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STUDY OF THE INFLUENCE OF OPERATING CONDITIONS ON HIGH PRESSURE ELECTROLYZER EFFICIENCY

Andrii A. Shevchenko
shevchenko84@ukr.net
ORCID: 0000-0002-6009-2387

Mykola M. Zipunnikov
ORCID: 0000-0002-0579-2962

Anatolii L. Kotenko
ORCID: 0000-0003-2715-634X

Iryna O. Vorobiova
ORCID: 0000-0002-1712-8831

Vitalii M. Semykin
ORCID: 0000-0001-5042-810X

A. Podgorny Institute of Mechanical Engineering Problems of NASU,
2/10, Pozharskyi St., Kharkiv, 61046, Ukraine

The high pressure of the gases being generated (hydrogen and oxygen) makes it possible to increase the efficiency of the electrochemical generator. Energy components of the process of liquid electrolyte decomposition under pressure are described. Dependencies of reducing energy costs per cell during the electrolysis of water under pressure at different temperatures are presented. It has been established that with increasing pressure, the processes of electrode depolarization by dissolved gases increase, however, their quantitative value and influence on the current efficiency depend on the design of electrolyzers, adopted electrolyte circulation scheme, and conditions for the penetration of the dissolved hydrogen into the anode space and of oxygen into the cathode. The pressure increase of the electrochemical process of producing hydrogen and oxygen is accompanied by an increase in their solubility in the electrolyte, which may be accompanied by the anode and cathode depolarization by dissolved gases. The transition of the electrolysis equipment to the operation from atmospheric pressure to the operation from pressures of the order of 0.1–4.0 MPa is most effective. The decrease in the voltage of electrochemical reactions is due to the processes of electrode depolarization, dissolved gases, as well as a decrease in gas filling due to a decrease in the size of gas bubbles. With increasing pressure, the value of the electrode potential increases, which should lead to an increase in the cell voltage, but the opposite is observed. This can be explained by a decrease in voltage loss during electrolysis. We conducted a comparative analysis of the existing technologies for the electrolysis of hydrogen and oxygen. To correctly compare the energy efficiency of a membrane-free technology for producing high-pressure hydrogen and oxygen as well as existing industrial electrolytic technologies, the energy costs for electrolytic hydrogen production by an industrial method and its subsequent compression should be summarized.

Keywords: *electrolyzer, hydrogen, oxygen, high pressure.*

Introduction

The volume of the gases being generated during the electrochemical decomposition of a liquid electrolyte relative to the volume of water needed to generate them increases approximately by 2,000 times. Carrying out electrolysis in a sealed vessel can provide an increase in the pressure of the hydrogen and oxygen being generated by the same amount. This peculiarity of water decomposition by the electrolytic method was used by electrochemists more than 140 years ago to create electrolyzers operating under high pressure, [1, 2].

At the A. Podgorny Institute of Mechanical Engineering Problems of the NAS of Ukraine (IPMash NAS of Ukraine), there have been developed modular high-pressure gas generating electrolyzers [3–6], providing the ability to obtain the required performance by combining unified electrolytic cells. As shown by the experimental and laboratory studies of the electrochemical decomposition of liquid alkaline electrolyte (25% KOH solution), the use of metals with variable valence as active electrodes reduces the energy intensity of the process, eliminating the need for using ion-exchange membranes [7], thereby increasing the reliability and durability of work. The pressure of the hydrogen and oxygen being generated is limited only by the strength characteristics of the structural elements of high pressure electrolyzers (HPE). The use of gas-absorbing electrodes makes it possible to eliminate the use of rare-earth materials and platinum group metals as catalysts for electrolysis [8], which increases the reliability, and reduces the cost of equipment. In addition, it was experimentally established that during electrolysis, with an increase in pressure, a decrease in the total voltage across the cell is observed [9, 10].

Purpose and Goal of the Study

The main objective of the study is to analyze the effect of the pressure of the gases being generated on the HPE efficiency, and determine its main distinctive peculiarities in comparison with electrolyzers operating under low overpressure up to 5.0 MPa. According to the results of the analysis, it is necessary to develop methods of reducing irreversible losses and increasing the energy efficiency of the electrochemical method of cyclic production of high pressure hydrogen and oxygen. The goal can be achieved by using rational design solutions, modern software to optimize the operating algorithm, and temperature conditions.

