



# Electrochemical biotechnologies minimizing the required electrode assemblies

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Microbial electrochemical systems (MESs) are expected to be put into practical use as an environmental technology that can support a future environmentally friendly society. However, conventional MESs present a challenge of inevitably increasing initial investment, mainly due to requirements for a large number of electrode assemblies. In this review, we introduce electrochemical biotechnologies that are under development and can minimize the required electrode assemblies. The novel biotechnologies, called electro-fermentation and indirect electro-stimulation, can drive specific microbial metabolism by electrochemically controlling intercellular and extracellular redox states, respectively. Other technologies, namely electric syntrophy and microbial photo-electrosynthesis, obviate the need for electrode assemblies, instead stimulating targeted reactions by using conductive particles to create new metabolic electron flows.

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## Introduction

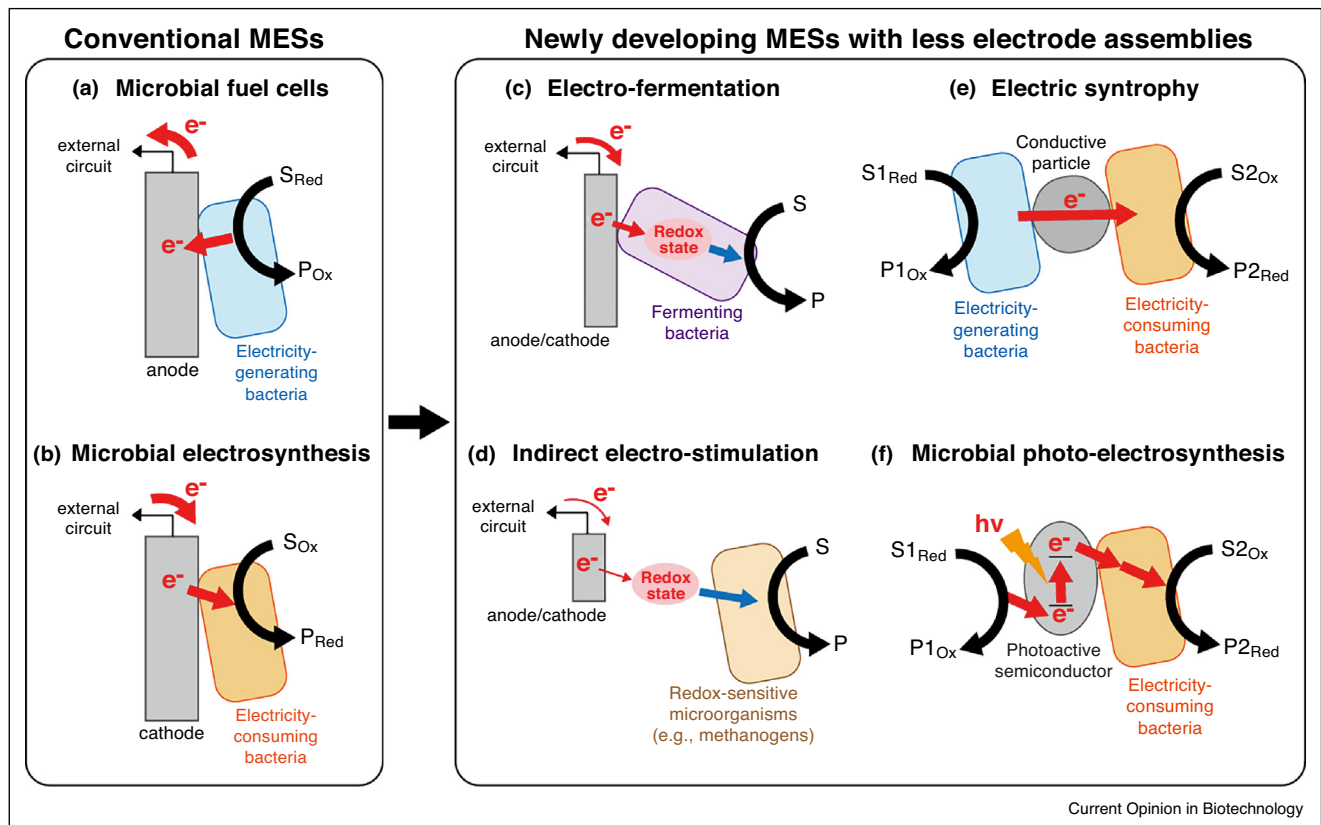
Some of the redox reactions that occur in living cells, such as organic-oxidation/oxygen-reduction in microbial respiration and water-oxidation/carbon dioxide-reduction in photosynthesis, are important from the viewpoint of

global environmental and energy issues. Although catalysts composed of rare elements such as platinum are generally required, microorganisms achieve these reactions at normal temperature and pressure using earth-abundant elements. In developing environmentally friendly energy systems that do not rely on fossil fuels, it is critical to use the energy and material conversion abilities inherent in microbial metabolism. However, microorganisms derived from nature do not necessarily perform the specific metabolic reactions desired by human beings. Hence it is necessary to develop biotechnologies for exploiting or controlling microbial metabolism.

