IMPLANTED BIOFUEL CELLS

Nathan Guillaumat
School of Engineering
EPFL
Lausanne, Switzerland
nathan.guillaumat@epfl.ch

Lucien Gallezot
School of Engineering
EPFL
Lausanne, Switzerland
lucien.gallezot@epfl.ch

Arno Douady
School of Engineering
EPFL
Lausanne, Switzerland
arno.douady@epfl.ch

***Abstract*—Recent advancements in implantable biofuel cells (IBCs) have sparked significant interest for the past decade. This article will review the techniques, considerations and challenges in designing IBCs. The complexity of IBC design will be highlighted through in-vitro and in-vivo studies over the years. This review will cover the technologies behind electrochemical reactions, electrode design and a summary of key papers in the field.**

***Index Terms*—Implantable Biofuel Cells, Bioelectrochemistry, Biocompatibility, Energy Harvesting, Sustainable Power Sources**

I. Introduction

In pursuit of the widespread development of implantable bioelectronics, a major challenge to tackle has always been the development of sustainable, biocompatible, and non-invasive power sources. Rather than relying on external energy sources to power electroceutical devices, harvesting power from human body has always been a matter of interest, from watch industry to implanted medical devices [1]. While many sources of energy critically rely on mechanical activity (muscle movements, blood flow, heart beats) or thermal gradients between the body and its environment, or even sweat, energy harvesting based on internal chemical activity provides a continuous and stable power supply. The main goal of implanted biofuel cell is to manage powering electrical devices, using the human body chemicals. The topic gained a lot of attention early 2000s continued in the years 2010s to be extensively explored, with great improvements in enzymatic biofuel cells and their applications. Based on the principle of the fuel cell, that is to create electrical energy from chemical reaction, the biofuel cell and especially implantable ones bring many more challenges and considerations to address in their design, due to their need to be biocompatible for human implantation. In this article, we will review the state of the art in the field of implanted biofuel cells for various applications, discuss technology's benefits and drawbacks as well as their performances.

II. Biofuel and fuel cells technology

A. General fuel cell technology

As for modern batteries, fuel cells aim to create electrical current as a result of controlled chemical reactions. Fuel cells are made of a rare metal anode and cathode separated by an electrolyte solution for ion transfer (+membrane). In a hydrogen fuel cell, dihydrogen (H_2) is injected in the anode through a catalyst (typically platinum) helping its oxidation [2]. The reaction producing electrons at the anode is

$$H_2 \rightarrow 2H^+ + 2e^-$$

The produced electrons travels through an conducting wire from anode to cathode, producing current, while protons travel through an electrolyte to the cathode, thus creating electrical current. At the cathode, injected oxygen (O_2) molecules react with protons and electrons. The reduction half-reaction produces water (H_2O) molecules (Fig. 1), creating the following reaction :

$$\frac{1}{2}O_2 + 2H^+ + 2e^- \rightarrow H_2O$$

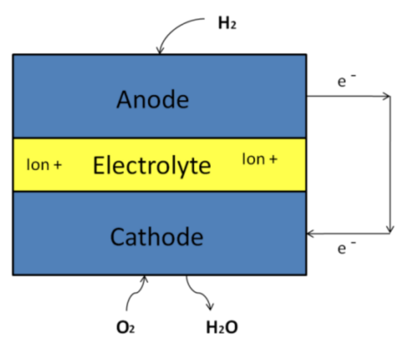

Fig. 1: *Block diagram of a fuel cell* [3]

This technology is promising and energy efficient, but it depends on hydrogen, which may be difficult to produce, store, and transport. Additionally, because precious metal catalysts are used, fuel cells are expensive and frequently function in extremely high pH environments.
Biofuel cells, on the other hand, provide an alternative by producing power from easily accessible organic materials, such wastewater or glucose. These cells provide a sustainable and possibly more accessible energy source by utilizing biological catalysts, such as enzymes or microbes, to transform chemical energy from biofuels into electrical energy.[4]

B. Microbial and enzymatic biofuel cells

Biofuel cells operate with the same principle as fuel cells, except that organic materials replace above-mentioned inorganic catalysts such as a biofuel, oxidized at the anode by the biological catalyst. The enzymatic fuel cells (EFCs) use enzymes to catalyze the oxidation of biofuels for example. The use of biological catalysts, mostly enzymes, has significantly improved the catalytic processes of glucose and oxygen degradation under physiological conditions, which has rekindled this very promising line of research in the past years.

