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Research Article

Experimental and Analytical Study of a Proton Exchange Membrane Electrolyser Integrated with Thermal Energy Storage for Performance Enhancement

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To peak carbon dioxide emissions and carbon neutrality, hydrogen energy plays a pivotal role in the energy system dominated by wind power and solar power. The proton exchange membrane (PEM) electrolytic hydrogen production technology has advantages of higher current density, higher hydrogen purity, higher load flexibility, and balanced grid load, becoming one of effective ways to consume renewable energy. Experimental analysis finds that the present PEM electrolyser cannot maintain a stable operating temperature as the input power changes; the polarization curve would distort with the change of temperature. This work proposes a PEM electrolyser coupled with the thermal energy storage device to meet power fluctuation and frequent start and stop caused by renewable resources. Through the involvement of the thermal storage device, electrolytic system is able to operate quickly and persistently in an efficient condition. The coupled system effectively reduces energy consumption in the process of start-stop or load changing, which can effectively adapt to the power fluctuation and frequent start and stop caused by renewable energy.

1. Introduction

The randomness and fluctuation of renewable energy are bringing great challenges to power network security nowadays. How to economically and efficiently use wind or solar power through energy storage technology has become a challenging topic in the global energy field [1]. Hydrogen has attracted widespread attention that it will play an important role in the future as a secondary energy carrier in addition to electricity. Hydrogen energy has the characteristics of large-scale reserves, environmentally friendly, and renewable, which meets the requirements of sustainable development of environment [2, 3]. The electrolysis system converts excess electricity into hydrogen, which can be reused as fuel or converted back into electricity by fuel cells when electric power is needed [4].

The principle technologies of hydrogen production by electrolysis focus on alkaline and proton exchange membrane (PEM). Troostwijk and Diemann first discovered electrolysis in 1789 [5]; so far, Alkaline Electrolysers (AWE) are long established in industry but involve hazardous chemicals and alkaline impurities, and purification is needed before use [6, 7].

In 1966, General Motors developed the first electrolytic cell based on the Solid Polymer Electrolyte (SPE) concept, overcoming the disadvantages of alkaline corrosion and pollution of alkaline electrolytic cells. Solid polymer membranes, also known as proton exchange membranes, provide high electrical conductivity, allowing compact design and high-pressure operation [8]. Figure 1 shows a cross-section of a PEM water electrolyser [9]. PEM cell is compact, mainly composed of anode and cathode plates, diffusion layers,

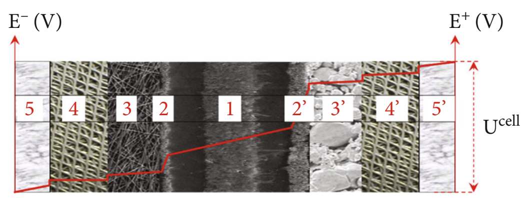

Figure 1: Schematic representation of a PEM water electrolysis cell: 1: proton exchange membrane, 2/2′: catalytic layer, 3/3′: porous transport layer, 4/4′: channel, and 5/5′: plate [9].

catalytic layers, and proton exchange membrane. The catalyst layer can be applied directly to the film or to the porous transport layer (3-3′) to equalize the current distribution. Liquid water is pumped through anodic channels to provide feedstock for the reaction and to expel heat generated during the reaction.

The thickness of proton exchange membrane is around 5~7 mm, which is beneficial to proton conductivity, allowing higher current densities to be achieved [10]. Due to low gas crossover rate of the polymer electrolyte membrane, PEM electrolyser is able to work under a wide range of power density (5%~120%) [11]. The PEM electrolytic hydrogen production technology has advantages, such as higher current density (generally 2~3 A/cm^2), higher hydrogen purity (up to 99.999%), higher load flexibility (5%~120%), and balanced grid load [12].

