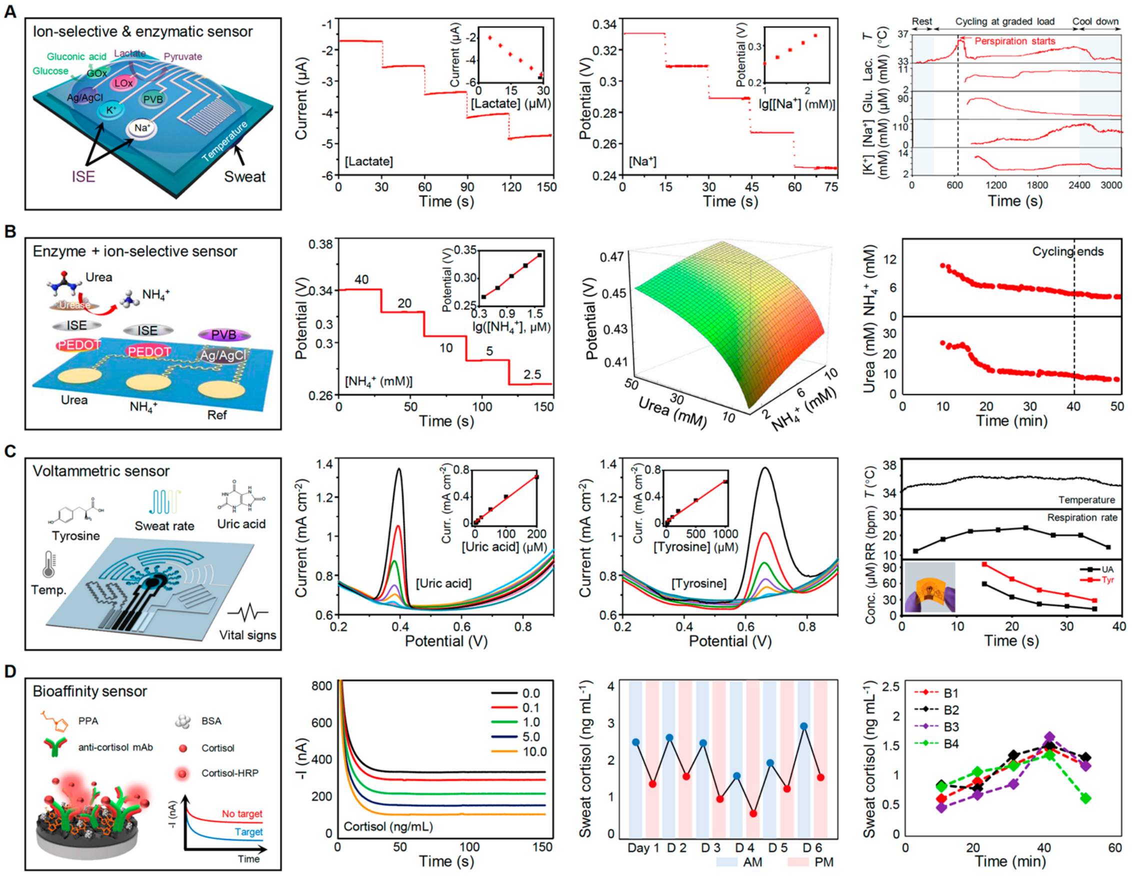

Figure 3. Wearable biochemical sensors. (A) Enzymatic and ion-selective sensors for a continuous analysis of metabolites and electrolytes. Reproduced with permission from ref 14. Copyright 2016 Springer Nature. (B) Enzymatic sensors based on ion-selective electrodes for a continuous urea sensing. Reproduced with permission from ref 17. Copyright 2020 American Academy for the Advancement of Science. (C) Voltammetric sensor for a sensitive detection of uric acid and tyrosine. Reproduced with permission from ref 15. Copyright 2020 Springer Nature. (D) Bioaffinity sensor based on the antibody−antigen interactions for a cortisol analysis. Reproduced with permission from ref 42. Copyright 2020 Elsevier.

(EMG), and electrooculography (EOG) noninvasively.[1,3] These informative signals can be used to monitor heart, brain, muscle, and eye activities, respectively. To acquire weak biopotential signals, wearable electrophysiological sensors with skin-compatible materials should perform with high signal-to-noise ratios, stable adhesion, and low motion artifacts for accurate signal processing.[33] Furthermore, functional materials with an optimal modulus, minimal skin irritation, and long-term usage offer compelling benefits to achieve a seamless and conformal contact with human skin. Son et al. employed self-healing electrodes as active components to fabricate a wearable ECG sensor composed of three electrodes (Figure 2E).[34] The sensor achieves ECG data acquisition through the percolated CNT network in a polymer matrix and is capable of returning to its original state within seconds even after the damage occurred.

Optical Vital Sign Monitor. Optical sensors capture the amount of light either transmitted or scattered and convert signal changes into electrical outputs. Wearable optical sensors have utilized nanostructured materials like quantum dots, nanocrystals, two-dimensional (2D) materials, and perovskite materials to provide clinically relevant information for disease diagnosis and treatment.[35] Optical analysis of the blood flow, for example, allows for the calculation of key physiological parameters such as arterial oxygen saturation via pulse oximetry and heart rate variability via photoplethysmography (PPG).[36] An example in Figure 2F shows a wearable optical sensor powered by a near-field communication (NFC) technology for a wireless optical characterization of the skin.[37] Arterial pulse waves temporally modulate the back-scattered light, and the measured signals reveal both systolic peaks and dicrotic notches, which are relevant to both remote diagnostics and health warnings. Wearable optical sensors require a high photoresponse sensitivity and flexibility to ensure an efficient transport of photogenerated carriers and minimize motion artifacts. Ultimately these sensors must be able to measure parameters such as flow rate, pulse wave velocities, and heart disease. Integrating a device with both wearable optical sensors and other biophysical sensors can facilitate a multimodal network of sensors across the body, allowing for a more complete assessment of one's health status.