In the case of biofuel cells powered by glucose and oxygen (Fig. 2), the Glucose oxidase (GOx) and glucose dehydrogenase at the anode are utilized catalyze the oxygenation of glucose to gluconolactone, producing hydrogen peroxide (H_2O_2) and releasing electrons (e^-) and protons (H^+)[5][6]. Moreover, Catalase is often co-immobilized with glucose oxidase at the anode to decompose the hydrogen peroxide (H_2O_2) produced during the oxidation of glucose. At the anode, we can find the reactions :

$$\text{Glucose} + O_2 \xrightarrow{\text{GOx}} \text{Gluconolactone} + H_2O_2$$

$$2H_2O_2 \xrightarrow{\text{Catalase}} 2H_2O + O_2$$

At the cathode, enzymes such as laccase (LAC), bilirubin oxidase (BOx), or peroxidase are commonly used [7]. These enzymes facilitate the reduction of oxygen (O_2), which combines with the electrons and protons to form water (H_2O), giving the following reaction at the cathode :

$$O_2 + 4H^+ + 4e^- \xrightarrow{\text{Laccase}} 2H_2O$$

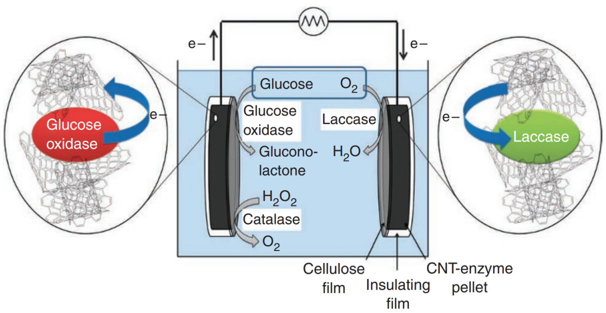

Fig. 2: *"Schematic presentation of the setup of a glucose biofuel cells (GBFC). At the anode, glucose is oxidized to gluconolactone, where the electrons are transferred from the GX to CT. Catalase decomposes hydrogen peroxide into oxygen and water. At the cathode, electrons are transferred from CT to laccase where dioxygen is reduced to water."*[6]

In ideal in-vitro conditions, this type of biofuel cells could reach a voltage of 0.95 V for open-circuit voltage, and significant power densities (1-1.54 $mWcm^{-2}$), thus could be a viable candidate of power source for implantable devices [6] [8] [9]. However, performances tend to be altered in in-vivo conditions, bringing further implementation problems.

On the other hand the Microbial fuel cells (MFCs) operate differently. In the anode chamber, microorganisms (such as bacteria) oxidize organic substrates (like glucose, acetate, or wastewater). A typical MFC contains anodic and cathodic chambers, which are separated by a proton exchange membrane (PEM) [10]. The microorganisms break down the organic matter, releasing electrons and protons. The general reaction at the anode can be represented as:

$$\text{Organic Matter} + H_2O \xrightarrow{\text{microbe}} CO_2 + H^+ + e^-$$

At the cathode, the electrons combine with protons (which have traveled through a membrane or salt bridge) and an electron acceptor (usually oxygen) to form water. The reaction at the cathode is:

$$O_2 + 4H^+ + 4e^- \xrightarrow{\text{microbe}} 2H_2O$$

resulting in the overall reaction [10] :

$$\text{Organic matter} + O_2 \xrightarrow{\text{microbe}} 2H_2O + CO_2$$

Due to their production of CO_2, the microbial fuel cells are challenging to implement. A wise management of the emission would be needed.

The EFCs have been mainly investigated in the context of research in implemented biofuel cells. In implantable bioelectrochemical systems, generally biofuel cells, natural biological components—typically enzymes—interfaced with electrodes have shown to be the most biocompatible and promising[11]. According to the literature, the engineered implanted biofuel cells used oxygen that was already present in large quantities in biofluids and physiologically created "fuel" to generate electricity using enzyme-modified electrodes. Most of current researches focus on glucose oxidation, which allows the usage of the abundantly available glucose in human blood as a source of energy for electronical devices, while operating in body-compatible environments[11]. One of the main benefits of glucose biofuel cells is their high conversion efficiency and ability to produce enough bioelectricity to power tiny electrical devices at room temperature and pH neutrality. The glucose and EFC is an intriguing path to investigate since glucose is often utilized as the "fuel" in the freely flowing hemolymph of invertebrates (snails, clams, lobsters, ...) and in the blood arteries of vertebrate animals (rats, rabbits, ...).

C. Abiotic Biofuel Cells

In implantable biofuel cells design, abiotic biofuel cells (ABCs) have been explored but are less common compared to EFCs. In contrast to biofuel cells that use biological catalysts (like enzymes or microorganisms), ABCs use inorganic catalysts to facilitate the oxidation of biological elements, such as glucose or lactate. The ABCs use catalysts like platinum, palladium or other metals to facilitate the oxidation of biofuels such as glucose, more suitable for long-term applications. Also, an important advantage of abiotic biofuel cell is their temperature resistance, allowing sanitation by sterilization[12]. Although the research on ABCs started in the 1970s, these cells were implanted in animals and demonstrated the potential to generate power from glucose in the body. Their development has been limited due to catalyst poisoning, low specificity and biocompatibility issues.[7]. Nevertheless, some research projects are underway on this subject [13].

