

A SURFACE MOUNTABLE GLUCOSE FUEL CELL FOR MEDICAL IMPLANTS

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Abstract: We present an enzymeless, surface mountable glucose fuel cell based on abiotic catalysts. The device is intended as an energy harvesting implantable power supply for low power medical implants and sensors. After 50 days of operation in physiological glucose solution the fuel cell exhibited a stable performance of $1.1 \mu\text{W}/\text{cm}^2$. The demonstrated lifetime of our glucose fuel cell therefore already exceeds the maximum lifetime reported for devices based on enzymatic catalysts. This renders the concept a promising approach for the development of autonomous and sustainable power supply systems for long-term medical implants.

Keywords: energy harvesting, fuel cell, medical implant, glucose

1. INTRODUCTION

The increasing number of low power MEMS implants currently being developed has stimulated research into energy harvesting power supply concepts [1]. Among these are implantable abiotically catalyzed glucose fuel cells that generate electricity from glucose and oxygen present in body fluids. In our earlier work we reported on an abiotically catalyzed glucose fuel cell [2], based on the original design of Rao et al. [3]. In this communication we present the fabrication and long-term performance of a surface mountable prototype, requiring body fluid access from only one side.

2. OPERATIONAL CONCEPT

Electricity is directly derived from body fluids by the spatially separated electrochemical reaction of glucose and oxygen. To facilitate the electrode reactions, abiotic catalyst materials like platinum alloys or activated carbon are used [3]. The governing electrode reactions are illustrated in Fig. 1. Both, glucose and oxygen are simultaneously present in body fluids, e.g. interstitial fluid. Since most noble metals catalyze both electrode reactions, a separation of reactants is necessary. This can be accomplished by a selective cathode consisting of activated carbon

that is arranged in front of the anode. This effectively depletes oxygen inside the fuel cell and glucose oxidation takes place under predominantly anoxic conditions (see Fig. 2). We chose this effective and elegant embodiment for our prototype, since the concept allows for mounting the fuel cell directly on the surface of medical implants, facilitating system integration and implantation.

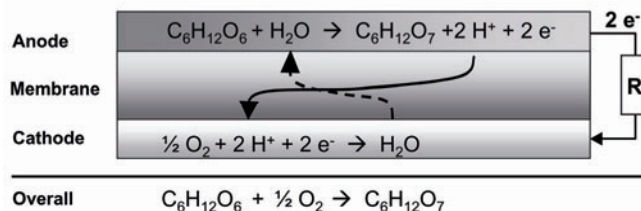


Fig. 1: Electrode reactions of an abiotically catalyzed glucose fuel cell.

The construction of our surface mountable glucose fuel cell is illustrated in Fig. 3. The electrode dimensions are $1.5 \text{ cm} \times 1.5 \text{ cm}$. For increased electrical conductivity a platinum mesh electron collector is embedded in both electrodes. Electrical connections are established by platinum wires connected to the electron collector with conductive carbon cement. The electrodes are electrically insulated by a glucose permeable hydrogel membrane. An epoxy resin casing gives structural stability to the membrane electrode assembly.

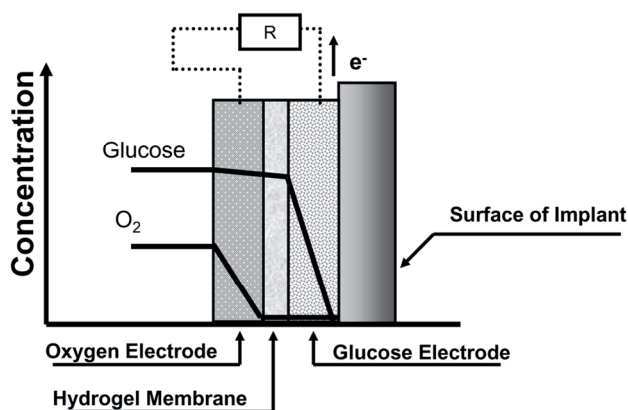


Fig. 2: Reactant separation by special electrode arrangement: Oxygen is consumed at the exterior cathode leading to low oxygen concentration inside the fuel cell.