2.2. Wearable Biochemical Sensors

Considering that wearable biophysical sensors only monitor vital signs and physical activities, wearable biochemical sensors are essential to assess the human health state at the biomolecular level. Biofluids, such as saliva, tears, sweat, and interstitial fluids, are ideal analytes, as they can be retrieved noninvasively and contain a wealth of physiological information.[8] With techniques including potentiometry, amperometry, voltammetry, and impedance spectroscopy, wearable biochemical sensors can continuously monitor dynamic variations of

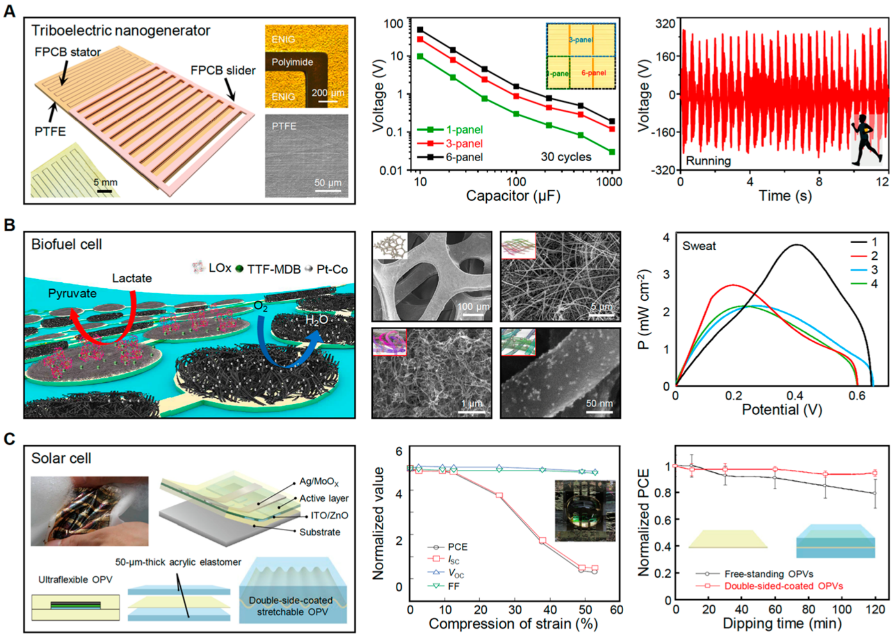

Figure 4. Wearable energy harvesters that convert various energy sources to electricity. (A) FTENG for scavenging mechanical motions energy. Reproduced with permission from ref 18. Copyright 2020 American Academy for the Advancement of Science. (B) Flexible nanoengineered biofuel cell array for chemical energy harvesting from human sweat. Reproduced with permission from ref 17. Copyright 2020 American Academy for the Advancement of Science. (C) Stretchable and waterproof organic solar cell as a textile-compatible power source. Reproduced with permission from ref 16. Copyright 2017 Springer Nature.

biomarkers in biofluids.[9] Biomarkers including ions, metabolites, amino acids, hormones, and drugs can be detected to monitor or diagnose conditions like cystic fibrosis, gout, mental disorders, and drug abuse.[8]

Enzymatic and Ion-Selective Sensors. Metabolites and electrolytes in biofluids are excellent indicators of a healthy state and can provide warnings for various diseases.[38] For example, an imbalance of glucose leads to severe threats to human health for individuals afflicted with diabetes mellitus, and increased lactate levels can correspond to cardiac diseases, endotoxic shock, or liver disease. Concentrations of ions including sodium, potassium, and calcium are also markers for dehydration during exercise activities. Key metabolites such as glucose and lactate can be monitored with amperometric enzymatic sensors, while a number of electrolytes (e.g., Na^+, K^+, NH_4^+, and Ca^{2+}) can alternatively be detected via potentiometric ion-selective sensors. Figure 3A demonstrates an electrochemical sensor array consisting of enzymatic and ion-selective sensors that can simultaneously monitor lactate and glucose as well as sodium and potassium ions in sweat via amperometric and potentiometric techniques, respectively.[14] In some special cases, a combination of enzymatic and potentiometric sensors is sometimes needed to realize an accurate detection of a given analyte. For example, an enzymatic urea sensor can be developed based on an NH_4^+ ion-selective electrode (Figure 3B).[17] The urease layer of the sensor converts urea to NH_4^+, which is subsequently detected. This combination of sensors allows for a real-time monitoring of urea in sweat.

Voltammetric Sensor. Voltammetric sensors are the most relevant methods for the rapid and accurate detection of electroactive analytes. Similar to amperometric sensors, voltammetric sensors adopt a three-electrode configuration. The measured potential dramatically increases at a redox potential within the oxidization/reduction range of the analytes. Electroactive analytes, such as certain drugs, amino acids, and vitamins, can be directly oxidized at a specific potential. To achieve a high sensitivity and low detection limits, pulsed techniques such as differential pulse voltammetry (DPV) are commonly used to measure analyte oxidation. Figure 3C illustrates a wearable voltammetric sensor for continuously monitoring the uric acid and tyrosine in human sweat.[15] The laser-engraved graphene (LEG) biochemical sensor exhibits great selectivity and sensitivity within the target's physiological concentration range in sweat. For on-body validation, the integration of a multi-inlet microfluidic module and the LEG-based voltammetric sensors ensures a reliable analysis process with a high accuracy and temporal resolution. Combined with a preconcentration process, voltammetric sensors can also be used to monitor heavy metal ions (e.g., Cu^{2+}, Zn^{2+}, and Pb^{2+}) in biofluids.[39]

