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Air bio-battery with a gas/liquid porous diaphragm cell for medical and health care devices

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ABSTRACT

Powering future generations of medical and health care devices mandates the transcutaneous transfer of energy or harvesting energy from the human body fluid. Glucose-driven bio fuel cells (bio-batteries) demonstrate promise as they produce electrical energy from glucose, which is a substrate presents in physiological fluids. Enzymatic biofuel cells can convert chemical energy into electrical energy using enzymes as catalysts. In this study, an air bio-battery was developed for healthcare and medical applications, consisting of a glucose-driven enzymatic biofuel cell using a direct gas-permeable membrane or a gas/liquid porous diaphragm. The power generation characteristics included a maximum current density of 285 $\mu\text{A}/\text{cm}^2$ and maximum power density of 70.7 $\mu\text{W}/\text{cm}^2$ in the presence of 5 mmol/L of glucose in solution. In addition, high-performance, long-term-stabilized power generation was achieved using the gas/liquid porous diaphragm for the reactions between oxygen and enzyme. This system can be powered using 5 mmol/L of glucose, the value of which is similar to that of the blood sugar range in humans.

1. Introduction

Beginning in the 1960s, extracorporeal pacemakers, various wearable devices and implantable medical devices have been developed in the fields of healthcare and medical science fields (Bandodkar and Wang, 2014; Hayes and Furman, 2004; Steinberg et al., 2016; Svechtarova et al., 2016). Several of these devices operate with an external power supply such as a lithium battery, leading to issues during the charging or exchanging of batteries (Nguyen et al., 2014). Even commercialized low-power-consumption pacemakers reportedly require a stable power supply of greater than or equal to 50 μW to power medical equipment (Ayano et al., 2013; Mallela et al., 2004). Hence, it is crucial to develop a novel battery capable of continuous power supply, which can maintain at least these levels for medical use.

On the other hand, enzymes and microorganisms can be cost-effectively procured in large amounts via genetic engineering. By using biochemical materials, genetic engineering can also be applied for portable power supplies and implantable-type power supplies (Deng et al., 2010; Miyake et al., 2011; Yu and Scott, 2010). Katz's group has reported the feasibility of embedding a biofuel cell in two living lobsters, which generates an electromotive force of 1.2 V (MacVittie et al.,

2013). Embedded fuel cell can be also operated an electronic watch with the generated power. In addition, embedded fuel cells can be operated by an electronic watch with the generated power. In addition, Gale's group has reportedly combined biofuel cells and contact lenses (Reid et al., 2015). The fabricated contact lens biofuel cells exhibit a maximum power density of $8.01 \pm 1.4 \mu\text{W}/\text{cm}^2$, which is obtained with synthetic tears. Along with these developments, biofuel cells have received increasing international attention from the research community (Matzeu et al., 2015). However, the research and practical applications of biofuel cells remains in the developmental stage, and several issues and disadvantages have been reported in terms of battery performance, such as output and lifetime, thus far (Moehlenbrock and Minteer, 2008).

Biological cells are generators that imitate the mechanism of living organisms and convert chemical energy in the body into electrical energy. For example, enzymatic fuel cells use an enzyme known as a biocatalyst. The enzyme generates electrical energy via the oxidation and reduction of sugar and acid, which are biological components; hence, it can be applied as a power source for the above-mentioned equipment (Miyake et al., 2011). Enzymatic batteries that generate electricity at glucose concentrations corresponding to blood sugar

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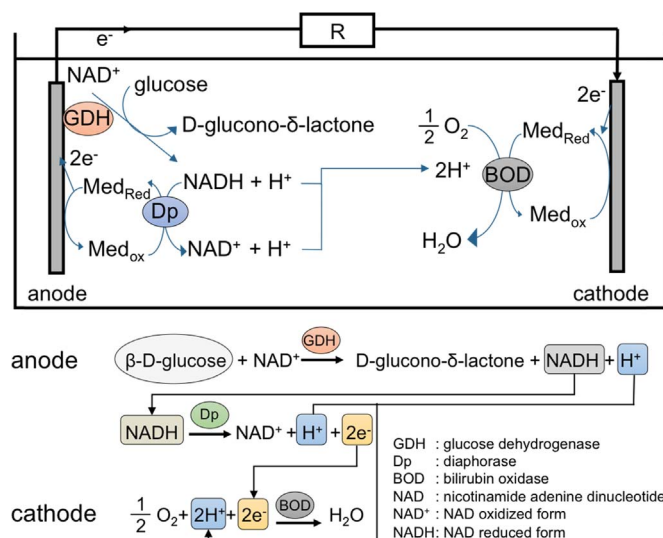


Fig. 1. Schematic illustration of the principles of used to develop the air bio-battery with glucose solution.

levels have already been reported; however, issues of low output and stability have been noted (Supplemental Table 1) (Habrioux et al., 2008; Milton et al., 2015; Pan et al., 2010; Scherbahn et al., 2014).

In this study, a high-performance enzyme-based bio-battery using a glucose solution as the source of the chemical energy supply was described. The enzyme and electron mediator were selected for highly efficient power generation. In addition, enzyme-immobilized electrodes under each condition were optimized. A novel enzyme bio-battery with improved power generation efficiency was reported by the fabrication of a gas/liquid porous diaphragm.