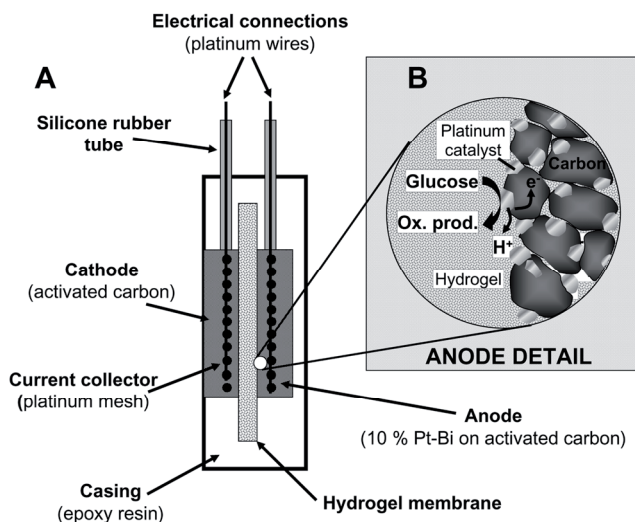


Fig. 3: Construction details of the fuel cell.

3. EXPERIMENTAL

3.1 Pre-hydrogel solution

As crosslinkable pre-hydrogel an aqueous solution containing poly(vinyl alcohol) (PVA, molecular weight: 72 kD) and poly(acrylic acid) (PAA, molecular weight: 240 kD) in the ratio of 8:1 by weight was prepared. The total polymer content of the pre-hydrogel solution was 7 wt %.

3.2 Electrode preparation

For the cathode, 526 mg of activated carbon (A 196, Degussa AG, Hanau/Germany) was mixed with 10 g of the pre-hydrogel solution for one hour under vacuum. The mixture was transferred into 2 ml centrifuge tubes and spinned in a fixed angle rotor at 16000 g for 35 min. After

removing the supernatant a spreadable catalyst paste could be obtained. Similarly the anode catalyst paste was prepared from 1052 mg of activated carbon impregnated with Pt-Bi alloy (CF 196, 5 % Pt + 5 % Bi, Degussa AG, Hanau/Germany) and 10 g of the pre-hydrogel solution.

Electrodes were fabricated by a doctor blade technique using adhesive tape as casting frame on a fluorinated ethylene propylene (FEP) sheet. A first layer of catalyst paste was spread to a thickness of 130 μm for the cathode and 260 μm for the anode. The platinum mesh current collector was introduced on top of the still wet catalyst layer. Subsequently a second layer of catalyst paste was applied. The casting frame thickness for the second step was 260 μm for both the anode and cathode. After drying the electrodes were removed from the FEP sheet and crosslinked at 145 $^{\circ}\text{C}$ for 80 min.

Electrodes saturated in PBS were framed with epoxy resin (Duopox AD 895, Delo, Landsberg/Germany) using a silicone rubber mold on a FEP sheet, as depicted in Fig. 4. During this step the cathode was protected with a silicone rubber sheet that would later be removed to enable access of glucose and oxygen.

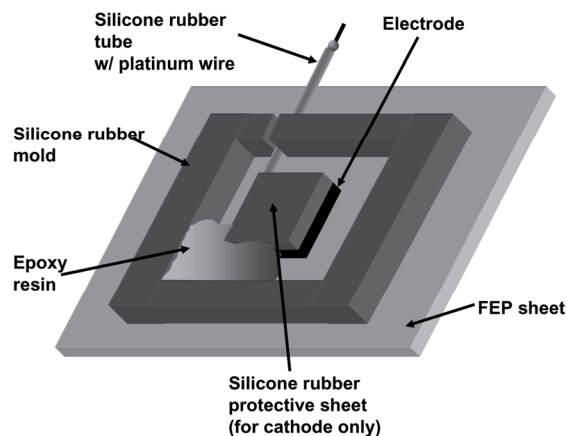


Fig. 4: Framing of electrodes with epoxy resin.

