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BIOFUEL CELLS: STATE OF THE ART AND PERSPECTIVES

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Abstract. On the basis of a comprehensive explanation of the principles of biofuel cells functioning, a review of the state of the art in this field is presented. Some of the most important achievements in developing the main elements of these new devices are described. The main biofuel cell peculiarities are presented. The crucial role of the biocatalysts is discussed. The types of cathodes and of anodes as well as of solid membranes are critically scrutinised. In this connection, the most important technological problems and their possible solving are discussed. The potentialities of implementating such devices in waste water treatment are assessed. The results of the research show that the biofuel cells can generate electricity from waste waters and other organic materials. However, the power density is still low for practical applications. The main goal of future R&D activity should be concentrated on a significant improvement of this parameter.

Keywords: biofuel cell, biocatalyst, solid membrane, power density, wastewater treatment

INTRODUCTION

A biofuel cell (biological fuel cell) is a device, which contentiously converts chemical energy of a reduced fuel into electricity with the help of a biological agent, i.e. a single strain or a consortium of microorganisms or isolated microbial enzymes (Sell, 2001, Shukla *et al.*, 2004). In fuel cells, a substance with reduction properties (fuel) is oxidized at the anode, while the produced electrons are transferred, via an external circuit, to a suitable electron acceptor molecule, or oxidant (such as oxygen) at the cathode (Sell, 2001).

Depending on the biocatalyst employed, biological fuel cells can be classified as enzymatic and microbial. Both types can utilize many different organic and inorganic substances as potential fuels, which is a major advantage of biofuel cells over other types of fuel cells (Sell, 2001). So far research in the field of *microbial fuel cells* has been mostly concentrated on biocatalysts which catalyzed the anodic reaction. (Sell, 2001, Shukla *et al.*, 2004, Rabaey *et al.,* 2005). On the other hand, in enzymatic fuel cells either the anodic or the cathodic reactions can be catalyzed by enzymes (Shukla *et al.*, 2004). A typical anodic biological fuel cell is shown in Fig. 1.

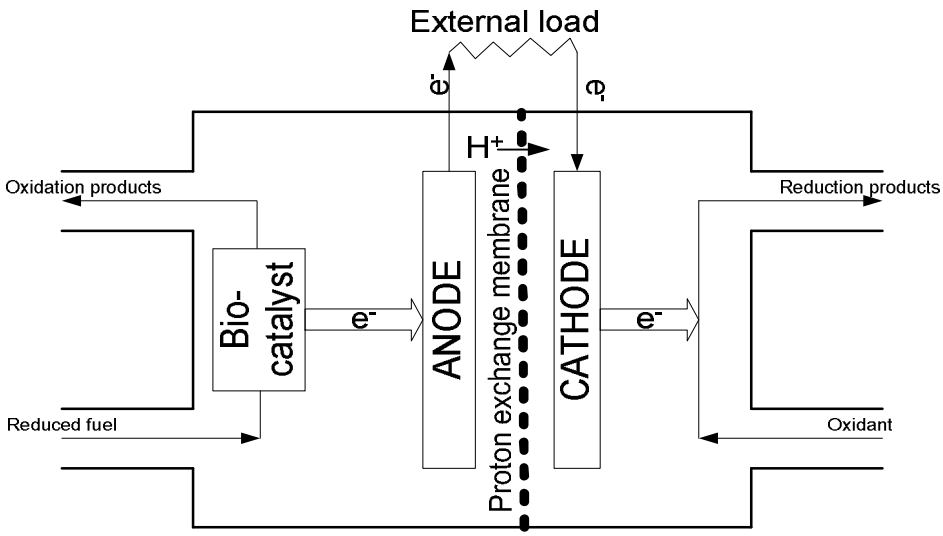

Fig. 1. Typical anodic biological fuel cell

Microorganisms and isolated enzymes can convert a vast variety of electron-rich compounds, which cannot directly react with the electrode of a fuel cell (e.g. hydrocarbons, lipids, etc.), into electroactive compounds, like hydrogen, methane and others substances, which can readily be oxidized at a conventional fuel cell anode (Sell, 2001). In this case the biocatalyst need not be in the same vessel as the anode, i.e. the biological reaction can take place in a separate bioreactor, and only the products of the biological reaction can be transported to a conventional fuel cell. This is the concept of *indirect biofuel cells*, and one of the earliest to be exploited in the beginning of the 1960s (Sell, 2001).

