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THE METHOD OF DETERMINING PEMFC FUEL CELL STACK PERFORMANCE DECREASE RATE BASED ON THE VOLTAGE-CURRENT CHARACTERISTIC SHIFT

METODA WYZNACZANIA SZYBKOŚCI SPADKU WYDAJNOŚCI STOSU OGNIW PALIWOWYCH TYPU PEMFC NA PODSTAWIE PRZESUNIĘCIA CHARAKTERYSTYKI NAPIĘCIOWO-PRĄDOWEJ*

The article presents mathematical model designed to estimate the rate of performance decrease in fuel cell stack. The fuel cell stack performance decrease rate is determined on the basis of stack average voltage measurements. The proposed model is used to determine power curve as well as exploitation indicators of fuel cell stack with a nominal power of 50 kW after 14 000 hours of continuous operation. The model is also used to determine the average voltage drop for the eleven-year fuel cell stack with a nominal power of 1,2 kW. In both studies, the values of exploitation indicators as well as their differences in relation to nominal values are determined.

Keywords: *fuel cells system exploitation, fuel cell stack exploitation indicators, stack performance decrease after years of operation, stack exploitation characteristics.*

Artykuł przedstawia model matematyczny przeznaczony do wyznaczenia szybkości spadku wydajności stosu ogniw paliwowych. Szybkość spadku wydajności stosu ogniw jest wyznaczana na podstawie wartości napięcia średniego stosu. Zaproponowany model wykorzystano do wyznaczenia krzywej mocy i wskaźników eksploatacyjnych stosu ogniw paliwowych o mocy nominalnej 50 kW po 14 000 h ciągłej pracy. Model wykorzystano także do wyznaczenia szybkości zmiany wartości napięcia średniego jedenastoletniego stosu ogniw paliwowych o mocy 1,2 kW. W obu badaniach wyznaczono wartości wskaźników eksploatacyjnych oraz ich różnice względem wielkości nominalnych.

Słowa kluczowe: *eksploatacja systemów ogniw paliwowych, wskaźniki eksploatacyjne stosu ogniw paliwowych, spadek wydajności stosu po latach eksploatacji, charakterystyka eksploatacyjna stosu.*

1. Introduction

Hydrogen fuel cells with Proton Exchange Membrane Fuel Cell (PEMFC) are considered one of the most promising and forward-looking technologies for generating electricity and heat. Their usage is foreseen for high-capacity power plants, small distributed sources [14] and transport sector [12, 30]. Fuel cells can be exploited in a wide range of electrical load variability, with favourable performance indicators such as: efficiency of the processing of fuel chemical energy into electricity, unit fuel chemical energy consumption, unit fuel consumption. Nevertheless, the reliability indicators of cell operation are still reaching unsatisfactory values and are the main reason for inhibiting the commercialisation of this technology on a large scale [21].

The improvement of reliability indicators and determination of operational indicators is now an up-to-date and important problem. The development of fuel cell technologies will be the main driver of the development of the hydrogen energy sector and hydrogen fuel-based transport. The research is being carried out on the use of hydrogen as an additional fuel for petrol and diesel engines [18, 20]. The analysis of the results of the study conducted by the authors of the work [20] showed the lack of appropriateness of using hydrogen as an additional fuel in compression ignition engines. Hydrogen power supply is mainly adapted by spark-ignition engines. However, the work [30] concluded that this is a temporary solution to pre-prepare and implement hydrogen storage and distribution infrastructure before the introduction of prospective fuel cells.

Due to continuously high costs of fuel cells, many studies of this technology are carried out using proprietary mathematical models [2, 34]. Work on modeling the fuel cell stack can be divided into two groups. The first includes works that present models designed to optimize stack parameters according to goal functions such as: minimizing construction costs, maximizing current density [1, 4, 6, 15, 27, 32].