Bioaffinity Sensor. Despite the current challenges for wearable implementation, bioaffinity sensors have emerged as an efficient and powerful analysis method for analyzing a broad spectrum of biomarkers, including protein, peptides, and hormones.[40−42] In general, bioaffinity sensors work via a specific detection of target-bioreceptor interactions (such as antigen−antibody binding). Figure 3D shows one example in which the antibody-based immunosensors are applied to monitor the dynamics of cortisol in biofluids.[42] Cortisol is linked to chronic stress, which enhances the threats of depression, anxiety, suicide, fragile immune system, and cardiovascular diseases. The fully integrated mHealth system is capable of measuring the cortisol diurnal cycle and the dynamic stress-response profile noninvasively on the body.

3. WEARABLE ENERGY HARVESTERS

The rapid rise of bioelectronics is facilitating advancements in wearable biosensors for monitoring body motion and electrophysiological signs as well as biomolecular information. One of the challenges with wearable biosensors is to ensure a prolonged use with a reliable power supply. Unlike traditional rigid batteries, which require intermittent replacement or recharging, novel wearable energy harvesters provide a battery-free approach to efficiently scavenge energy from human motion and the ambient environment. They can also greatly enhance the feasibility and convenience of wearable biosensors in continuous health monitoring.[43] Emerging wearable energy harvesters can be categorized by their energy sources, including mechanical energy, biochemical energy, and solar energy.[11] This section presents a focused introduction of various kinds of energy harvesters tailored for wearable applications.

3.1. Triboelectric Nanogenerator

Triboelectric nanogenerators (TENGs) harvest mechanical energy by utilizing contact electrification coupled with electrostatic induction for energy conversion from dynamic stimuli.[44] The mechanical motion between the two electrified materials with opposite triboelectric charges drives the flow of induced charges and produces a voltage. TENGs are low-cost, biocompatible, and have great adaptability. Their efficiency largely depends on the differences in the electron-attracting capability of the tribomaterials and the morphology of the contact surfaces.

Figure 4A shows a freestanding-mode TENG (FTENG) consisting of an interdigital stator and a grating-patterned slider integrated with a commercial flexible printed circuit board.[18] The FTENG converts mechanical energy into electrical energy via the coupling effect of the tribo-electrification and electrostatic induction. Surface charge density is a crucial factor in electricity generation and is determined by materials with different charge polarities. The relative movement of two electrodes made of different triboelectric materials (e.g., poly(tetrafluroethylene) (PTFE) and copper) induces a flow of electric charges to maintain the electrostatic equilibrium. To further enhance the capability of power output, TENG can adopt a novel design with multiple panels to meet the high energy demands of wearable sensors. When fixed on the side torso, the wearable FTENG shows remarkable mechanical stability with reliable outputs during exercise.

Wearable nanogenerators are a highly promising option for energy production from body movements. Similar to TENGs, electromagnetic generators (EMGs) and piezoelectric nanogenerators (PENGs) could also potentially be used to power future wearables.[45] PENG is based on the piezoelectric effect,[46] and there is an induced piezoelectric potential difference when an external force is applied to the piezoelectric material. The exploration of new materials with higher charge densities will enable improved power outputs of these nanogenerators. Meanwhile, the utilization of microstructured surfaces such as pyramids, pillars, and wrinkle shapes can also increase their contact area, thereby improving the output power densities as well.[47]

3.2. Biofuel Cell

Miniaturized flexible biofuel cells (BFCs) offer an attractive innovation for wearable and implantable power sources, as they could directly harvest energy from our body fluids.[48] BFCs convert chemical energy into electricity by utilizing redox enzymes as catalysts. To ensure the optimal performance of BFCs, it is crucial to maximize the electrochemical transduction between the enzyme and the electrode surface. The use of nanomaterial-modified electrodes with a large electrochemically active surface area (ECSA) enables a high loading and promotes the power generation.

Figure 4B shows a nanoengineered BFC array consisting of lactate oxidase immobilized bioanodes and alloy nanoparticle-decorated cathodes.[17] On the one hand, a monolithic integration of the hierarchical Ni microstructures, reduced graphene oxide (rGO) films, and Meldola's blue-tetrathiafulvalene-modified carbon nanotubes (MDB-TTF-CNT) on the enzymatic anode increase the ECSA by 3000 times. On the other hand, a Pt–Co alloy nanoparticles-decorated MDB-modified CNT network (MDB-CNT) is used to immobilize the BFC cathode. Co dopants could enhance the cohesive energy, stabilize the nanoparticles, and then reduce biofouling, leading to a remarkable long-term stability in body fluids. The BFC array can efficiently catalyze the lactic acid to pyruvate in human sweat and generate a stable current output with a high-power density (as high as 3.6 mW cm^{-2}).

The development of wearable and flexible BFCs will require materials that can offer conformability and mechanical compliance against the body while providing highly efficient biocatalytic chemical energy conversion efficiency. Their efficiency strongly depends on the dynamically changed compositions of the biofluids during physical activities and surrounding environments (temperature, pH, humidity, and oxygen level). It is essential to immobilize the enzyme on the electrode surface reliably to maintain a stable performance in the complex biofluids during a long-term operation.

