Battery-free wearable electrochemical sweat sensors

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Abstract**—** Broad adoption of wearable sensor devices that continuously and unobtrusively monitor physiochemical biomarkers can enable personalized healthcare through big data and predictive algorithms. Wearable sweat sensing offers an attractive means of biochemical screening among large populations as sweat is rich in biomarkers. However, sustainable powering of wearable sweat sensor devices is a challenge as autonomous sweat extraction, multiplexed biomarker detection, data processing, and data communication consume significant energy and typically require bulky lithium batteries. Various energy harvesting modules such as triboelectric nanogenerators (TENGs), biofuel cells (BFCs), and solar cells (SCs) have been judiciously designed and integrated into wearable platforms for battery-free analysis of sweat biomarkers.

I. INTRODUCTION

The shift towards personalized and remote healthcare has accelerated the development of wearable devices that can continuously monitor physical vital signs as well as biochemical markers [1], [2]. Analyzing various biomarker levels in sweat among vast populations can provide a comprehensive understanding of an individual's health. As datasets compile, big data and predictive algorithms can be used to achieve personalized healthcare, where healthcare interventions can be tailored to meet the individual's specific needs. However, to achieve this vision, it is fundamental to develop unobtrusive wearable sweat sensor devices that can achieve (1) continuous access to sweat, (2) accurate detection of a wide range of biomarkers, (3) efficient data processing and communication, and (4) sustainable powering over long periods and among large populations.

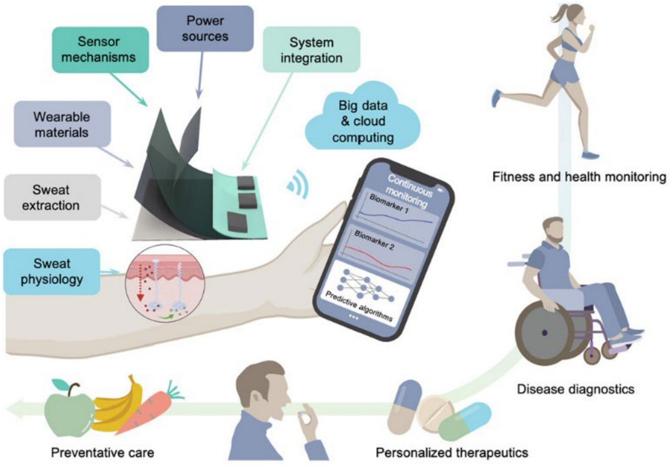

Figure 1. Overview of wearable sweat sensors [1].

Firstly, sweat sensors require a continuous stream of flowing sweat to perform continuous measurements. Sweat can be induced through physical exercise for fitness tracking applications, or chemically through iontophoresis for health monitoring during sedentary activities. Natural sweat from the fingertips can even be sampled in some cases, but in very low volumes [3]. While exercise-induced sweat requires high exertion from the individual, iontophoresis-induced sweat requires high power consumption from the wearable electronic system. Secondly, most wearable sweat sensors are based on potentiometric ion-selective electrodes or amperometric enzyme electrodes, limiting the range of detectable biomarkers. Novel sensors incorporating alternate recognition elements such as MIPs or aptamers, and more complex electrochemical detection mechanisms such as voltammetry and impedimetry can expand the range of detectable biomarkers [4], [5]. Furthermore, individualized factors such as sweat rate or sweat pH can influence the secretion of other biomarkers or influence response of electrochemical sensors, confounding sensor readings. Therefore, multiplexed detection of various biomarkers can enable the cross-calibration of measurement results for more accurate biomarker analysis. Incorporation of more complex measurement techniques and increasing the number of measurements channels, however, increase the power demand for the electronic circuit.

