*Sensors & Transducers, Vol. 203, Issue 8, August 2016, pp. 1-7*

**Sensors & Transducers**



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# Characterization of a Self-powered Glucose Monitor

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*Received: 7 July 2016 /Accepted: 8 August 2016 /Published: 31 August 2016*

**Abstract:** Glucose substrates are successfully harnessed to generate electricity in a membraneless biofuel cell with a mesh network of carbon nanotubes pyroquinoline quinone glucose dehydrogenase-modified anode and a laccase-modified cathode. Using glucose as a substrate, this glucose-oxygen biofuel cell is able to produce a steady current density of 337.5 μA/cm² and an open circuit voltage of 524 mV in 360 mg/dL glucose solution. Interestingly, the fuel cell in combination with a capacitor as the transducer element can also be utilized as a glucose monitor while generating electricity simultaneously to power small electronic devices, such as light emitting diode (LED). Moreover, the self-powered glucose monitor exhibited a linear dynamic range of 9 mg/dL to 630 mg/dL glucose. These results and device demonstrations suggest that further research into self-powered glucose monitors can provide major benefit in developing a novel autonomous implantable glucose monitor platform to greatly improve the quality of life for individuals living with diabetes. 

**Keywords:** Glucose monitor, Diabetes, Biofuel cell, Voltage boosting.

## 1. Introduction

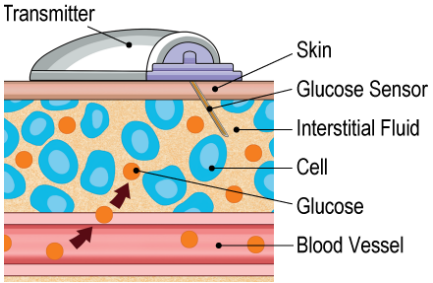
The current epidemic of diabetes and its potential growth is a public health risk that is unsustainable and must be addressed. According to the Center for disease control (CDC) 2014 report, 29.1 million people in the U.S. suffer from the disease diabetes, which is the seventh leading cause of death. This number is predicted to be doubled or tripled by the year 2050. The cost incurred to keep diabetes under control was 245 billion US dollars [1]. This cost is attributed to the complications that arise due to poor maintenance of blood glucose levels. Some of the complications include retinopathy, neuropathy, gastroparesis, foot complications, ketoacidosis, kidney disease, etc. [2]. Diabetes is a metabolic disorder caused by abnormal blood glucose level and is a result of either insufficient production of hormone insulin by the pancreas or the cells in the body do not respond correctly to the insulin produced thus, defining Type I and Type II diabetes respectively. In addition, gestational diabetes is common in pregnant women during their second trimester. This type of diabetes at times complicates the pregnancy [3]. There is a significant growth in the number of people suffering from Type II diabetes due to the unhealthy lifestyle and stress in their daily life. Normal blood glucose levels for a healthy individual as well as an individual suffering from diabetes is provide in Table 1.

**Table 1.** Normal blood sugar levels for non-diabetic as well as diabetic people.

| Non-diabetic people | | Diabetic people | |
|---|---|---|---|
| **Test** | **Blood glucose (mg/dL)** | **Test** | **Blood glucose (mg/dL)** |
| **Normal** | 79.2-110 | **Pre-meal** | 90-130 |
| **Fasting** | 70-100 | **Post-meal** | < 180 |

Current technology for monitoring blood glucose levels involves the use of invasive devices, such as the finger prick test using a glucometer as illustrated in Fig. 1(a). However, this device is bulky and at times, depending on different batches of the test strips, the meter must be recalibrated. The blood glucose reading can drift by as large as 72 mg/dL. This drift in the blood glucose level can be fatal at times. In addition, tight glucose monitoring may involve pricking the finger multiple times per day, which may prove painful and tedious. Continuous glucose monitoring (CGM) devices (Fig. 1(b)) is another invasive technique used to monitor blood glucose level.