In *direct biofuel cells*, the biological agent directly contributes to the electron transfer (Sell, 2001). In the case of anodic microbial fuel cells, for example, bacteria switch from a natural electron acceptor, such as oxygen or nitrate to an insoluble acceptor – the fuel cell anode. Usually, due to steric hindrance the electrical communication between the biological agent and the anode is highly ineffective (Shukla *et al.*, 2004, Barton *et al*., 2004, Sell, 2001). In order to overcome that problem biological fuel cells may employ *mediated electron transfer* /MET/. In MET, a small-molecular-weight redox active species (*mediator*) is used to shuttle electrons from the biocatalyst to the electrode (Barton *et al*., 2004). Usually, enzymatic fuel cells work in the presence of a mediator (Shukla *et al.*, 2004), unless the enzyme is immobilized on the anode surface, in which case *direct electron transfer* is facilitated (Barton *et al*., 2004). In that case, the electrode surface acts as a cosubstrate for the enzymatic reaction. The catalytic effect of the enzyme is the reduction of the overvoltage for the reaction of the substrate (Barton *et al*., 2004). As for microbial fuel cells, these two mechanisms also imply, however, in some cases the microorganisms can produce electron-shuttling substances, and such don't need to be added to the system (Rabaey, 2005).

The comparison between enzymatic and microbial fuel cells is difficult due to their different nature. It can be noted that enzymatic fuel cells can produce higher power and current densities, but they are still generally suitable for low-power applications such as biosensors (Barton, 2004, Skukla, 2004). For this reason we are going to concentrate our review mostly on microbial fuel cells.

The history of microbial fuel cells starts in 1960s and 1970s (Sell, 2000, Rabaey, 2005c). However, only recently (at the end of the 20th century and the beginning of the 21st) more significant interest in the area has been noted. Particularly rapidly developing sub-area is the production of electricity from waste water in biofuel cells. At present at least three groups are working on the topic: Korneel Rabaey's group from Ghent University, Belgium; Bruce Logan's group (Pennsylvania State University, USA); and a group form Saint Louis University (USA).

Rabaey *et al.* (2004) list the following crucial parameters for the operational effectiveness of a fuel cell:

- Bacterial metabolism
- Bacterial electron transfer
- Performance of the proton exchange membrane
- Internal resistance of the electrolytes
- Efficiency of the cathode-oxygen electron transfer.

Nevertheless the most important process is the transfer of electrons from the bacteria to the electrode (Rabaey et al., 2004). In batch processes this parameter is controlled by addition of electron shuttling mediators, but for continuous regime this mechanism does not lead to increased performances (Rabaey et al., 2005b). Another method is to use bacterial strains that can produce electron mediators, such as phenzine (Rabaey et al., 2005d).

In order to facilitate biofilm formation, needed for effective waste water treatment, the anodic compartment of a fuel cell can be designed as a packed-bed reactor, with graphite granules used as anodic matrix and biofilm support (Rabaey et al., 2005b). This concept has been developed further in a design configuration of a cylindrical fuel cell (Rabaey et al., 2005a) and studied in a fuel cell stack (Rabaey 2006).