Significant efforts have been made to improve the performance of the PEM electrolyser by modeling works to discuss the hydrogen crossover through the membrane and difference, such as temperature, mass transfer coefficient, hydrogen concentration in oxygen, and liquid/gas diffusion layer (LGDL) compression and operating pressure [12]. Babar et al. [13] developed an equivalent electrical model for the PEM electrolyser, an input current–voltage (I–V) characteristic for a single PEM electrolyser cell was modeled through a series of experiment, and it has been observed that the electrolysis energy efficiency was in the range of 65–68% at steady-state conditions. Atlam et al. [14], through a single-channel-based three-dimensional CFD model, studied the influence of the key performance parameter of a PEM water electrolyser. The CFD model successfully predicted the current–voltage polarization curve. Carmo et al. [9], Han et al. [15], and Deshmukh and Boehm [16] used similar mathematical models to analyse the effect of current density, temperature, pressure, and membrane properties on the performance of the PEM electrolyser. Their modeling results can help to improve electrolysis system performance. In addition, Ma et al. [17] illustrated temperature distribution along the membrane-electrode assembly under different voltages through the simulation of the internal structure of the electrolytic cell. Three-dimensional (3D) models were also used by Upadhyay et al. [18] and Xu et al. [19]. It is found by Upadhyay et al. that the stack temperature is one of the most influencing parameters and a higher temperature (313 K to 353 K) is optimal for the PEM electrolyser [18]. Through studying the detailed distribution of two phases in anode flow channel, it is found by Xu et al. that the cell performance is improved by 0.171 V at 3 A cm^{-2} by replacing the traditional parallel flow with the new flow field [19].

Another part of the research is based on a combination of experiments and simulations. The thermal effects in the development of electrolyser models were also considered. The common method is to improve or verify the accuracy of the simulation model through experimental test results or empirical formulas [20]. Kim et al. [21] proved the effect of chemical reaction, chemical component thermodynamics, external ambient temperature, and Joule effect on the PEM electrolyser through experimental measurements. The results presented that the dynamic temperature impacted for both current and voltage. Aouali et al. [22] compared modeling results and lab-scale experimental data, and an acceptable mismatch of temperature dynamic performance was found in the PEM electrolyser system.

Water quality is another one of the influencing parameters to the performance of PEM electrolysis. Li et al. [23] investigated long-term Fe^{3+} ion contamination effects on the performance of single PEM cell. Though measuring membrane thickness and fluoride count on the cathode side, it is proved that membrane was attacked by radicals formed from hydrogen peroxide due to the existence of Fe^{3+} ions. The contamination effects of many other cations were also investigated by many researchers, such as Ca^{2+} [24, 25], Na^{+} [24, 26], and Cu^{2+} [27].

Drawing on the research history of the PEM electrolyser, there are few studies on power fluctuation and frequent start-stop of electrolytic cell system. This work will discuss the impact of power changes on the PEM electrolyser system and proposed a coupled PEM electrolyser with an energy storage device. Through the combination of solar heat collection device and thermal energy storage device, heat can be used in electrolysis process and keep its efficient operation when input power is fluctuant.

2. System Description

It is found that the PEM electrolyser cannot be maintained in the high-efficiency operating range, when renewable energy is connected to the hydrogen production system, resulting in the increase of overvoltage and instantaneous energy consumption (see details in Section 4.1). A thermal energy storage device can maintain the operating temperature of the PEM electrolyser and drives system in a higher performance. Figure 2 shows the layout of a PEM electrolyser system integrated with thermal energy storage device. The novel system can operate in two modes, namely, normal mode and power fluctuation mode, respectively.

