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REVIEW

Discover the possibilities: microbial bioelectrochemical systems and the revival of a 100-year–old discovery

Uwe Schröder

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Abstract Microbial fuel cells (MFC) can be considered as archetype microbial bioelectrochemical systems that exploit the bioelectrocatalytic activity of living microorganisms for the generation of electric current. "Microbial fuel cells represent a novel technology that is still in its infancy" is a typical statement in the current articles to describe the state of research on microbial fuel cells. It is a quite absurd statement for a technology that to the date has a history of 100 years. At the same time, it is quite correct since the actual, systematic MFC development has started only about a decade ago. The history of the MFC development reads like a science fiction novel—we read about space research and autonomous robots ("gastrobots"). But yet, the current tremendous interest in this technology is rather down to earth—it is about energy recovery and wastewater treatment. To understand the presence and to predict and actively shape the future, it is a good opportunity to look into and learn from the past. This article shall go on a short journey through the past 100 years of microbial fuel cells. This is not to be understood as a comprehensive and complete survey of the MFC literature and development. It is rather a personal view on critical aspects in the past development and the possible future of this technology.

Keywords Microbial bioelectrochemical system · Microbial fuel cell · Bioelectrocatalysis

U. Schröder (✉)
Technische Universität Braunschweig,
Institute of Environmental and Sustainable Chemistry,
Hagenring 30,
38106 Braunschweig, Germany
e-mail: uwe.schroeder@tu-braunschweig.de

The past

The year 2011 marks the 100th anniversary of a study generally considered as the first microbial fuel cell publication. In 1911 the occurrence of an electromotoric force between electrodes immersed in bacterial or yeast cultures and in sterile medium in a battery-type setup was reported by Michael C. Potter [1]. In this communication Potter came to the conclusion that electric energy can be liberated from the microbial disintegration of organic compounds. Twenty years later Cohen confirmed these results and reported a stacked bacterial fuel cell delivering a voltage of 35 V at a current of 0.2 mA [2]. Although these publications may be considered as the hours of birth of microbial fuel cells, the early resonance was rather skeptical. Thus, in 1963, Davis stated laconically: "… he (Dr. Cohen) reported only briefly on his bacterial battery and then presumably turned his attention to other, more interesting things" [3] and "what are you going to do with such a small amount of electrical energy" [3]. In the 1960s the idea of microbial electricity generation was picked up again, in the framework of the NASA space program—as an opportunity to recycle human wastes to electricity during

space flights, e.g., [4–8]. Unfortunately, the resulting activities were only short lived. The rapid advances in other energy technologies like photo voltaics and later also the low prices for mineral oil again lead to a decreasing interest in microbial fuel cells. In the 1980s again, a slow but steady interest in microbial fuel cells was established. This interest was particularly inspired by the work of Bennetto [9, 10], who also brought this fascinating technology to public attention [11]. During this period the idea of microbial power generation was picked up by Wilkinson who proposed the application of microbial fuel cells (MFC) for the powering of self–sustaining (eating) robots—so-called gastrobots [12–14].

Although this development caused public interest, the actual MFC development stagnated at comparatively low publication level until the turn of the century. Thus, between 1980 and the year 2000, annually an average of only three publications, published by a few active groups, appeared (see Fig. 1). To understand the reasons for the rather sluggish development of microbial fuel cells in the past century, one may discuss the following reasons:

1. *MFC performance* In 1931 Cohen stated that although anaerobically grown bacterial cultures may exhibit a strongly negative potential, the produced current was generally minute [2]. Cohen ascribed this low current generation capacity to a lack of "electromotively active oxidation–reduction products." As a solution to this problem, he proposed the use of inorganic or organic substances to facilitate the electron transfer from the microorganisms to the MFC anodes. The use of these so-called redox mediators indeed facilitated the transfer of electrons from the microbial metabolism to the MFC anode. The approach was reanimated in the 1980s especially by Bennetto and co-workers [9, 10]. In the following, a large number of compounds (mostly phenazines, phenothiazines, phenoxazines, and quinones) were investigated for their suitability and behavior as MFC mediators, e.g., [9, 10, 15, 16]. Although the average current densities were comparatively low and usually well below 100 μA cm^{-2}, there are studies in which current densities of up to 170 μA cm^{-2} (with 2D electrodes) and about 1.3 mA cm^{-2} (with a packed bed anode) were reached [17–19]—values that were already in the range of current MFC. Thus, the often discussed poor performance of these "early" systems may not be the sole reason for the low acceptance of MFC at that time.
2. *Practicability and sustainability* The use of artificial mediators has several crucial disadvantages. First of all, at higher concentrations, most redox mediators are toxic to bacteria. Thus, the concentration level (that also determines the limiting current density) has to be kept in the low μ–molar range; possibilities to increase the anode performance via an increased mediator level are thus very limited. Further, it has been shown that the current generation in mediated MFC substantially inhibited the growth of the used microorganisms. The reason is that the mediator diverts ("steals") electrons from the metabolism of the bacteria and thus limits the energy gain for the microorganisms. Especially for the already low-energy gain of the, at that time, mostly used fermenting bacteria (e.g., *Escherichia coli*, *Proteus vulgaris*, *Clostridia*, *Actinobacillus*) or yeasts, this energy loss did not allow a sustainable growth.