3.3. Solar Cell

Solar cells are one of the most promising green-energy harvesters.[49] Their ability to convert freely accessible incident light into electrical energy makes them attractive energy generators for wearable devices. Both materials development and functional modifications are crucial for a high-efficiency energy conversion. Among the various materials applied for solar cells, perovskite, dye-sensitized, and inorganic and organic photovoltaics (OPVs) are of particular interest.

Figure 4C shows mechanically flexible OPVs that exhibit a high power conversion efficiency by combining stable active layers and an inverted architecture.[16] The washable and lightweight OPVs retain superior stretchability and stability even in water, with an efficiency of 7.9%. The implementation of such OPVs provides a feasible solution to wearable biosensors for a continuous and long-term health monitoring in the presence of light.

For the optimal application in bioelectronics and wearable systems, both experimental and theoretical studies need to be conducted to provide fundamental guidelines on the best choices of functional materials and specific designs of solar cells. It is essential to design the cells that operate stably on the body and harvest enough energy to power or charge an entire wearable device for a sufficient time.

3.4. Hybrid Energy Harvester

Hybrid energy harvesting enables one system to convert multiple sources of energy into electricity simultaneously or sequentially.[50] It provides a simple but effective method for sustainable energy harvesting and a promising means to effectively increase their energy production efficiency.[51] Hybrid energy harvesters can either harvest one type of energy, like using TENGs with PENGs or EMGs to harvest mechanical energy, or utilize various different energy harvesters to enable a favorable energy production in any environment.

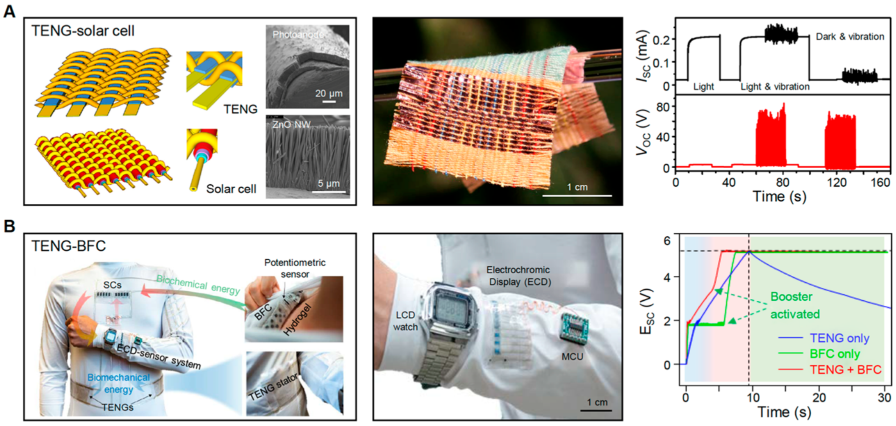

Figure 5. Hybrid energy harvesters. (A) Microcable structured textile for simultaneously harvesting solar and biomechanical energy. Reproduced with permission from ref 52. Copyright 2016 Springer Nature. (B) Wearable e-textile microgrid system that unites BFCs and TENGs for hybrid biochemical and biomechanical energy harvesting. Reproduced with permission from ref 53.

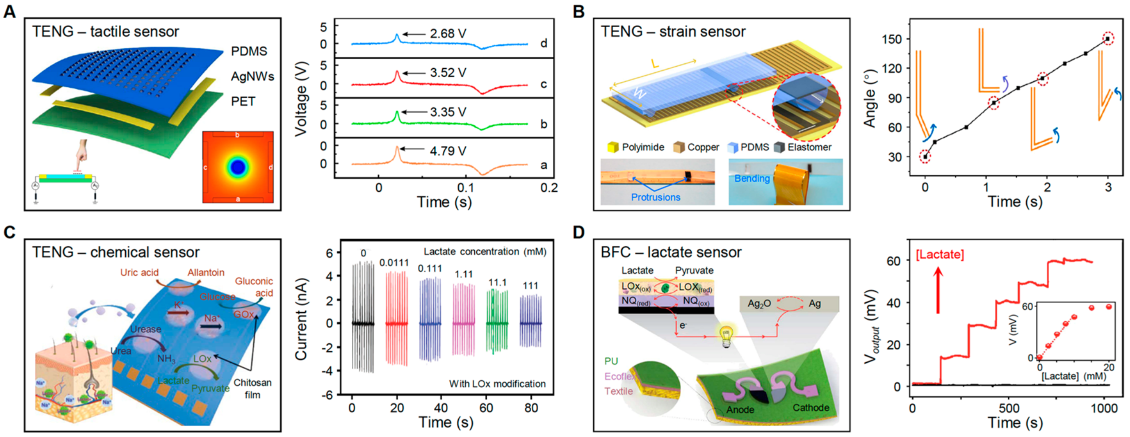

Figure 6. Wearable energy harvesters for active sensing. (A) Analogue smart skin as a tactile sensor for detecting location with high resolution. Reproduced with permission from ref 21. Copyright 2016 American Chemical Society. (B) Digitalized strain sensor with high linearity for static and dynamic measurement. Reproduced with permission from ref 55. Copyright 2017 Elsevier. (C) Electronic skin as a chemical sensor for real-time perspiration analysis. Reproduced with permission from ref 23. Copyright 2018 Royal Society of Chemistry. (D) Biofuel cell as a lactate sensor and the generated voltage signal for lactate sensing. Reproduced with permission from ref 22. Copyright 2016 Royal Society of Chemistry.

Chen et al. presented a microcable hybrid textile that integrates a TENG and a solar cell for converting biomechanical energy and ambient solar energy into electricity (Figure 5A).[52] Made of lightweight and low-cost polymer fibers, the hybrid power textile is highly deformable and breathable in response to both solar irradiance and human motion. Under sunlight exposure with mechanical excitation, it is capable of generating sub-milliwatt energy continuously.