The heat storage is temporarily out of work at normal mode. It should be noted that purified water is required to fill the water loop before the system can be started. When the electrolysis process begins, purified water is continuously pumped through the heat exchanger to the electrolytic cell; in the process of electrolysis, hydrous oxygen is produced at the anode and hydrous hydrogen at the cathode. Hydrogen is stored in a high-pressure tank after separation and purification. The water returns to the relay water tank from

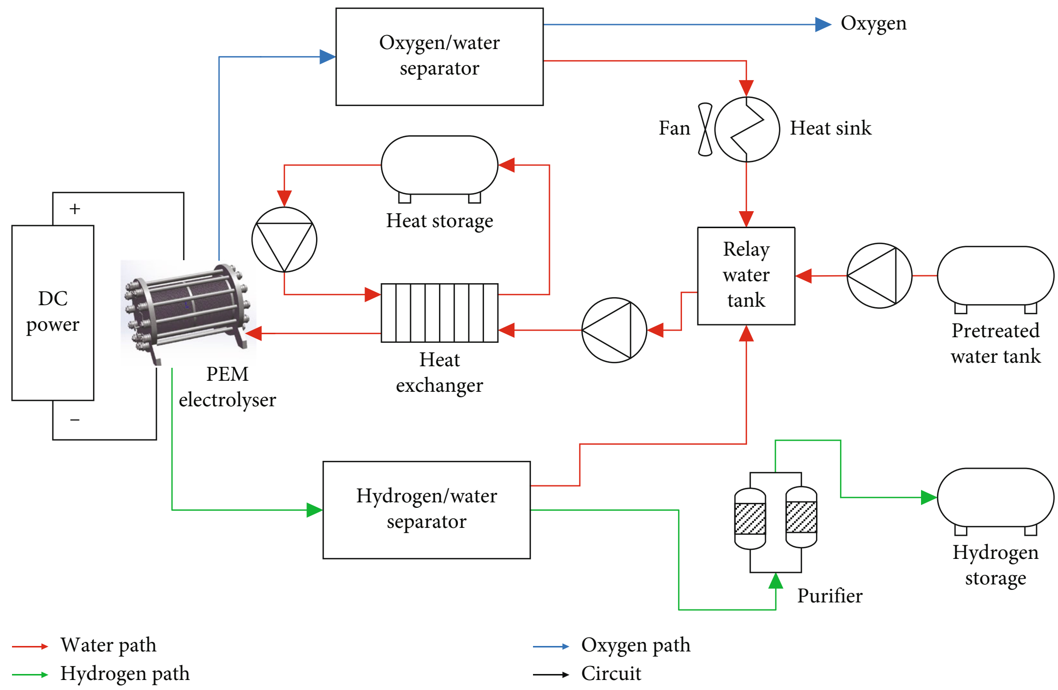

Figure 2: Schematic diagram of the PEM electrolyser system coupled with energy storage device.

the bottom of the separator after oxygen separation, and the oxygen is collected or exhausted from the upper part of the separator.

Power fluctuation mode launches when power fluctuates or system starts. The medium (water) in the energy storage device transfers the heat to the purified water, forcing its temperature to rise rapidly to reach the optimal temperature of the electrolytic cell. Two points should be clarified; one is that pretreated water is pumped to the relay water tank only when its level is low, thus replenishing the PEM electrolyser with the water consumed during the reaction; the second is that the fan is launched when the water is close to the temperature barrier. Due to the characteristics of the catalyst and membrane, the operating temperature of the PEM electrolyser is commonly lower than 90°C, although there are differences of operating temperature in these studies [5, 9, 12].

3. Establishment of the Coupled PEM Electrolyser

According to the coupled PEM electrolyser system selected in this work, the corresponding PEM electrolyser system (hydrogen production test bench) is established, mainly including the PEM electrolyser, water separation subsystem, purification subsystem, heat storage subsystem, pressurizing subsystem, and controlling and monitoring subsystem, as shown in Figure 3. The rated power of the PEM electrolyser system is about 60 kW, affording 3 stacks operating synchronously, and it should be noted that only 1 PEM electrolyser stack is used during the whole experiment. designed hydrogen yield 4 Nm^3/h, and purity is about 99.99%. To ensure the reliability of the experimental system, a throttle valve is used to reduce pressure at the outlet of the oxygen path. Concentration detection and alarm devices are also installed around the platform to ensure the test safety.

