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MEMS Ascorbic Acid Fuel Cells with Flexible Electrodes of Enzyme-Coated Graphene Carbon Fiber Cloth

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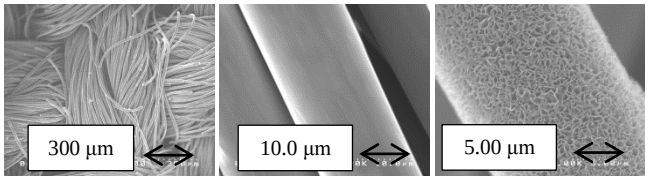
Abstract

Miniaturized fuel cells utilizing ascorbic acid (AA) as a fuel are considered to be a good candidate as a portable and flexible power sources for wearable or portable electronic systems. AA known as vitamin-C is very safe to human body and biologically friendly. In this work, we fabricated an ascorbic acid fuel cell (AAFC) using a graphene coated carbon cloth (GCC) as electrodes because the GCC electrodes have much larger effective areas and are expected to maintain more bilirubin oxidase (BOD) enzymes. The power density of the fabricated AAFC by immersing the fuel cells in a phosphate buffer solution with 100 mM AA at room temperature was 21.3 μW / cm^2 at 0.34 V. The power density was more than 20 times larger than that of an AAFC using conventional porous carbon as the electrode material.

1. Introduction

Recently, great attention has been paid to the appearance of miniaturized power sources, especially for wearable or portable electronic systems. Ascorbic acid (AA) known as vitamin C is considered to be a nonflammable, biologically friendly, and safe fuel for wearable fuel cell power systems [1]. Fujiwara et al. reported an ascorbic acid fuel cell (AAFC) without catalysts on the anode [2-4]. However, these AAFCs were not portable because they needed heavy fuel circulation systems and ion exchange films between the anode and cathode. Recently, Mogi et al. reported a miniaturized AAFC with an area of 1 cm^2 fabricated on a flexible polyimide (PI) film using micro electro mechanical (MEMS) system technology [5]. Their fuel cells utilized an enzyme bilirubin oxidase (BOD)-coated cathode to selectively reduce oxygen dissolved in an aqueous solution of AA. Thus, the AAFC did not require any ion exchange layers and the fuel circulation systems, which realized the miniaturized AAFC. However, they utilized porous carbon layers as the electrodes, and the porous carbon electrodes were not flexible, which frequently caused peeling off of the electrodes from the substrate after repeated bending of the fuel cells.

On the other hand, a research group of the Incubation alliance Inc. reported a (a) graphene-coated carbon fiber cloth (GCC) {Fig. 1(a)} [6]. The GCC consists of carbon fibers with a diameter of approximately 10 μm {Fig. 1(b)}, and their surfaces were covered by forest like graphene layers called graphene flowers {Fig. 1(c)}.



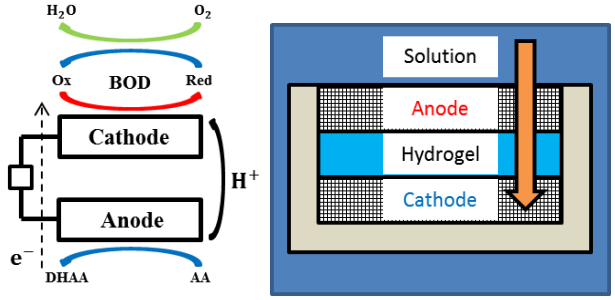
(a) (b) (c)

Fig. 1 SEM micrographs of (a) graphene-coated carbon fiber cloth (GCC), (b) carbon fiber without graphene layers, and (c) carbon fiber coated with forest like graphene layers.

Therefore, it is considered that the GCC can maintain more BOD enzyme for improved fuel cell performance and good flexibility by using GCC as the electrodes of the AA fuel cells. In this study, we report on fabrication and characterization of AAFCs fabricated using GCC as flexible electrodes.

