



Microbial electrolysis cell platform for simultaneous waste biorefinery and clean electrofuels generation: Current situation, challenges and future perspectives



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ABSTRACT

Microbial electrolysis cell (MEC) holds the flexible potentials for waste biorefinery, pollutants removal, CO₂ capture, and bioelectrosynthesis of clean and renewable electrofuels or valuable chemical commodities, dealing with the depletion of fossil fuels and environmental deterioration issues. Although substantial advances in process design and mechanisms exploration have greatly promoted the development of MEC platform from a concept to a technology, how to virtually utilize it in real-world scenario remains a big challenge. There are numerous technical issues ahead for MEC to be tackled towards up-scaling and real implementations. This review article presents a state-of-the-art overview of the fundamental aspects and the latest breakthrough results and accomplishments obtained from the MEC platform, with a special emphasis on mapping the key extracellular electron transfer (EET) mechanisms between electroactive microorganisms and electrode surface (including *i*: cells ↔ anode; and *ii*: cathode ↔ cells). A unified discussion of different process design: inoculation methods for rapid start-up, role of membranes, modification of cathode materials, cathodic catalysts (i.e. noble, un-noble metal catalysts and biocatalysts) as well as designs and configurations of versatile bioelectrochemical cells, is also involved. Finally, the major challenges and technical problems encountered throughout MEC researches are analyzed, and recommendations and future needs for the virtual utilization of MEC technology in real waste treatment are elaborated.

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1. Introduction

Global energy supply continues to be problematic. The United States Energy Information Administration (EIA) reported that global energy needs will increase by 57% during the 2004–2030 period [1], which aggravates energy shortage and global warming. Today's planet is at a critical stage in its efforts to combat energy crisis and environmental deterioration. A growing awareness and concerns over energy security promote the scientists worldwide to seek new energy supplies. Renewable energy sources represent sustainable and carbon-neutral alternatives to carbon-based fossil fuels to tackle this issue, alleviating the dependence on fossil fuels and ensuring the energy security. As per International Energy Agency (IEA)'s latest report [2], it is predicted that nearly 60% of all power generation capacity by 2040 will come from renewable energy. Bio-energy technologies that use renewable sources to produce biofuels or chemical commodities will play a role in our planet's energy security [3].

Bioelectrochemical systems (BES) are regarded as an emerging and promising strategy for sustainable energy production. According to the direct of electron transfer, BES includes two categories, i.e. microbial fuel cells (MFC) and microbial electrolysis cells (MEC). In an MFC, electrochemically active microorganisms (i.e. exoelectrogens) on the anode oxidize biodegradable organic compounds (mainly acetate) in renewable energy sources such as wastewaters, extracellularly transferring electrons to the anode and producing electrical current [4,5]. In contrast, an MEC, as does photosynthesis [6], utilizes metals or electroactive biofilms (i.e. "electrotrophs") attached to the cathode as catalysts [7] to convert chemical energy in low-grade biomass into hydrogen gas (referred to as "electrohydrogenesis") while remediating contaminants [8], or to electro-synthesize a wide variety of the carbon-natural electrofuels or industrially useful chemicals from the wasted CO₂ (named "electromethanogenesis") [4,9] which captures, recycles and utilizes CO₂ while mitigating the greenhouse effect (GHE) [10]. Slightly different from MFC, the bioelectrocatalysis reaction in such a system does not occur spontaneously, and a small power supply (0.2–0.8 V in practice) needs to be supplied between two electrodes to reduce thermodynamic barrier for the wastes biorefinery and electrofuels bioconversion. Regarding the external power supply, except the commonly used potentiostat or DC power, other alternative renewable energy sources such as solar, wind, geothermal or tide can also be applied in this system, via which both natural energy and bioenergy are stored in the form of transportable fuels simultaneously (Fig. 1). Notably, the MEC system possesses great potential as a green, sustainable and flexible energy producer. These distinct,

unique features of MEC clearly align this process with the principles of environmental sustainability and green chemistry.

The MEC research is a highly new cross-discipline involving environmental engineering, biochemistry, bioelectrochemistry, microbiology, materials science, and molecular biology as well [11]. The advances made in the relevant fields have motivated the great research interests in this concept. During the past decade, MEC as a new platform technology for producing the highly versatile electrofuels and/or valuable chemicals such as hydrogen, methane, formate, acetate, alcohols and hydrogen peroxide (H₂O₂), or for the removal of different organic and inorganic contaminants has gained the ever-growing concerns in the scientific community [10,12–19], inducing the tremendous achievements in both mechanisms exploration and journal publications. To date, there have been several excellent review articles that summarized the current advances in this field, with the specific emphasis related to substrates [20], electrode materials [21], cathodic catalysts [22,23], electroactive biofilm and microbial community [24,25], extracellular electron transfer (EET) mechanisms [7,26,27], reactor architectures, functions and operational performance [3,28–30], as well as commercial viability [31,32]. These reviews provide critical appraisal and in-depth insights, and propose new ideals and recommendations for future directions, making outstanding contribution to MEC development. However, the present efforts hardly keep pace with the encouraging accomplishments, as indicated by the exponential growth in research papers available. Moreover, the majority of previous reviews give grant concerns to H₂ harvest from wastewaters [3,30], with obviously few attentions paid to the versatile capabilities and potentials of MEC platform in CO₂ electromethanogenesis or electro-synthesis for other multi-carbon electrofuels, as well as in seawater desalination, etc. For instance, Albo et al. [10] recently evaluated the economic and environmental benefits of CO₂ electroreduction process, but with particular focus on methanol electrosynthesis. Nowadays, the huge amounts of CO₂ emission (e.g. 35.7 Gt CO₂ in 2014) and resultant global warming further necessitate the comprehensive understanding of the CO₂ electrochemical reduction to diverse valuable products. In addition, the thermodynamic working principles and electrocatalytic routes underlying the different bioelectrochemical processes still remain not clearly recognized, although some efforts have already been devoted [7,13,25,26].

In this review, the working principles and present progress of MEC in the electroproduction of a variety of electrofuels from waste streams are overviewed. The routes and rates of extracellular electron transfer (EET) between electroactive microorganisms and electrode surface are the decisive factors influencing the electrofuels yields and conversion efficiencies. Thus, this article will also

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