In the article [4], the authors present a three-dimensional multiphase model of the PEM fuel cell designed to study the effect of assembly pressure on the contact resistance between the gas diffusion layer (GDL) and the bipolar plate. The correct selection of the mounting pressure allows you to make a stack with a longer life.

In the paper [6], the authors present a model designed to optimize fuel cell stack dimensions according to an improved version of the seagull optimization algorithm. The model allows to determine the operational characteristics of the stack, however, it does not take into account its displacement after the operational time. In turn, in the paper [1], the authors present a 500-watt fuel cell stack model with ion exchange polymer membrane implemented in Matlab/Simulink environment. The model is used to determine the reference value of electric current for any steady state.

In this paper [27], the authors present the processes taking place in the fuel cell stack and the developed numerical models aimed at mini-

(*) Tekst artykułu w polskiej wersji językowej dostępny w elektronicznym wydaniu kwartalnika na stronie ***www.ein.org.pl*** mising the stack production costs and maximising current density. These models are designed to support the design of fuel cell stacks.

New techniques of fuel cell modeling are being developed, e.g. with the use of the so-called bonding diagrams [32]. Model tests are also carried out to increase energy efficiency by connecting a stack of cells with a thermoelectric generator into a hybrid system [15].

All the above mentioned mathematical models are built to support the design of the fuel cell stack. However, these models do not take into account the decrease in stack efficiency after years of operation.

The second group of publications on stack modelling include works that model the degradation processes taking place in the stack during its work. The authors focus on determining the influence of one of the structural elements, such as gas diffusion layer (GDL) [23], bipolar plates, polymer membrane [26] and catalyst layer on the electrodes of the stack [31], on the voltage-current characteristics. These models are used to minimize the source of degradation and increase the stack life.

The paper [26] presents a numerical model of fuel cells designed to test mechanical membrane degradation caused by local, non-pressurized membranes. In order to reduce the stresses, the authors suggest using an additional seal in connecting the electrode-electrolyte with GDL.

Subsequent works concern the determination of the probability of degradation of a given element [25, 35]. The authors of the article [25] proposed to use a foul three analysis tree to determine the probability of degradation of fuel cell stack elements. In turn, the article [35] proposes an innovative model of forecasting the degradation of a fuel cell with proton exchange membrane using the molecular filter and data extrapolation method.

Unfortunately, the above mentioned models and proposed methods are not oriented towards practical application because they do not show how the progressing degradation processes will affect the efficiency of the fuel cell stack after many hours or years of operation and, most of all, how the values of the performance indicators will change. The knowledge of changes in performance values will have a decisive impact on the correct determination of stack operating costs.

Publications on modelling hybrid power generation systems [16, 19, 28] confirm that the demand for a model showing the impact of fuel cell stack ageing on performance indicators is currently very high. The modelling techniques of hybrid systems presented in the above mentioned works do not take into account the decrease in stack efficiency after years of operation. Consequently, in such a hybrid system, energy imbalance may occur [24], which will result in increased consumption from the power grid [9].

Similarly, if a fuel cell is used in the transport sector [3, 11, 29], failure to take into account the decrease in stack efficiency may lead to an erroneous estimate of the vehicle's range or fuel demand, resulting in an erroneous estimate of the vehicle's operating costs.

In order to correctly determine the operating costs of a vehicle or a fuel cell hybrid electricity generation system, mathematical models should be built to determine the loss of performance of the cells after years of operation.

The main purpose of this article is to present a mathematical model which allows to determine the rate of decrease of fuel cell stack efficiency and the shift of the stack's operational characteristics in relation to the characteristics for catalog data after a given period of use. The model allows to determine the values of performance indicators, i.e. electrical efficiency, specific fuel chemical energy consumption for electricity production, specific fuel consumption for electricity production and to compare them with the values of indicators corresponding to the characteristics of the stack catalogues.

2. The mathematical model of the fuel cell

The mathematical models of fuel cells described in the literature make it possible to determine the operating characteristics for rated conditions (catalogue parameters). The proposed models do not take into account the decrease in stack efficiency after years of life and operation.