Effective data communication is crucial for practical use of wearable sweat sensors. Colorimetric sensors exhibit a color change in response to a target analyte that can be detected by the naked eye, thereby eliminating the need for any electronic circuitry [6]. While they can provide information regarding biomarker levels at a specific point in time, they don't have the capability to track biomarker trends autonomously and continuously over time. NFC-based communication is attractive for wearable sweat sensor devices because mobile phones can be used as NFC readers to both power the wearable circuitry and acquire sensor readings without the need for a battery [7]. While these devices can be battery-free, they require a mobile phone to be in close proximity to the wearable device for operation, making it inconvenient for long-term continuous measurements. Bluetooth-based communication provides the most attractive means of data communication because sensor data can be wirelessly and autonomously recorded on a mobile phone over farther distances. However, Bluetooth low energy (BLE) communication consumes a significant amount of power, typically requiring a battery for continuous operation. While most wearable devices are currently powered by lithium batteries, lithium batteries are bulky, require frequent charging, and can pose a toll on the environment. Through judicious system integration, energy harvesting devices such as triboelectric nanogenerators (TENGs), biofuel cells (BFCs), and solar cells (SCs) can serve as attractive alternatives to batteries for sustainable powering of wearable devices [8]–[11].

II. HARVESTING ENERGY FOR WEARABLE SWEAT SENSORS

A. Harvesting energy from human motion

TENGs can be used to convert the mechanical energy from human motion to electrical energy via the triboelectric effect. A highly efficient wearable freestanding-mode TENG was paired with low-power electronic circuitry and a microfluidic sweat sensor patch for the battery-free and wireless (BLE) monitoring of sweat biomarkers including pH and Na+ during exercise [8]. A freestanding-mode TENG, fabricated via commercial FPCB manufacturing processes, consists of two main components: a stator and a slider. The stator, which is directly connected to the energy harvesting circuitry, is placed on the side torso, and the freestanding slider is fixed on the inner arm. Arm swing motion during exercises such as running or rowing lead the stator and slider to rub against each other, generating power in the form of a high voltage AC signal.

The high voltage AC output is rectified to a DC voltage using a bridge rectifier to charge an energy storage capacitor to an upper threshold voltage, initiating the low power circuitry to perform sensor measurements. However, as the TENG power output during exercise can be erratic and dependent on arm swing frequency, it is difficult to perform measurements and BLE data transmission at a fixed interval. Instead, an intermittent operation scheme was implemented, where the low power circuity is typically shut off, and intermittently started up to perform a measurement and transmission operation whenever the energy storage capacitor gets charged to an upper threshold voltage. Therefore, the measurement and data transmission interval of such a system is dependent on exercise intensity and consistency.

For onbody validation of the TENG-powered wearable sweat sensor, a healthy subject ran on a treadmill for an hour, during which the system was able to perform measurement and transmission events at an interval of 2~4 minutes, depending on how fast the energy storage capacitor was charged. Furthermore, the device was used to successfully monitor sweat pH and sodium levels. Such a system is very robust and cost efficient because it can be mass manufactured using commercial FPCB technology. It is therefore suitable for monitoring and alerting dehydration in runners. However, as it requires vigorous exercise for operation and operates discontinuously, its applications may be limited to fitness monitoring.