Pairing biochemical and biomechanical harvesters is one means of scavenging energy efficiently and reliably from human activities, as shown in Figure 5B.[53] During human movements, the TENGs harvest biomechanical energy to generate motion-induced charge instantly, and the activated BFCs harvest biochemical energy from enzymatic reactions of human sweat for a continuous power delivery. With a judicious layout design, this wearable microgrid significantly enhances the sustainability and practicality of the self-powered wearable systems.

Through the optimization of both performance and layout, the hybridization of different energy harvesters will be beneficial to maintain a long-term operation for wearable devices. Since the outputs from hybrid energy harvesters are variable with distinct characteristics, an efficient power management strategy is highly desired to modulate the generated electricity.

4. WEARABLE HARVESTERS FOR ACTIVE SENSING

The advent of personalized healthcare requires the independent operation of multifunctional wearable biosensor networks. Emerging wearable energy harvesters are highly attractive for wearable applications: in addition to being the power sources for sensing networks, they can also act as active sensors. Electrical signals acquired from the energy harvesters can serve as indicators for physiological events.[13] This greatly simplifies the resulting biosensor network, making wearable devices more feasible for a broader range of applications. In this section, we will introduce the power outputs of wearable energy harvesters for the active sensing of both biophysical and biochemical parameters.

4.1. Nanogenerators for Active Sensing

TENGs can convert mechanical energy into electricity and can be easily fabricated from a variety of functional materials. They can be integrated into wearables to measure electrical outputs such as voltage, current, or frequency. These measurements can be used to obtain information about an individual's

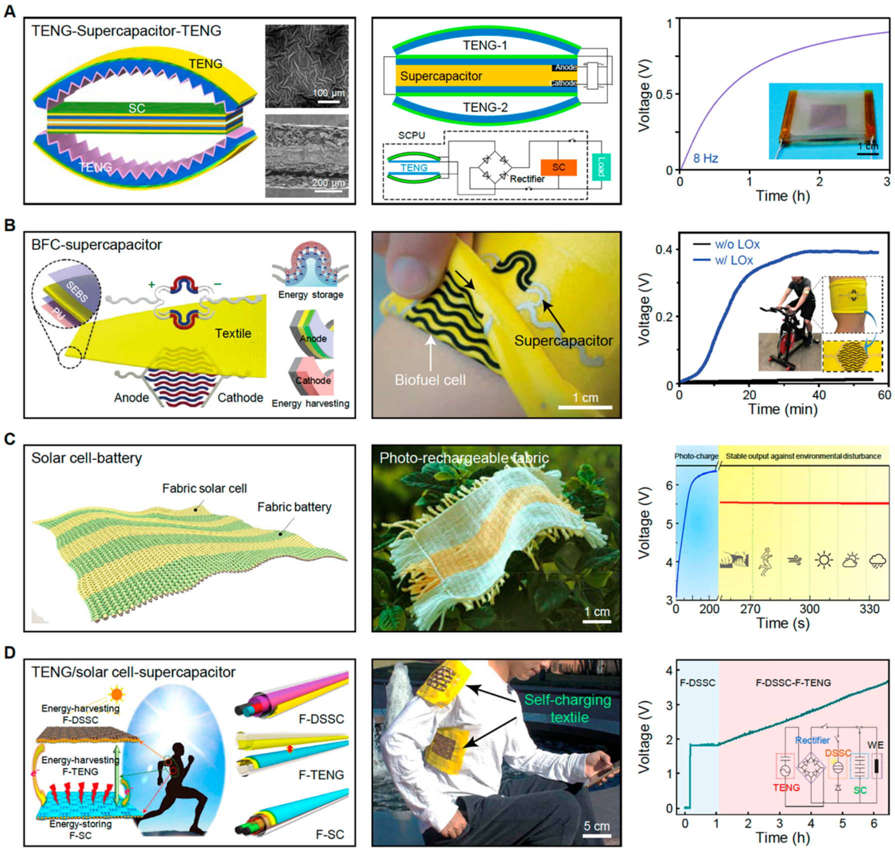

Figure 7. Wearable self-charging power units. (A) Sandwiched TENG-supercapacitor-TENG integrated power unit with efficient mechanical energy harvesting. Reproduced with permission from ref 60. Copyright 2016 Royal Society of Chemistry. (B) Wearable BFC-supercapacitor textile and real-time voltage charged during a constant cycling exercise. Reproduced with permission from ref 61. Copyright 2018 Royal Society of Chemistry. (C) Renewable energy fabrics based on a solar cell and rechargeable battery for powering body area sensor networks. Reproduced with permission from ref 62. Copyright 2020 Elsevier. (D) Fiber-shaped hybrid power textile with different operation modes under outdoor and indoor conditions. Reproduced with permission from ref 64. Copyright 2016 American Academy for the Advancement of Science.

position or frequency of movement and can be applied to health monitoring or human−machine interfaces.[54]

Enhanced spatial resolution and motion recognition are essential for TENGs when used as tactile sensors. Figure 6A shows one such sensor, an analogue smart skin used to detect the pressure and velocity of an object from a single-electrode TENG paired with an analogue localizing method.[21] Voltage ratios, obtained from an electrostatic induction between two electrodes, reflect the relative position and hence the direction of motion, with peak voltages indicating touching velocities. This analogue smart skin exhibits an especially high resolution and sensitivity.