The analysis method proposed by the authors allows to determine the operational characteristics of the fuel cell stack for the rated data and the characteristics after "n" years of operation. The input variable of the model is, in this case, the rate of decrease of the average voltage value of one cell (one stack cell). On the other hand, the proposed algorithm can be used to determine the rate of drop in performance of the stack based on the shift between the catalogue characteristics and the stack characteristics after a given service life.

The method allows to quickly determine the efficiency and other performance indicators of the fuel cell stack.

The efficiency of converting the chemical energy of a fuel into electricity through a stack of fuel cells can be presented using formula (1) [7]:

$$\eta_{el} = \frac{P_{el}}{\dot{n}_{H_2} \cdot Q_{W_{H_2}}} = \frac{U \cdot I}{\dot{n}_{H_2} \cdot Q_{W_{H_2}}} \quad (1)$$

where: P_{el} – electrical power of the fuel cell stack, U – fuel cell stack voltage, I – electric current, $\dot{n}_{H_2}$ – hydrogen mole stream, $Q_{W_{H2}}$ – hydrogen calorific value related to 1 mole respectively.

The indicator of the unit chemical energy consumption of the fuel for the production of electricity through the stack is defined by formula (2):

$$q_{el} = \frac{\dot{n}_{H_2} \cdot Q_{W_{H_2}}}{P_{el}} \left[\frac{kJ}{kJ}\right] \quad (2)$$

The unit fuel consumption ratio for electricity production can be calculated using formula (3):

$$b_{el} = \frac{V_{H_2}}{P_{el}} \left[\frac{Nm^3 H_2}{kWh}\right] \quad (3)$$

where: V_{H_2} – hydrogen volume flow rate.

It should be noted that manufacturers of PEMFC fuel cell systems specify two efficiency values for converting the chemical energy of the fuel into electricity: the efficiency value related to the calorific value of hydrogen (Low Heating Value) and the efficiency value related to the heat of hydrogen combustion (High Heating Value).The analysis presented in this paper is carried out in relation to the calorific value of hydrogen, because the value of the q_{el} index is defined, in the classical theory of exploitation of generating sources, in relation to this value.

Defining the efficiency of a fuel cell in relation to its calorific value allows to compare its value with other technologies of electricity generation using the chemical energy of the fuel, e.g. conventional steam units, gas and gas-steam units, gas engines or technologies using biomass.

The hydrogen molar flux formula can be determined from the Faraday II electrolysis law (4):

$$q = n \cdot z \cdot F \quad (4)$$

where: q [C] – electric charge, n – number of gas moles, z [-] – number of electrons needed to release the molecule for H_2 = 2, i.e. 2 electron moles are needed to release 1 mole of H_2, for O_2 = 4, F [C/mol] – Faraday constant.

By dividing equation (4) by the time t, the relations to the electric current (5) is obtained:

$$I = \frac{n}{t} \cdot z \cdot F \tag{5}$$

From equation (5) the molar flow marked $\dot{n}$ (6) can be determined:

$$\dot{n} = \frac{n}{t} = \frac{I}{z \cdot F} \tag{6}$$

For a fuel cell stack, the hydrogen molar stream is proportional to the number of cells in the stack, hence:

$$\dot{n}_{H_2} = \frac{I \cdot n_{celek}}{z \cdot F} \tag{7}$$

where: n_{celek} – number of cells (single cells) from which the stack is built.

In the theory of fuel cells, a size called thermoneutral potential, which is defined according to formula (8) [22], applies in practice:

$$E_t^0 = -\frac{\Delta H^0_{H_2 0(g)}}{z \cdot F} \tag{8}$$

where: E_t^0 – thermoneutral potential [V], $\Delta H^0_{H_2 0(g)}$ – standard enthalpy of water formation in gaseous phase [kJ/mol], index “0” means standard conditions (T = 298 K, p = 10^5 Pa).