3.1. Separation Subsystem. The gas-water separator is used for separating liquid droplets from the gas when the removal rate of liquid impurities in the gas is very high. This subsystem consists of hydrogen separators and oxygen separators, and their structure and function are similar. Hydrogen and oxygen flow out from cathode and anode, respectively, with a certain proportion of liquid water, and enter the primary separator for gas-liquid separation. The function of the separator is to make the gas change direction suddenly in the flow, separating water droplets from the gas. The water droplets gather at the bottom of the separator and flows back to relay water tank. The function of the secondary separator is to further remove water in hydrogen or oxygen after the primary separator; thus, it has the function of gas-liquid separation and cooling and dehumidification. The design parameters of primary and secondary hydrogen separators are listed in Table 1, and Figure 4 shows the real pictures of primary and secondary hydrogen separators and primary and secondary oxygen separators, from left to right, respectively.

3.2. Purification Subsystem. The hydrogen produced by the PEM electrolyser removed almost all liquid water after the separation processes, and the purification subsystem is used to further remove gaseous water from hydrogen to ensure the purity. The hydrogen purification subsystem consists of two adsorption towers in parallel. One adsorption tower is in an adsorption state, and the other is in a regeneration state. The adsorbent is a combination of alumina and molecular sieve, the upper layer of adsorption tower uses molecular sieve, and the lower part uses activated alumina. The

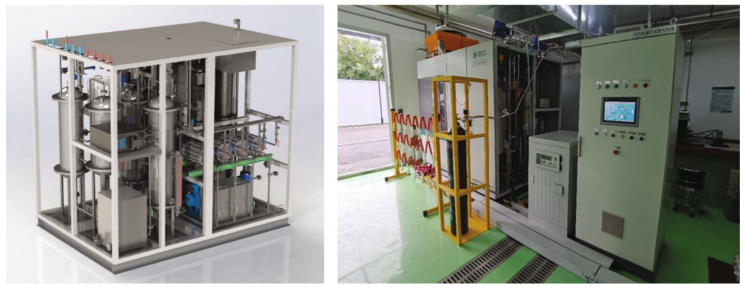

Figure 3: 60 kW PEM electrolyser test bench internal 3D model drawing and actual external view.

Table 1: Main parameters of primary and secondary separators.

Name	Value			
	Primary hydrogen separator	Secondary hydrogen separator	Primary oxygen separator	Secondary oxygen separator
Pressure (MPa)	0.1-3.5	0.1-3.5	0.1-3.5	0.1-3.5
Temperature (°C)	65	20	65	20
Design separation rate (%)	90	90	90	90
Size (mm)	$\Phi 300 \times 1800$	$\Phi 300 \times 1600$	$\Phi 300 \times 1800$	$\Phi 300 \times 1600$

Figure 4: Primary and secondary separators.

rated parameters of purification subsystem are listed in Table 2.

3.3. Heat Storage Subsystem. Heat storage subsystem is used to simulate the process that solar energy or other renewable energy participates in the performance improvement of the PEM electrolyser. Solar or other renewable energy is stored as heat in a subsystem, and the stored heat, through a plate heat exchanger, can be used for PEM electrolysis when input power is fluctuant. The design parameters of circulating water pump are listed in Table 3, and Figure 5 shows the real pictures of the circulating water pump and plate heat exchanger.

3.4. Pressurizing Subsystem. The circulating water pump is a pressurization equipment, through the impeller rotating to form the pressure difference between the inlet and the outlet. It is used to overcome the resistance loss of circulating water system, leading to water continuously flowing in the loop. In a PEM electrolyser system, the circulating water pump is one of the key equipment to steadily provide reaction water,

Table 2: Main parameters of the purification subsystem.

Name	Value
Maximum flow (Nm^3/h)	20
Outlet purify (%)	≥99.99
Pressure range (MPa)	3-3.5
Adsorbent	Alumina, molecular sieve

Table 3: Main parameters of the heat storage subsystem.