   The greatest disadvantage of the use of exogenous redox mediators, however, is the necessity of a regular addition of the exogenous compound to the substrate solutions. This is certainly technologically and economically unfeasible. Further it is environmentally questionable since the mediators are not consumed or degraded during the bioelectrochemical processes and would thus have to be removed from the effluent solution. These disadvantages not only made any large-scale application unthinkable but lead to the general abandonment of the approach.
3. *Missing application focus* Before the year 2000, microbial fuel cells were commonly seen as a scientific peculiarity. For most scientists it was fascinating to see the possibility of producing electric current directly from the metabolic activity of microorganisms. Potential applications were, as mentioned above, seen in the space program of the NASA as well as in the robotics. Whereas the former target was already abandoned at the end of the 1960s, the development of microbial fuel cell technology for the powering of autonomous robot systems is still followed in some labs [20, 21]. Further, the potential of microbial fuel cells as biosensors was

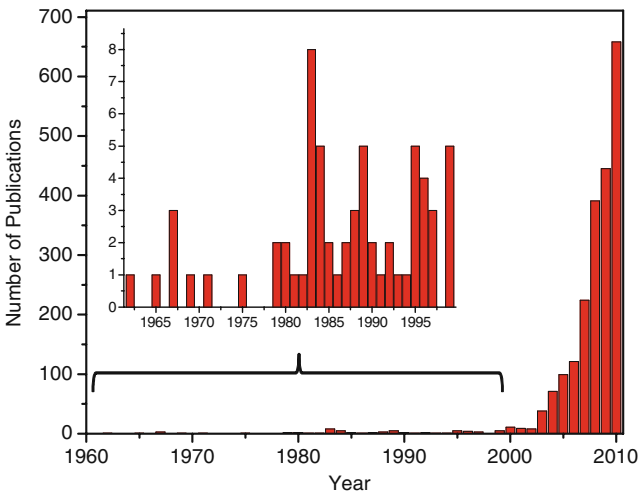

Fig. 1 Number of publications from 1960 to 2010 containing the phrase "microbial fuel cell" (SciFinderScholar, February 2011)

discussed [22–25]. The prominently mediator-based MFC, or MFC based on artificially immobilized microorganisms [26–28] did not allow to think about large-scale applications. On the other hand, medical applications like implantable fuel cells, as a major driver for the development of enzymatic fuel cells [29], were (and still are) certainly out of question.

These examples illustrate that before the year 2000, microbial fuel cells did not have a tangible application focus. There was practically no "killer application"—to speak in computer technology terms. And without such focus and application target, the overall interest in this technology remained very limited.

4. *"It was the wrong time"* Right from the first reports, it became obvious and acknowledged that microbial fuel cells offer the possibility to convert a virtually unlimited spectrum of organic matter into electricity. Yet, at that time, before the growing awareness of upcoming climate changes and recourse depletion, the environmental importance of this ability—namely to recover energy from organic waste in order to save energy and to reduce CO_2 emissions—remained unrealized, or was practically not in demand.

The present

Without exaggeration one can state that since the turn of the century, microbial fuel cells have entered a new stage of development. The technology is no longer considered a scientific peculiarity and side issue but as a serious research subject. This is nicely illustrated in the number of publications that contain the phrase "microbial fuel cell" (see Fig. 1): Between the years 2001 and 2010, in total, 2,193 papers have been published on MFC (the majority, i.e., 1840, in the past five years, between 2006 and 2010). In the years 1960–2000, the overall number was just 75.

How can this impressive change be interpreted? Well, first of all, one may state that for the development of any alternative means of energy production and conversion, the global political conditions have changed:

1. *"It is now the right time"* Based upon the growing awareness of exhausting resources of fossil fuels and the emerging environmental consequences of their use, global scientific efforts to develop technologies for a sustainable handling of our environment and our planet's resources have been triggered—efforts that also explain an interest in microbial fuel cells.