Thermalneutral potential is the theoretical value of the voltage that a fuel cell will reach with the theoretical assumption that 100 % of the supplied chemical energy stream will be converted into electricity.

The standard enthalpy of water formation in the gaseous phase ΔH¬H2O(g) energetically corresponds to the calorific value of hydrogen, assuming that water is the product in the gaseous state (9) [17]:

$$-\Delta H^0_{H_2 0(g)} = Q_{W H2} \tag{9}$$

By substituting equations (4), (8), (9) for equation (1), the formula for the efficiency of electricity generation by the fuel cell stack takes the form of formula (10):

$$\eta_{el} = \frac{U}{n_{celek} \cdot E_t^0} \tag{10}$$

The average voltage of a fuel cell stack is defined as the ratio of the voltage of the stack to the number of cells (single cells) in the stack:

$$U_{av} = \frac{U}{n_{celek}} \tag{11}$$

After substituting relation (11) to formula (10), the formula for stack efficiency can be presented with the help of the relation:

$$\eta_{el} = \frac{U_{av}}{E_t^0} \tag{12}$$

Formula (12) for the efficiency of converting the chemical energy of the fuel into electricity through the stack is very practical, because to determine the efficiency it is sufficient to measure the voltage of the stack and know the number of individual cells forming the stack. There is no need to measure the hydrogen flux used.

The change in stack efficiency is measured by changing the average stack voltage. The change in efficiency is, according to relation (13), directly proportional to the change in average stack voltage:

$$\Delta\eta_{el} = \frac{\Delta U_{av}}{E_t^0} \tag{13}$$

The change of average stack voltage is defined by formula (14):

$$\Delta U_{av} = U_{av} - \frac{dU_{av}}{dt} \cdot t \tag{14}$$

where: $\frac{dU_{av}}{dt}$ – the rate of change of average stack voltage over time [μV/h], t – fuel cell stack lifetime [h].

The power change of the stack can be determined on the basis of the relationship (15):

$$\Delta P_{el} = \Delta\eta_{el} \cdot \dot{n}_{H_2} \cdot Q_{WH_2} \tag{15}$$

The algorithm presented above allows to quickly determine the rate of change in the average voltage of the stack, and thus the rate of decrease in the generated electric power and the efficiency of converting the chemical energy of the fuel into electric power, as well as the increase in the values of the indicators of specific fuel consumption and specific chemical energy consumption of the stack.

The input data of the model are the catalogue parameters of the stack and the value of the stack voltage determined from the measurements. Based on formulas (1)-(15), the values of the stack's operating indicators are determined and the operating characteristics are plotted for the catalogue parameters and after a given period of use. In order to determine the rate of decrease in the stack's efficiency, the algorithm performs a series of simulations for different values of the rate of change of the average voltage of the stack with a given step and evaluates, with a given accuracy, the adjustment of the actual characteristics to the simulated one.

3. Determination of performance indicators based on the rate of change of average stack voltage values

The change of the average stack voltage ΔUav can be approximated by the linear function [33]. The rate of change in average voltage over time for systems of several tens of kilowatts is included, based on operational experience, in the range from about 3 to 9 μV/h [33]. This value depends on many factors, such as the culture of stack operation (observance of operating procedures, starting and stopping the stack), stack operating conditions (weather conditions, ambient temperature), the nature of stack operation (continuous, intermittent), etc.

A pilot plant built of 50 kW PEM cells (12 stacks of 4,2 kW) in the Netherlands, in Delfzijl, worked 14 000 hours without interruption.

Measurements during operation showed that the average stack voltage decreased at an average rate of 8 μV/h [33].

Based on equations 1 - 15 and the proprietary code developed in the Matlab environment, simulations were carried out to determine the power curve of the stack and the values of the performance indicators. The results of the simulation are shown in Figure 1 and Table 1.