Name	Value
Heat transfer medium	Water
Storage temperature (°C)	80-90
Pressure range (MPa)	0.1-0.5
Outlet flow (L/min)	20-40

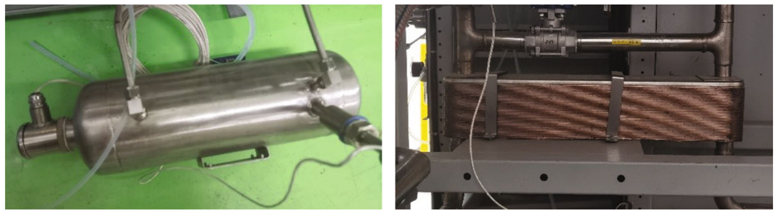

Figure 5: Heat storage and plate heat exchanger.

Table 4: Main parameters of the circulating pump.

Name	Value
Flow range (L/min)	10-50
Temperature range (°C)	5-90
Pressure range (MPa)	0.1-3.5
Lift (m)	10
Motor type	Variable-frequency
Rated power (kW)	1.5

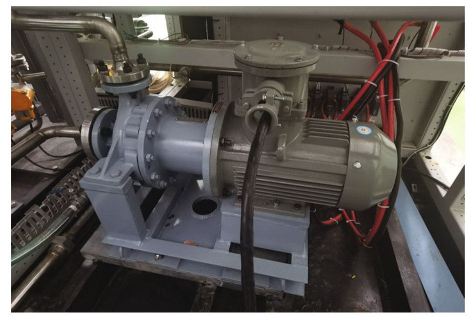

Figure 6: Circulating water pump.

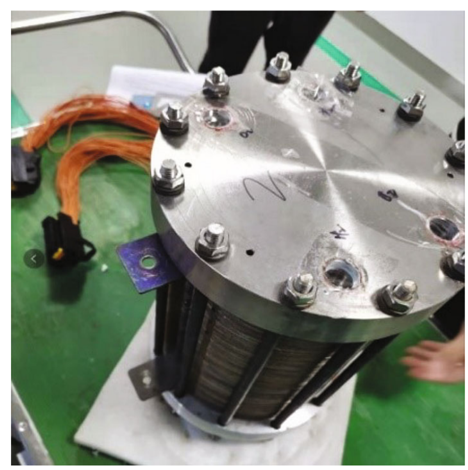

Figure 7: The PEM electrolyser used in the experiment.

meanwhile, and circulating water can also take heat out of the PEM electrolyser, which is mainly generated by ohmic overpotential [5]. Because the working pressure range of electrolytic cell is 0.1-3.2 MPa, the selected circulating water pump must withstand pressure. The design parameters of the circulating water pump are listed in Table 4, and Figure 6 shows the real pictures of the circulating water pump.

60 kW PEM electrolyser system is mainly used for performance testing of electrochemical reactor polarization curve, electrochemical impedance, electrochemical adsorption area, and hydrogen current density and can monitor the current and voltage of the PEM electrolyser in real time and monitor hydrogen concentration and oxygen concentration to prevent safety problems.

4. Results and Discussions

The testing process of the PEM electrolyser system was divided into two parts: PEM electrolytic process and thermal energy-involved electrolytic process. This section analysed the changes of parameters such as operating temperature of substances at the entrance of the PEM electrolyser during these two processes. Meanwhile, the relevant polarization curves were measured during the two processes. Figure 7 shows the PEM electrolyser used in the experiment, and the details of membrane electrode assembly (MEA) used in the test are shown in Table 5.