In recent years, biotechnologies combining microbiology and electrochemistry, namely microbial electrochemical systems (MESs), have attracted considerable attentions [1\*,2]. The representative technologies among MESs are microbial fuel cells (MFCs) and microbial electrosynthesis cells (MECs) (Figure 1a,b). In MFCs, the respiratory electrons of microorganisms are transferred to an electrical circuit via an anode, under conditions providing the coexistence of an appropriate cathode reaction such as an oxygen reduction reaction, thereby forming a battery circuit [3,4\*]. MFCs are particularly appealing as a novel technology for energy-saving wastewater treatment systems. On the other hand, in MECs, high-energy electrons are injected into the microorganisms from a cathode, resulting in efficient microbial production of valuable substances [5]. This mechanism can be regarded as a process for the conversion of electrical energy to chemical energy; indeed, recent efforts have permitted the production of high-energy chemicals from carbon dioxide using high-energy electrons derived from an MEC cathode [6,7]. In addition, developments have lately yielded hybrid technologies that couple MECs and anaerobic wastewater treatment systems to stimulate degradation of recalcitrant substances on the electrodes [8–11].

In recent years, on-site and scale-up experiments with MFC/MEC technologies have made large advances, and research on the scaling-up of these technologies has reached a mature state [12\*,13,14]. However, these conventional MES technologies rely on interfacial electron transfer at the electrode surfaces, and therefore inevitably require a large number of electrode assemblies. Currently, constituents of electrode composites such as electrode materials, electrocatalysts, and various functional membranes remain too expensive to be economically

Figure 1



Schematic diagrams of the conventional and next-generation microbial electrochemical systems (MESs) introduced in this review. Microbial fuel cells (a) and microbial electrosynthesis cells (b) are representative of conventional MESs, which require a large numbers of electrode assemblies to exchange metabolic electrons with external circuits. In electro-fermentation (c), microbial metabolism is stimulated by electrochemical modification of the intracellular redox state, a process that can decrease the required number of electrode assemblies. In indirect electro-stimulation (d), there is no electron exchange between microorganisms and electrodes; microbial metabolism is controlled indirectly, via redox states in bulk solution. Electron exchanges with external circuits are no longer necessary in electric syntrophy (e) and microbial photo-electrosynthesis (f); in both of these processes, new electron flows are created by supplementation with (semi)conductive particles. Red and black arrows represent electron and carbon flows, respectively. Blue arrows represent promotive and/or suppressive effects. S, substrates; P, products; Red, reductive forms; Ox, oxidative forms.

feasible. This cost challenge represents a major obstacle, necessitating both scientific and technological breakthroughs for practical application of MFC/MEC technologies.

In this review, we introduce some developing biotechnologies that, which based on a knowledge of microbial electrochemistry, permit the use of smaller numbers of electrode assemblies. The first emerging technology is 'electro-fermentation', in which the desired metabolic pathways are stimulated by electrochemically controlling the microbial intracellular redox state (Figure 1c). The second such technology is 'indirect electro-stimulation', in which targeted microbial activities are promoted or suppressed by controlling the redox state of the bulk solution (Figure 1d). We also present examples of electrochemical biotechnologies that do not require the use of any electrodes, namely electric syntrophy and microbial

electro-photosynthesis, in which new metabolic electron flows are created by supplementation with conductive materials (Figure 1e,f).

### Electro-fermentation: stimulation of microbial metabolism by electrochemical control of intracellular redox states

Microorganisms alter their gene expression patterns and metabolic pathways in response to shifts in the intracellular redox balances. Based on this knowledge, several laboratories have generated biotechnologies that stimulate a specific metabolic pathway by electrochemically controlling the intracellular redox states of microbial cells; these processes are called 'electro-fermentation' [15,16,17]. Compared with the conventional MESs, these technologies require smaller numbers of electrodes, since the quantities of electrons that need to be exchanged with the external circuits are smaller than

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