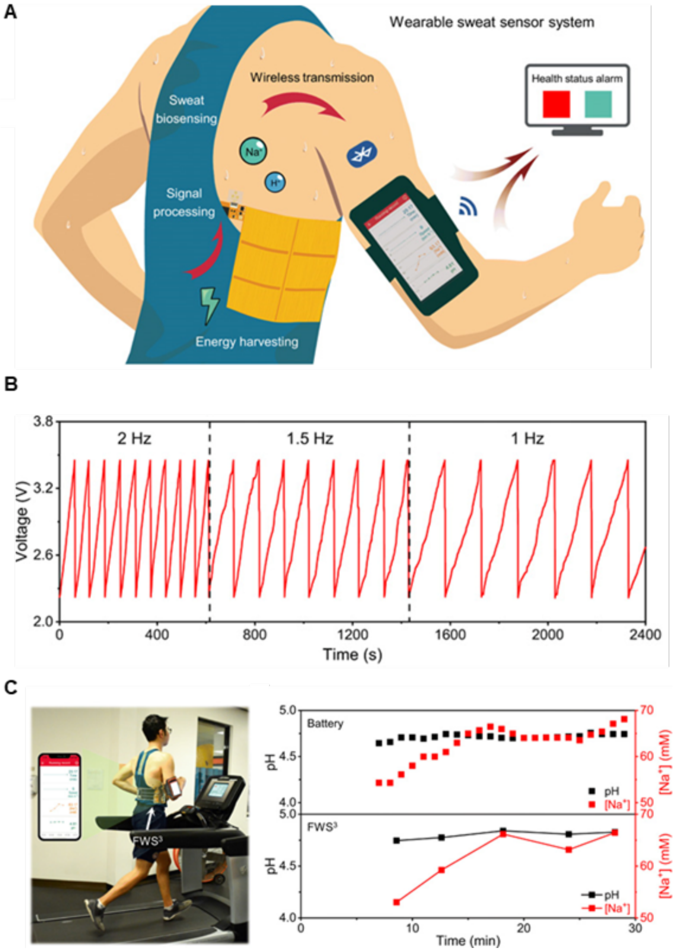

Figure 2. a) Illustration of FTENG powered wearable sweat sensing platform for monitoring sweat pH and Na^+ during exercise [8]. b) Real-time potential of energy storage capacitor when charged by the FTENG under varying working frequencies. c) Onbody validation of the wearable platform for monitoring sweat pH and Na^+ while running on a treadmill.

B. Harvesting energy from human sweat

BFCs employ enzymes as catalysts to generate electricity from redox-active metabolites such as lactate and glucose. For powering sweat sensors, lactate BFCs are an ideal candidate as sweat is rich in lactate. Lactate biofuel cells comprise an anode that is immobilized with a redox mediator and LOx enzyme for oxidation of lactate into pruvate, and a complementary cathode that facilitates the reduction of oxygen into water. A highly stable and efficient wearable lactate BFC that can generate up to 3.5 mW cm–2 from natural sweat was developed through the monolithic integration of zero-dimensional (0D) to 3D nanomaterials [9]. This BFC was able to continuously power a battery-free electronic skin system for multiplexed biosensing and Bluetooth data transmission.

As the BFC outputs a low voltage DC signal, a power management circuit was used for boost converting the BFC output with maximum power point tracking to charge an energy storage capacitor. One the capacitor was charged to an upper threshold voltage, the electronic system was continuously powered to perform multiplexed potentiometric measurements and data transmission at a fixed interval of 15 s, as long as the capacitor didn't discharge below the lower threshold voltage. The BFC-powered e-skin system was validated through onbody biking trials, where the device was able to monitor two analytes along with skin temperature continuously and simultaneously. In an intake study, urea and ammonium were monitored two hours before and after protein intake.

Biofuel cells offer a sustainable and environmentally friend approach to generating electricity. Furthermore, BFC powered battery-free systems can operate continuously as long as there is exposure to sweat with sufficient lactate concentrations. However, BFCs still require significant sweat volumes produced by exercise to initiate the energy harvesting, therefore making it difficult to monitor biomarkers continuously throughout the day even while performing sedentary tasks. While BFCs scavenging energy from natural perspiration have been developed, low sweat volumes limit their power output.