TENG-based strain sensors can be used to detect vigorous human motion. Different from conventional resistive or capacitive strain sensors, active strain sensors can yield dynamic measurements by utilizing TENGs. These sensors are however sensitive to a different set of parameters and have unstable output signals, limiting their accuracy and long-term durability. Instead of analyzing voltage or current outputs during the stretching process, Su et al. proposed a digitalized strain sensor consisting of a flexible grated electrode and a stretchable elastomer with two specially designed protrusions (Figure 6B).[55] Digitalizing the obtained periodic signals from the TENG yields strain measurements with high sensitivity and accuracy. This active strain sensor allows for a real-time detection of elbow joint motion and an analysis of posture, indicating potential in prosthetic and biomedical applications.

With a proper surface modification and structural design, the TENGs can act as active chemical biosensors to monitor the concentrations of target biomarkers. Figure 6C depicts a TENG-based biosensing unit matrix functionalized with various enzymes for real-time perspiration analysis.[23] With the synergistic effect of triboelectrification and enzymatic reactions, this TENG-based chemical sensor can selectively analyze biomarkers (such as glucose, lactate, uric acid, urea, and ions) in sweat when driven by an elbow bending. Further integration of such enzymatic TENG-based biosensors with a visualization system will facilitate the development of a visualized healthcare warning and point-of-care diagnosis system.

4.2. BFC for Active Sensing

Biofuel cells convert biochemical energy in biofluids to electrical outputs through redox reactions with immobilized enzymes.[56] The generated power or current is usually proportional to the analyte concentration, so BFCs can simultaneously monitor the real-time level of specific biomolecules.[56] Figure 6D describes a stretchable textile-based BFC for noninvasive sweat monitoring.[22] With the synergistic effects of nanoengineered inks and serpentine designs, this printable textile-BFC can withstand repeated severe mechanical deformations with a minimal impact on its structural integrity. The output signal of the stretchable BFC is proportional to the lactate level in human sweat, which acts well as a selective and quantitative lactate sensor. Such a capability offers great promise for noninvasive self-powered continuous metabolic monitoring.

The combination of BFC and other energy harvesting strategies (TENG, PENG, and solar cell) can further promote the development of active biosensors for personalized health monitoring. It is of vital importance to improve the durability and output stability, which ensures the long-term use and sensitive monitoring for various signals. Meanwhile, through adopting novel structural designs, the integration of energy harvesters with other components (signal detection and data transmission) will eventually enable the applications of active sensors in wireless sensor networks. It should also be mentioned that, although such energy harvester-based active sensors could be self-powered, the data collection, transmission, or display will still require additional system-level configurations.

5. WEARABLE SELF-CHARGING POWER UNITS

The demands for wearable, sustainable, and environmentally friendly power supplies have subsequently increased over the last few decades. Frequently recharging or replacing traditional power supplies leads to an inconvenience and a high maintenance cost during a long-term operation. To address this, the integration of wearable energy harvesters with energy storage devices (e.g., supercapacitors and lithium ion batteries)[57] to create self-charging power units (SCPUs) has drawn tremendous attention.[58] This section introduces the progress of SCPUs by integrating single or hybrid energy harvesters with energy storage devices suitable for wearable electronics.

5.1. TENG-Based SCPU

TENG paired with a supercapacitor is a promising combination for harvesting and storing mechanical energy. Supercapacitors provide high power density, have a long lifespan, and can be integrated onto flexible substrates.[59] Since TENGs have an alternating current output, the general strategy is to convert the alternating current into a direct current with a bridge rectifier before charging an energy storage device. However, most of the SCPUs proposed are separate units, which reduce the power density and limit practical applications. Though the TENG and supercapacitors are not available to share electrodes, novel designs are developed to share the package or substrate to achieve an all-in-one layout with a high degree of integration. For example, Song et al. reported an SCPU with a wrinkled PDMS-based TENG and a CNT-paper-based supercapacitor (Figure 7A).[60] When compressive stress is applied, the SCPU can directly convert mechanical energy into electrochemical energy and store it in the supercapacitor. Such mechanical energy harvesters are extremely versatile in terms of material choice and structure design, making the SCPU wearable, biocompatible, and applicable to various service environments. Effectively harvesting and storing energies from daily human activities will further expand the longevity of wearable biosensors and make their application more realistic.

5.2. BFC-Based SCPU

Integration of enzymatic BFCs with suitable energy storage devices, like supercapacitors, will enable self-charging and promote overall device efficacy.[56] Figure 7B describes a textile-based stretchable BFC-supercapacitor SCPU, capable of harvesting and storing energy from human sweat.[61] Fabricated through the screen-printing method on both sides of the textile, the BFC on the inner side facing the skin is capable of harvesting sweat-lactate to produce biochemical energy. This energy is then stored in the supercapacitor on the outer side. Directly laminated on the arm of the human subject, the BFC can charge the supercapacitor to 0.4 V in 37 min during a cycling exercise. With a seamless integration and unique architecture, this textile-based SCPU opens future opportunities for healthcare, fitness, and security monitoring applications.

5.3. Solar Cell-Based SCPU

As solar energy is widely accessible, solar cell-based SCPUs represent an attractive solution to obtain a stable, continuous, and high-performance power during daily life for on-body wearable sensing. For example, Zhang and colleagues report a solar cell-based SCPU consisting of photorechargeable fabrics (with wire-type ZnO nanoarrays photoanode) and rechargeable Zn/MnO_2 battery textile (Figure 7C).[62] The fabric is able to deliver a high power of 0.1 mA for 10 min after 1 min of charging under sunlight, sufficient to power a body area sensor network including a humidity sensor, a temperature sensor, and a motion sensor.