(a)




(b)

**Fig. 1.** Blood glucose monitoring devices: (a) Glucometer, and (b) Continuous glucose monitor (CGM).

This device consist of a disposable needle that acts as an in-vivo sensor and an external transmitter and receiver. The receiver at the receiving end displays the blood glucose level information. The receiver section consist of a potentiostat circuit, which makes the entire device bulky. This device measures the glucose in interstitial fluid as opposed to blood vessel and often suffers a lag of as much as 15 – 20 minutes [4]. This device also needs recalibration every 12 hours and relies on traditional glucose monitoring technique previously described. Moreover, all these devices requires a battery to power the potentiostat circuit shown in Fig. 2. A glucose biosensor typically consist of a reference electrode, a counter electrode and a working electrode at which the glucose is oxidized by a glucose selective enzyme to release electrons. A potential is applied by the battery through the potentiostat circuit across the working electrode and the reference electrode and the current is measured in terms of the potential drop across the load resistor. This current is directly proportional to the glucose concentration.

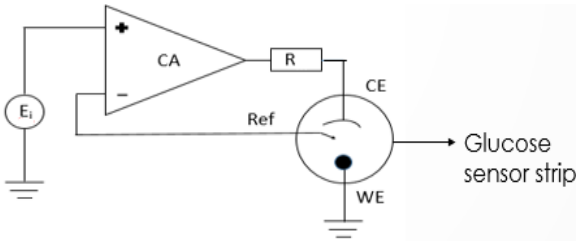



**Fig. 2.** Schematic of a potentiostat circuit.

Since the potentiostat circuit relies on battery as its source of power supply, the battery needs to be replaced or recharged from time to time once all the stored chemical energy is utilized. This type of glucose biosensor can be used for up to 7 days before replacement. There have been several attempts made to design a noninvasive glucose monitoring device. Of all the attempts, the only device that was approved by U.S. Food and Drug administration (FDA) was the "GlucoWatch G2 Biographer" (Fig. 3), which was designed by Cygnus Inc. This device operated on reverse iontopherosis principle in which glucose molecules are removed from within the body for detection [5]. However, the inconsistencies in the measurement of glucose levels as a result of perspiration made this device a fad on the market. The above shortcomings of the current glucose monitoring technology must be addressed by developing a closed loop system that would be minimally invasive and flexible and easy to use. All the devices described above require a battery as a source of power and due to the disadvantages of the batteries, research is being conducted on alternative source of power for bioimplantable devices.

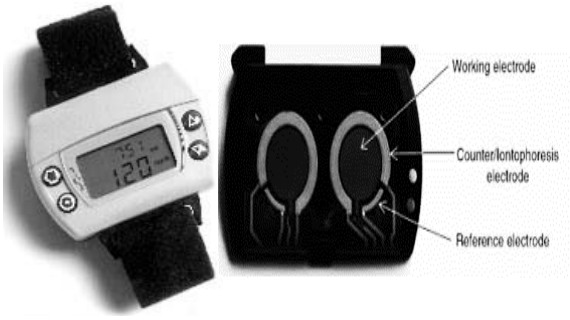



**Fig. 3.** A GlucoWatch G2 Biographer designed by Cygnus, Inc.

Early research to explore alternate power source that would overcome the drawbacks of batteries led to the development of non-enzymatic fuel cells. Although these fuel cells have demonstrated high electrical density values, they use precious non-metal catalyst to oxidize fuel [6]. The costly metal catalyst has shown to produce metal oxide byproduct from the oxidation of fuel that is poisonous and affects the cell stability and longevity [7]. The above shortcomings of non-enzymatic fuel cells led to the rise of enzymatic fuel cells. These fuel cells use naturally occurring enzymes that are derived from living organisms as catalyst to oxidize fuel which makes it a clean, economical and easily replenished source of catalyst.

Glucose selective enzyme, such as glucose oxidase have been used to oxidize glucose to form Gluconolactone. However, the oxidation reaction produces harmful hydrogen peroxide [8] which affects the sensor's stability and life. On the contrary, many glucose monitors strive to use glucose dehydrogenase enzyme because they do not produce any byproducts. Moreover, glucose dehydrogenase enzyme have also been used to fabricate a glucose biofuel cell in prior works [9-10]. Significant research has been conducted on the design and characterization of enzymatic glucose biofuel cell and demonstration of its use as a potential replacement for batteries. Recently, the focus has shifted to the applications of these glucose biofuel cell. One such example involved a theoretical demonstration of an enzymatic biofuel cell as a power source to detect glucose using a glucose sensing contact lens [11]. In this research, a mathematical equation was used to demonstrate that sufficient power can be produced from a human tear solution by the biofuel cell that could drive the glucose sensing circuit embedded on the contact lens. Another novel design that led to two biofuel cells stacked such that the overall thickness of the system was only 425 μm and produced sufficient parameters which were then amplified to drive a wireless circuit [12]. This enabled the transfer of temperature data from a sensor to a nearby computer. Several such instances have been recorded where the biofuel cell produces sufficient electrical parameters to drive an amplification circuit that produces sufficient power to power micro-electronic devices such as LED, stopwatch and digital thermometer [13-15]. Here we use glucose dehydrogenase enzyme to build a glucose monitor assembly consisting of glucose biofuel cell and a capacitor transducing element acting as a glucose sensor [16]. We also show improved stability of our system compared to the previous work done [10]. This device has the potential to overcome the shortcomings of the existing glucose monitors.