   Yet, only in combination with a suitable application focus that attracts sufficient interest in the research communities, a sustainable and lasting development can take place.

2. *Application focus* Beside the conservation of the future energy supply, clean water represents a major challenge for human kind. Both challenges are strongly connected. Thus, increasing amounts of energy are consumed to provide clean water and to clean wastewater before it is released into our environment. For example, in industrial countries, the treatment of wastewater causes out 10% of the municipal energy consumption (see, e.g., [30] for the example of Stuttgart). In the United States of America, wastewater treatment causes about 1.5% of the total electricity consumption [31]. At the same time, with an average of 3.3 kWh m^{-3} of stored chemical energy [32], the energy content of municipal wastewater is considered to be nine times higher than the energy demand necessary for its treatment [33]. These numbers illustrate that despite the growing efforts to recover energy from wastewater (e.g., via anaerobic digestion), energy is afforded to abolish the valuable energy contents in our wastewater.

   In 2001 it was for the first time proposed to use wastewater as a fuel to power microbial fuel cells [34]. Such a process would allow to produce electric current and to simultaneously remove organic matter from the wastewater. Since that, this possibility to directly combine wastewater treatment and energy recovery has become the major driving force in the development of microbial fuel cells, e.g., [31, 34–38], and surely, this new target application is responsible for the upturn of this technology. But certainly, without a practical and technically feasible concept to wire microbial activity to a fuel cell anode, such an upturn would not have happened.

3. *Practicability and sustainability* The greatest misconception in the past development of microbial fuel cells was the assumption that microorganisms are evolutionarily not designed to transfer electrons to a fuel cell anode. The assumption was actually true for most of the, usually fermentative, microorganisms tested for microbial fuel cell application at that time. Thus, artificial means to divert electrons from the microbial metabolism were established—with all the above-described constraints and disadvantages.

   It was primarily the credit of Prof. Byung Hong Kim who realized that in wastewater or wastewater sludge, plenty of bacteria are present that are able use a plain electrode (e.g., graphite) as an electron acceptor—without the help of an artificial mediator [34, 39, 40]. He named this MFC type a "mediator–less microbial fuel cell" [40]. Derek Lovley, who studied microbial metal oxide reduction [41–43], discovered the importance of metal-reducing bacteria like *Geobacter sulfurreducens* as electroactive bacteria

that are capable of performing extracellular electron transfer, for microbial fuel cells [44, 45]. In the upcoming years, more and more of these electroactive bacteria (often also referred to as electricigens, electrode respiring bacteria or exoelectrogenic bacteria) have been discovered [46, 47], a development that is still ongoing. These discoveries can doubtlessly be considered as a turn of the tide in the development of microbial fuel cells. It now became obvious that extracellular electron transfer does not require artificial means but is a process that in nature occurs in many anoxic compartments, such as in sediments, in soil, and in biofilms. The electroactive bacteria perform anaerobic respiration, and the electrons that are liberated in the substrate oxidation are stoichiometrically transferred to an extracellular electron acceptor—the anode in case of the MFC. It is a win–win situation: the microorganisms get the opportunity to respire and dispose the resulting electrons to the anode, and we receive them as electric current. This situation is crucial for the sustainable operation of the microbial bioelectrochemical systems (BES). For the description of the individual electron transfer mechanisms, the reader may be referred to one of the recent review publications [48].

Another very simple measure and at the same time major breakthrough was the use of natural sources as inoculum for the MFC anode [39]. Now it was not anymore required to select isolated bacterial strains for their potential ability to decompose a given substrate but simply use natural sources like sewage or soil as inoculum. Based on the two selection criteria—the ability to digest the provided substrate and the ability for an extracellular electron transfer—efficient electrocatalytically active microbial biofilms are formed that even do not require operation under sterile conditions [49].

In the past, the major MFC research was focused on the understanding and engineering of the microbial anode processes. In this course, the cathode reaction was almost neglected and often experimental, easy–to–handle cathode systems (e.g., based on ferricyanide) were used. During the last years, the community has become aware that these unsustainable cathodes are no option aside the lab bench [50]—an awareness that triggered considerable research efforts towards cheap and sustainable cathodes for microbial bioelectrochemical systems. For the details of these developments, the reader may be referred to one of the recent review articles [51, 52].

