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Flat enzyme-based lactate biofuel cell integrated with power management system: Towards long term *in situ* power supply for wearable sensors

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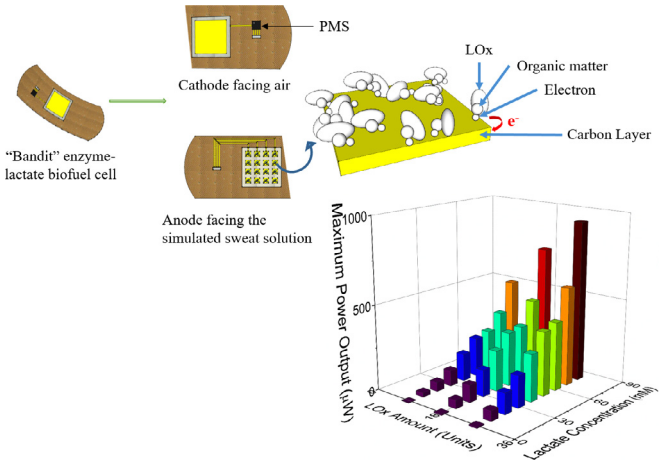
HIGHLIGHTS

- Maximum power output of ELBC (27 U LOx with 80 Lactate) reached at 938 μW.
- Michaelis-Menton kinetic models of ELBCs were established.
- LOx (6U-54U) was determined for lactate concentration (10–40 mM) in human sweat.
- Data transfer frequency (1/h to 1/min) can be supported by the ELBC-PMS entity.
- Batch-mode ELBCs exhibited a stable voltage output and a long lifespan (2 weeks).

GRAPHICAL ABSTRACT



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ABSTRACT

Flat enzyme-based lactate biofuel cell (ELBC) integrated with power management system (PMS) was developed as a potential power supply for wearable sensors. Kinetic models of power output and chemical reagent concentration (lactate oxidase LOx and lactate) were developed to determine the limiting factor of the ELBC performance. Given the lactate concentration (0–40 mM) in human sweat, the optimum LOx amount coated on the anode ranged from 6U to 54U based on the experimental maximum power output. In the ELBC-PMS entity simulation test, power discharge/recharge frequency was calculated for self-sustained sensing and data transmission, indicating that the ELBC could support wearable sensors (power requirement: 1–1350 mW; signal transfer frequency: 1–1320 times/h). The electrochemical activity of LOx coated anode was validated using the cyclic voltammetry (CV). The coefficient of variance and regression statistical analysis revealed the high stability and long lifespan (2 weeks) of ELBC without the need of lactate refill.

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1. Introduction

Wearable sensors (e.g. skin/chest electrodes, arm cuff-based monitor, and pulse oximeter) have been used to collect physiological (e.g. blood pressure, respiration rate and oxygen saturation) and movement data and enable the real-time monitoring of human health [1]. Durable and stable power supply for wearable sensors is a critical challenge [2–4], since existing rechargeable battery has three major problems [5–7]. First, due to the finite lifespan (500–800 charge cycles) of rechargeable battery [8], its replacement could destroy the whole sensor device. Second, it is inconvenient to periodically take the sensors off for recharging. Specially, the frequency of present wearable devices charging is high (once per 3–12 days) [9–11]. Third, charging battery could be harmful for the patients with wearable sensors to monitor heart rate and blood pressure. Therefore, there is an urgent need to explore self-sustained stable power supply for wearable sensors.

Last decade has seen a rapid development of harvesting tiny amount of energy in the form of human body's heat, movement, metabolism and vibrations [12–18]. Bio-electromechanical systems (BES) has a great potential to extract human-generated energy through chemical degradation (e.g. glucose, carbohydrate, lactate) [14,15,19]. However, there are three challenges of BES as power supply for wearable sensors [14,15,20–29]. First, BESs generally utilize anaerobic electrogenic bacteria (e.g. *Geobacter sulfurreducens, Escherichia coli* and *Shewanella putrefaciens*) [26,29,30] as the anodes, which cannot be directly applied on human skin. Second, BESs have the size of at least 10–400 mL scale with rigid configurations, including single chamber, two chambers and/or tubular chamber [21–23,29,31]. This bulky structure is unsuitable for deployment on human body. Third, the power output of BES ranges from 1 to 900 mW/m^2 [14,15,26,32], meaning that the total power output is less than 810 μW (with the electrode area of 9 cm^2), which is insufficient to support wearable sensors (power retirement: 1–100 mW) [24,25].

