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SIMULATION OF THE PROCESS OF IMPROVING THE OXYGEN-HYDROGEN FUEL CELL

МОДЕЛЮВАННЯ ПРОЦЕСУ ВДОСКОНАЛЕННЯ КИСНЕВО-ВОДНЕВОГО ПАЛИВНОГО ЕЛЕМЕНТА

Modeling the process of improving the oxygen-hydrogen fuel cell is a relevant task, since such modeling in line with the theory of inventive problem solving allows one to better see the ways of searching for strong inventive solutions. In addition, such modeling helps the researcher and engineer to overcome psychological inertia, to increase the efficiency of scientific research and design and technological developments. Instead of intuitive search and enumeration of many options, such modeling of the inventive process allows to significantly reduce its duration.

The purpose of this study is to model the process of improving the oxygen-hydrogen fuel cell. The second goal of the research is, with the help of modeling, solving the problem of improving the technical characteristics of the fuel cell and reducing its cost.

The paper presents a retrospective analysis of the process of modernization of oxygen-hydrogen fuel cells from the standpoint of the theory of inventive problem solving, and proposes qualitative models of the technical system under study, as well as the process of its improvement. The obtained qualitative mathematical models are presented in the form of traditional diagrams in accordance with the theory of inventive problem solving. Such methods as «vepol» («matter-field») analysis, search and overcoming of technical contradiction are applied.

Based on high-quality models, specific solutions have been developed to improve the technical characteristics of the oxygen-hydrogen fuel cell. In particular, by using gas flow dividers and replacing expensive platinum catalysts with cheaper catalysts made of nickel nanopowder in a corrosion-resistant shell, it was possible to reduce the cost of the fuel cell while slightly increasing its specific power. This was confirmed by preliminary tests of the fuel cell model. In the future, the authors plan to conduct more detailed modeling of the problem of reduced service life of fuel cells due to catalyst deactivation.

***Keywords**: inventive process, fuel cell, catalysts, technical contradiction.*

Моделювання процесу вдосконалення киснево-водневого паливного елемента є актуальним завданням, оскільки таке моделювання в руслі теорії вирішення винахідницьких задач до-

зволяє краще бачити шляхи пошуку сильних винахідницьких рішень. Крім того, таке моделювання допомагає досліднику та інженеру подолати психологічну інерцію, підвищити ефективність наукових досліджень і конструкторсько-технологічних розробок. Замість інтуїтивного пошуку і перерахування багатьох варіантів таке моделювання винахідницького процесу дозволяє суттєво скоротити його тривалість.

Метою даного дослідження є моделювання процесу вдосконалення киснево-водневого паливного елемента. Другою метою дослідження є вирішення проблеми покращення технічних характеристик паливного елемента та зниження його вартості за допомогою моделювання.

У роботі проведено ретроспективний аналіз процесу модернізації киснево-водневих паливних елементів з позиції теорії вирішення винахідницьких задач, запропоновано якісні моделі технічної системи, що досліджується, а також процесу її вдосконалення. Отримані якісні математичні моделі представлені у вигляді традиційних діаграм, відповідно до теорії вирішення винахідницьких задач. Застосовано такі методи, як «вепольний» («речовина-поле») аналіз, пошук і подолання технічної суперечності.

На основі якісних моделей розроблені конкретні рішення для покращення технічних характеристик киснево-водневого паливного елемента. Зокрема,використання правила добудови «веполей» дозволило глибше осмислити заміну рідкого електроліту на протонообмінну мембрану (твердополімерний електроліт). А використання правил добудови і розвитку «веполей» дозволило розробити розсікачі потоків водню і кисню, та запропонувати заміну платинових каталізаторів на каталізатори з нанопорошку нікелю в корозійностійкій оболонці (із вуглецю).

