Biocomputing Systems Based on Electrically Contacted Enzyme Assemblies

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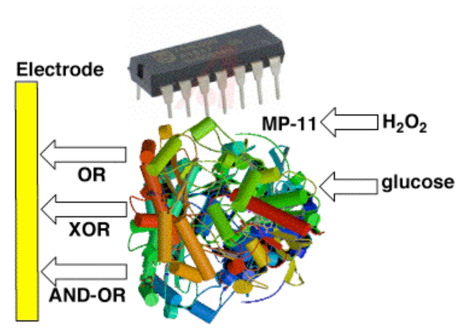
Biochemical computing is an exciting new field, which shows great promise, but at the same time faces substantial challenges. Recently, research efforts were directed at unconventional chemical computing based on a chemical "soup" where data are represented by varying concentrations of chemicals [1]. The computations are performed by naturally occurring chemical reactions that can proceed in a solution or at an interface functionalized with catalytic sites. The computing problems can be solved at the level of a single molecule, resulting in dramatic miniaturization and allowing parallel computation performed by numerous molecules [2]. So far, the field is still in a very early experimental stage, but may have great future potential [3]. Biochemical computing (biocomputing) is a special kind of chemical computing based on enzyme-catalyzed reactions and DNA recognition/reaction processes. Recently designed logic gates based on enzyme-catalyzed reactions allowed logic operations and simple computing in solutions with the optical read-out of the output signals [4-7].

The present paper reports on the logic gates based on electrically wired enzymes associated with electrode surfaces [8]. Logic gates "AND", "OR", "XOR" were developed using bioelectrocatalyzed reactions of redox-enzymes and their assemblies with electron transfer mediators and co-factors, Scheme 1. The electrochemical read-out of the output signals from the immobilized enzyme-assemblies was achieved, thus converting the biochemical logic gates into real bioelectronic systems. Future application of multi-electrode arrays functionalized with various enzyme-based logic gates is envisaged. This will finally result in bioelectronic computing chips that will interface biological and electronic systems.

Another example of the enzyme-based logic gates includes magnetic nanoparticles functionalized with biocatalytic units [9]. Magneto-controlled OR, AND and INHIB logic gates were designed using cobalt ferrite magnetic nanoparticles ($CoFe_2O_4$, saturated magnetization ca. 70 emu g^{-1}, 17±2 nm diameter) functionalized with microperoxidase-11. Tunable magnetic field generated by three external permanent magnets (NdFeB) upon moving them below the electrochemical cell resulted in translocation of the biofunctionalized magnetic nanoparticles between conductive and non-conductive domains of a solid plate. This resulted in electrochemically readable output signals with the Boolean logic controlled by the magnetic input signals. The current corresponding to the reversible redox process of the heme measured at -0.4 V (vs. SCE) was considered as "1" output signal, while a small background current obtained from the conducting interface in the absence of the magnetic nanoparticles was considered as "0" output signal. Addition of H_2O_2 to the solution resulted in the generation of a cathodic catalytic current when the microperoxidase-11-functionalized magnetic nanoparticles are associated with the conductive domain of the support. This resulted in the amplification of "1" output signal and the increased difference between "1" and "0" signals generated by the cell, thus reducing the possibility of errors in the Boolean logic operations.

The enzyme-based system can be adapted to various chemical signals and their combinations to perform logic operations of different kind and complexity by integrating different enzymes with the electrochemical interface. Integration of several enzymes with the electrode support will allow scaling up the biocomputing network where the connections between the enzymes will be maintained through the exchange of substrates/products of the biochemical reactions, and the final output signal will be transduced into the electrical current.



Scheme 1. The electrode functionalized with glucose oxidase (GOx) and microperoxidase-11 (MP-11) performs various Boolean logic operations (OR, XOR, AND-OR) upon addition of glucose and/or H_2O_2 and application of different potentials. The output signal coming from the electrically wired enzymes is electrochemically readable allowing interfacing of biochemical computing systems with ordinary electronics

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