Abstract

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«Application, structure and functional properties of coatings based on titanium compounds for electrodes of electrochemical devices with a proton exchange membrane»

Relevance of the research topic

The development of modern energy systems integrating renewable energy sources requires the creation of highly efficient, durable, and cost-effective technologies for energy conversion and storage. One of the most promising devices in this field are reversible fuel cells with a proton exchange membrane, capable of operating in fuel cell mode for energy generation and in water electrolyzer mode for hydrogen production. However, their large-scale commercial implementation is hindered by limited operational lifetime, the key reason for which is the degradation of inorganic components, primarily the carbon gas diffusion layers of the oxygen electrode, under conditions of high oxygen concentration, high potentials (1.5 − 2 V), and temperature (~50 − 90°C).

Increasing electrode durability without a significant rise in cost is a critically important task for ensuring the competitiveness of this technology. Traditional solutions based on protective coatings of noble metals (Pt, Ir) are characterized by high efficiency but are unacceptable from an economic point of view for mass production due to their high cost. In this regard, a relevant direction is the development of technology for applying protective coatings based on transition metal compounds (such as oxides, nitrides, carbides), which possess high chemical stability in aggressive environments and significantly lower cost.

Magnetron sputtering appears as a promising method for obtaining such coatings. This technology is based on the use of low-temperature plasma and provides precise control over the formation of the structure, morphology, and composition of the deposited layers, allowing materials with varying density and porosity, controlled stoichiometry, and optimal electrical conductivity to be obtained. Using the magnetron sputtering method for coating formation enables the creation of an optimal design for carbon electrodes with enhanced operational characteristics. The application of modified electrodes will increase the service life of electrochemical devices and reduce their production costs by replacing expensive coatings based on precious metals.

Thus, the aim of this work is to develop a technology for forming protective coatings based on titanium compounds and their multicomponent analogs by magnetron sputtering to protect electrodes in reversible fuel cells. Achieving this goal includes studying the influence of magnetron sputtering modes and deposition parameters on the structure, phase composition of the applied coatings, and their functional characteristics within reversible fuel cells. The realization of this task will create scientific and technical foundations for producing new efficient components of electrochemical devices with improved resource performance.

Degree of topic development

Despite a significant number of works devoted to the development and application of magnetron sputtering for obtaining various types of protective coatings, the literature has practically not studied the influence of magnetron sputtering mode parameters on the structure, phase composition, and functional properties of the obtained coatings, nor their application for protecting carbon materials in a reversible fuel cell.

Object of research

Development of a deposition technology and composition for an inorganic protective coating based on titanium compounds with specified technical parameters (structure, morphology, and composition) aimed at increasing the durability of carbon materials in particular, and extending the operational lifetime of electrochemical devices in general.

Research tasks

To achieve the set objectives, the following tasks were formulated:

1. Development of a technology for coating deposition by magnetron sputtering, searching for the best operating mode and its parameters for sputtering.

2. Deposition of coatings based on titanium compounds onto carbon material with specified structure, morphology, and composition.

3. Investigation of the structure, morphology, composition, physicochemical, and electrochemical properties of coatings based on titanium compounds deposited onto the surface of carbon material.

4. Testing the carbon material with applied coatings in a reversible fuel cell to find the most suitable titanium compound that provides an increase in the device's service life.

5. Modification of the architecture and composition of the oxygen electrode of a reversible fuel cell using a protective coating of a known composition as a sublayer to increase overall efficiency.

Scientific novelty

1. A protective coating of TiC_xN_y composition for protecting carbon material was obtained for the first time by magnetron sputtering. The deposition method is based on the application of a direct current mode with an operating power of 90 W and a negative bias voltage of -200 V applied to the substrate. Applying a bias potential of -200 V enhances the adhesion of the deposited particles and promotes the formation of a multicomponent titanium carbonitride compound. The 90 W power ensures a uniform particle flux and coating homogeneity.

2. The effect of a TiC_xN_y coating on the durability of electrodes based on carbon materials was investigated for the first time under accelerated stress testing conditions in a reversible fuel cell. An increase in the degassing resistance of carbon electrodes was demonstrated due to the shift of oxidative reactions to the protective sublayer.

3. A multilayer architecture of the oxygen electrode for a reversible fuel cell was created using a sublayer based on a TiC_xN_y coating, which allows increasing the durability and efficiency of the reversible fuel cell. Separating the catalytic layers by means of the sublayer makes it possible to increase their individual efficiency and protect them from mutual degradation.

Theoretical and practical significance

1. The parameters of magnetron sputtering that allow creating highly effective wear-resistant protective coatings for electrodes based on carbon material have been determined. Such electrodes can be used in many electrochemical devices, including a reversible fuel cell.

2. It has been shown that the application of a TiC_xN_y protective coating as a sublayer within an oxygen electrode can increase the number of operation cycles when switching a reversible fuel cell between fuel cell and water electrolyzer modes. The research results can be used in the development of highly efficient electrodes for various types of electrochemical devices.

Research methodology and methods

The methodological basis of the work consists of the analysis of modern scientific publications, as well as standard methods of laboratory experiment and processing of its results. The information base for the research included literary sources from leading domestic and foreign specialists in the field under consideration. The studies were conducted using physicochemical and electrochemical methods of analysis. The following methods were used in the course of the work: magnetron sputtering, X-ray phase analysis, elemental analysis, scanning electron microscopy, impedance spectroscopy, voltammetry, accelerated stress testing methods, cyclic voltammogram methods, as well as calculation methods based on equations for electrochemical systems.

Provisions for the defense:

1. Method for applying protective coatings using magnetron sputtering with a specified structure and phase composition.

2. Results of physicochemical studies of the obtained coatings based on titanium compounds of various compositions, determination of the best coating for protecting carbon-based components.

3. Results of investigating the durability of carbon components coated with a protective coating of TiC_xN_y composition in a reversible fuel cell, including conducting electrochemical and resource tests.

Degree of result reliability

The reliability of the obtained results is ensured using modern physicochemical and electrochemical methods, high-precision equipment, statistical data processing, and the consistency of experimental data with literary sources.

Work approbation

Based on the dissertation materials, reports were prepared for the following conferences: XV and XVII International A.A. Kurdyumov School for Young Scientists and Specialists; II International Scientific and Practical Conference "Rare Metals and Materials Based on Them: Technologies, Properties and Application"; XXIX, XXX and XXXI International Scientific and Technical Conference of Students and Postgraduates "RADIO ELECTRONICS, ELECTRICAL ENGINEERING AND ENERGY"; XVII Kurchatov Youth Scientific School; X All-Russian Conference with International Participation "Fuel Cells and Power Plants Based on Them".

Publications

Based on the dissertation materials, 12 works have been published, including 5 publications in peer-reviewed scientific journals included in international citation databases, and 1 patent for an invention has been obtained.

Dissertation scope and structure

The dissertation consists of an introduction, 5 chapters, a conclusion, a list of publications on the dissertation topic, a list of abbreviations and conventional designations, and a list of references. The work is presented on 120 pages of typewritten text, contains 9 tables and 36 figures. The list of references includes 169 sources.