За допомогою розсікачів газових потоків та заміни дорогих платинових каталізаторів на більш дешеві каталізатори з нанопорошку нікелю в корозійностійкій оболонці вдалося здешевити паливний елемент,а також дещо підвищивши його питому потужність. Це підтвердили попередні випробування макету паливного елементу. Надалі автори планують провести більш детальне моделювання проблеми зниження терміну служби паливних елементів через дезактивацію каталізатора.

***Ключові слова**: винахідницький процес, паливний елемент, каталізатори, технічна суперечність.*

Problem’s Formulation

Fuel cells have been under development for almost two centuries, but there are still many opportunities to improve this promising chemical current source. Fuel cells are used in spaceships, airplanes, water transport, ground vehicles, forklifts, and uninterruptible power supply systems. A fuel cell is an electrochemical converter that directly converts chemical energy into electrical energy. However, unlike batteries, fuel cells function as long as the fuel and oxidant come from an external source, while the chemical composition of the electrolyte does not change during operation [1, 2]. Unfortunately, catalysts based on platinum, palladium, and gold, which are used in fuel cells, are expensive and do not work efficiently enough [3]. The process of developing and improving fuel cells is very complex and requires consideration of many factors, in particular, the cost of catalysts, corrosion resistance of materials. Such studies require a lot of time and intellectual effort. But inventive activity can be intensified by modeling according to the theory of inventive problem solving, which was created by Heinrich Altshuler [4]. In the last decades, the theory of solving inventive problems began to be applied by some international companies, for example, the South Korean group of companies «Samsung», which is known for the production of complex electronic products and systems, and which actively uses this theory during the creation of innovations [5]. This means that the formal techniques and methods of the theory, which can be analyzed as qualitative mathematical models, are successfully used to create complex intelligent systems, including control systems, electrochemical systems, and others. Therefore, modeling the process of developing and improving fuel cells in order to increase the efficiency of this process and improve the technical characteristics of devices and reduce the price of products is an urgent scientific and practical task.

Analysis of recent research and publications

Most sources claim that the fuel cell was first proposed in 1839 by the English amateur Sir William Grove (W.R. Grove), who observed the electrolysis of water in sulfuric acid solutions on platinum electrodes and discovered that after saturating the electrodes with oxygen and hydrogen and disconnection of the external current, a direct current is generated in the electrolytic cell [1, 2]. However, it was not possible to implement this idea of a fuel cell. The first samples of working fuel cells appeared only in the 1930s, but they had low current density and low durability. In 1921, German-Swiss researcher Emil Baur built the first fuel cell with molten carbonate. The first fundamental scientific monograph devoted to fuel cells was published in 1947 by the Soviet researcher O.K. Davtyan. The first large installation based on oxygen-hydrogen fuel cells with a capacity of 5 kW was created by the Englishman F. Bacon in 1958. In the 1960s of the 20th century, during the time of active space exploration, a boom around fuel cells began. Oxygen-hydrogen fuel cells with a capacity of up to 1 kW were created for the US «Gemini» and «Apollo» space programs. Later, in the 1980s, PEs of 10 kW were created for the American «Shuttle» and the Soviet «Buran». Most often, platinum catalysts were used in oxygen-hydrogen fuel cells, but already in the middle of the 20th century fuel cells with non-platinum catalysts (skeletal nickel and silver) appeared in Germany. In the 1960s 20th century 100 kW power plants were also built on oxygen-hydrogen fuel cells that use phosphoric acid as an electrolyte (phosphoric acid fuel cells). The «Apollo» spacecraft had oxy-hydrogen fuel cells that used the alkaline electrolyte KOH. Liquid electrolyte (phosphoric acid or alkali) in fuel cells is usually tried to be enclosed in some kind of porous matrix. However, there are products with free liquid electrolyte. Fuel cells with a liquid electrolyte have a rather complex design [1, 2].