Development of the HPE Layout Scheme

At the hydrogen energy department of the IPMash NAS of Ukraine, a number of gas-generating HPEs have been developed. Fig. 1 shows the layout of their main structural elements: 1– electrochemical cell; 2 – hydrogen separator; 3 – oxygen separator; 4 – technological H_2O storage tank; 5 – electromechanical gas-liquid flow switch.

The design of the electrolyzer that implements the proposed technology includes an electrolysis unit, a gas-liquid flow separation system, an electronic control unit for monitoring the operating modes of the electrolysis unit, and a power supply. A serial electrical connection of electrolytic cells allows reducing the phase current, which significantly reduces the consumption of copper in the buses for supplying power to the electrolyzer.

The HPE parameters were monitored using the SCADA supervisory control and data acquisition software, and the electrolyzer was controlled using the ZelioSoft 2 controller software package (fig. 2).

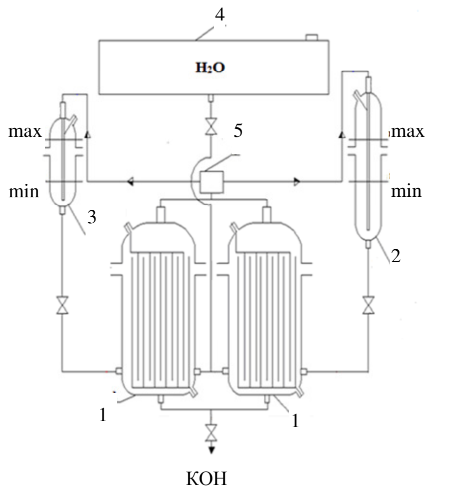
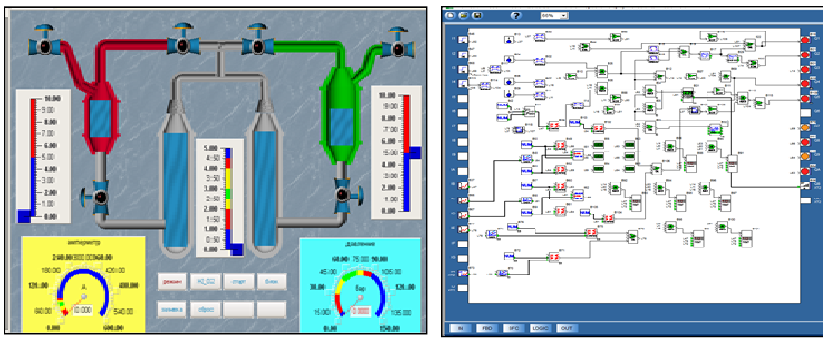


Fig. 1. Schematic layout of the placement of the main mechanisms in a series connection of cells

A peculiarity of modern integrated electrochemical production complexes is the combination of the possibilities of controlling a real process according to the “here and now” principle, which made it possible to remotely fix the parameters being measured with high accuracy, and ensure the control of an HPE at the hardware level, thereby achieving reliable operation of the equipment and ensuring the safety of the study.



a b

Fig. 2. Main software packages for creating HPEs:
a – SCADA supervisory control and data acquisition software;
b – ZelioSoft 2 controller software package

Energy Components of the Process of Decomposition of a Liquid Electrolyte Under Pressure

The decrease in the voltage of electrochemical reactions can be explained by the processes of electrode depolarization, dissolved gases, and a decrease in gas filling of the electrolyte due to a decrease in the size of gas bubbles. An increase in current density leads to an increase in gas filling, and to reduce it, it is necessary to ensure forced electrolyte circulation.

Fig. 3 shows graphs of voltage changes depending on pressure and current density.

It can be seen from the figure that the most significant decrease in voltage is observed in the framework of pressure increase to 4.0 MPa. With a further increase in pressure, the voltage change becomes insignificant. This is clearly manifested at various current densities.

Currently, filter-press electrolyzers with a working pressure of 0.05–2.6 MPa are widely used, which is not enough for the compact buffer storage and use of hydrogen. For the operation of hydrogen filling stations, the hydrogen pressure should be 35.0 to 70.0 MPa [12]. To further increase the pressure to the value specified, a low-efficiency compressor technique is used, which requires additional energy consumption. In the table below are the energy costs for the adiabatic compression of hydrogen at different temperatures.