## 2. Experimental Section

### 2.1. Materials and Method

10 mm×2 mm strips of Buckypaper were prepared according to previously established protocols [10]. Briefly, the cleaned buckypaper strips (anodic and cathodic electrodes) were incubated with 1-Pyrenebtanoic acid, succinimidyl ester (PBSE) in 180 mg/dL DMSO solution, where noncovalent π - π stacking occurred between the aromatic ring on the PBSE molecule and the series of aromatic rings that compose buckypaper. The excess PBSE were removed by rinsing the electrodes in 180 mg/dL PBS (pH 7.0), followed by DMSO rinse. Pyroquinoline quinone glucose dehydrogenase (PQQ-GDH) was immobilized at the anode and laccase was immobilized at the cathode. The immobilized electrodes were preserved by coating the active surface with 2 μL of Nafion. Finally, the enzyme modified electrodes were stored in their respective buffer.

### 2.2. Voltage Amplification Circuit

The anode and cathode electrodes were assembled together to realize a biofuel cell. The electrical current and voltage produced by this single biofuel cell is not sufficient of powering any device (i.e., a transducer). To improve the electrical output, multiple biofuel cells have been stacked together [17] at the expense of complexity and the bulkiness of the circuit. Here we use a charge pump integrated circuit (IC) to excite the low input voltage from 300 mV to 1.8-2.4 V depending on the current and voltage requirements of the electronic device. The amplification of electrical parameters from a single biofuel cell is shown in Fig. 4.

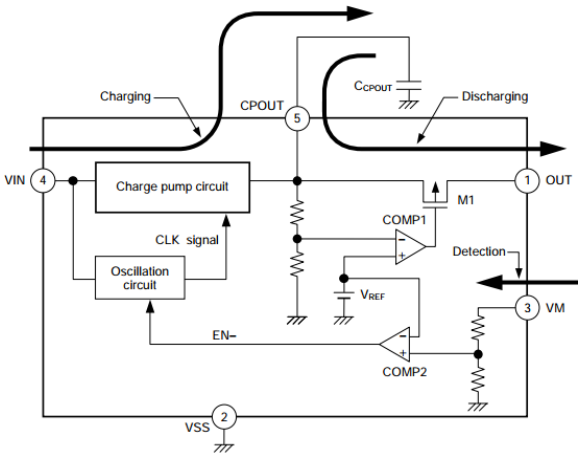



**Fig. 4.** Internal working of a charge pump IC (S882z datasheet).

Charge pump IC takes an input power of 0.3 V or higher from a single biofuel cell to trigger the oscillation circuit, which gives a clock signal output that drives the charge pump circuit. Power from VIN pin is converted to step up electric power in the charge pump circuit. This electric power is gradually charged up to the startup capacitor CPOUT pin and its voltage

rises gradually. When the CPOUT voltage reaches the discharge start voltage (VCPOUT1) which is 1.8 V, the output signal of the comparator (COMP1) changes from high level to low which enables the discharge control switch M1. As a result, the stepped up electric power to CPOUT is discharged from the OUT pin. When CPOUT voltage drops to discharge stop voltage (VCPOUT2) which is 1.2 V, M1 switches off and the discharge is stopped. When the VM pin voltage (VVM) reaches or exceeds the shutdown voltage (VOFF), the output signal (EN−) of the comparator (COMP2) changes from low level to high. As a result, the oscillation circuit stops operation and the shutdown state is entered. If VVM does not reach the shutdown voltage, the stepped up power from the charge pump circuit is recharged to CPOUT. Thus, charging and discharging can be observed via the output transducing capacitor where the voltage toggled between 1.8 V and 1.2 V.