Today, the most advanced technologies are oxygen-hydrogen fuel cells with a solid polymer electrolyte. Most often, a proton exchange membrane of the Nafion type, manufactured by the well-known «DuPont» company, is used as such an electrolyte. These fuel cells have the following advantages: high specific power (per unit mass and volume); compactness; simplicity of design; absence of liquid electrolyte (ie prevention of carbonation, prevention of leakage); separation of water as a reaction product is facilitated. (For the first time, a solid polymer electrolyte with an ion exchange membrane was used in 1965 on the American spaceship «Gemini», but the characteristics of that membrane were not yet sufficiently satisfactory.) The proton exchange membrane must have high electrical conductivity due to the movement of hydrogen ions (protons), as well as chemical stability under the conditions of operation of fuel cells. Attempts to further improve the characteristics of the proton exchange membrane, which are carried out in the best laboratories of countries such as the USA, Germany, Japan, and Canada, do not stop. The fact is that aliphatic perfluorinated sulfonic acid ionomers of the Nafion type work only with some moisture (which is almost always realized in fuel cells), which limits the temperature range of their performance to 80—90 °C. However, increasing the efficiency of fuel cells with a solid polymer electrolyte is associated with an increase in the operating temperature range to 100—200 °C [1, 2, 6].

The first fuel cells, both with liquid electrolyte and solid polymer, were characterized by a high content of platinum — up to 200—300 g/m^2 (0.02—0.03 g/cm^2). Until the beginning of the 80s 20th century it was possible to significantly reduce the content of platinum — to 1—10 g/m^2 (0.1—1.0 mg/cm^2). In modern oxygen-hydrogen fuel cells, the platinum content is approximately 0.2—0.5 mg/cm^2, so they are still expensive. Researchers have tried other metals that are not as expensive as platinum, for example: Ni, Co, Cr, but they are not chemically stable enough, especially on the oxygen electrode [1, 2, 7, 8].

Therefore, attempts are also being made today to increase the efficiency of using platinum catalysts by creating nanocomposites and various alloys [1, 7, 8]. For example, work [7] describes a method of preparing highly active stable catalysts containing ultra-low platinum content, using cobalt or bimetallic cobalt and zinc zeolitic imidazolate frameworks as precursors. The interaction between platinum-cobalt nanoparticles and platinum group metals free sites improves the activity and durability of the catalysts.

Since the 1990s, the development of fuel cells with a capacity of 1 kW to 10 MW for stationary autonomous power generation is still ongoing. Today, portable power sources of less than 100 W are also being developed for computers, mobile phones, cameras, etc. Methanol (alcohol) is used as a fuel in them, from which hydrogen is obtained for fuel cells [1].

In fuel cells with a liquid acidic electrolyte, as well as with a solid polymer electrolyte based on a proton exchange membrane, hydrogen is supplied by a highly porous anode and enters the electrolyte through micropores in the anode material. At the same time, hydrogen molecules are decomposed into atoms, which, as a result of chemisorption, giving up one electron each, turn into positively charged ions — protons. Oxygen entering the cathode passes into the electrolyte and also reacts on the surface of the electrode with the participation of the catalyst. When it interacts with hydrogen ions (protons) and electrons coming from the external chain, water is formed. Similar chemical reactions occur in fuel cells with an alkaline electrolyte [1].

A schematic representation of the construction of oxygen-hydrogen fuel cells with a solid polymer electrolyte is shown in Fig. 1.