5.4. Hybrid SCPU

Considering that multiple energy sources commonly coexist in real applications of wearable sensors, hybrid SCPUs provide an effective approach to optimize the power outputs and broaden the scope of applications.[63] Wen et al. presented a hybrid SCPU that can simultaneously collect energy from ambient light and random human motion that can subsequently be stored as chemical energy (Figure 7D).[64] On the basis of the all-fiber-shaped configuration, the SCPU can be easily woven into textiles to develop smart clothes. With impedance matching among solar cells, TENGs, and supercapacitors, the hybrid SCPU can achieve further improvements in the charging efficiency to sustainably operate wearable electronics.

The SCPU is available to harvest energy from common types of renewable energy resources and has shown the viability of an in situ storage, making it a promising approach to sustainably power sensors and systems.[65] Different integration strategies with diverse mechanisms allow more practical applications of SCPU, ranging from wearable sensors and personal healthcare to implantable electronics. In the future, power management strategies should be investigated to enable efficient energy transfer as well as innovative structure designs to improve system integration and allow for a large-scale fabrication.[65]

6. SELF-POWERED BATTERY-FREE WEARABLE SENSOR SYSTEMS

Wearable biosensors show great promise in the realm of noninvasive health monitoring, as they are able to detect various physiological indicators including both biophysical and biochemical signals. The realization of data acquisition, processing, and transfer from the biosensor interface typically requires a bulky and rigid battery, which is an inconvenience and is inadequate for long-term usability. A fully self-powered wireless wearable sensor system, which harvests energy from renewable and sustainable sources (e.g., human motion, light, body heat, and biofluids), is an attractive approach to enable the sustainable operation of biosensors. These integrated systems can communicate with the user through a visual display or modern wireless transmission technologies.

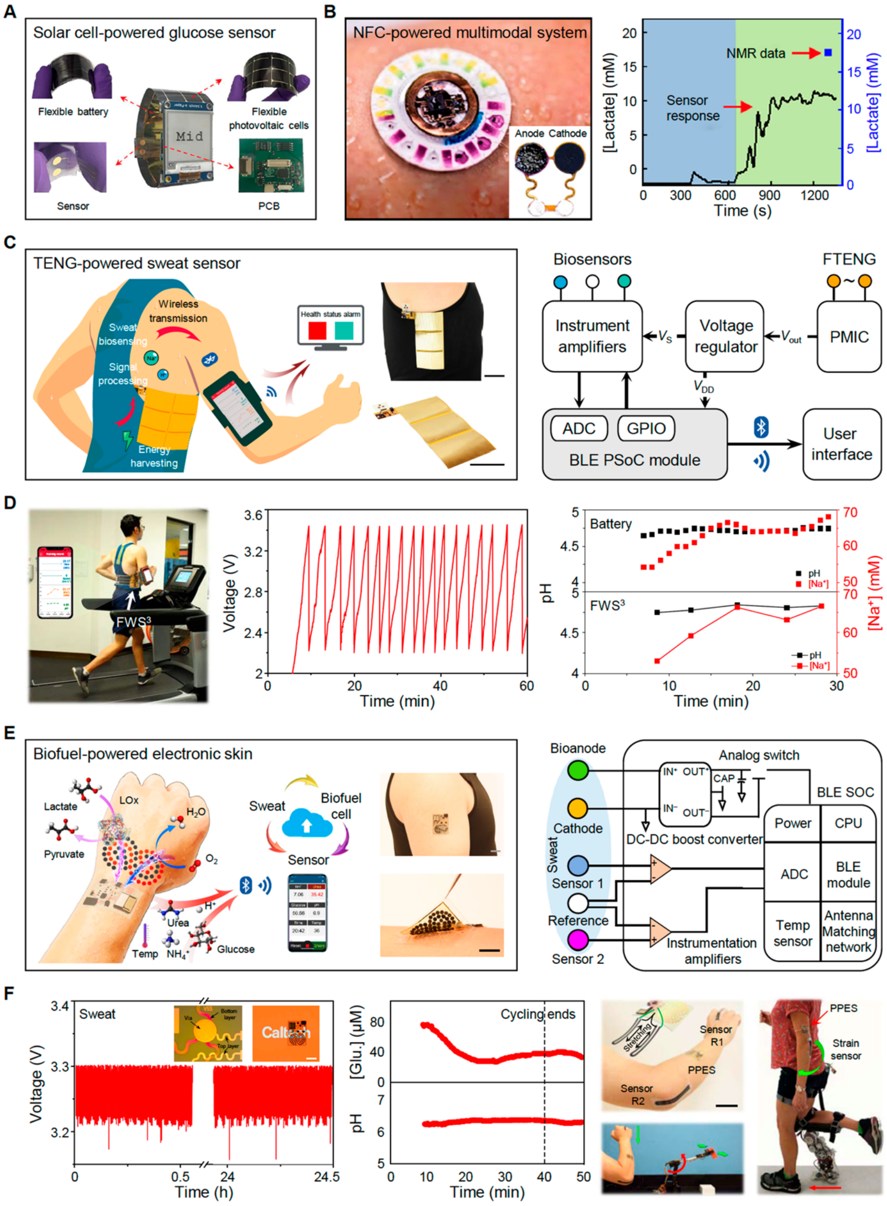

Figure 8. Fully integrated self-powered battery-free wireless wearable sensor systems. (A) Solar cell-powered sweat glucose sensor with visual e-ink display. Reproduced with permission from ref 66. Copyright 2019 American Chemical Society. (B) Battery-free, skin-attachable chemical sensing platform. Reproduced with permission from ref 19. Copyright 2019 American Academy for the Advancement of Science. (C) Wireless wearable sensor powered by human motion to analyze sweat biomarkers for personalized healthcare. (D) On-body evaluation of the FTENG-powered wearable sweat sensor system for wireless, dynamic perspiration analysis. (C, D) Reproduced with permission from ref 18. Copyright 2020 American Academy for the Advancement of Science. (E) Biofuel-powered soft electronic skin for multiplexed wireless sensing and human–machine interfaces. (F) On-body validation of the biofuel-powered electronic skin for personalized metabolic monitoring and robotic assistance. (E, F) Reproduced with permission from ref 17. Copyright 2020 American Academy for the Advancement of Science.