ELECTRODES FOR MICROBIAL FUEL CELLS

Types of Cathodes

The cathode is the electrode on which the electrochemical reaction of the oxidant reduction reaction takes place. There are several different configurations of cathodes which have been used in microbial fuel cells. Most electrodes are made from carbon due to its high conductivity and low reactivity. Microbial fuel cells have been demonstrated using cathodes constructed from plain graphite, graphite coated with platinum, reticulated vitreous carbon, carbon paper, graphite felt, carbon granules as well as several variations of these materials. For any interface chemical reaction, a large surface area is desirable. That is why many of the materials chosen for electrodes are fibrous, such as carbon paper and graphite felt. These materials have a very large surface area and make the overall surface reaction rates higher. Many cathodes make use of a potassium ferricyanide solution in the cathode compartment as an intermediate redox couple. As the ferricyanide is reduced by the fuel cell reaction, it is reduced to ferrocyanide. Therefore, the ferricyanide solution must be constantly replenished in order to maintain high current densities. An advantage of using a ferricyanide catholyte is that a noble metal electrocatalyst such as platinum is not needed on the electrodes. This substantially affects the cost of the fuel cells as the electrodes typically account for 70-80% of the cost of a hydrogen fuel cell which uses a platinum catalyst.

Types of Anodes

The anode is the electrode on a surface of which the oxidation reaction of the fuel occurs. The anode chamber is typically where the organic matter such as wastewater is digested by microorganisms. Many different anode configurations have been used to demonstrate microbial fuel cells including plain graphite electrodes, platinum coated and plain carbon paper, vitreous carbon, graphite felt with bound electron mediators, carbon cloth, and graphite granules. In general, the greater the surface area, the better performance will be observed from the fuel cell.

Comparison of current densities

There are many factors which affect the final current density of a microbial fuel cell. The electrode design is one of the most important because current density is very sensitive to fuel cell internal resistance. Internal resistance is directly related to the design of the anode and cathode. Although most microbial fuel cells are being designed with the intention of generating power from wastewater, relatively few research groups use actual wastewater to test their fuel cells. Most use simulated wastewater consisting of a dilute glucose or acetate solution. While this is useful for the purpose of setting benchmarks, the performance of a fuel cell will be lower when running on real wastewater compared to simulated wastewater.

Park et al. (2003) developed a single chambered microbial fuel cell with graphite felt electrodes covalently bonded with electron mediators to eliminate the need to constantly supply them with the influent. The electrodes were made from 80 cm^2 pieces of graphite felt which were estimated to have a total surface area of up to 1.27 m^2. The best results they achieved occurred when using graphite felt bonded with Mn^{4+} on the anode and graphite felt bonded with Fe^{3+} on the cathode. The power density they achieved with this configuration was 787.5 mWm^{-2}.

He *et al.* (2005), developed a continuous flow microbial fuel cell to generate electricity from wastewater. The electrodes were made of reticulated vitreous carbon (RVC), which has a highly porous, rigid, open cell carbon structure. The RVC used in the anode had greater porosity to help prevent clogging due to a biofilm build-up. The maximum power density they achieved using this reactor was 170 mWm^{-2}.

Bruce Logan's research group at Pennsylvania State University have reported using a number of different microbial fuel cell designs to generate electricity from wastewater sludge, domestic wastewater and simulated wastewater (Oh et al., 2004; 2006). They constructed a simple reactor consisting of two

media bottles joined by a tube containing a proton exchange membrane. They used this reactor to test the effectiveness of different types of electrodes on the same system. They used plain carbon paper electrodes, carbon paper with deposited platinum, and plain carbon electrodes with ferricyanide in the cathode compartment in order to compare the effect various electrodes would have on the power output of the fuel cell. The plain carbon paper electrodes had the lowest power density, followed by the platinum electrode which had 0.097 mWm^{-2}, approximately three times higher than the plain carbon electrode. The best results were obtained from the plain carbon paper electrodes with ferricyanide in the cathodic compartment. Using this configuration a power density of 0.12 mWm^{-2} was achieved (Bookimin et al., 2004).

The same group has also reported generating electricity from simulated and domestic wastewater using a single chambered microbial fuel cell. In a single chambered fuel cell instead of having a cathodic compartment with an electron acceptor, such as aerated water or ferricyanide, the cathode is exposed to the air directly. This simplifies fuel cell design and decreases size. The electrodes used in this fuel cell were constructed of plain carbon paper. This fuel cell resulted in a power density of 506 mWm^{-2} when running on acetate (Liu et al., 2005). Another test was done using platinum coated carbon paper as a cathode with a PTFE diffusion layer on the air side of the cathode, which resulted in a power density of 766 mWm^{-2} (Cheung et al., 2006).