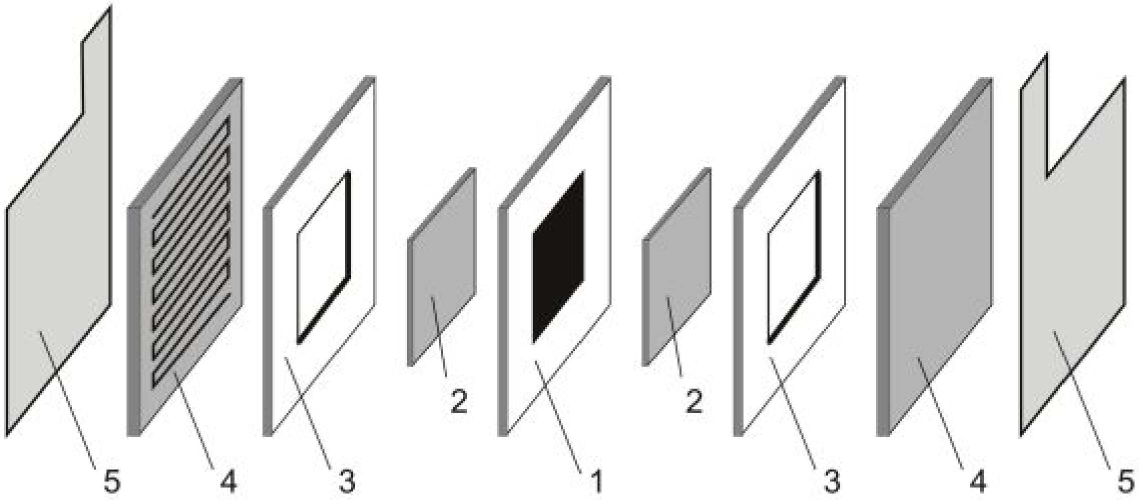

Fig. 1. Schematic representation of the construction of a fuel cell with a solid polymer electrolyte, according to [9], with changes

In fig. 1, the number «1» denotes a membrane with layers of catalysts, the number «2» denotes porous gas diffusion layers (they provide supply and removal of gases, and also work as electric current collectors), the number «3» — Teflon gaskets, the number «4» denotes graphite electrodes (anode and cathode), metal current collectors marked with the number «5».

Research and development of oxygen-hydrogen fuel cells is also underway in Ukraine. For example, the patent [10] describes a proton exchange membrane for a fuel cell, which can potentially compete with the well-known Nafion membrane. The patent [11] describes the operation of a power plant for the production of electricity based on oxygen-hydrogen fuel cells.

A recent analysis of global patent developments and scientific publications in the field of fuel cells indicates that fuel cells have attracted considerable attention from researchers due to their environmentally friendly properties, and that they are seen as one of the most promising renewable technologies for the future hydrogen economy. However, the high cost, low durability and low productivity of fuel cells limit the potential of their use in the economy. An effective energy transition of society is highly dependent on the support of politicians and requires joint efforts of all stakeholders, including the promotion of awareness and common knowledge to engage all stakeholders in more conscious behavior for better support and adoption of innovations. The greatest efforts of fuel cell developers in the period from 1973 to 2022 were aimed at improving the structure of fuel cells, the technology of creating electrodes and modernizing the electricity production system. In particular, the fuel cell cathode still needs to improve its corrosion resistance (if made of metal) and mechanical strength (if made of carbon), and the power generation system needs to address the issue of efficient and safe hydrogen storage. The main drawback of oxygen-hydrogen fuel cells has not been overcome — expensive catalysts that use platinum and palladium [12, 13].

According to the authors of the analytical study [14], there is constructive and useful competition between two promising technologies that will provide an alternative to fossil fuels — lithium batteries and fuel cells. Both the first and the second are considered as effective accumulators of electrical energy from renewable energy installations. But hydrogen-oxygen fuel cells have an energy-to-weight ratio about ten times higher than lithium-ion batteries, giving them an advantage for electric transportation applications, particularly electric vehicles. That is, electric cars and pure hydrogen electric transport can take the leading position [14].

Formulation of the study purpose

The purpose of this study is to model the process of improving the oxygen-hydrogen fuel cell, according to the theory of inventive problem solving, and also, with the help of modeling, to solve the problem of improving the technical characteristics and reducing the cost of the fuel cell. The study is a continuation of work [3].

Presenting main material

The functional scheme of the oxygen-hydrogen fuel cell, which takes into account its design, can be depicted as follows (Fig. 2). In Fig. 2, we abstract from the design details, but only illustrate the purpose of the main parts. The structure of the fuel cell contains: electrodes (anode and cathode); catalysts; electrolyte; devices for supplying gases (hydrogen, oxygen (or air)); water drainage device.