Fig. 4 shows graphs of the adiabatic compression of hydrogen at different temperatures.

Taking into account the gas compressor efficiency from 20 to 50%, the energy saving for the compression of 1 m^3 of hydrogen and 0.5 m^3 of oxygen during liquid electrolyte electrolysis under pressure with taking into account different temperatures is from 0.5 to 1.0 kW·h/m^3.

The increase in the pressure of the electrolysis process made it possible to expand the range of operating temperatures. Fig. 5 shows graphs of the reduction of energy costs per cell during the electrochemical water decomposition under pressure at different temperatures.

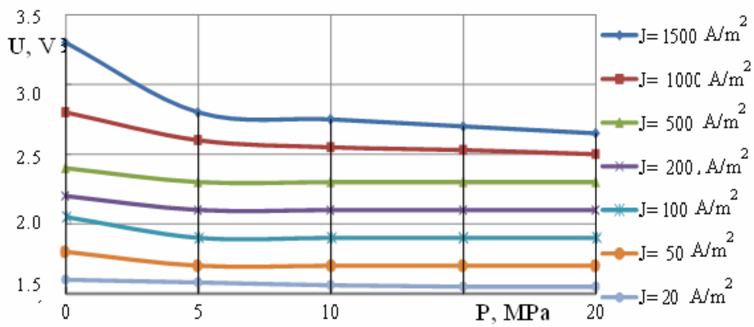


Fig. 3. Dependence of the electrolyzer voltage on pressure and current density [11]

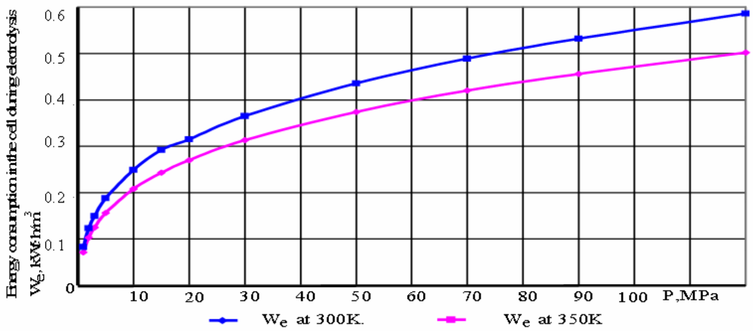


Fig. 4. Energy costs for the adiabatic compression of hydrogen at different temperatures

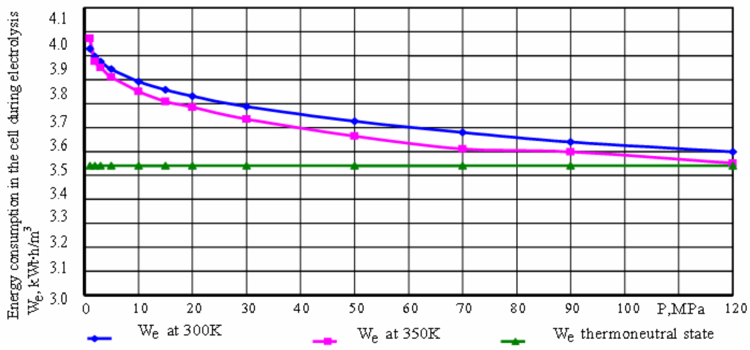


Fig. 5. Dependence of the reduction in energy costs on the electrolytic cell pressure

Costs for the adiabatic compression of hydrogen

P, MPa	1	2	3	5	10	15	20	30	50	70	90	120
W_e, kW·h/m^3	*T*=300 K											
	0.071	0.103	0.125	0.156	0.208	0.243	0.270	0.313	0.374	0.420	0.460	0.502
	T=350 K											
	0.083	0.123	0.149	0.188	0.249	0.292	0.315	0.365	0.436	0.489	0.532	0.586

As can be seen in the figure, to realize the advantages of electrolysis under pressure, it is advisable to work at a temperature of 350 K.

The determination of the regularities of the change in voltage across a cell with increasing pressure has been studied by many researchers [13, 14].