4. *MFC performance* At the beginning of the recent MFC development, the electrocatalytic performance of microbial biofilm anodes was low in comparison to approaches that used bacterial suspensions (see Fig. 2). Especially the use of suspensions of hydrogen producing fermentative bacteria in combination with tailored electrocatalytic anodes [53–58] clearly outperformed the biofilm anodes. However, the necessity to use additional anode electrocatalyst and the disadvantages of suspended biocatalysts for a continuous MFC operation did not help this approach to a breakthrough. At the same time, the biofilm anodes have run through a remarkable development: During the past decade, their performance basically increased from originally microampere per square centimeter level up to well above 1 mA/cm^2 (Fig. 2). This development can be attributed to the improvement of the microbial biofilms moiety, e.g., via sophisticated enrichment and acclimatization procedures [59–61], as well as to improved electrode biocompatibility via surface modification [62–64] and the development of sophisticated three–dimensional electrodes [65–67].
5. *From MFC to MXC—the versatility of microbial bioelectrochemical systems* The vast majority of previous BES-related research dealt with MFCs. Thus, for a long time, the cathode focus was put almost exclusively on the oxygen reduction reaction [68]. Recently, the expansion from pure electricity generation to the reductive formation of energy-rich chemicals, i.e., of H_2, CH_4, and H_2O_2 in microbial electrolysis cells to the use for reductive remediation processes (which one might denote as a microbial remediation cell) and even to microbial desalination cells [69] allowing the desalination of salt water opened novel venues. Thus, microbial bioelectrochemical systems are now much more than microbial fuel cells—MFC have got compa-

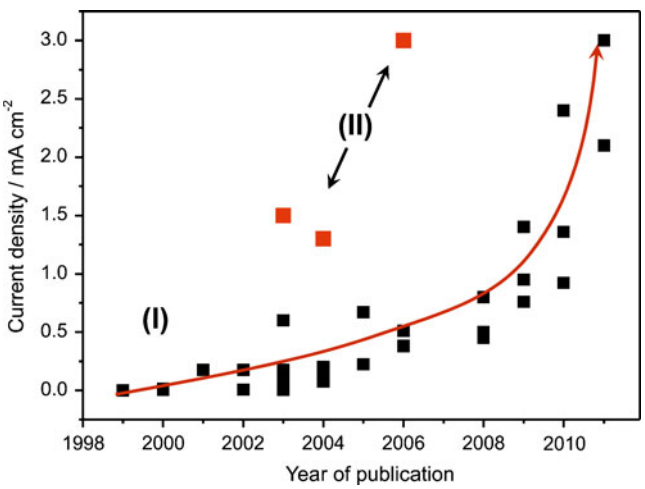

Fig. 2 Schematic and exemplary illustration of the development of the anode performance in microbial fuel cells. *(I)* Electroactive biofilm anodes, *(II)* use of suspended, fermentative bacteria in combination with electrocatalytic anode materials

ny from a multitude of new concepts of bioelectrochemical cells, which one could denote as "MXC." All these BES create additional functionality that open new application aspects (see "The future").

The future

Are we already where we want to be? Are microbial bioelectrochemical systems already an established technology? The answer is a clear "no." Some people may even argue that we do not even exactly know where we want to be and where the journey goes. In terms of technological realization, the technology is indeed still in its infancy. We may state that microbial fuel cells and related BES are on the threshold from the lab bench to technological implementation. But, the number of actual pilot projects is still very limited and the list of issues to be solved is remarkable. Thus, whereas the performance of individual MFC components like that of the anode has improved impressively over the past years, the overall fuel cell performance (i.e., the performance of the assembled cells) considerably lacks behind this progress. Here, innovative fuel cell designs that allow low internal resistances and enable upscaling (at low material costs!) are required. The apparently inevitable pH splitting between anode and cathode [70]—is there any way to solve this problem, or should we rather try to exploit it [71]? How shall we integrate microbial BES in the wastewater treatment [72, 73]? Is the classical MFC (with the primary function of electricity generation and COD removal from wastewater) the ultimate goal? A success is far from certain. In this discipline MFC will always have to compete with a technically mature and established technology like anaerobic digestion that requires low investment and maintenance costs and high conversion rates [74]. Applications will have to be identified in which microbial bioelectrochemistry is unrivalled. Thus, there are further exciting applications emerging, among which the use of microbial BES for the bioelectrocatalytic carbon dioxide reduction [75–77] and bioelectrosynthesis [78]. These are certainly very exciting new concepts that make me look forward to the upcoming future developments.

Despite the uncertainties, microbial bioelectrochemical systems like microbial fuel cells are a fascinating research subject. Their beauty lies in the inherent multidisciplinarity, which joins different scientists as microbiologists, electrochemists, and environmental engineers in one venture.

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