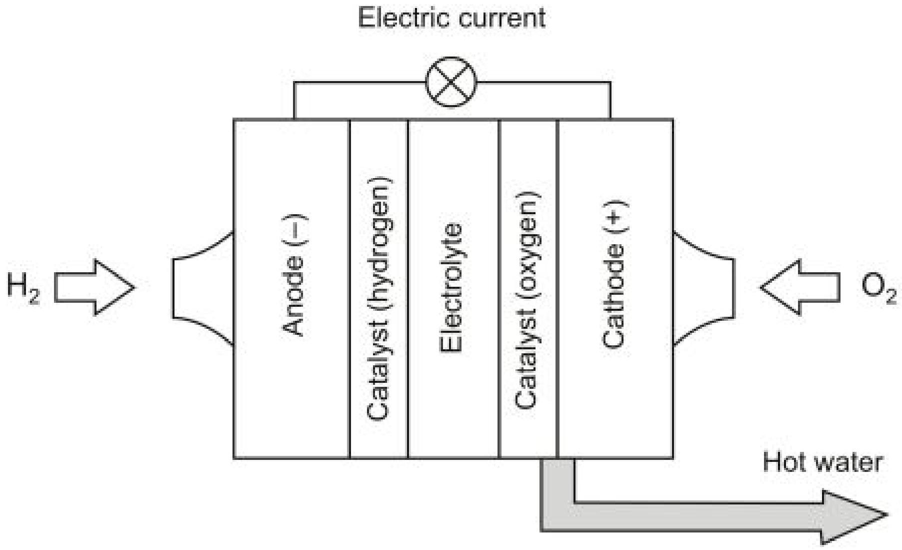

Fig. 2. Functional scheme of the fuel cell, according to [1], with additions

If the design and functional scheme of the oxygen-hydrogen fuel cell and the main flows of substances/energy are translated into the language of «vepol», according to the theory of inventive problem solving (G. Altshuler) [4], then the qualitative model of the fuel cell looks like this (Fig. 3). Let us recall that «vepol» in this theory denotes the simplest technical system and is an abbreviated translation of the pair of words «matter-field».

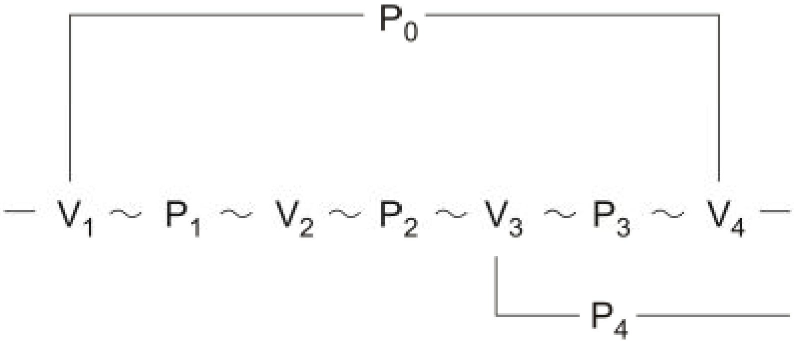

Fig. 3. Qualitative model of the fuel cell, according to «vepol» analysis

In Fig. 3 the «matters» V_1 and V_4 denote the anode and cathode, respectively. «Field» P_0 denotes electric current, «field» P_4 denotes hot water flow. The straight lines (segments) near V_1 and V_4 indicate the incoming flows of hydrogen and oxygen, respectively. And the straight lines (segments) near P_0 and P_4 indicate sufficiently satisfactory characteristics of the «fields» P_0 and P_4 and their connections with the corresponding elements of the model. Wavy segments in Fig. 3 indicate unsatisfactory interactions between the corresponding elements of the model. In particular, the flow of hydrogen P_1 after the anode V_1 has unsatisfactory characteristics — it is uneven, it also interacts poorly with the catalyst V_2. A similar situation with the flow of oxygen P_3 after the cathode V_4 and its interaction with the catalyst V_3. The flow of ions through the electrolyte is depicted as the «field» P_2. In a fuel cell with a liquid electrolyte, the flow of ions does not satisfactorily interact with the catalysts due to the complex design of the fuel cell. That is, the interaction between V_2 and P_2, as well as the interaction between P_2 and V_3 is not satisfactory enough.