4.1. PEM Electrolytic Process. Preparatory works, such as pipeline cleaning or water circulation flowing, should be finished before launching the PEM electrolyser, and this preparatory time was not accounted into the whole test. It should be noted that the PEM electrolyser runs in constant current mode, and the power supply adjusted the voltage continuously according to the characteristics of the PEM electrolytic cell, so that the current was approximately fixed. The water flow into the electrolyser was maintained at 200 mL/min per cell. Figures 8 and 9 present the operation process of the test which almost lasts 55 minutes. The current density increased by around 0.2 A/cm^2 each time as the voltage gradually stabilizes until it reached to about 0.715 A/cm^2; the electrolyser continued to operate for 1600 s. During the process of 1600 s operation, load variation tests were done twice at 1800 s and 2250 s, respectively. Then, the current density was gradually increased to the peak value of 0.881 A/cm^2, and after continuous operation for 200 s, the current density was gradually reduced to 0 to complete the whole test.

It can be found that there was no remarkable increase in the water outlet temperature of the cell during the early stage of the test (from 0 to 500 s); obviously, 200 mL/min water flow was excessive for the electrolytic cell at this current density. When the current density increased to a relatively high value, the outlet water temperature gradually rises. In

Table 5: Details of membrane electrode assembly.

Name	Type/value
Proton exchange membrane	Nafion 115
Active area of MEA	160 cm^2
Cathode	Carbon paper
Anode	Titanium fiber felt
Pressure range	0.1-3.2 MPa
Water flow	200 mL/min

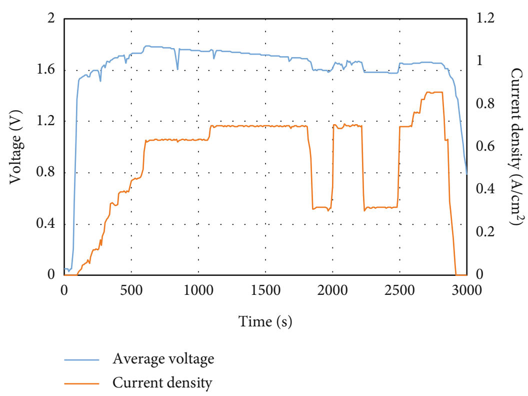

Figure 8: Trends of average voltage and current density of the PEM electrolyser.

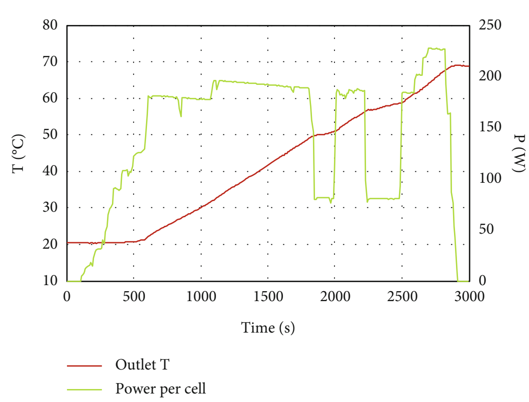

Figure 9: Trends of outlet water temperature and power of the PEM electrolyser.

constant current mode, the voltage dropped slightly as the temperature increases, which was mainly because the activation overpotential and ohmic overpotential presented a significant drop as the temperature increases [5–7], as shown in Figure 10. However, due to the characteristics of the catalyst and membrane, the PEM electrolyser cannot operate persistently without a temperature barrier [5, 10]. It can be concluded that higher temperature is beneficial to reduce the energy consumption of electrolytic cell as long as it is within the temperature limit.

In order to suit the randomness and fluctuation of wind power and solar power, hydrogen production system had to run under variable power for a long time, and its start-stop

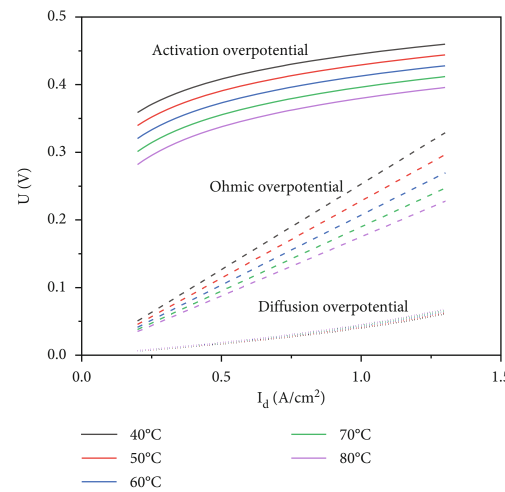

Figure 10: Effect of current density and temperature on activation overpotential, ohmic overpotential, and diffusion overpotential.