Finally, Rabaey et al. (2003) have reported results using a wide range of microbial fuel cell designs including continuous flow tubular fuel cells and stacked multi-cell fuel cell batteries. They have reported a fuel cell that uses plain graphite electrodes with ferrocyanide in the cathodic compartment to generate electricity from glucose with a relatively high power density of 3600 mWm^{-2}. Later they reported a new configuration of microbial fuel cell which had a tubular design with the anodic chamber in the center, filled with graphite granules which acted as the anode, surrounded by a proton exchange membrane and a woven carbon fiber mat for the cathode. The maximum power density achieved with this fuel cell was 90 mWm^{-3} using acetate (Rabaey et al., 2005). More recently they have also published a report of a stacked microbial fuel cell which also used graphite granules as the electrode in the cathode compartment in addition to the anodic compartment. The graphite granules made a very good electrode material because of their high surface area to volume ratio. This fuel cell had a power density of 258 mWm^{-3}. It also ran on acetate (Aelterman et al., 2006).

SOLID MEMBRANES IN BIOFUEL CELLS

For the time being there are several types of biological fuel cells (Shukla et. al, 2004), namely microbial and enzymatic fuel cells. The latter ones, in most cases, are membraneless or non-compartmental, whereas microbial ones are very similar to conventional PEM fuel cells (Figure 1) and consist of cathode and anode separated by an ion exchange membrane.

A distinctive feature of a biofuel cell is that electrode reactions are controlled by microorganisms or enzymes, while in the conventional (chemical) ones catalysts play a key role. Beyond that, biological fuel cells operate under mild reaction conditions, mainly ambient temperature and pressure that is more appealing in terms of operation and, consequently, maintenance.

PARAMETERS AFFECTING FUEL CELL PERFORMANCE

First of all, it should be noted that biological fuel cells are much more complicated system compared to chemical ones because they involve biological and electrochemical processes, which are closely related to each other, and thereby the power generated by this type of cells is dependant on both these processes. Therefore, satisfactory performance of the cell is possible provided that very careful tuning of the overall system.

Thus, the main parameters that must be taken into consideration at designing biofuel cells are the following (Rabaey and Verstraete 2005c):

- Substrate conversion rate;
- Overpotentials at the cathode and the anode;

- Proton exchange membrane performance;
- Internal resistance in the biofuel cell.

Requirements for a good membrane separator in fuel cells

Since, along with electrodes, the membrane is one of the core components of a fuel cell, it has great impact on the overall fuel cell efficiency, and ideally it should be able to inhibit the transfer of the electron donor (fuel) or electron acceptor (catholyte) while conducting protons to the cathode with low resistance. In this regard, in order to achieve high fuel cell performance, the following properties of a membrane are required (Dunwoody and Leddy, 2005):

- High ionic (protonic) conductivity to support high currents with minimal resistive losses, but low electronic conductivity;
- Low fuel and oxidant permeability;
- Limited crossover and solvent transport;
- Thermal, chemical and electrochemical stability at operating conditions;
- Low cost;
- Good mechanical properties and easy fabrication of fuel cells.

In turn, the ionic conductivity depends greatly on a membrane hydration, i.e. the higher hydration results in a higher conductivity. However, from the other side, excessive membrane hydration is not desirable in terms of slowing down oxidation reaction due to dilution of catholyte in close proximity to the membrane. For example, Nafion 117 membrane equilibrated with water vapour has the electro-osmotic drag coefficient equal to unity, which is defined as the number of water molecules transported per one proton, whereas that immersed in water has the drag coefficient of about 2.5 (Smitha et al, 2005).

One of the ways to avoid excessive water crossover is to use thinner membranes, that in parallel, allows for improving overall fuel cell performance owing to lowering membrane resistance. However, like in previous case, it is not versatile way, because membrane thickness can be reduced up to the certain extent, beyond that, problems with durability and fuel by-pass begin to appear.

Another way for enhancing conductive properties of the membrane is a spatial control of acidic regions and charge density in its chemical microstructure. For this reason, scientists have been studying various types of conductive materials, which can be used for membrane production and could be able to overcome existing shortcomings.