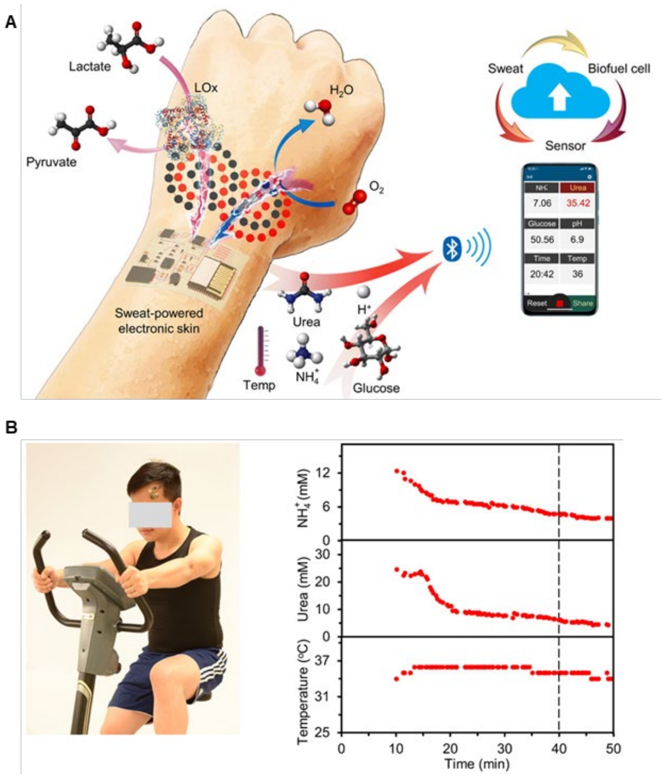

Figure 3. a) Schematic of a BFC powered e-skin for continuous monitoring of sweat biomarkers [9]. b) Onbody validation of the BFC powered e-skin for continuous monitoring of sweat NH_4^+, sweat urea, and skin temperature during cycling.

C. Harvesting energy from the environment

While TENG and BFC-powered wearable sweat sensor devices were able to sustainably harvest energy from body motion and body fuel, they require exercise for power generation and have limited power output. Such systems are limited to performing simple electrochemical measurements such as potentiometry during exercise, making it difficult to monitor a wide range of biomarkers while performing various activities and account for personalized factors such as sweat rate or sweat pH. Power from ambient light such as sunlight and indoor light is ubiquitous and readily accessible during daily activities. Silicon-based solar cells are commonly used for harvesting energy from sunlight but are often fragile and rigid. Additionally, while they perform great under sunlight, their efficiencies are low under indoor lighting because indoor lighting sources have a narrower emission spectrum that don't align well with the external quantum efficiency of typical crystalline silicon solar cells.

To effectively power a multi-functional wearable system, a flexible perovskite solar cell (FPSC) was designed to generate a high power density under various lighting conditions, and be flexible and robust enough to endure the mechanical stress and exposure to sweat during on-body wear and tear [10]. The judiciously engineered FPSC achieved a high power conversion efficiency (PCE) of around 15% under sunlight, and a record breaking PCE of 31% under 600 lux indoor lighting. Furthermore, the FPCB was able to withstand thousands of bending cycles (5 cm bending radius) with minimal reduction in performance and negligible leakage of lead.

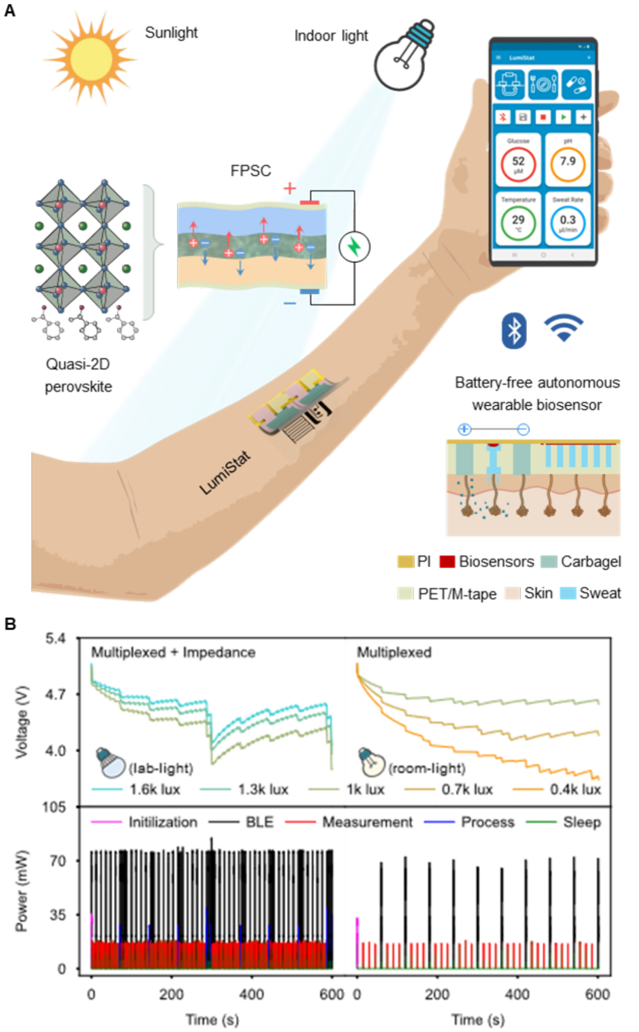

Figure 4. a) Illustration of FPSC powered autonomous wearable sweat sensor device [10]. b) Power consumption profile of wearable electronic system while performing multiplexed biomarker measurements (8 s interval) and sweat rate

measurements (5 min interval), and multiplexed biomarker measurements (1 min interval) (bottom). Corresponding real time potential of energy storage capacitor while system is exposed to different indoor light intensities (top).