The theoretical decomposition voltage increases with increasing pressure. According to the Nernst equation, the reversible potential [11]:

– for the hydrogen electrode

$$e_{0H_2} = -\frac{RT}{nF}\ln\frac{P_{H_2}}{p^2_{H^+}},$$

– for the oxygen electrode

$$e_{0O_2} = -\frac{RT}{nF}\ln\frac{P_{O_2}}{p^2_{O^-}}.$$

When the gas pressure changes from P_1 to P_2, the value of the reversible electrode potential should change:

– for the cathode by

$$\Delta e_{0H_2} = -\frac{RT}{nF}\ln\frac{P_2}{P_1},$$

– for the anode by

$$\Delta e_{0O_2} = -\frac{RT}{nF}\ln\frac{P_2}{P_1}.$$

The change in the theoretical water decomposition voltage depending on pressure is given below [11]

$$\Delta E_0 = \Delta e_{0O_2} - \Delta e_{0H_2} = 3/4\,\frac{RT}{nF}\ln\frac{P_2}{P_1}.$$

where ΔE_0 is the electrode potential, V; Δe_{0H_2} is the reversible potential for the hydrogen electrode, V; Δe_{0O_2} is the reversible potential for the oxygen electrode, V; R is the universal gas constant equal to 8.314 J/(mol·K); T is the absolute temperature, K; F is the Faraday constant equal to 96,485.33 C·mol $^{-1}$; n is the number of electrons involved in the process; P_1, P_2 are the initial and final values of the gas pressure, MPa.

By substituting the numerical values R=8.316 V·k (volt·pendant) into this expression and passing to decimal logarithms, we obtain

$$\Delta E = 0.00015T\,\lg\frac{P_2}{P_1}.$$

Analysis of Industrial Designs and Promising Developments of Electrochemical Hydrogen Generators

In modern technological schemes, filter-press electrolyzers with the following technical characteristics are widely used:

– range of working pressures from 0.05 to 2.6 MPa;
– operating temperature from 333 to 353 K;
– current density from 1200–2500 A/m^2.

In this case, energy consumption depends on the process temperature, pressure of the gases being generated, electrode material, electrolyzer design, and varies from 4.3–5.8 kW·h for the production of 1 m^3 of hydrogen and 0.5 m^3 of oxygen [15–19].

A promising direction in the development of electrolytic technology is the use of the design of electrolyzers with proton exchange membranes (PEM), which makes it possible to operate under relatively high pressures up to 1.0–5.0 MPa [20–22].

The membrane-free technology (with a gas-absorbing electrode) makes it possible to reduce energy consumption by 10–15%, compared with the existing industrial counterparts. To generate 1 m^3 of hydrogen and 0.5 m^3 of oxygen, 3.9–4.0 kW·h of energy are consumed. The operating pressure range is from 1.0 to 70.0 MPa [23–26].

Fig. 6 presents the characteristics of the existing and promising electrolyzers: 1 – modern industrial electrolyzers: PV, SEU (Russia), Weuder Model, DeNora (Italy), Norsk (Norway), StuartImet (Belgium), Teledyne (USA), Loncza, Demag (Germany) (293–363 K; 0.1–3.0 MPa); 2 – promising alkaline electrolyzers (363–413 K; 0.1–7.0 MPa): Hamilton Sunsdstranl (USA), Proton Energy Systems Inc. (USA), H – TEC (Germany), RRC "Kurchatov" Institute (Russia); 3 – solid polymer electrolyte electrolyzers (423 K; 0.1 MPa): Matsushita Electric Works, Ltd, Fuji Electric Co, Ltd (Japan), Norwegian University of Science and Technology (Norway), David Systems and Technology (Spain); 4 – high-temperature electrolyzers (1373 K; 0.1 MPa): General Electric Co (USA), Brown Boveri (Switzerland); 5 – gas absorbing electrode electrolyzer (353–423 K; 1.0–20.0 MPa): IPMash NAS of Ukraine (Ukraine).

The figure shows that the electrolyzer with a gas-absorbing electrode (position 5), developed at the hydrogen energy department of IPMash NAS of Ukraine, in terms of energy efficiency, is at the level of electrolyzers with a solid polymer electrolyte (position 3) and high-temperature electrolic cells (position 5).

To correctly compare the energy efficiency of the membrane-free technology for producing high-pressure hydrogen and oxygen with the existing industrial electrolytic technologies, the energy costs for the electrolytic production of hydrogen by an industrial method should be summarized with its subsequent compression.

