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Short communication

A membraneless single compartment abiotic glucose fuel cell

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HIGHLIGHTS

- Selectively catalyze glucose in the presence of oxygen in abiotic fuel cell.
- Abiotic catalyst Al/Au/ZnO prepared using hydrothermal method.
- Abiotic glucose fuel cell possesses an open-circuit voltage of 840 mV.
- A maximum power density of 16.2 μW cm^{-2} at a cell voltage of 495 mV was obtained.

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ABSTRACT

A simple energy harvesting strategy has been developed to selectively catalyze glucose in the presence of oxygen in a glucose/O_2 fuel cell. The anode consists of an abiotic catalyst Al/Au/ZnO, in which ZnO seed layer was deposited on the surface of Al/Au substrate using hydrothermal method. The cathode is constructed from a single rod of platinum with an outer diameter of 500 μm. The abiotic glucose fuel cell was studied in phosphate buffer solution (pH 7.4) containing 5 mM glucose at a temperature of 22 °C. The cell is characterized according to its open-circuit voltage, polarization profile, and power density plot. Under these conditions, the abiotic glucose fuel cell possesses an open-circuit voltage of 840 mV and delivered a maximum power density of 16.2 μW cm^{-2} at a cell voltage of 495 mV. These characteristics are comparable to biofuel cell utilizing a much more complex system design. Such low-cost lightweight abiotic catalyzed glucose fuel cells have a great promise to be optimized, miniaturized to power bio-implantable devices.

1. Introduction

Bio-implantable devices, such as implantable glucose biosensors, require a power source, which may be provided by charging of a battery. The two basic and most immediate challenges facing bio-implantable devices include (1) a high desire that bio-implantable devices are self-powered without using a battery and (2) the power source that can drive bio-implantable devices must not add much weight to the bio-implantable device. Therefore, it is important to exploit innovative nanotechnologies that harvest energy from the environment for self-powering these bio-implantable devices. Self-powering bio-implantable devices have an enormous potential to improve individual's well-being. A key advantage of self-powered bio-implantable devices is that they usually operate at extremely low power (nW to μW) [1]. As a result, the biochemical energy harvested from within the human body is sufficient to power these systems. By harvesting a small fraction of this energy into electricity, sufficient energy can be generated for self-powering wireless bio-implantable devices by harvesting energy directly from the biological fluid without using external battery sources. The realization of glucose as an ideal fuel source for bio-implantable devices such as miniaturized biosensors [2], microactuators [3], and pacemakers [4] has received significant attention in the development and characterization of enzymatic biofuel cells which convert the biochemical energy from glucose into electrical energy [5].

In enzymatic biofuel cells, glucose is oxidized at the anode by glucose oxidase and oxygen is reduced at the cathode by laccase or bilirubin oxidase [6]. In non-compartmentalized enzymatic biofuel cells, the presence of oxygen and glucose mixture results in the reduction at the cathode as well as at the anode, which results in a decrease in the overall power output of such a device [7]. Moreover, there are significant problems with extending the lifetime and durability of these biocatalysts and hence further decreasing the efficiency of enzymatic biofuel cells since these enzymes have different optimum operating pH and temperature conditions and may require the use of electron transfer mediators [8]. To overcome

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these problems several approaches have been considered such as the use of enzyme with reconstructed active centers that are less sensitive to oxygen [9], inorganic materials such as osmium containing redox polymers [10,11] and carbon nanotubes [4,12–15] that act as mediators or the combination of inorganic mediators and biocatalyst [16] in order to facilitate the efficient electron transfer between the anode and cathode. Glucose dehydrogenase is frequently used in enzyme-biofuel cells and requires the immobilization of NAD^+ cofactor. The co-immobilization of NAD^+ cofactor requires the utilization of complex procedures [17], which limits its application in the bio-implantable devices [18]. Thus, in order to achieve kinetically preferential electron transfer, O_2 dependent and NAD^+ dependent enzymatic biofuel cells must employ complex multimolecular arrangement in the fabrication of the cells. These multimolecular ensembles are not readily adaptable for batch fabrication and are insufficient to provide the long-term power that bio-implantable devices require. Although abiotic glucose fuel cells exhibit higher stability and lifetime than enzymatic biofuel cells, abiotic glucose fuel cells have received very little attention for being used as a potential power source for bio-implantable devices. Generally, abiotic glucose fuel cell are used to convert the chemical energy of glucose and oxygen in biological fluid into electricity using noble metal as the catalyst to abiotically catalyzed glucose because they are inert and biocompatible [19]. In this contribution, we refocus the attention on the development of a simple approach to harvest the excellent electrochemical properties of the Al, Au, and ZnO by combining them via sputtering and hydrothermal methods to fabricate an abiotic glucose/oxygen fuel cell. We utilized the Al/Au/ZnO as the anode and platinum as the cathode material to selectively catalyze glucose oxidation and oxygen reduction in the presence of oxygen.

