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Membrane contactors for glucose/O₂ biofuel cell

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1. Introduction

Considering alternative energy supply sources, miniaturized bioelectronic devices as biofuel cells can be attractive bioelectrocatalytic systems [1]. Biological fuel cells utilize microorganisms or enzymes as biocatalysts [2] to convert biochemical energy into electricity. In order to elaborate miniature and implantable biofuel cells, a compartment-separating membrane is excluded [3,4]. Immobilization of enzymes and mediators on the conducting support are thus required. A variety of immobilization methods on different supports have been used for enzyme applications in biotechnology and environment. However, incorporation of enzymes in electropolymerized films such as polypyrrole impregnated with electron-transfer mediators has never been carried out in biofuel cell design. This technique largely developed for a variety of biomolecules provides very convenient and stable biocatalyst interfaces investigated for biosensor development [5]. The biomolecule immobilization on electrode surfaces

can be carried out either by entrapment or by covalent binding to electrochemically polymerized films.

The aim of this work was to elaborate operational membrane contactors based on enzymes entrapment within electrochemically polymerized polypyrrole films (Fig. 1) to develop a glucose/O₂ biofuel cell. Carbon porous tubes were used as original conducting membrane support for enzyme incorporation and for transport of dissolved dioxygen solution via convective flow, through the porosity. Such biofuel cell was based on laccase and glucose oxidase enzymes (Fig. 2). Laccase reduced dissolved dioxygen in presence of mediator such as 2,2'-azinobis (3-ethylbenzothiazoline-6-sulfonate) diammonium salt (ABTS²⁻) at the cathode of the biofuel cell, whereas Glucose Oxidase oxidised glucose in presence of 8-hydroxyquinoline-5-sulfonic acid hydrate (HQS) at the anode.

Carbon porous tubes were used as original conducting membrane support for enzyme incorporation and for transport of dissolved dioxygen solution via convective flow, through the porosity.

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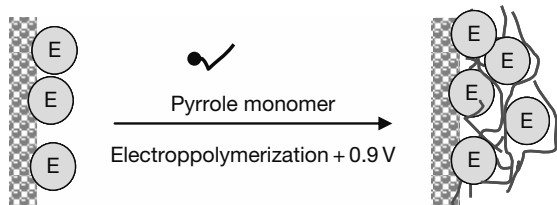


Fig. 1. Elaboration of membrane contactors: enzyme adsorption on carbon tube followed by entrapment in polymer matrix.

2. Results and discussion

Experiments were conducted to determine the activity and the stability of the enzymes immobilized on the membrane contactor. Results revealed that the immobilization method confers high enzyme activity with restrictions of loadings and accessibility of the active sites.

Operational conditions and performances of the membrane contactors have been studied by electrochemistry. The feasibility of each enzyme contactors was demonstrated by chronoamperometry and current–voltage measurements using electrochemical half-cells. For example, as depicted in Fig. 3, dioxygen is reduced on laccase-modified cathode electrode at several hundred millivolts higher than on Pt at pH 3. High densities of current were obtained depending on the pH. We also showed the potentialities of the carbon tube to the diffusion of dissolved oxygen by circulating an oxygen flow inside the inner cavity of the tube.

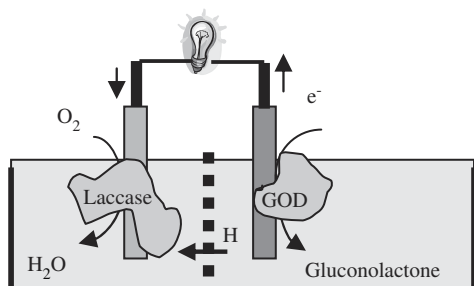


Fig. 2. Schematic O₂/glucose biofuel cell.

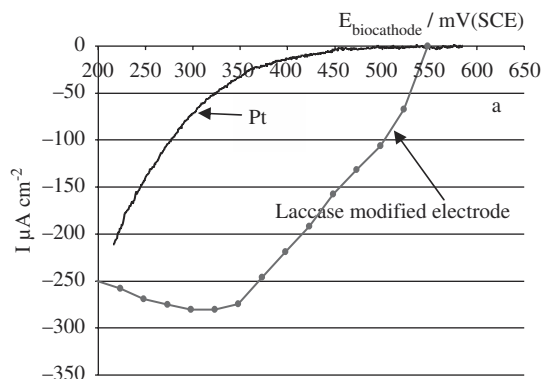


Fig. 3. Polarization curves of biocathodes and platinum electrode in pH 3.0 citrate–phosphate buffer 0.2 M solution under O₂ bubbling.

It was observed that the open-circuit voltage and the current densities increase with the oxygen bubbling time, ensuring efficient oxygen diffusion through the porosity of the tube.

Performances of the glucose/O₂ biofuel cell were demonstrated by current–voltage curves operating at variable external loads and in solutions of different pH.

3. Conclusions

The membrane contactors presented good and stable current densities that established the feasibility of the co-immobilization of both enzyme and its mediator within electropolymerized films and of an operative glucose/O₂ biofuel cell.

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