2. Experiment

Figure 2 illustrates a possible power generation mechanism of an AAFC and the conceptual structure of the fabricated fuel cell.



(a) (b)

Fig. 2. (a) Possible power generation mechanism for an ascorbic acid fuel cell (AAFC), and (b) structural con-

cept of the AAFC investigated in this work.

The AAFC consisted of an anode with no catalysts and a BOD adsorbed Cathode {Fig. 2(a)}. On the Anode, AA was oxidized on the GCC electrode's surface and dehydroascorbic acid was generated [7]. As a result, the electrons and protons were produced on the anode. The electrons were transferred from the anode to the cathode via an external circuit to extract electricity. Oxygen (O_2) in the fuel solution was selectively reduced to water (H_2O) by BOD enzymes immobilized on the increased cathode areas of the GCC electrode on reaction with electrons and protons. The electrode areas of the anode and cathode were 3 cm × 10 mm^2 {Fig. 2(b)}. The cathode and the anode were separated by agarose hydrogel with a thickness of 2 mm, and these electrode structures were embedded in a flexible polydimethylsiloxane (PDMS) substrate. The PDMS on the top of the anode was removed to expose the anode surface to a 100 mM AA phosphate buffer solution with naturally dissolved oxygen. Figure 3 shows the photograph of the fabricated AAFC.

Fig. 3 Photograph of the fabricated AAFC.

3. Result and discussion

The power was measured by immersing the fuel cells in the phosphate buffer solution with 100 mM AA at room temperature. Load resistances with different values were connected to the wirings of the anode and cathode to measure the relationships between the generated voltage and the current. Figure 4 shows comparison of the relationship between the power density and output voltage of the AA fuel cells with (a) GCC electrodes (this work) and (b) porous carbon electrodes (Mogi et al. [5])

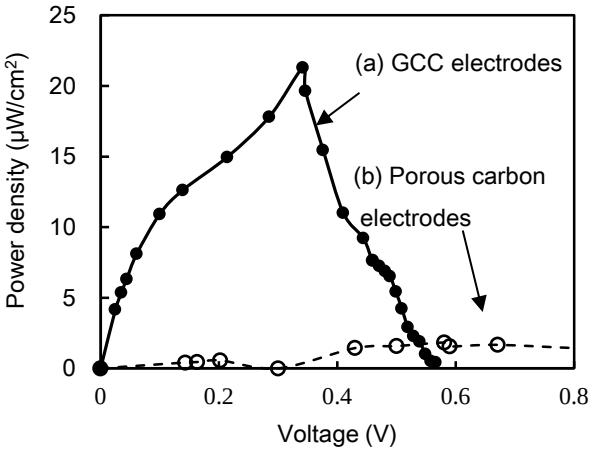


Fig. 4 Relationships between the power density and output voltage of AAFCs with (a) GCC electrodes (this work) and (b) porous carbon electrodes (Mogi et. al. [5]).

The maximum power density for the AAFC with the GCC electrodes was 21.3 μW / cm^2 at 0.34 V. The power density of the AAFC with porous carbon electrode was also shown for comparison, and the power density of the AAFC with the porous carbon FC was only 1 μW / cm^2 at 0.5 V. The remarkable increase of the power density of the GCC AAFC was considered due to the increased surface areas of the GCC anode for increased oxidation rates of AA and increased coverage of BOD on the cathode to reduce oxygen.

4. Conclusion

Miniaturized fuel cells utilizing ascorbic acid as a fuel are considered to be a good candidate as portable and flexible fuel cells for wearable or portable electronic systems. AA is known to be a very safe and environmentally friendly fuel material for fuel cells. In this work, we fabricated an AAFC using a GCC as electrodes because the GCC electrodes have much larger effective areas and are expected to maintain more BOD enzymes. The power density of the fabricated AAFC by immersing the fuel cells in a phosphate buffer solution with 100 mM AA at room temperature was 21.3 μW / cm^2 at 0.34 V. The power density was more than 20 times larger than that of an AAFC using conventional porous carbon as the electrode material.

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