The excited voltage is sufficient to provide the drive strength for an LED [17]. A capacitor incorporated in our circuit (Fig. 5) to function as the transducing element, wherein the charging/ discharging frequency of the capacitor can be correlated to the changes in glucose concentration.

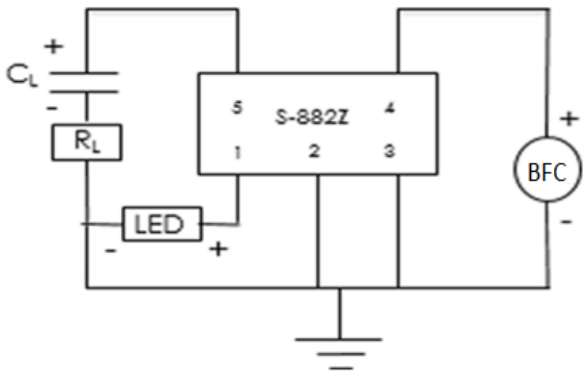



**Fig. 5.** Schematic of a charge pump circuit.

between the active sites of the enzyme and the 3D CNT substrate.

An increase in the peak power density was observed compared to our previous work [17] and this is attributed to decrease in internal resistance of the system thereby, allowing better electron transfer between the active sites and the buckypaper. Moreover, the system exhibited a stable peak power density for 96 days which improved from previously reported [17]. At the end of 96$^{th}$ day of operation, the overall drop in the peak power density was approximately 87 % and 79 % in 180 mg/dL and 360 mg/dL glucose, respectively. It is important to note that the enzyme PQQ-GDH does not produce poisonous hydrogen peroxide byproduct on glucose oxidation thus, prevents bioelectrode deterioration [23]. This further ascertains the stability of enzymes even after over three months which surpasses all data previously reported [24-28]. The experiments were performed in triplicates and the standard deviation values of 4.69 and 5.68 in 180 mg/dL and 360 mg/dL glucose solution confirms the stable operation of this device after 96 days.

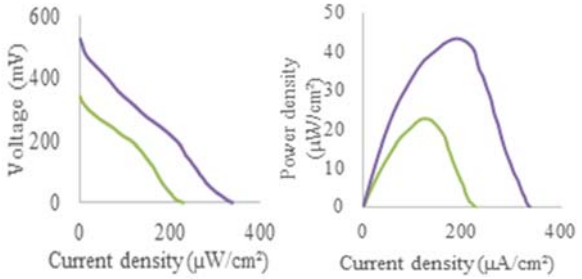


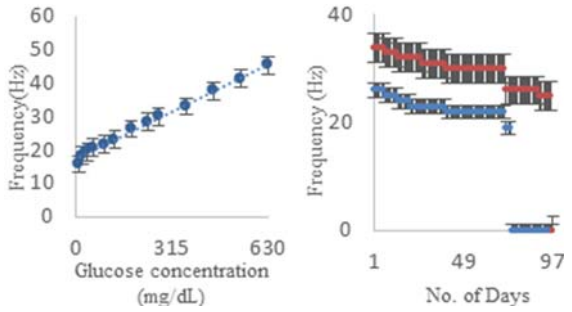

**Fig. 6.** Glucose biofuel cell characterization: (a) Polarization curve, and (b) corresponding power curves (gray) performed in 180 mg/dL glucose solution and (yellow) in 360 mg/dL glucose solution in 0.1 M PBS (37 °C, pH 7).

## 3. Results and Discussion

The biofuel cell system was tested in 180 mg/dL and 360 mg/dL glucose standard solutions at a neutral pH and 37°C. The assembly produced an open circuit voltage and short circuit current density of 339.2 mV and 524 mV in 180 mg/dL and 360 mg/dL glucose solution and 228.75 μA/cm² and 337.5 μA/cm², respectively (Fig. 6). The peak power density produced by the biofuel cell was 22.74 μW/cm² and 43.41 μW/cm² in 180 mg/dL and 360 mg/dL glucose solution, respectively which is greater than the previously reported [19-21]. The use of highly conductive 3D multiwalled carbon nanotube (CNT) buckypaper provided an improved surface area for enzyme immobilization process [22]. Also, the use of oxygen independent and highly glucose selective enzyme pyroloquinoline quinone glucose dehydrogenase allows for direct electron transfer