Types of membrane materials

The variety of membranes used in fuel cells can be split into several main groups based on their chemical origin. To date, the first group of perfluorinated membranes is the best studied and the most widely used with its most vibrant representative – Nafion membrane – in the lead. These membranes possess high conductivity and very good chemical and electrochemical stability. However, this type of membranes has not been successful at applying to biofuel cells, in particular enzymatic ones, since at immobilizing enzymes Nafion forms acidic membrane, which reduces activity and lifetime of enzymes (Akers et al, 2005). Beyond that, conductivity of membrane is greatly dependant on the operational temperature that makes its use, in some kinds of fuel cells, very ineffective, and finally, these membranes do remain too expensive and their impact on the environment is still questionable (Lojoiu et al, 2005). In this regard, several attempts have been made on modifying Nafion membrane by incorporating silica and titanium dioxide particles, by doping with heteropolyacids (Smitha et al, 2005). However, the most remarkable result was obtained after incorporation of thiophene in Nafion 117 membrane.

From the other side, partially fluorinated hydrocarbon membranes are less expensive compared to Nafion membrane and their structure allows for introduction of polar sites as pendent groups, which plays a critical role in increasing water uptake (Smitha et al, 2005).

Nasef et al. (2006) proposed the technique for preparation of polystyrene sulfonic acid pore-filled poly(vinylidene fluoride) membrane by simultaneous electron beam irradiation. The synthesized

membrane exhibited superior performance characteristics compared to Nafion membrane at testing in direct methanol fuel cell.

However, mechanical and thermal stability of the above-mentioned kind of membranes is not very good, and in order to enhance these parameters the incorporation of aromatic hydrocarbons into the backbone polymer is applied (Lojoiu et al, 2005).

Membranes prepared on the basis of acid-base complexes, that is membranes with acid components incorporated into an alkaline polymer bas, show fairly high conductivity in the wide temperature range, including elevated temperatures. Moreover, conductivity of these membranes is not dependant on humidity in contrast to Nafion membrane. Another very important and remarkable thing is that electro-osmotic drag coefficient for that was found to be zero (Bouchet et al, 2001).

Although it is quite difficult to compare efficiencies of existing membranes because each of them has its inherent advantages, from the information available in the literature it is believed that Nafion is the leader in the group of perfluorinated membranes, the sulfonated poly(4-phenoxy benzoyl-1,4-phenylene) in the non-fluorinated group and very promising is the acid-base membrane doped with phosphoric acid (Smitha et al, 2005).

As it was already mentioned above, to date Nafion membrane is the best one in terms of conductivity and stability, however, it is not suitable for using in biological systems due to its very high sensitivity to biofouling.

Rabaey et al. (2004, 2005) have carried out quite extensive research in finding the most appropriate membrane for biosystems and came to the conclusion that Ultrex membrane (Membranes International) can give the best results in biofuel cell applications. Unfortunately, there is no information available in the literature on this membrane except that concerning performance characteristics with its employing in biological fuel cells. So, it was impossible to make a deep analysis of its advantages and disadvantages in detail.

SIMULTANEOUS ELECTRICITY GENERATION AND WASTEWATER TREATMENT

Aerobic treatment processes are among the most popular ones for wastewater treatment. Only in the U.S. over 126 billion liters of domestic wastewater is treated aerobically each day at an annual cost of over $25 billion (Liu et al. 2004). Anaerobic treatment technologies provide potential for reducing costs, but many wastewaters are too dilute to make current anaerobic treatment technologies, based on methane production, economical (Logan, 2004). One method to reduce the cost of wastewater treatment is finding new useful products other than methane, from wastewater treatment. Hydrogen and electricity are the most promising products that can be produced from wastewater.