The high power output of the FPSC under various illumination conditions enabled the development of a miniaturized wearable electronic system (LumiStat) that can perform autonomous iontophoretic sweat extraction throughout the day while performing multimodal sweat analysis using a wide range of electrochemical measurement techniques including potentiometry, amperometry, voltammetry, and impedimetry. The capacity for employing a wide range of measurement techniques enabled the simultaneous monitoring and cross-calibration of sweat glucose, sweat sodium, sweat pH, skin temperature, and sweat rate.

The power management scheme of the LumiStat is similar to that of the BFC powered system, where the FPSC output is boost converted to charge a capacitor to an upper threshold voltage, whereafter the system can operate continuously. However, as the FPSC can output high power without the need for exercise, it was possible to incorporate a constant current iontophoresis module for carbachol-based autonomous sweat induction. Moreover, the microfluidic system was optimized for maximal and prolonged sweat extraction. Furthermore, due to the extremely low power consumption of the electronic system, the LumiStat was able to perform wireless and multiplexed measurements of sweat glucose, sweat pH, sweat sodium, and skin temperature at an interval of 8s, as well as sweat rate measurements at an interval of 5 min under indoor lighting as low as 1000 lux. With extended measurement and transmission intervals, the system was able to perform multiplexed measurements under indoor lighting as low as 400 lux.

As such, the LumiStat can adjust its data measurement and transmission interval to tune to power consumption according to light exposure. For onbody validation, the battery-free wearable device was used to simultaneously monitor various sweat biomarker levels throughout the day under various lighting conditions ranging from outdoor sunlight to indoor room light while a subject performed various daily activities ranging from exercise to sleep. Throughout the 12 hour period, the system was able to continuously extract a steady stream of sweat and acquire accurate biomarker trend cross-calibrated in real time based on personalized factors. Glucose sensor readings were calibrated by sweat pH and skin temperature readings, and the sweat rate sensor readings were calibrated by ionic strength and skin temperature readings to improve the accuracy of biomarker analysis.

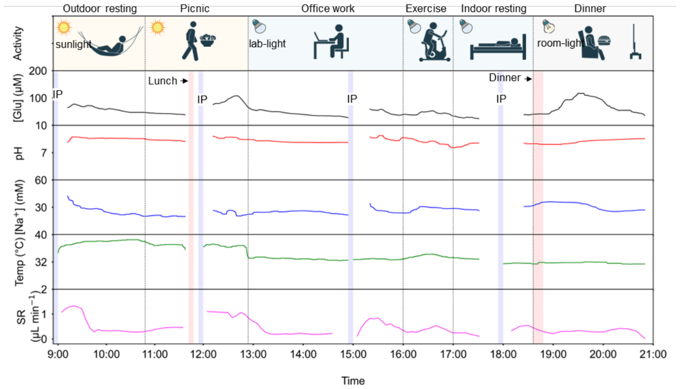

Figure 5. Full-day cross-activity multimodal sweat monitoring with the wearable device while exposed to various light conditions [10].

III. CONCLUSION & OUTLOOK

To conclude, various energy harvesting modalities have to potential to sustainably power wearable devices for the continuous detection and wireless transmission sweat biomarkers. TENG and BFC powered devices can effectively harvest energy from our body, but generally require exercise for sweat extraction and operation. Solar cells, on the other hand, can power wearable systems during sedentary activities to autonomously induce sweat for continuous sweat sensing. However, solar cells require light exposure for operation, but certain activities or scenarios may block out light exposure. To address this, it may be interesting to combine various energy harvesting modalities to extend the operation window of battery-free devices. Another approach would be to utilize energy harvesting modules for recharging next-generation batteries that are environmentally friendly and flexible to meet the demands of sustainable powering and user friendliness.

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