frequency is often more than that of hydrogen production system connected to the power grid. Meanwhile, intermittent operation may lead to changes in operating temperature that affect the efficiency of the PEM electrolyser. Figure 11 illustrates the effect of operating temperature on the voltage of the PEM cell, the temperature changes from 20°C to over 60°C in the process of the current density rising from small to large, and the performance improvement was caused by the higher temperature of large current density. It is found that the electrolytic cell cannot maintain a stable operating temperature as the input power changes; the polarization curve would distort with the change of temperature. Therefore, with renewable energy linked to hydrogen production systems, the PEM electrolyser cannot be maintained in the high-efficiency operating range, further resulting in the increase of overvoltage and instantaneous energy consumption. Ignoring errors between simulation and experimental results, dashed lines in Figure 11 clarify the polarization curve of electrolytic cell at constant temperature. In the case of input power fluctuations or continuous start-stop for several times, a renewable energy storage device can maintain the operating temperature of the PEM electrolyser and may keep the system at a higher efficiency.

4.2. Thermal Energy-Involved Electrolytic Process. Before the thermal energy-involved experiment, energy storage device stored solar or other renewable energy in the form of heat in a medium (using purified water in this process) beforehand. The initial temperature of the energy storage device was set at around 85°C. Figures 12 and 13 described the thermal energy-involved electrolytic process. The circulating water temperature reached ~58°C in 30 seconds due to the heat received from the heat storage device. As the current density increases (around 0.63 A/cm^2 in ~380 seconds), the heat generated by the electrolytic cell can maintain the operating temperature, and the heat storage device stops heating. Meanwhile, in order to prevent the electrolytic cell overheating, cooling fan starts to cool down the circulating water.

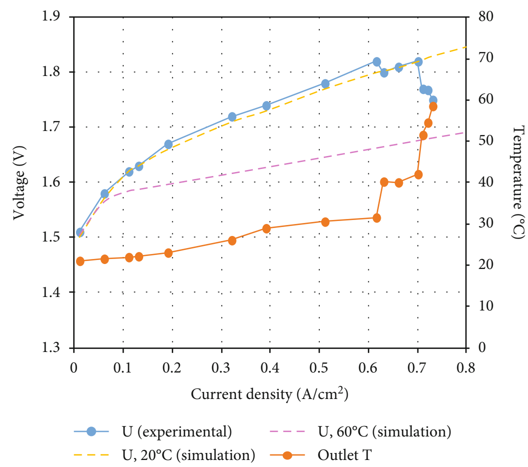

FIGURE 11: Polarization curves and the effect of operating temperature on the voltage of the PEM cell.

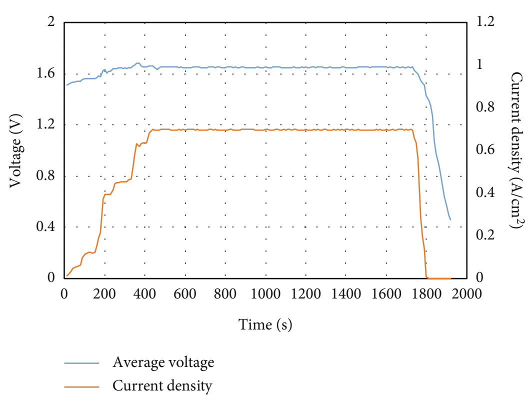

FIGURE 12: Trends of average voltage and current density of the PEM electrolyser with heat storage involved.