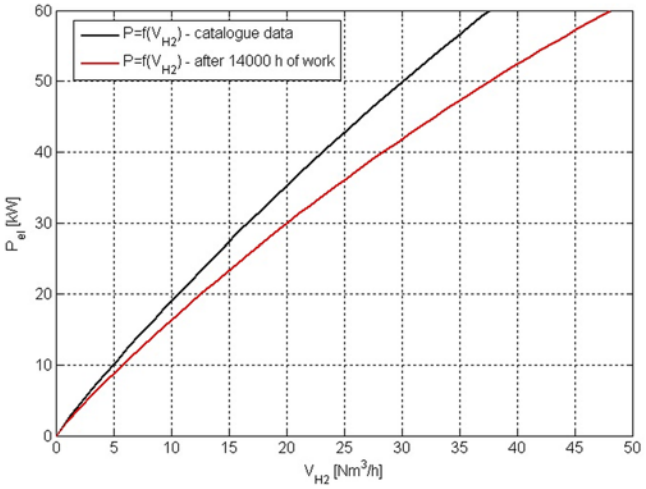

Fig. 1. Effect of U_{av} change on 50 kW fuel cell stack power curve - simulation tests

Table 1. Fuel cell stack performance indicators 50 kW - simulation tests

	Rated data	After 14 000 hours of operation
P [kW]	50	42.4
ΔP [kW]	−7.6	
η_{el} [-]	0.55	0.46
$\Delta\eta_{el}$ [-]	-0.09	
q_{el} [kJ/kJ]	1.81	2.16
Δq_{el} [kJ/kJ]	0.35	
b_{el} [Nm^3H_2/kWh]	0.60	0.72
Δb_{el} [Nm^3H_2/kWh]	0.12	

After 14,000 hours of continuous operation, the efficiency of the plant's electricity generation decreased by 16.36% from 55% to 46%. The effect of the decrease in efficiency is a decrease in the value of electrical power generated by the stack and an increase in the value of the indicators of specific chemical energy consumption of fuel and specific fuel consumption. The nominal capacity of the stack decreased by 15.2%. This means that the additional 7.6 kW of hydrogen chemical energy stream will be converted by the installation into thermal power at the expense of the value of generated electrical power. The value of the q_{el} index increased by 19.3% compared to the nominal value, while the value of the b_{el} index increased by 20%.

4. Determination of the rate of change of the average stack voltage from the measurement of the operating characteristics

In order to verify the model, it was used to match the voltage and current characteristics of the 1.2 kW fuel cell stack and to determine the rate of change of the average stack voltage. The tested fuel cell stack is an element of the NEXA training system, which is located in the energy conversion laboratory of the Wrocław University of Technology.

The NEXA system is a device designed for emergency power supply of both fixed and alternating current devices. Apart from the 1.2 kW stack, the system is made of [8]:

- hydrogen supply system – 20 MPa compressed hydrogen cylinders, hydrogen pressure regulator, hydrogen pressure regulator, pressure relief valve, solenoid valve to cut off fuel supply during system shutdown, hydrogen leakage detector,
- air supply system – a Roots blower,
- stack cooling system – the stack of cells in the NEXA system is cooled with air using a cooling fan,
- electronic control system – control computer, measuring sensors.

The age of the fuel cell stack is 11 years. Based on the simulation performed, the specific value of the average voltage change rate is 0,34 μV/h. For this value the best fit of the simulated and measured characteristics is obtained. The low value of the mean voltage change rate compared to a 50 kW system is due to the fact that the 1,2 kW cell stack consists of 46 cells. Additionally, the NEXA system operates in intermittent mode and is only used for research and teaching purposes.

Figure 2a shows the characteristics of the fuel cell stack: nominal, measurement-based characteristics (measured in March 2020) and simulation-based characteristics. Figure 2b shows the matching of measured and simulated characteristics.

