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Designing PEM Electrolysis-Based Hydrogen Reactors in The Area of Baron Beach of Yogyakarta, Indonesia

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ABSTRACT

This study aimed to design a PEM electrolysis-based hydrogen reactor and the potential for hydrogen production at Baron Beach, Gunung Kidul, Yogyakarta. Based on the calculation done at the initial process, the electrical energy potentially generated from renewable energy, such as wind, waves, and solar, reached 10.7 MW. This study also investigated the effect of reactor operating temperature on reactor efficiency and hydrogen production. A numerical thermodynamic approach was applied in the design process. The model, validated by laboratory experiments by other institutions, was in good agreement with previous research with an error value of 13%. The temperature range was dynamically limited from 30 to 80°C. The optimum operating conditions occurred when the temperature was set at 80 °C with a reactor efficiency, a water consumption rate, and a hydrogen production capacity of 76.3%, 2.817 kg/hour, and 250.42 kg/hour, respectively. The raw material, namely seawater, was processed using the reverse osmosis method. Ten reactors (with 13 cells per reactor) were installed in parallel.

I. INTRODUCTION

Baron Beach in Yogyakarta, Indonesia, has a unique contour with a basin surrounded by rocks. It is suitable for constructing power plants. The potential for electrical energy from wave, wind, and solar power in the vicinity of the area reaches 10.7 MW. This value, if produced in the absence of local demand and export, can be a potential commodity to be converted into hydrogen as an environmentally friendly fuel through water electrolysis that has been widely applied as fuel cells in the industrial and automotive fields [1][2]. Hydrogen itself is a substance that has many strategic functions in the chemical industry, like being a raw material for ammonia, methanol, oil refining, and various other chemicals [3].

Many researchers have designed renewable energy-based water electrolysis reactors with various approach methods, including modeling with an empirical and computational approach using Aspen and MATLAB software. [4] modeled a combination of an 18 kW wind turbine and 12 kW solar cell-Alkaline Water Electrolyzer with the help of MATLAB software.

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The parameters studied were the effect of hydrogen reactor temperature and on hydrogen production. They concluded that the higher the temperature, the efficiency of hydrogen production increased, which was accompanied by an increase in current density and a decrease in electric voltage. [5] modeled a PEM Electrolyzer with Aspen Custom Modeler software. In such modeling, they did not consider reactor geometry but did the effect of operating conditions, namely temperature and pressure, on hydrogen production. They found that an increase in operating pressure will only increase the voltage, while an increase in temperature will increase the current density only.

In 2018 and 2020, Sánchez [7] modeled a 15 kW-Alkaline Water Electrolyzer with the help of MATLAB software and the Aspen Custom Modeler. In his two studies, he developed a semi-empirical model compiled with MATLAB to be a computational model with the Aspen Custom Modeler. His modeling included a hydrogen reactor and its supporting process units. He found that as the operating temperature rose from 50 to 80°C, the voltage dropped, as did the stack power required for electrolysis. Meanwhile, the hydrogen production rate decreased when the temperature increased because the Faraday efficiency decreased and more hydrogen molecules crossed to the oxygen side. At a pressure of 5-9 bars, the stack stress did not increase significantly and did not affect the rate of hydrogen production. However, the higher the operating pressure, the lower the purity of the hydrogen. The purpose of this study is effect of operating temperature on the PEM Electrolyzer performance was studied using an empirical approach.

II. METHODOLOGY

Water electrolysis means separating water molecules into hydrogen and oxygen through an electrochemical reaction with the addition of electrical energy [6]. An electric current flow between two separate electrodes immersed in the electrolyte to increase the ionic conductivity. The reactions that occur at the electrodes are formulated in Eq (1) - (3) [8]. However, these reaction are occured in base environment.

Anode : $2OH^- \rightarrow H_2O + 1/2O_2 + 2e^-$ (1)

Cathode: $2H_2O + 2e^- \rightarrow H_2 + 2OH^-$ (2)

Overall : $H_2O \rightarrow H_2 + 1/2O_2$ (3)

There are several types of water electrolysis technologies, including Alkaline Water Electrolyzer (AEL), Proton Exchange Membrane (PEM) Electrolyzer, and Solid Oxide Electrolyzer (SOEL) [9]. PEM is the most attractive technology to be applied in the industrial sector because it has a compact design, high current density, higher stability, higher gas purity, and high hydrogen production rate and is the most suitable to be developed commercially. Therefore, in this study, the PEM Electrolysis-based hydrogen reactor was chosen.