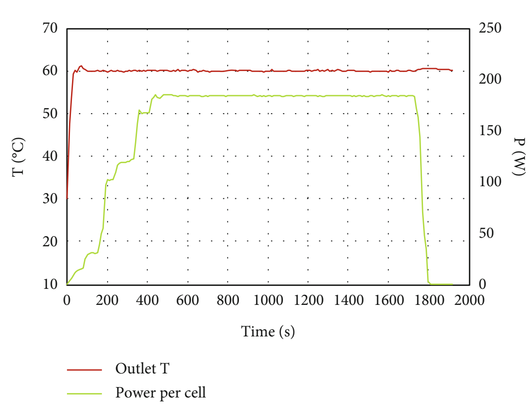

FIGURE 13: Trends of outlet water temperature and power of the PEM electrolyser with heat storage involved.

The power of the cell was 185 W, and the voltage was about 1.67 V under stable operation condition.

The performance improvement of the PEM electrolyser with heat storage involved is found in Figure 14. With the participation of heat storage device, the polarization curve

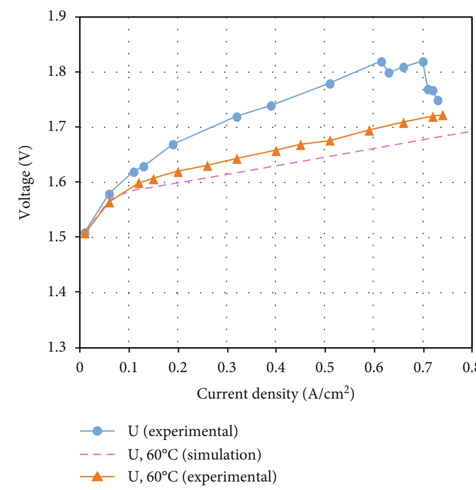

FIGURE 14: Polarization curves of the PEM cell with thermal storage involved.

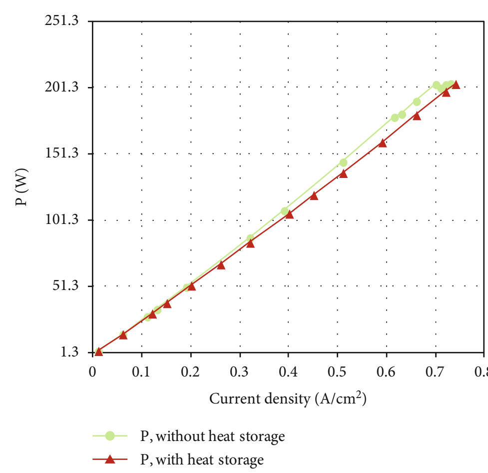

FIGURE 15: Power consumption comparison with or without energy storage device for the single PEM cell.

of the PEM electrolytic cell showed a similar trend to the simulation results; their errors were less than 3%. The maximum difference of power consumption per cell can reach ~12 W, as shown in Figure 15. The PEM electrolyser installed with heat storage presented a higher current density (more charge transfer) under the same power consumption, which means a higher hydrogen yield (maximum increment of 5.04%). It is concluded that the PEM hydrogen production system coupled with the thermal energy storage device can effectively adapt to the power fluctuation and frequent start and stop caused by renewable energy, so that the hydrogen production system operated in a higher performance range.

5. Conclusions

In this work, a PEM hydrogen production system coupled with the thermal storage device was proposed to meet power fluctuation and frequent start and stop caused by renewable resources. It is found that the present PEM electrolyser cannot maintain an approximately fixed temperature as the input power changes; the polarization curve would distort with the change of temperature. By optimizing the structure of the PEM system, the inlet water temperature of electrolyser reached ~58°C in 30 seconds and promoted the electrolytic system to run quickly and persistently in an efficient condition. Meanwhile, the coupled system can effectively reduce electrolytic voltages during the process of start-stop or load changing, and the maximum difference of power consumption per cell can reach ~12 W. Therefore, the PEM hydrogen production system coupled with the thermal storage device can effectively adapt to the power fluctuation and frequent start and stop caused by renewable energy.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

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