2. Experimental methods

2.1. Materials

Pure Aluminum foil (99.9999%, 250 μm thick) was purchased from Alfa Aesar. Zinc chloride (99.99%), triethenamine (TEA, 99.99%), and all chemical reagents were purchased from Sigma–Aldrich and all supplementary chemicals were of analytical grades and used without further purification. All solutions were prepared with 18.2 MΩ cm Milli-Q water.

2.2. Fabrication

2.2.1. Al/Au/ZnO anode fabrication and Pt cathode

Fig. 1 illustrates the anode fabrication steps for glucose oxidation. Briefly, rectangular (6 mm × 5 mm) strips of pure Al foils were used as substrates and were cleaned to remove contaminants by standard cleanroom procedures prior to use. Thin layers of gold (40 nm) were sputtered onto the surface of the aluminum substrate using the magnetron sputtering process. ZnO seed layers were coated on the Al/Au sputtered substrate by a simple sol–gel process [20] under mild conditions. The ZnO precursors were prepared using zinc chloride and propanol. Briefly, the reaction solution for ZnO seed layer fabrication was prepared by mixing appropriate quantities of propanol and 0.4 M zinc chloride solution under constant stirring at 75 C. Equimolar of triethenamine was added dropwise to stabilized the precursor solution and produce a final 0.1 M homogeneous ZnO nanosol. The ZnO nanosol was covered and maintained under constant stirring at 85 C until a homogeneous solution was observed. The reaction mixture was aged at room temperature until the desired consistency was obtained. The Al/Au/ZnO seed layers were established by coating the Al/Au substrate with the ZnO nanosol using dip-coating method followed by

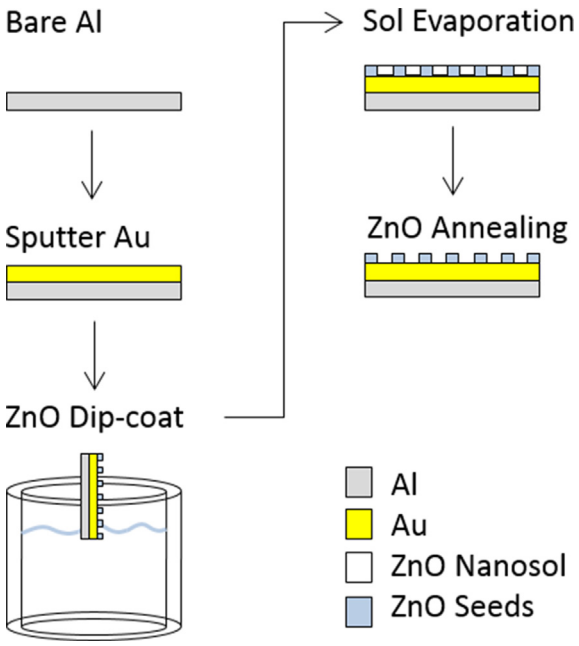

Fig. 1. Cross-section process flow diagram of the fabrication process used in the design of the Al/Au/ZnO anode.

natural solvent evaporation to form a thick layer of seeds. Between coatings, the substrate was annealed at 150 C for 1 h to ensure nanoparticles adhesion to the substrate surface. The dip-coating and annealing process was repeated until a thick, uniform seed layer was obtained on the surface of the electrode. Subsequently, the Al/Au/ZnO substrates were repeatedly rinsed using deionized water to remove unbound salt and dried at 30 C in a convection oven overnight. The cathode for oxygen reduction was achieved by utilizing platinum rod (ϕ = 500 μm) since platinum has been show to exhibit the highest oxygen reduction potential when compared to palladium, gold and silver in phosphate buffer pH = 7 [21]. The morphology and structural characteristics of the as-fabricated, annealed Al/Au/ZnO were observed by means of scanning electron microscopy (JEOL JSM-5600 SEM).

2.3. Experimental setup

2.3.1. Glucose fuel cell setup and measurements

The abiotic glucose fuel cell was built using the as-fabricated Al/Au/ZnO as the anode and a platinum rod as the cathode positioned 20 mm apart in the cell. Fig. 2 shows the schematic diagram of the experiment setup. All experiments were performed at physiological glucose and pH levels (5.0 mM glucose in 0.1 M phosphate buffer solution (pH = 7.4)) at 22 °C. The Al/Au/ZnO selectively catalyzes glucose oxidation in the presence of oxygen and the highly specific platinum catalyzes oxygen reduction reaction at the cathode, thereby enabling the use of a membraneless single compartment design for testing. The current–voltage outputs of the cell were obtained under various loads with 5 mM glucose in phosphate buffer. The load was connected directly in parallel with the glucose fuel cell. Fluke 87V True RMS multimeter was used to capture the glucose fuel cell voltage and current readings.

3. Results and discussion

Recently, zinc oxide nanostructures have received significant attention in the construction of sensors [22] and piezoelectric