Biocatalysts in fuel cells-towards an implantable power source

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We are interested in mediated enzyme reactions for electrochemical applications such as fuel cells, novel bleaching agents and chemical sensors. Our recent efforts have focused on tyrosinase and laccase enzymes, copper oxidases that catalyze the four-electron reduction of oxygen to water in the presence of chemical mediators. We have successfully demonstrated that the natural substrates of these enzymes (phenols) can be replaced by organic or inorganic one-electron mediators, either in homogeneous solutions or co-immobilized with the enzymes in a redox hydrogel [*Anal. Chem*., 69, 882 and 4108]. These enzymes have been shown to turnover oxygen at relatively high potentials (0.2V) and current densities (200mA/cm^2).

We have recently commenced a project on the development of anodes and cathodes that can be combined in an implantable biofuel cell that yields power. The anode will electro-oxidize glucose in a half-cell such as that developed in the Heller group [J. Electrochem. Soc., 147 (2000) 2780]. This will be coupled to the electro-reduction of molecular oxygen, using the immobilized redox mediated enzyme catalytic system developed in the Leech group, at the cathode. The overall cell reaction is thus the oxidation of glucose, to gluconolactone, and reduction of oxygen to water. Both fuels are continuously available *in vivo* at sufficiently elevated levels to provide the required power output.

The thermodynamic redox potentials of the glucose/gluconolactone and O_2/H_2O couples are such that an output voltage of over 1V is possible from a fuel cell using these components. The kinetics of these complex reactions at bare electrode surfaces (carbon) is however slow. Use of enzymes and mediators can help improve on these slow reactions rates to provide appreciable currents at voltage outpouts above 0.5V. We use glucose oxidase at the anode to catalytically oxidise glucose and laccase at the cathode to catalytically reduce oxygen. The output potential is thus now dependent upon the difference in the redox potentials of these enzymes. Because the active sites of these enzymes, responsible for electron transfer, are buried in the insulating protein shell, direct electron transfer between these active sites and an electrode is difficult and slow (if possible at all). An electron transfer catalyst (mediator) consisting of a redox polymer can improve electron transfer kinetics. We use redox polymers based on osmium and ruthenium immobilized with the enzyme using a crosslinker. The redox potential of the polymer now determines the cell output and will be tuned to ensure maximum current density, at voltage outputs above 0.5V. Tuning of the redox polymer potential is accomplished by varying the substituents on the ligands and/or the ligands themselves.

Preliminary results to be presented show that we can catalytically oxidize glucose and reduce oxygen using co-immobilized enzymes and redox polymer. These results demonstrate that an output can be achieved close to neutral (physiological) pH and in saline solution (a problem with initial studies on alternate biofuel cells by other groups).