Thermodynamically, the minimum energy required to break a water molecule can be calculated from the Gibbs free energy as a function of the reversible voltage. The reversible voltage itself is the minimum voltage of the cell for allowing the electrolysis process to occur at standard temperature and pressure.

$$V_{rev} = \frac{\Delta G°}{nF} = 1.23V \quad (4)$$

where:

$\Delta G°$ = standard Gibbs free energy (J/mol)

n = the number of electrons involved

F = Faraday constant (96500 C/mol)

V_{rev} = reversible voltage (V)

However, when the water molecules separate, a certain amount of entropy is released, so the total energy required for the electrolysis process is the enthalpy change of the process. So Equation (4) changes to:

$$\Delta G° = \Delta H° - Q = \Delta H° - T.\Delta S°$$

$$V_{rev} = \frac{\Delta H°}{nF} = \frac{\Delta G°}{nF} + \frac{T\Delta S°}{nF} = 1.48V \quad (5)$$

where:

$\Delta H°$ = enthalpy change (J/mol)

$\Delta S°$ = entropy change (J/mol.K)

T = operating temperature (K)

In fact, the reversible voltage is not only affected by temperature and pressure but also by the catalytic properties of the electrode, the diffusivity of the membrane, and the internal resistance of the cell. So the cell voltage follows the following equation:

$$V_{cell} = V_{rev}(T) + \eta_{electrode} + \eta_{\Omega} \quad (6)$$

where:

Vcell = Cell voltage (V)

ηelectrode = Kinetic resistance

η_Ω = Ohmic loses

According to Carmo [10], the typical operating temperature of a PEM electrolyzer ranges from 20 to 80°C. In this study, variations in operating temperature on the performance of the PEM electrolyzer reactor were studied using a thermodynamic approach to determine the optimum operating conditions. The thermodynamic approach was carried out empirically with the following assumptions:

- The effect of pressure was ignored
  According to Marangio *et al.* [11], pressurized PEM electrolyzers have several weaknesses, including: the phenomenon of cross-permeate or water that is not converted because it crosses directly through the membrane to the cathode side, corrosion, brittleness of reactor material by hydrogen, and cell instability.
- Uniform temperature in every cell.
- The heat lost to the surroundings was ignored.
- The mass transfer resistance or gas diffusivity to the membrane was ignored.
- The catalytic or kinetic properties of the electrode were ignored.
- Ohmic losses were constant
  In this study, the type of membrane used was Nafion 117 with the value of ohmic losses considered constant and not a function of temperature. According to Slade *et al.* [12], Nafion 117 membrane has an ohmic resistance value of 0.15 V

So Equation (6) can be simplified to Equation (7)

$$V_{cell} = V_{rev}(T) + 0.15 \tag{7}$$

where V_{rev} is a function of temperature at constant pressure by following the following equation.

$$V_{rev} = 1.5184 - 1.5421.10^{-3}T + 9.523.10^{-5}T\, lnT + 9.84.10^{-8}T^2 \tag{8}$$

By combining Equations (7) and (8), the equation to calculate the cell voltage for the electrolysis process is as follows:

$$V_{cell} = 1.6684 - 1.5421.10^{-3}T + 9.523.10^{-5}T\, lnT + 9.84.10^{-8}T^2 \tag{9}$$

The hydrogen production capacity of an electrolyzer is closely related to cell current and Faraday efficiency. Faraday efficiency is the relationship between the actual and theoretical hydrogen production rates caused by standby current losses or parasitic currents. According to research by Barbir [13] and Gorgun [14], Faraday's efficiency value for PEM electrolyzer is more than 99%. Hydrogen production capacity and electrolyzer feed water requirements can be calculated by the following equation:

$$fH_2 = \frac{n_{cell} I_{cell}}{2F} \eta_F \tag{10}$$

$$fH_2O = 1.25 \frac{n_{cell} I_{cell}}{2F} \eta_F \tag{11}$$

where:

f_{H2} = hydrogen production rate

f_{H2O} = feed water consumption rate

n_{cell} = number of cells

I_{cell} = cell current

η_F = Faraday efficiency

F = Faraday constant (96485 C/mol)

where the cell current can be calculated by the following equation, where P is the power generation (Watts):

$$I_{cell} = \frac{P}{V_{cell}} \tag{12}$$

After calculating with a thermodynamic approach, the results of these calculations were validated with experimental results. Validation was done by comparing the calculated Vcell value and experimental results at various temperatures. Validation accuracy was calculated based on error following the following equation:

$$Error = \frac{experiment - calculation}{experiment}\ x\ 100\% \tag{13}$$

The experimental results were taken from the PEM Electrolyzer facility for Hydrogen production at UPCT (Technical University of Cartagena) with the following specifications [15].

- Capasity = 1 kW
- Number of cells = 12
- Membrane = Nafion, 50cm2
- Number of reactor = 1 unit

III RESULTS AND DISCUSSION

As shown in equation (9), the cell voltage (Vcell) is the reverse voltage required to start an electrolysis process. The Vcell itself is the voltage required to counter resistance such as thermodynamic resistance (operating conditions), kinetic resistance (catalytic properties of the material), and mass transfer resistance (membrane diffusivity), and ohmic losses. In this study, the kinetic resistance-mass transfer was neglected and the ohmic losses were considered constant so that Vcell was considered as a function of temperature only. The results of calculating the Vcell and Icell values at various operating temperatures is presented in Figures 1 and 2.

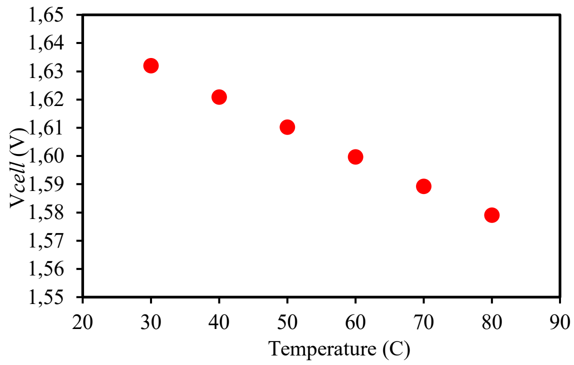

Figure 1. Effect of Temperature on Cell Voltage

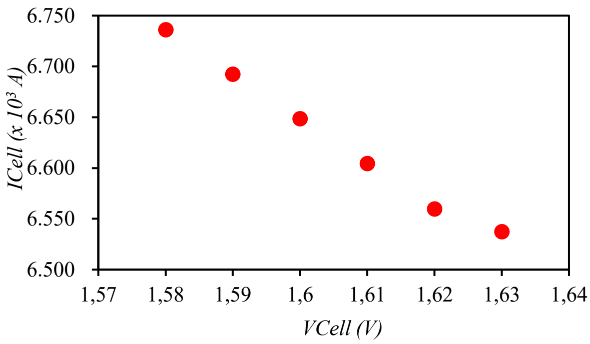

Figure 2. Effect of Cell Voltage on Cell Current

Figure 1 and 2 show that the increase in temperature decreases Vcell and increases Icell so that the rate of hydrogen production also increases according to Equation 10 as presented in Figure 3.

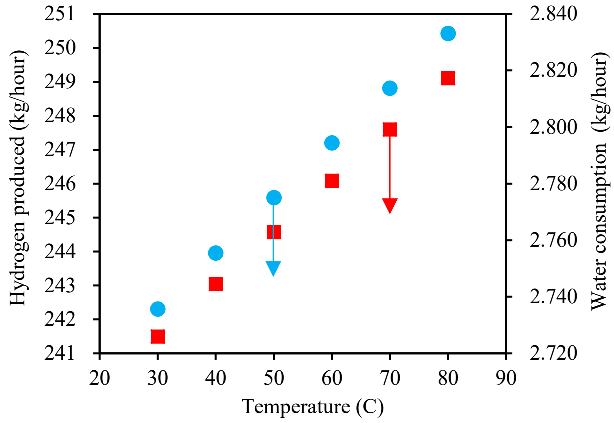

Figure 3. Hydrogen Production Rate and Water Consumption Rate at various operating temperatures

These data were following the report of the research conducted by Colbertaldo et al. [5], Sánchez et al. [7], and Sánchez et al. [16]. However, there was a difference between the results of the thermodynamic approach and those of the experiments, as presented in Figure 4. At the same temperature, the experimental Vcell was higher than the calculation one using the thermodynamic approach due to the presence of obstacles such as kinetic resistance and mass transfer, which were taken into account in the experiment. Thus, to carry out the electrolysis process, a higher Vcell value was needed. Figure 4 also shows that the higher the temperature rise, the smaller the difference in the Vcell values based on the experimental results and the approach results.

