Arkadeva Irina Nikolaevna

Mathematical modeling and optimization of processes occurring in a biofuel cell

Abstract

Relevance of the research. In accordance with Decree of the President of the Russian Federation dated July 7, 2011 No. 899, technologies of new and renewable energy sources are included in the list of critical technologies of the Russian Federation. One of the promising areas of modern alternative energy is biofuel cells (BFC) - devices for converting the chemical energy of a fuel into electrical energy using biological catalysts (enzymes, microbes, etc.). BFCs are alternative devices to traditional fuel cells (FCs) with catalysts based on precious metals, since they have a number of advantages compared to them: they use renewable catalysts and operate in mild conditions - a temperature range of 20-40°C and a pH close to neutral, when using glucose as a fuel, while the most common low-temperature hydrogen-air fuel cells with a solid proton-conducting polymer electrolyte and platinum-based catalysts operate in the range of pH ~ 1-3, temperature 60-80°C. The main field of application of BFCs is the possibility of their use as a power source, including for implantable devices (for example, a pacemaker, an artificial kidney, etc.). In addition, BFC-based biosensors are being developed for a wide range of substrates for medical and environmental purposes.

To promote a deeper understanding of the basic laws of the functioning of such systems and expand the potential for their practical application, it is necessary to formulate a mathematical apparatus capable of predicting the parameters that determine the output characteristics of the developed BFC.

One of the most promising designs for BFC is the one without a membrane and without any mediator, in which the participants and products of electrode reactions are renewable and environmentally friendly. Glucose and oxygen were chosen as substrates because they are the most accessible, widespread, and environmentally friendly substances. Laccase, an active and stable enzyme that accelerates the electroreduction of oxygen to water, was chosen as a catalyst for the cathode. Gold-modified carbon black was used as an anode catalyst, on which glucose is oxidized under conditions favorable for the operation of a laccase-based cathode.

The purpose of the work – to develop and investigate materials for electrodes of glucose-oxygen BFC, to obtain experimental data characterizing the operation of BFC, and to form on their basis a mathematical apparatus that describes this system and allows identification of the main patterns that ensure its efficiency.

Tasks:

1) The choice of catalytic systems for current-forming reactions in the composition of glucose-oxygen BFC based on experimental studies under model conditions. Establishment of the main electrochemical characteristics that determine the efficiency of the functioning of the reaction of O_2 reduction and glucose oxidation.

2) Development of mathematical models of the processes occurring on the selected catalytic systems, determination of the parameters that affect the electrode processes, including laccase immobilization.

3) Study of the processes occurring in the BFC model without a separating membrane using selected catalytic systems.

4) Development of a mathematical apparatus that describes the processes occurring in the BFC, establishing the basic laws of the functioning of the BFC and determining the parameters that affect its efficiency.

Scientific novelty. Effective catalytic systems and a glucose-oxygen biofuel cell without a separating membrane and without mediators based on them have been developed.

For the first time, based on the mathematical apparatus of fractional differentiation, the mathematical models have been developed: 1) immobilization of the enzyme, taking into account the porous structure of the carbon support; 2) direct bioelectro-catalytic reduction of oxygen by laccase; 3) electrooxidation of glucose, taking into account changes in the number of active centers of the catalyst, which made it possible to establish the basic patterns of the occurrence of physicochemical processes in the studied systems, to optimize the amount of carbon material on the electrodes and glucose in the feed solution.

Theoretical and practical significance. In this work, experimental and theoretical studies of physicochemical processes and phenomena occurring in BFCs without a separating membrane and without mediators with catalysts based on laccase (cathode) and gold-modified soot (anode) were carried out. The main regularities of the processes were

determined: spontaneous adsorption immobilization of laccase on carbon materials (CM) with different structures (carbon nanotubes (CNT) and soot); electroreduction of oxygen under conditions of direct electron transfer (without a mediator) on an electrode with a laccase-based catalyst; electrooxidation of glucose on an electrode with a gold-modified soot catalyst at a pH close to neutral.

Were developed: 1) A mathematical model of spontaneous adsorption immobilization of laccase on CMs of different nature. Based on the data obtained, the structural parameters of CMs that affect the efficiency of adsorption were determined. 2) Mathematical models of the processes of oxygen electroreduction at the cathode based on laccase and glucose electrooxidation at the anode with the gold-modified soot catalyst, capable of predicting the electrochemical characteristics of the studied electrode processes. 3) A mathematical model of BFC, capable of predicting the electrochemical characteristics of the system under study.

The mathematical apparatus formulated on the basis of extensive experimental studies made it possible to create a theoretical basis for the subsequent expansion of the possibilities of practical application of BFC without a membrane and without mediators.

The main provisions for defense:

1. Results of experimental studies of cathode and anode catalytic systems based on adsorption-immobilized laccase on various CMs and the gold-modified soot catalyst, respectively.

2. Mathematical models: the process of adsorption of laccase on CM; electroreduction of O_2 by laccase; electrooxidation of glucose on an electrode with gold-modified soot; processes of functioning of BFC without a membrane and without any mediator using the developed catalysts.

3. Results of calculations using the indicated mathematical models and their comparison with experimental data.

4. Results of experimental studies of the BFC model without a membrane and without any mediator using the created catalysts. Investigation of the influence of the composition of the active layers of electrodes, the concentration of substrates and the conditions for testing on their activity when using glucose as a fuel and an oxidizing agent - oxygen.