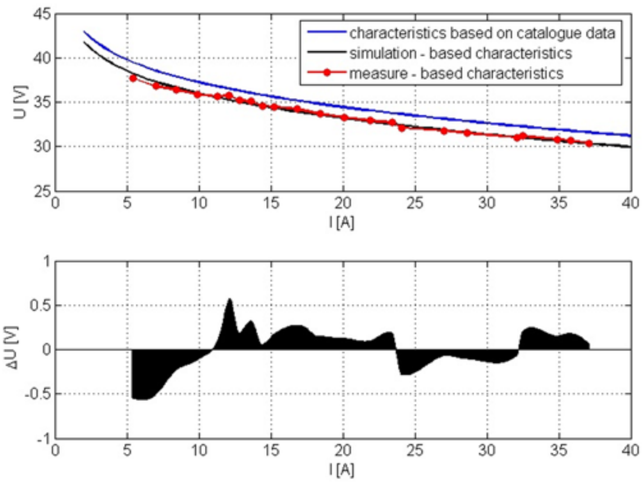

Fig. 2. a) Fuel cell stack characteristics: catalogue, simulated, measured, b) ΔU between simulated and measured characteristics

Deviations visible on the measurement characteristics are the effect of the anode flushing system during measurements [5]. At this point, the difference between the measured and simulated quantity is about 0.6 V. In most of the ohmic area [10, 13] of the voltage and current characteristics the difference between the points is about 0.2 V.

Table 2. Fuel cell stack performance indicators 1.2 kW

	Rated data	After 11 years
P_{el} [kW]	1.2	1.16
ΔP [kW]	−0.040	
η_{el} [-]	0.550	0.521
$\Delta\eta_{el}$ [-]	−0.029	
q_{el} [kJ/kJ]	1.840	1.920
Δq_{el} [kJ/kJ]	0.080	
b_{el} [Nm^3H_2/kWh]	0.610	0.640
Δb_{el} [Nm^3H_2/kWh]	0.030	

Table 2 shows a comparison of performance indicators calculated on the basis of catalogue data and simulation results. The low value of the average stack voltage change rate results in smaller changes in stack power and operating indicators. The nominal power of the fuel cell stack decreased from 1.2 kW to 1.16 kW after 11 years, reducing its value by 3.33%. The conversion efficiency of hydrogen chemical energy into electricity decreased by 5.27%. The fuel chemical energy conversion rate for electricity production increased by 4.35%. The specific fuel consumption for the production of one kWh of electricity increased by 4.92%.

Given the fact that the stack works under laboratory conditions with a high exploitation culture, the results should be considered as correct.

5. Summary

Updating the performance characteristics of fuel cells with ion exchange polymer membranes is a key issue in the development of hydrogen-based technologies, i.e. distributed generation of electricity and electromobility.

The proposed method allows to easily and quickly determine the decrease in efficiency of the fuel cell stack, which makes it possible to determine the current values of the performance indicators of the stack. On the other hand, the performance indicators determined after years of using the stack allow for a more accurate estimation of the operating costs of the fuel cell system.

The proposed method is a useful tool for carrying out a feasibility study of a project (e.g. hybrid power generation system with hydrogen storage, hydrogen car or hydrogen bus) on the basis of which the investor will be able to more accurately assess the risks associated with a given project by making technical and financial estimates a reality. The proposed model may facilitate the planning of long-term operation of fuel cell stacks in both distributed generation systems and hydrogen vehicles.

The advantages of the proposed method are its simplicity, short calculation time and the fact that it is sufficient to measure the voltage generated by the stack to determine the efficiency of the fuel cell stack.

The conducted simulations have shown that the value of the pile performance decrease depends on its nominal power (indirectly on the number of targets) and the mode and conditions of operation (continuous operation, intermittent operation). The model presented in the article contributes to the development of methods to update the performance characteristics of fuel cell stacks. These issues will be increasingly important in view of the expected decentralization of the electricity generation sector, the development of hybrid fuel cell stack generation systems and electromobility. The loss of fuel cell stack efficiency over the years of operation is an extremely important aspect for investors interested in new hydrogen technologies.

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