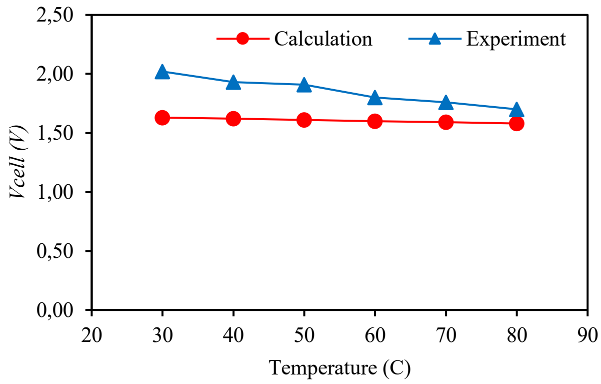

Figure 4. Comparison of Vcells based on Experiment vs Calculation

Figure 5 presents a process flow diagram for the hydrogen reactor design in this study. From this figure, the reactor heat and mass balance can be arranged so that the reactor efficiency can be calculated by the following equation:

$$\eta = \frac{n_{H2}\, HHV_{H2}}{P + Q_{cell} + Q_{H2O}}\ x\ 100\% \quad (14)$$

where

n_{H2} = Mol of hydrogen produced

HHV_{H2} = High heating value of hidrogen (286 kJ/mol)

P = Power required (kW)

Q_{cell} = Heat required for electrolysis (48.6kJ/mol H_2O) [12]

Q_{H2O} = The heat required to raise water temperature

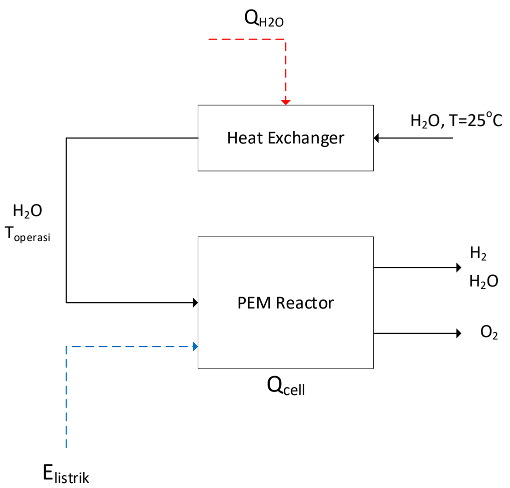

Figure 5. Reactor Design Process Flowchart

Through Equation (14), the calculation of the efficiency of the electrolysis process at various temperatures is presented in Figure 6. The highest efficiency was obtained at an operating temperature of 80°C. Thermodynamically, at this temperature, the electrolysis process should be carried out.

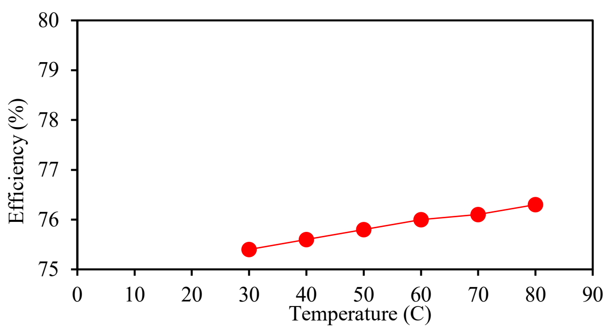

Figure 6. Reactor efficiency at various temperatures

Based on the calculation of the thermodynamic approach, the hydrogen reactor designed in this study was as follows:

Reactor type	= Proton Exchange
Membrane Toperating	= 80°C
Number of reactors	= 10 units
Configuration	= parallel
Number of cells	= 13
Membrane type	= Nafion 117
Active Area	= 50 cm^2
Current Density	= 1.2 A/cm^2

IV. CONCLUSION

From this research's results, it can be concluded that,

a. The PEM Electrolysis-based hydrogen reactors designed with the empirical thermodynamic approach were quite appropriate, as evidenced by an error value of 13%.

b. The optimum production could be obtained at an operating temperature of 80oC with a hydrogen production capacity, system efficiency, and an electric power input of 250.42 kg/hour, 76.3%, and 10.7 MW, respectively.

c. The designed hydrogen reactors consisted of 10 units installed in parallel. The number of cells in each reactor was 13.