3.3 Hydrogel membrane fabrication

Membranes of ~ 20 μm thickness were manufactured by spreading the pre-hydrogel solution onto a silanized glass plate. A 500 μm casting frame made from adhesive tape was used to control the thickness. Dried membranes were crosslinked at 145 $^{\circ}\text{C}$ for 80 min and equilibrated in PBS.

3.4 Fuel cell assembly

The complete fuel cell was assembled with the wet PBS-saturated hydrogel membrane placed in between the electrode frames. Fig. 5 schematically shows the assembly procedure: (1) The components are arranged in PBS solution to prevent the entrapment of air pockets; (2) A clamp is used to fixate the assembly; (3) Upon removal from the PBS solution the edges of both electrode frames are immediately joined with epoxy resin.

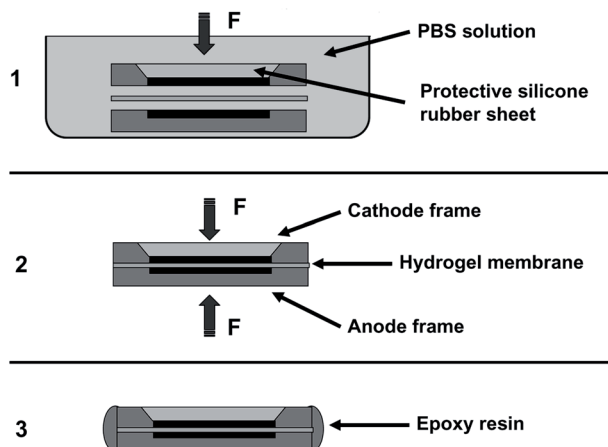


Fig. 5: Fuel cell assembly procedure. See text for explanations.

After curing the protective silicone rubber sheet was removed from the cathode and the complete fuel cell was equilibrated in PBS solution containing 0.1 wt% glucose.

3.5 Electrochemical characterization

Electrochemical characterization of the fuel cell was carried out at 37 °C in 700 ml aerated PBS solution containing 0.1 wt % glucose (pH = 7.4). A Keithley 2400 source meter operating in galvanostatic mode served as current sink. Electrode potentials were recorded with a Keithley 2700 datalogger system in 2 min intervals against a saturated calomel electrode (SCE) in the same solution.

4. RESULTS & DISCUSSION

4.1 Long-term performance

Fig. 6 shows the fuel cell performance at a constant current density of 5 $\mu\text{A}/\text{cm}^2$. Exhibiting an initial peak performance of 1.6 $\mu\text{W}/\text{cm}^2$ after two days of operation, the performance

approached a stable power output of 1.1 $\mu\text{W}/\text{cm}^2$ after 40 days. The corresponding cell voltage amounted to 218 mV. In the following 10 days no performance degradation could be observed. A closer look at the individual electrode potentials reveals that the initial performance degradation stems from a change in anode potential to more positive values (Fig. 6).

When after 32 days of operation the aeration was interrupted ("B" in Figs. 6 and 7) the reduced oxygen concentration induced a shift towards more negative potentials at both electrodes. The change in potential was more pronounced at the anode and consequently both cell voltage and power output increased. This indicates that under full aeration conditions the reactant separation is incomplete and the oxygen diffusing into the fuel cell is not fully consumed at the cathode. Consequently, the presence of oxygen at the non-selective anode leads to an electrochemical short-circuit and decreased cell voltage.

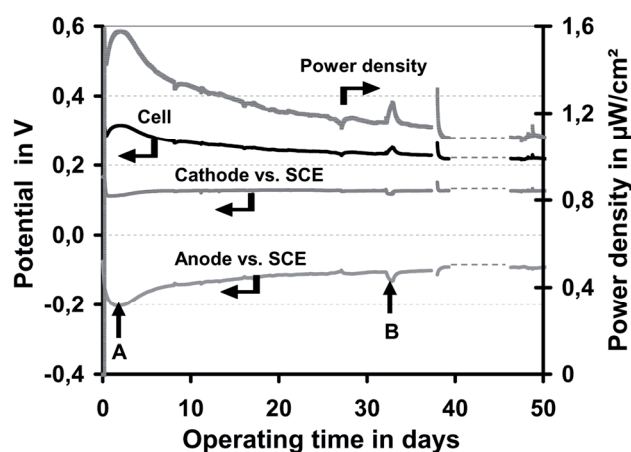


Fig. 6: Long-term fuel cell performance at a current density of 5 $\mu\text{A}/\text{cm}^2$. A): Initial peak performance. B): Increased performance due to interrupted aeration and the associated lower oxygen concentration.