III. Electrodes design for implants

A. Enzymatic cells

As electrodes act as the interface between the electrical system and biological environment, they must ensure biocompatibility and electricity production. One way to achieve such capapilities is the use of enzyme modified electrodes. Enzymes serve as catalyzers, oxidizing glucose and reducing oxygen respectively at the anode and cathode, thus creating a voltage difference.[14]

Furthermore, many parameters have to accounted for during electrode design and fabrication, such as electron transfer method. The latter element is critical as it defines how electrons will be moved from the enzyme to the electrode, thus defining system's electric density and efficiency. The pathway of electrons has to be shortened to allow transfer, leading to two possible methods : direct electron transfer (DET) and mediated electron transfer (MET). The main difference between these methods are the presence (or not) of a mediator allowing electron transfer, such as a polymer macromolecule or CNT structures[15].

Enzymes are immobilized to the metallic electrode surface using different techniques, such as absorbtion (which is less stable) or C-C covalent bonding. The latter method utilizes specific functional groups on the electrode surface, such as graphene and Carbon Nanotubes[15].
An widely used enzyme set often found in literature[16][17] uses biocatalytic electrodes (PQQ-GDH-anode and laccase-cathode) for implantation in living animals. Both laccase and PQQ-GDH (*pyrroloquinoline quinone dependent glucose dehydrogenase*) are well-studied enzymes commonly used for sugars and oxygen half reaction catalyzing [18].
However, a major limitation for implementation of enzyme modified electrodes are their limited lifetime, as they tend to degrade over time[19].

B. Abiotic cells

On other hand, one can take advantage of noble metals properties for abiotic fuel cell design. Promising results were observed using complex nanostructures, for example Platinum Nanoflowers as catalysts for glucose [20]. This technology utilizes porous structure having 3D molecular accessibility, hence enabling high chemical reductions and high electrical yield. Other materials are also currently being developped and enhanced, such as Titanium dioxide (TiO_2)[21] or other transition materials. However, major drawbacks occur for implantability of such biofuel cells, such as biocompatibility : recent results show a rapid poisoning due to the presence of amino acids[12].

IV. Voltage and power considerations

Most researches address technology improvements and power density increasing. However, as current can be increased with systems improvements, produced voltages are limited and barely reach 0.7V in practice[22], which usually is below minimal voltage for electronics applications.

Several approaches were developped to tackle this issue, such as series-connection of the electrodes or by charging capacitors in order to release bursts of higher-voltage energy.

The first approach was for instance tested in-vivo on lobster subjects[16]. As a single electrode could not produce enough voltage and was connected to an another electrode in series. Unfortunately, lobster tissues' low impedance caused short circuits limiting voltage increases. Final solution of the study was to implant single electrodes in two lobsters in series, enabling a 1V output voltage for optimized external load.

In addition, second approach is mainly limited by energy losses. This technology is already well-documented for non biological applications [23]. Main problem for this method is related to the fact that implanted biofuel cells produce very small currents, too small to power a charge pump. Moreover, implementation of storage systems overcomplicates the overall design.

V. State of the art of IBCs

A. Challenges of Implantable Biofuel Cells

Implantable biofuel cells (IBCs) are a great alternative to battery powered medical devices but several challenges hinder their practical application. Sterilization is a major issue as conventional methods like autoclaving or chemical treatments can denature enzymes which are essential for enzymatic biofuel cells. This could be avoided by specialized packaging materials that can withstand sterilization and maintain the structural and functional integrity of the cell components [24]. Another major concern is biocompatibility: implanted devices must not cause either an immune response or biofouling. Protein accumulation, cells and other biological debris on the electrode surface can block electron transfer and reduce energy generation efficiency. Moreover, the placement of the IBC is important: in blood vessels for example, non-flat devices can lead to thrombosis or embolism which are a serious safety risk [5]. The stability of the enzymes inside IBCs is another challenge, as enzymes like glucose oxidase are likely to degrade under physiological conditions due to pH fluctuations, temperature and presence of reactive oxygen species. Incomplete oxidation can also produce by-products like hydrogen peroxide that can degrade enzymes and shorten the operational life of the devices [24][5]. On top of that, achieving sufficient power output in a small form factor is a challenge. Low concentration of glucose and oxygen in physiological environment limit the energy available for bioelectrochemical reactions and efficient electron transfer between enzymes and electrodes is hard to optimize [5].

