

Preparation of Enzyme and Mediator co-immobilized  
Electrodes Using a Poly-Ion Complex  
for a Glucose Air Bio-fuel Cell

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1. We have studied a bio-fuel cell by using oxidation of sugars and reduction of oxygen. Co-immobilization of enzyme and mediator on an electrode may give a high current density because of increased concentration of enzyme and mediator on the electrode surface. Such immobilization systems allow separator-less bio-fuel cell, because of substrate specificity of enzyme. In this paper, we will describe a glucose air bio-fuel cell using glassy carbon electrode (GCE) (or carbon paper electrode) co-immobilized with enzyme and mediator by using poly-ion complex.

2. For glucose electrode, glucose oxidase (GOD) and tetrathiafulvalene (TTF, as an electron transfer mediator) (Fig. 1a) were used. On the other hand, bilirubin oxidase (BOD) and 2, 2'-azino-bis (3-ethylbenzo-) thiazoline-6-sulfonic acid (ABTS, as a mediator) (Fig. 1b) were used to reduce oxygen. Cyclic voltammetry was carried out on an electroanalytical system. A GCE ( $\phi = 3$  mm) was used as a working electrode. A platinum plate and an Ag/AgCl (saturated KCl) were used as counter and reference electrodes, respectively. The enzyme and mediator were co-immobilized onto the GCE by casting the poly-ion complex composed from poly-styrene sulfonic acid (PSS) and poly-L-lysine (PLL). Finally, the electrode surface was coated with a phospholipid polymer. The battery performance was measured for a cell composed of two enzyme electrodes (active area is  $1\text{ cm}^2$ ) in an air-saturated 0.1 M phosphate buffer solution with 0.1 M glucose by changing a loaded resistance ( $1-10^5\Omega$ ).

3. Among the poly-ion complexes examined, the best performance and stability were obtained for a PSS/PLL poly-ion complex. The phospholipid polymer membrane made the electrodes bio-compatible. At TTF and GOD co-immobilized on a GCE, glucose oxidation occurred at ca. 0 V with a peak potential of ca. 0.15 V (Fig. 2). On the other hand, ABTS and BOD co-immobilized electrode by using poly-ion complex, oxygen reduction took place at ca. 0.6 V in an  $\text{O}_2$  saturated phosphate buffer solution (pH 7) (Fig. 3). Using these two electrodes, glucose-air fuel cell was constructed. For the bio-fuel cell, carbon paper electrode was used instead of GCE to obtain larger surface area. Open-circuit potential of the cell was ca. 0.5 V, the short circuit current of ca.  $600\ \mu\text{A cm}^{-2}$ , and the maximum power of  $75\ \mu\text{W cm}^{-2}$  was obtained at the cell voltage of ca. 0.25V (Fig. 4).

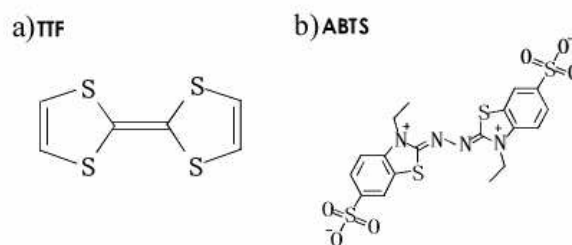


Fig. 1 Structures of TTF and ABTS.

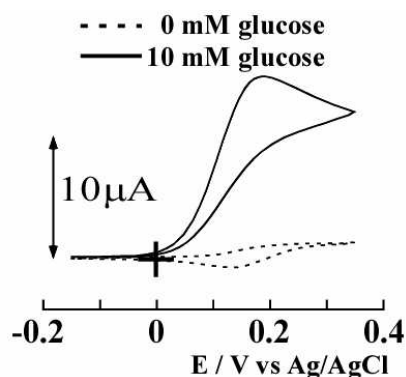


Fig. 2 Cyclic voltammograms of glucose on a TTF/GOD/PSS/PLL/MPC modified GCE in a phosphate buffer solution (pH 7) (5mV/s).

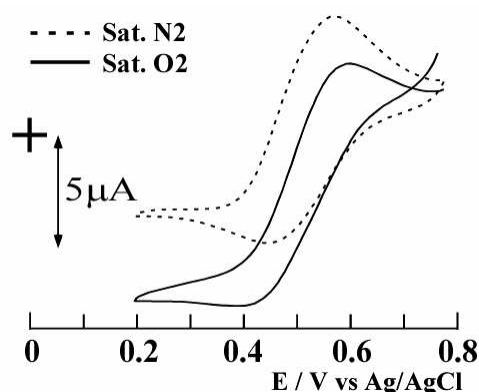


Fig. 3 Cyclic voltammograms of oxygen on an ABTS/BOD/PLL/PSS/MPC modified GCE in a phosphate buffer solution (pH 7) (5mV/s).

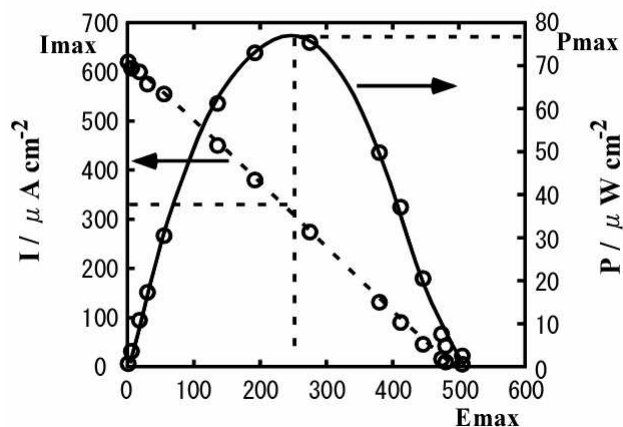


Fig. 4 Power output of glucose air bio-fuel cell