We use the rule of addition of «vepols», as indicated in Fig. 4. In this case, the theory of inventive problem solving recommends adding «matter» V_5, which will ensure, firstly, a satisfactory interaction between V_2 and P_2, and secondly, a satisfactory interaction between P_2 and V_3. The developers of fuel cell with a solid polymer electrolyte did just that. A thin Nafion-type proton exchange membrane acts as a V_5 «matter» on which catalyst layers (V_2 and V_3) can be easily applied on both sides, and the membrane provides a satisfactory flow (P_2) of protons (ions) through it. It is clear that a satisfactory proton exchange membrane was not created immediately. As indicated above, the first ion exchange membrane was imperfect. Only then was it possible to create a Nafion membrane. In order to solve the problem of unsatisfactory interaction of hydrogen and oxygen flows (P_1 and P_3) with catalysts (V_2 and V_3), the authors of this article used methods of completion and development of «vepols». Completing «vepols» means introducing one more «matter» for each of them — into the «matter-field» pair (Fig. 5). The additional «matter» must improve the interaction of the «field» with the first «matter». That is, the flows of hydrogen P_1 and oxygen P_3 must begin to interact effectively with catalysts V_2 and V_3, respectively. Thus, the authors of this article proposed the introduction of hydrogen and oxygen flow breakers to increase the efficiency of their interaction with catalysts. This was done by us according to the method of the theory of inventive problem solving [4]. As for the method of development of «vepols», according to the theory [4] we proposed to replace the «matters» V_2 and V_3 in such a way that the new «matters» V_2 and V_3 were more dispersed, i.e. more effective catalysts, or cheaper while maintaining efficiency. In practice, this means that we replaced expensive platinum catalysts with cheaper nickel catalysts, while using nickel nanopowders, which are protected by a shell of a corrosion-resistant substance (carbon). This should significantly reduce the cost of fuel cells.

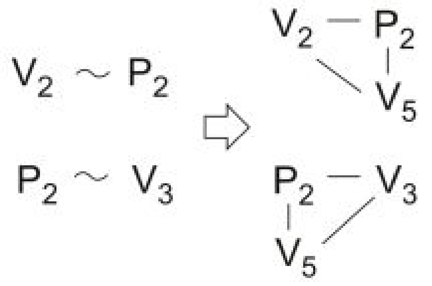

Fig. 4. The «vepol» completion rule for the transition of fuel cells to a solid polymer electrolyte

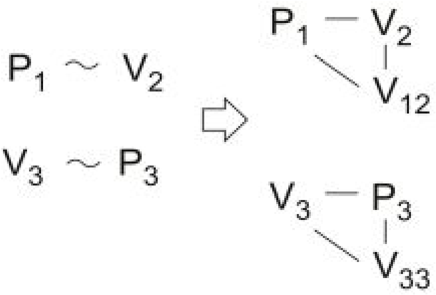

Fig. 5. Rules for completion and development of «vepol» for the interaction of gas flows with catalysts

The volt-ampere characteristic U = U(J) of a fuel cell can be written by the following equation:

$$U = U_0 - J \cdot (\rho + \eta_a + \eta_c),$$

where U_0 is the open circuit voltage, ρ is the specific ohmic resistance; η_a, η_c are the specific polarization resistances of the anode and cathode, respectively. Open circuit voltage is expressed through the change in free energy of reactants ΔG during electrochemical processes:

$$U_0 = -\Delta G / \Delta Q,$$

where ΔQ is electrical charge that flows during electrochemical processes. The change in free energy is the sum of the changes in free energies ΔG_j of all reactants involved in the process:

$$\Delta G = \sum \Delta G_j .$$

For a fuel cell involving gaseous reactants, the free energy G_j of each reactant is expressed by the equation:

$$G_j = G_j^0 + R \cdot T \cdot \ln a_j ,$$

where G_j^0 is the tabular value of free energy of the j reactant; R is the universal gas constant; T is the temperature; a_j is the thermodynamic activity of the j reactant.