It was discovered only several years ago that electricity can be generated during wastewater treatment in a microbial fuel cells. These microbial fuel cells are based on an anaerobic microbial process in the anodic chamber of a biofuel cell. The bacteria growing on the anode oxidize the organic components in the wastewater and transfer the electrons to the anode. In order to maintain the charge balance, protons are also created and these protons must be able to migrate to the cathode so that they can combine with electrons and oxygen to form water. The generation of electrical current and the potential difference between the cathode and the anode chambers create the basis of the microbial fuel cell. These microbial fuel cells have the maximum potential of 0.5 to 0.8 V, which is similar to that generated in a hydrogen fuel cell. This low voltage can be transformed to produce a higher voltage or converted from DC to AC (Logan, 2004). In the first published studies on electricity generation in microbial fuel cells, the electricity generation was very low, but in the recent years there have been substantial improvements in power generation.

Liu and Logan (2004) reported a power output of 146 mW/m^2 in a batch system using domestic wastewater. A maximum power output of 26 mW/m^2 was recently achieved in a continuous system using wastewater (Liu et al.

2004). The system that was used to generate power at this level was a cylindrical plexiglass chamber containing eight graphite rods arranged in a concentric form around an inner cylindrical direct-air cathode. In this system along with electricity generation, up to 80% of BOD was removed.

In a tubular, single-chamber, continuous microbial fuel cell, the maximum power outputs were 59 and 48 W/m^3 net anodic compartment for digester effluent and domestic wastewater respectively. In this microbial fuel cell using feed stream based on acetate and glucose, power outputs of 90 and 66 W/m3 net anodic compartment were obtained. (Rabaey et al. 2004).

CONCLUSION

As a conclusion, biofuel cells have been shown to be able to generate electricity from wastewater and other organic materials. However, the power density is still too low for practical applications. The main goal of the future work should be concentrated on a significant improvement of that parameter.

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ÁÈÎÃÎÐÈÂÍÈ ÊËÅÒÊÈ: ÑÚÑÒÎßÍÈÅ È ÏÅÐÑÏÅÊÒÈÂÈ

Ê. Ïåíåâ, Â. Ïóðêåâè÷, Á. Áàô÷åõñàðàè, Í. Ãðîóáóðã, Ä. Êàðàìàíåâ

Ðåçþìå: Íà áàçàòà íà îáÿñíåíèå íà ïðèíöèïèòå íà äåéñòâèå íà áèîãîðèâèçèðàùèòå êëåòêè å ïðåäñòàâåí îáçîð íà ñúñòîÿíèåòî íà ïðîáëåìèòå â òàçè îáëàñò. Ï îêàçàíè ñà íÿêîè îò íàé-âàæíèòå ï îñòèæåíèÿ â ðàçâèòèåòî íà òåçè óñòðîéñòâà. Ðàçãëåäàíè ñà íàé-ñúùåñòâåíèòå îñîáåíîñòè íà ôóíêöèîíèðàíåòî è ì. Èçêëþ÷èòåëíàòà ðîëÿ íà áèîêàòàëèçàòîðà å äèñêóòèðàíà. Òèï îâåòå íà àíîäèòå è êàòîäèòå, êàêòî è íà òâúðäèòå ìåìáðàíè ñà ðàçãëåäàíè ïîäðîáíî. Íà òàçè áàçà ñà îáúäåíè íàé-âàæíèòå òåõíîëîãè÷íè ïðîáëåìè. Îöåíåíè ñà âúçìîæíîñòèòå çà èçïîëçâàíåòî íà áèîãîðèâèòå êåòèò â ïðå÷èñòâàíåòî íà îòïàäíè âîäè è èçïîëçâàíåòî íà äðóãè îðãàíè÷íè ìàòåðèàëè çà ñóðîâèíè. Ðåçóëòàòèòå ïîêàçâàò, îáà÷å, ÷å êîëè÷åñòâîòî åëåêòðè÷åñêà åíåðãèÿ, êîåòî ñå ïîëó÷àâà å âñå îùå ìíîãî ìàëêî çà äà ñå çàïî÷íå ïðàêòè÷åñêîòî è ïðèëîæåíèå. Ãëàâíàòà öåë íà áúäåùèòå èçñëåäâàíèÿ å çíà÷èòåëíî ïîâèøåíèå íà êîëè÷åñòâîòî äîáèâàíà åëåêòðîíåðãèÿ.