B. Current research and Performances

The first implantable biofuel cells (IBCs) were described in the 1970s [25][26], but the use of expensive and poisoning noble metals as catalysts hindered their further development [7]. Recent research on IBCs using other catalysts (e.g. enzymes) has made them more suitable for biomedical applications. One of the early major examples comes from Cinquin et al. [27], who implanted in 2010 a biofuel cell in the retroperitoneal space of rats, generating a power output of $6\mu W$. This was one of the first in vivo energy harvesting from glucose and oxygen using enzymatic biofuel cells. In 2012, Halamkova et al. [17] implanted enzymatic biofuel cells in snails to harvest glucose as a biofuel. The cells produced a maximum power density of $30\mu W/cm^2$. Altough the electrical output decreased rapidly when using the cell, the snails' natural ability to regenerate glucose through feeding and resting allowed to reiterate the operation after less than an hour. Halamkova et al. also demonstrated the use of buckypaper to limit biofouling, reproducing the experiment after a period of 2 weeks. The same year, Rasmussen et al. [28] implanted a BFC in the abdomen of a female cockroach, achieving a maximum power density of $55\mu W/cm^2$ at $0.2V$, decreasing of only 5% after $2.5h$ of operation. In 2013, Katz et al.[16] used two pairs of Glucose BFCs in two lobsters connected in series and in parallel and generated enough power to supply a cardiac pacemaker.

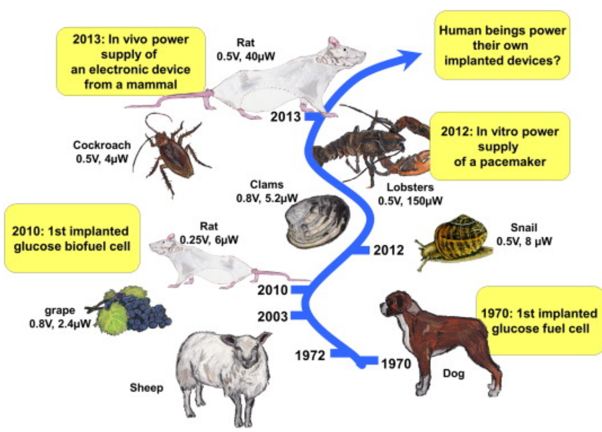

Fig. 3: *"Timeline of development of implanted biofuel cells/biosensors operating in vivo in living organisms"* [14]

A few years later, in 2016, El Ichi et al. [29] reported the longest in vivo study of a biocathode in a rat (almost 6 months), and demonstrated the use of a chitosan coating film for an increased biocompatibility and long term stability. The same group continued their research and they implanted not only the biocathode but complete G-BFCs in the abdominal cavity of a rabbit [30]. After being sterilized with gamma radiation, they remained active for almost 2 months. Most of the studies have been done on insects, mollusks and for mammals, rats and rabbits. But in 2021, Lee et al. [31] successfully implanted an EBC in a pigeon, reaching a maximum power of $0.08mW$, allowing to operate a brain stimulator in the flying bird. Recently, some innovative flexible designs have also expanded the use of IBCs. Indeed, stiff devices are not mechanically matching soft tissues in living creatures. Guo et al. [32] presented a fiber BFC based on carbon nanotube fiber, highlighting three main advantages: flexibility, anti-biofouling and small immune response. The device stably generated power under repeated deformations, with a maximum power density maintained over 90% during 100 cycles. Moreover, it showed a negligible immune response while generating long term power output, with less than 50% of loss after 1 month in vivo ($4.4\mu W/cm^2$ at the beginning versus $2.5\mu W/cm^2$ at the end). An even more recent study by Lee et al. [33] demonstrated EBCs integrated into artificial blood vessels using stretchable buckypaper electrodes. Lee et al. achieved with these devices an open circuit voltage of $0.59V$ and a power output of $26\mu W$ (under simulated physiological conditions), and showed potential for use in dynamic environments like pulsatile blood flow.

C. *Future research and usage*

As research in implantable biofuel cells encounter major challenges and drawbacks, promising technologies continuously emerge and bring new perspectives to the field. In 2022, MIT researchers [34] successfully created a ceramic-based electrolyte for glucose fuel cells. While reaching in-vitro satisfactory power output values, the usage of new ceramics open new hopes for abiotic fuel cell development, as used ceramics show biocompatible and temperature resistance characteristics.

VI. Conclusion

As the topics still remains an active focus of research, new papers still comes out with great and novel ideas. The technologies of biofuel cells are various and gives many opportunities to investigate for ways to create implantable devices, as many promising research such as the enzymatic biofuel cells that already presented their functioning on animals. However, some remaining challenges in biofuel cells make this technology complicated to develop. Even though the topic of research showed massive improvements over the past decades, the technology still lacks of power, or biocompatibility, and a significant performance gap persists between *in-vitro* and *in-vivo* conditions, thus limiting their implementation in humans in the next years. This technology could be useful, for various application in the future such as powering pacemakers. Energy harvesting for implanted applications open promises various medical electronic implants, pushing further researches towards machine-human hybrids.

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