References

[1] Yeh, S., D. H. Loughlin, C. Shay and C. Gage., "An Integrated Assessment of the Impacts of Hydrogen Economy on Transportation, Energy Use, and Air Emissions", *Proceedings of the IEEE,* 94 pp 1838-1851, 2006.

[2] Veziroglu, A. and R. Macario., "Fuel cell vehicles: State of the art with economic and environmental concerns", *International Journal of Hydrogen Energy,* 36 pp 25-43, 2011.

[3] Arregi, A., M. Amutio, G. Lopez, J. Bilbao and M. Olazar, "Evaluation of thermochemical routes for hydrogen production from biomass: A review", *Energy Conversion and Management*, 165 pp 696-719, 2018.

[4] Huang, P.-H., J.-K. Kuo and Z.-D. Wu., "Applying small wind turbines and a photovoltaic system to facilitate electrolysis hydrogen production", *International Journal of Hydrogen Energy,* 41 pp 8514-8524, 2016.

[5] Colbertaldo, P., S. L. G. Alaez and S. Campanari., "Zero-dimensional dynamic modeling of PEM electrolyzers", *Energy Procedia,* 142 pp 1468-1473, 2017.

[6] Burton, N. A., R. V. Padilla, A. Rose and H. Habibullah., "Increasing the efficiency of hydrogen production from solar powered water electrolysis", *Renewable and Sustainable Energy Reviews*, 135, 2021.

[7] Sánchez, M., E. Amores, L. Rodríguez and C. Clemente-Jul., "Semi-empirical model and experimental validation for the performance evaluation of a 15kW alkaline water electrolyzer", *International Journal of Hydrogen Energy,* 43 pp 20332-20345, 2018.

[8] Shiva Kumar, S. and V. Himabindu, "Hydrogen production by PEM water electrolysis – A review", *Materials Science for Energy Technologies,* 2 pp 442-454, 2019.

[9] Buttler, A. and H. Spliethoff., "Current status of water electrolysis for energy storage, grid balancing and sector coupling via power-to-gas and power-to-liquids: A review", *Renewable and Sustainable Energy Reviews,* 82 pp 2440-2454, 2018.

[10] Carmo, M., D. L. Fritz, J. Mergel and D. Stolten., "A comprehensive review on PEM water electrolysis", *International Journal of Hydrogen Energy,* 38 pp 4901-4934, 2013.

[11] Marangio, F., M. Santarelli and M. Cali, "Theoretical model and experimental analysis of a high pressure PEM water electrolyser for hydrogen production", *International Journal of Hydrogen Energy,* 34 pp 1143-1158., 2009.

[12] Slade, S., Campbell, S. A., Ralph, T. R. & Walsh, F. C., "Ionic Conductivity of an Extruded Nafion 1100 EW Series of Membranes", *Journal of The Electrochemical Society*, 149 pp A1556-A1564, 2002.

[13] Barbir, F., "PEM electrolysis for production of hydrogen from renewable energy sources", *Solar Energy,* 78 pp 661-669, 2005.

[14] Gorgun, H., "Dynamic modelling of a proton exchange membrane (PEM) electrolyzer", *International Journal of Hydrogen Energy,* 31 pp 29-38, 2006.

[15] Maeda, T., Ito. H, Akai. M, Hasegawa. Y, Hasegawa. R and Hada. Y., "Control method and basic characteristics of photovoltaic PEM electrolyzer system", *Proc Renewable Energy Chiba*, pp 22, 2006.

[16] Sánchez, M., E. Amores, D. Abad, L. Rodríguez and C. Clemente-Ju., "Aspen Plus model of an alkaline electrolysis system for hydrogen production", *International Journal of Hydrogen Energy,* 45 pp 3916-3929, 2020.