4.2 Current density-potential plot

In Fig. 7 the current density – potential plot of the fuel cell recorded after the long-term experiment is given. By increasing the current density to 10 $\mu\text{A}/\text{cm}^2$ an improved performance of 1.4 $\mu\text{W}/\text{cm}^2$ could be obtained. This shows that a further performance increase is possible by operating the fuel cell at optimized load current.

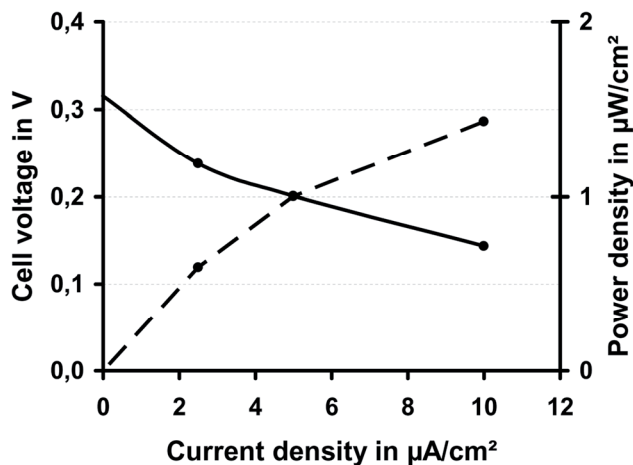


Fig. 7: Current density – potential plot in the useful cell voltage range, recorded after 50 days of operation. Data points represent the stable cell voltage (voltage drift < 0.3 mV/h) at a given load current.

5. CONCLUSION

We could demonstrate that our surface mountable glucose fuel cell (Fig. 8) is able to deliver electrical energy with power densities exceeding $1 \mu\text{W}/\text{cm}^2$ over a period of 50 days. A device $3 \text{ cm} \times 3 \text{ cm}$ in size and applied to the surface of an implant would therefore already be sufficient to supply the recently reported $8 \mu\text{W}$ low power pacemaker circuit [4].

The observed stabilization in power output after 40 days suggests that a stable operation of our prototype beyond the 50 days shown in this work is possible. This already exceeds the maximum lifetime reported for devices based on enzymatic catalysts [5] and renders the concept a promising approach for the development of autonomous and sustainable power supply systems for long-term medical implants.

A first analysis of oxygen sensitivity showed that fuel cell performance increases at lower oxygen concentrations, indicating an incomplete separation of reactants. A more detailed investigation of this phenomenon is currently underway in our laboratory.

We are also working on the optimization of the electrode structure to further improve performance and long-term stability in a simulated physiological environment.

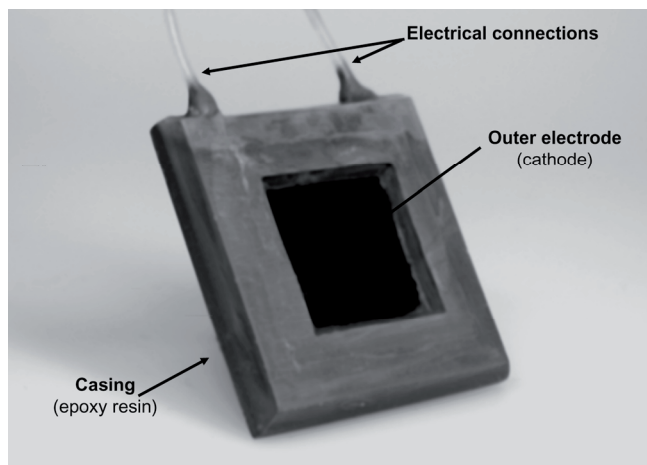


Fig. 8: Picture of the prototype fuel cell with a geometrical electrode area of 2 cm^2 .

6. ACKNOWLEDGEMENTS

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