A new enzyme-based biofuel cell (lactate oxidase enzyme, LOx) named as "tattoo biosensor" with a flat configuration was developed to solve the wearable battery problem and can be directly applied to human skin [14,17,18,33]. The voltage of the tattoo biosensor was generated by LOx oxidizing lactate contained in human sweat, and closely related with lactate concentration. The mechanism of the tattoo biosensor was based on the half-reaction [34]:

$$L-lactate \overset{LOx}{\rightarrow} pyruvate + 2H^+ + 2e^- \quad (1)$$

Although the tattoo biosensor avoided the need for external chemical besides human sweat, the power output of the tattoo biosensor was low (less than 4 μW, with power density of 50–100 μW/cm^2 and anode area of 6 mm^2) [14,17,18,35] due to the conventional three-electrode structure (working, counter and reference electrodes) inefficient at power generation and the small anode area (about 6 mm^2) less than 0.5% of the whole biosensor area (around 20 cm^2). In addition, the LOx amount was about 12U, while a wide range (2U-50U) of LOx had been reported for enzyme-based biofuel cells [17,34,36], and the power densities of tattoo biosensors were obtained at different concentration of lactate (8–20 mM) [17,33,35]. However, no model between the maximum power densities and the lactate concentration/ LO_X amount has been established, and the optimum amount of LOx on the anode for the maximum power supply has not been determined yet.

The scientific novelty of this study was to investigate the concept of enzyme-based lactate biofuel cell (termed as ELBC) integrated with power management system (PMS) that enables the effective harvest of the energy generated from human sweat as the *in situ* power supply. None of the existing lactate-based biofuel cell reaches the high power output as the ELBC. The novel flat "bandit" enzyme-based biofuel cell with enzyme-loaded anode facing to the simulated sweat and cathode facing to air (Fig. 1a) capable of converting sweat to *in situ* power supply for wearable sensors was expected to solve the problem of tattoo biosensors. In this study, current knowledge gap of converting sweat to *in situ* power supply for wearable sensors [20–31,37] was narrowed by establishing a model between LOx amount/lactate concentration in the simulated human sweat and the maximum power density, simulating the power discharge/recharge frequency of the ELBC-PMS entity and validating the stability and lifetime of the ELBC. Furthermore, due to the high cost of LOx (e.g. 500U LOx $857), the model developed would provide an accurate estimation for LOx usage based on the power requirement.

There were four main tasks in this study. First, the voltage and power output of ELBCs were correlated with LOx amount and lactate concentration. Kinetic models of power output and enzyme amount and/or lactate concentration were established to elucidate the limiting factor of ELBC performance. Second, the power discharge/recharge frequency of the ELBC-PMS entity was simulated to explore the long-term self-sustained sensing and data transmission. Third, the stability and the lifetime of the batch-mode ELBC were determined using the statistical analysis. Fourth, the electrochemical activity of LOx coated anode was determined using the cyclic voltammetry (CV) and the electrode surface characterization.

2. Materials and methods

2.1. Simulated sweat solution and LOx loaded anode

In this study, all the tests were conducted in the simulated sweat solution instead of real human skin. Carbon cloth (CCP10, 0.38 mm thickness, Fuel Cell Earth Inc.) was used as anode/cathode electrode material. The electrolyte solution contained 84 μM creatinine (Sigma-Aldrich Co.), 10 μM ascorbic acid (>99.9%, Fisher Scientific Co.), 0.17 mM glucose (Fisher Scientific Co.) and 59 μM uric acid (≥99%, crystalline, Sigma-Aldrich Co.) to simulate the human sweat as previously reported [38]. The concentration of lactate (Syrup, 60% w/w, Sigma-Aldrich Co.), the predominant organic constituent in human sweat [14,17,33,39–41] was tested at 0–80 mM. Lactate oxidase (LOx) (activity, 20 U/mg) solution was made by mixing LOx (40 mg mL^{-1}, EC #232-841-6, L-1175, AG Scientific Co.) with BSA (Bovine Serum Albinum) solution (stabilizer 10 mg mL^{-1}, ≥96% agarose gel electrophoresis, Sigma-Aldrich Co.) and then dropped on carbon cloth (anode: 1 cm × 1 cm).

2.2. Fabrication of ELBC

The anode and cathode of the flat compact ELBC were fabricated on carbon cloth (non-wet proofing, Type-A, E-Tek, FL), which was widely used in the fuel cell. The anode side of the carbon cloth (1 cm × 1 cm) was coated by LOx solution and then immobilization as previously reported [14,17,33]. Three types of anodes were made with three LOx amounts (15, 30 and 45 μL that were referred to as 12, 24 and 36U in the following sections). The cathode side of carbon cloth (3 cm × 3 cm) was made by coating 4-layer polytetrafluoroethylene (PTFE) on the outside side facing air and coating Platinum (Pt: 0.5 mg cm^{-2}) on the inner side facing the simulated sweat solution [29]. A non-conductive filter paper (3 cm × 3 cm, P8, Fisher Science) was inserted between the anode carbon cloth and the cathode carbon cloth to prevent short circuit. The anode carbon cloth, filter paper and cathode carbon cloth were stacked tightly to form a compact ELBC entity (Fig. 1b), and was then inserted into a chamber (working volume: 30 mL) made by

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