6.1. Self-Powered Sensor System with Visual Display

Recent studies have shown that such wearable energy harvesters, when integrated with a digital watch or a light-emitting diode (LED) or e-ink screen, can realize a self-sustainable wearable sensing system with a visual display. For example, a fully integrated smart watch powered by a solar cell was demonstrated to monitor sweat glucose continuously during exercise (Figure 8A). A Zn-MnO_2 battery was used to store the harvested solar energy and power an electronic ink screen to display the qualitative sweat glucose data from an enzymatic glucose sensor.[66]

6.2. Self-Powered Sensor System with near-Field Communication (NFC)

NFC is an effective strategy to realize a battery-free wearable system, as it could enable wireless power delivery as well as data transmission. To continuously obtain quantitative physiological information, Bandodkar et al. reported a battery-free multimodal wearable sensor system that can simultaneously perform electrochemical, colorimetric, and volumetric sweat analyses (Figure 8B).[19] The detection of sweat glucose and lactate was achieved using BFCs as active sensors. Signal processing and wireless data transmission between the wearable sensor and the user interfaces (e.g., smartphone) was enabled by NFC.

6.3. Self-Powered Sensor System with Bluetooth Communication

Considering that NFC could suffer from a short operation distance, Bluetooth is a more attractive wireless communication choice for a large number of wearable applications. For such self-powered wireless systems, although signal processing and sensor operation could consume relatively low power (∼μA level), Bluetooth communication usually leads to high power consumption (∼mW level) and poses a high requirement in energy harvesting and power management efficiencies.[17,18] Song et al. proposed a fully self-powered wearable system consisting of a multipanel FTENG array, a microfluidic sweat sensor patch, and a power management circuit module (Figure 8C).[18] The FTENG array generates a high power output of ∼416 mW m^{-2}, which is temporarily stored in a capacitor. The system can be periodically woken for performing a biochemical measurement and transmitting the collected data. Through a seamless integration and efficient power management, this TENG-powered wearable sensor system allows for a real-time detection of the molecular level (sodium and pH) in sweat wirelessly during on-body evaluation (Figure 8D).

BFCs are another energy conversion method gaining popularity in the bioelectronic field due to their capability of on-site power generation. Molecular biomarkers in sweat can also serve as an ideal and sustainable energy source for BFCs. Figure 8E describes a fully integrated BFC-powered electronic skin for the continuous monitoring of key metabolic analytes (i.e., glucose, pH, urea, and NH_4^+).[17] The biochemical information collected from the biosensors can be wirelessly transmitted to the user interface via Bluetooth Low Energy. As shown in Figure 8F, this battery-free biofuel-powered system demonstrates stable performance in energy harvesting and health monitoring during prolonged physical activities. In addition, the BFC-powered electronic skin can serve as a human−machine interface for robotic assistance when integrated with soft strain sensors.

Self-powered wearable sensor systems based on various mechanisms of energy harvesting allow them them to operate continuously without even needing a battery to address the challenge of energy requirements. The development of multimodal, fully integrated, and self-powered wearable sensor systems paves a feasible way to personalized healthcare and prosthetic assistance.

7. CONCLUSIONS AND OUTLOOK

In this Account, we have reviewed and highlighted the recent advances in self-powered wearable sensors. Novel structural designs of wearable biosensors coupled with functional micro/nanomaterials enable a real-time monitoring of health status at both biophysical and biochemical levels. The seamless integration of wearable energy harvesters and biosensors allows for self-powered wearable sensor systems. The advances of self-powered wearable sensor systems shed a light on personalized healthcare, from real-time health monitoring and human−machine interfaces to on-demand therapeutics.

Despite the considerable achievements made recently in this field, important challenges remain to be addressed for self-powered wearable sensors in practical biomedical applications. First, further improvements on the selectivity, sensitivity, repeatability, and stability as well as the mechanical reliability of biosensors are highly desired to real-life multimodal biosensing, especially toward human activity tracking and continuous health monitoring. Second, innovations in materials, structural designs, and system integration are crucial to make the key components applicable for a scale-up fabrication. For example, functional nanomaterials and design configurations compatible with advanced printing technologies such as inkjet printing, 3D printing, and roll-to-roll printing should be explored toward mass-producible, high-performance, and low-cost self-powered wearable systems. Third, the hybridization of different energy harvesters and high-efficiency power management strategies will be beneficial in enhancing the adaptability of self-powered wearable sensors in complicated conditions. At the same time, it is important to adopt both circuit modules and biosensors with low power consumption to further decrease the required power capacity. An integrated circuit design, in this regard, is an attractive solution to achieve miniaturized ultralow-power systems with efficient energy harvesting and signal processing capabilities.[67] Last but not least, an evaluation of the integrated self-powered wearable sensor systems in large-scale human trials is crucial to validate the usability of these systems in practical applications. Interdisciplinary collaborations across materials, chemistry, engineering, and clinical medicine fields will be crucial to realize the full potential of self-powered wearable sensors. The large-scale multimodal data collected from the cohort studies, coupled with modern data mining approaches, could open the door to numerous personalized healthcare applications.

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