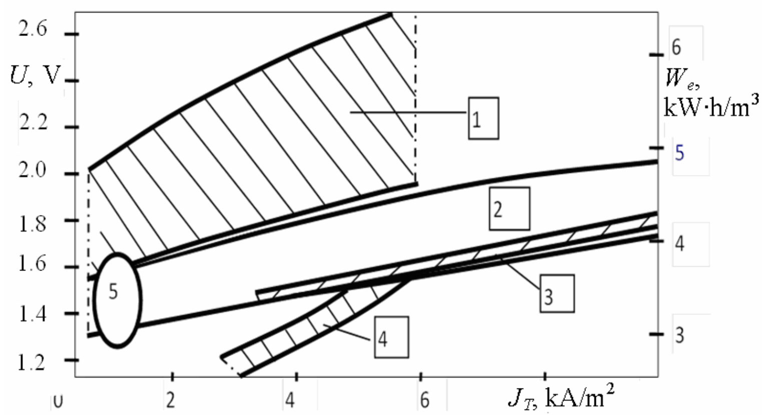


Fig. 6. Current-voltage characteristics of the existing and promising industrial electrolyzers [11]

Conclusions

The analysis of the main distinguishing operating peculiarities of an HPE in comparison with electrolyzers operating under low overpressure showed the following.

1. The reason for the decrease in voltage loss in the electrolyte with increasing pressure may be:

– a reduction of gas filling (a reduction in the size of gas bubbles) of the electrolyte;

– an increase in its operating temperature (a decrease in the ohmic resistance of the electrolyte due to an increase in electrochemical activity);

– a reduction in voltage during electrolysis.

2. The reduction in energy consumption in the production of hydrogen by 10–15% compared with the existing analogues (specific energy consumption is 3.8–4.1 kWh with generating 1 m^3 of hydrogen and 0.5 m^3 of oxygen).

3. The absence of separation membranes ensures the generation of gases with the pressure that is limited only by the structural strength of housing elements, which increases the reliability and safety of the system.

4. The electrochemical decomposition of a liquid electrolyte under high pressure is associated with an increase in the size of its structural elements in order to ensure their strength characteristics, which leads to an increase in metal consumption as a whole.

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Дослідження впливу режимних параметрів на ефективність роботи електролізера високого тиску

А. А. Шевченко, М. М. Зіпунніков, А. Л. Котенко, І. О. Воробйова, В. М. Семикін

Інститут проблем машинобудування ім. А. М. Підгорного НАН України,
61046, Україна, м. Харків, вул. Пожарського, 2/10

Високий тиск газів, що генеруються (водню і кисню), дає можливість підвищити ефективність роботи електрохімічного генератора. Описано енергетичні складові процесу розкладання рідкого електроліту під тиском. Наведені залежності зниження витрат енергії на комірці під час електролізу води під тиском за різних температур. Встановлено, що з підвищенням тиску посилюються процеси деполяризації електродів розчиненими газами, проте їх кількісне значення і вплив на вихід за струмом залежить від конструкції електролізерів, прийнятої схеми циркуляції електроліту та умов проникнення розчиненого водню в анодний простір і кисню – в катодний. Зростання тиску електрохімічного процесу отримання водню і кисню супроводжується збільшенням їх розчинності в електроліті, що може супроводжуватися процесами деполяризації анода і катода розчиненими газами. Перехід роботи електролізного обладнання від атмосферного тиску до тисків близько 0,1–4,0 МПа найбільш ефективний. Зниження напруги протікання електрохімічних реакцій обумовлено процесами деполяризації електродів, розчиненими газами, а також зниженням газонаповнення внаслідок зменшення розміру газових бульбашок. З ростом тиску збільшується значення електродного потенціалу, що має призвести до збільшення напруги на комірці, але спостерігається протилежне. Це можна пояснити зниженням напруги втрат в процесі електролізу. Проведено порівняльний аналіз існуючих технологій електролізного одержання водню та кисню. Для коректного порівняння енергоефективності безмембранної технології отримання водню та кисню високого тиску та існуючих промислових електролізних технологій слід підсумувати енерговитрати на електролізне виробництво водню промисловим способом і наступне його компримування.

Ключові слова: *електролізер, водень, кисень, високий тиск.*

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