The power from the biofuel cell was supplied to the charge pump IC, which amplified the voltage to 1.8 V which was sufficient to power the LED and the charge cycle across the transducer element (capacitor) was monitored. The open circuit voltage at 180 mg/dL and 360 mg/dL was sufficient to drive the charge pump circuit which required a minimum voltage of 0.3 V to begin its operation. The output voltage from the charge pump IC was observed to toggle between 1.2 V to 1.8 V which provided burst of power to light the LED. The LED utilized was non-bright light emitting diode that typically requires a voltage of 1.7 V for proper operation. Thus, the output voltage from the charge pump IC caused the flickering of the LED. The LED glowed for 10 msec after every 70 msec. All the tests were performed in static environment with a limited supply of glucose fuel and thus, the LED continued to flicker for over 40 minutes until all the glucose fuel in 180 mg/dL glucose solution was consumed after

which the biofuel cell failed to produce a voltage over 0.3 V due to lack of fuel supply. Fig. 7(a) shows a calibration curve in which the average frequency of charge/ discharge cycle of the capacitor was observed. The linear dynamic range observed is 1.8 – 630 mg/dl glucose with a regression coefficient of 0.993. The charge cycle across the transducing element capacitor was observed over a period of one second. Response time is an essential property of glucose biosensor and needs to be as quick as possible. The observed system's response time was 1 sec including the time required to process the signal. Increase in frequency of charge cycle was observed with an increase in glucose concentration due to more glucose molecules being present resulting into higher electrical parameters thus, producing rapid charge cycle across the transduction element capacitor. The linear trend in the charge cycle with increase in glucose concentration confirms that this system can be used as a glucose monitoring system. This system is capable of detecting hypoglycemic, normal, and hyperglycemic glucose levels. In addition to detecting different glucose concentration via charge/ discharge cycle of the transducer, the system exhibited a stable operation for 96 days after which the voltage dropped below 300 mV for 360 mg/dL glucose (Fig. 7(b)).




(a) (b)

**Fig. 7.** (a) A calibration curve for glucose monitor system (error bars indicate RSD). (b) 96 day stability profile in the presence of 180 mg/dL (blue) and 360 mg/dL (orange) Glucose (37 °C, pH 7; error bars indicated the RSD).

The charge pump IC was no longer able to amplify the small voltage produced since the minimum voltage requirement of the charge pump IC was not satisfied. The charge cycle across the capacitor was also monitored in 180 mg/dL glucose solution for 74 days without recalibration which is still higher than the previously seen [17]. In the presence of 180 mg/dL and 360 mg/dL glucose concentration, the system exhibited stable operation for first week followed by 9 % reduction in its activity at the end of over two months. The overall drop in the performance of the system was approximately 15 % and 24 % at the end of 96$^{th}$ day indicating stable operation of the glucose monitor, which surpassed the previous known results without recalibration [29-30]. The glucose monitoring system described here could greatly reduce the need to device recalibrate. The performance of the glucose monitor could be further improved by improving the performance of the glucose biofuel cell which will extend the life of the sensor. This sensing system's stability along with its stable operation at various pH and temperature demonstrated in our prior work [16] serves a strong candidate for a potential glucose monitoring system.

## 4. Conclusions

We successfully demonstrated the stable functioning of the glucose monitoring system. Our system demonstrated a stable operation in the presence of 360 mg/dL glucose solution for over 3 months which enabled effective functioning of the sensing circuit. The successful functioning of the charge pump IC enabled sufficient drive strength to light a LED. The system had an overall drop of 15.38 % and 26.47 % in the presence of 180 mg/dL and 360 mg/dL glucose concentration solution after 74 and 96 days of continuous operation, respectively. The stability of the system indicated that this device continues to sense glucose without the need for calibration for over three months. This glucose sensing microsystem has a potential to replace the currently available technology on the market if the durability of the system can be extended. The current work involves testing at neutral pH and the performance of the system drops considerably at physiologic pH (7.4) due to the optimal pH of oxygen reducing laccase enzyme being 5.5 – 6. Future work will focus on prototyping the sample which will involve minimizing the overall dimensions of our system as well as exploring enzymes that will enable the system's operation at physiologic pH and temperature.

## Acknowledgements

This work was supported by National Science Foundation (Award ECCS-#1349603).

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