2. Experimental setup

2.1. Fabrication and characterization of enzyme-immobilized electrodes

Fig. 1 shows the structure and enzymatic reaction formula for an enzyme-based bio-battery in a glucose solution. In our bio-battery, reduced NADH and hydrogen ions are generated by a catalytic reaction using glucose as the substrate and oxidized nicotinamide adenine dinucleotide (NAD⁺)-dependent glucose dehydrogenase (GDH) as the anode. Furthermore, diaphorase (Dp), which oxidizes reduced NADH, is used to generate oxidized NAD⁺, hydrogen ions, and electrons because a large overvoltage is required to oxidize NADH and extract electrons. On the cathode, bilirubin oxidase (BOD), a multicopper oxidase that catalyzes oxygen reduction, is used to generate water by the catalytic reaction of electrons, hydrogen ions, and oxygen. Using the above three enzymes, power is generated from glucose.

For the anode and cathode, a disk-type carbon electrode with a wide potential window and chemical stability, as well as polishing capability, was selected. The end face of the carbon disk electrode was polished by a polishing device (PK-3 Electrode Polishing kit, ALS Co.). First, dry polishing was carried out using rough polishing diamond (polishing particle size: 1 μm), followed by wet polishing using polishing alumina (particle diameter: 0.05 μm) to mirror-finish the electrode end faces. Two types of enzymes GDH (274 units/mg, TOYOBO CO.) and Dp (191 units/mg, ORIENTAL YEAST CO.) were entrapped and immobilized on the anode electrode using ultraviolet curable resin PVA-SbQ (Biosurfine SPH (10.2 wt%), Toyo Gosei Co., Ltd.) (Jeanty and Marty, 1998; Marquette et al., 2003). In addition, BOD was immobilized using PVA-SbQ or a biocompatible polymer PMEHL (Poly (MPC-co-EHMA) (Chu et al., 2009; Ishihara et al., 1990; Kudo et al., 2008) on the cathode electrode. A mixed solution of PVA-SbQ and BOD (2.51 units/mg, Amano Enzyme In. -> Amano Enzyme Inc.) was applied to the

electrode surface using a desalinated solution to reduce aggregation. Next, the enzyme was immobilized by ultraviolet irradiation. For the immobilization using PMEHL, a mixed solution of a 10 wt% PMEHL solution and ethanol as the solvent was applied to the electrode surface, followed by drying to entrap and immobilize the enzyme. To evaluate the efficiency of the electron transfer between the enzyme and the electrode, we also evaluated the electrode on which the enzyme and the electron mediator were concurrently immobilized was also evaluated. The electron mediator is a low-molecular-weight redox substance; therefore, it is not suitable for entrapment immobilization within polymers. Hence, the immobilizing method for the enzyme and electron mediator by electrostatic interactions is investigated. The enzyme and electron mediators were adsorbed via electrostatic interactions and immobilized on the electrode using a positively charged poly(sodium-4-styrenesulfonate) solution (PSS, M.W. 200000, 30 wt% in H₂O, 561967, Sigma-Aldrich) and positively charged poly diallyldimethyl ammonium chloride (PDAA, M.W. 240000, 17338, Polysciences) on the anode electrode. PDAA solution (2% (w/v)), GDH, Dp, NADH and Vitamin K₃ (VK₃, 36405-84, NACALAI TESQUE, INC.) served as the electron mediator, the PSS solution (2%) was deposited, and finally dried (40 °C, 10 min) to prepare an enzyme electrode. The CV potential using the VK₃ mediator was applied as described previously (Togo et al., 2007). For the cathode electrode, potassium ferricyanide (K₃[Fe(CN)₆] (167-03722, Wako) was used as the electron mediator. As BOD and K₃[Fe(CN)₆] exhibit a negative charge in a buffer solution of pH 7.0, the positive charge of PDAA was utilized. The immobilized enzyme and electron mediator were coupled via electrostatic interactions. Cyclic voltammetry (CV) for of three electrodes type (counter electrode: Pt, reference electrode: Ag/AgCl) was evaluated, and the potential was swept using a potentiostat with each of the enzyme immobilized electrodes as the working electrode (10 mV/s) with the output current was measured (Supplemental Fig. 2). For examining the anode, the enzyme electrode was immersed in a beaker filled with a glucose solution (50 mmol/L) and the cathode electrode was filled with a phosphate buffer solution (pH 7.0, 50 mmol/L). The potential was swept and the CV characteristics were investigated

2.2. Construction of the air bio-battery

An enzymatic fuel cell was constructed by combining the enzyme immobilized electrodes prepared as shown in Supplemental Fig. 2. Power generation characteristics were investigated in a glucose solution (5 mmol/L: 90 mg/dL) corresponding to the human blood glucose concentration. A variable resistor was connected between both electrodes, the resistance was varied, and the output voltage was measured with an electrometer (8240, ADCMT). Current density (A/cm²) and power density (W/cm²) during power generation were calculated from the measured values of resistance, recorded voltage, and power generation.

2.3. Improvement of gas/liquid porous diaphragm cell

For increasing the output of the enzyme-based bio-battery, a gas/liquid porous diaphragm cell was used to efficiently deliver oxygen molecules from the gas phase to the liquid phase (Fig. 2). As the reduction of oxygen occurs at the cathode, improved output can be expected with an efficient oxygen supply to the electrode surface. Hence, a gas/liquid exchange bio-battery using a porous diaphragm as the organic engine, which can convert chemical and mechanical energy, is developed (Kato et al., 2010; Munkhjargal et al., 2015). The gas/liquid porous diaphragm, serving as the gas-permeable membrane, served as the diaphragm in a two-phase reaction cell between the gas and liquid phases. The face of the cathode enzyme electrode was in contact with the diaphragm, whereby oxygen molecules in the air were efficiently transmitted to the diaphragm. The power generation characteristics were compared using the diaphragm, a dialysis membrane (UC 36-32-

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