Many of the above quantities exhibit complex behavior during fuel cell operation, so in practice it is often necessary to use empirical and semi-empirical formulas.

Preliminary tests of a mock-up of a fuel cell, which was created according to the improvements made, showed encouraging results.

The volt-ampere characteristic of a mock-up of a fuel cell demonstrates three sections:

– section 1 corresponds to a current density J (A/cm^2) from 0 to 0.3;

– section 2 corresponds to a current density J (A/cm^2) from 0.3 to 1.01;

– section 3 corresponds to a current density J (A/cm^2) from 1.01 to 1.2.

The voltage $U(V)$ at the current terminals of a mock-up of a fuel cell decreased from 1.05 to 0.42.

We used dimensionless variables: $y = U/U_{\max}$; $x = J/J_{\max}$. Then we approximated the volt-ampere characteristic (in dimensionless variables) with three empirical formulas:

$$y = a \cdot e^{-bx} + c; \quad (1)$$

$$y = d - f \cdot x; \quad (2)$$

$$y = -g \cdot e^{hx} + q, \quad (3)$$

in accordance with sections 1, 2 and 3. The empirical dimensionless constants a, b, c, d, f, g, h, q take values equal to or greater than zero.

Constants a, b, c describe (1) the rapid voltage drop in section 1, and constants g, h, q describe (3) the rapid voltage drop in section 3. The increase in technical characteristics, in particular the specific energy of the fuel cell, corresponds to the increased values y in section 2, where a practically linear slow decrease in voltage is observed (2). This means that the constant d can increase, the constant *f* can decrease compared to the prototype mock-up of a fuel cell. In our preliminary experiments, an increase in the constant d from 0.81 to 0.82 was observed, and the immutability of the constant *f*. Overall this means a slight increase in specific energy.

We also established that the requirements for fuel cell electrodes contain a technical contradiction: the anode and cathode must be mechanically strong and corrosion-resistant (especially the cathode). Mechanical strength is required so that the electrodes can fasten the entire structure of the fuel cell. For mechanical strength, the electrodes must be made of stainless steel. On the other hand, the electrodes must be corrosion resistant, as they interact with gases, especially the cathode, which interacts with oxygen under heating conditions. Therefore, the electrodes must be made of carbon material. However, electrodes made of carbon material are not mechanically strong enough, and stainless steel electrodes are not sufficiently corrosion resistant and subject to harmful hydrogen embrittlement. The use of titanium electrodes could satisfy the conflicting requirements, but titanium is an expensive metal.

As a promising solution to the technical contradiction, you can use composite materials that will satisfactorily solve the problem. The authors of this article suggest the use of a grid (armature) made of stainless steel or titanium with little waste of material, on which carbon material is applied by stamping.

Conclusions

The proposed mathematical model of the process of improving the oxygen-hydrogen fuel cell can reduce the time of searching for a technical solution and corresponding development. It is clear that the appropriate result can possibly be achieved by a thorough research search and sorting out many options. But the use of a qualitative mathematical model, in particular, «vepol» analysis, significantly reduces the time of research and search. The use of methods of the theory of inventive problem solving helps to overcome the psychological inertia of scientists and engineers. Formal methods, such as searching for and overcoming technical inconsistencies, «vepole» analysis and other operations cannot replace specific design and technological solutions. Nevertheless, the usefulness of such modeling is beyond doubt.

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