



# Effects of operating conditions on performance of high-temperature polymer electrolyte water electrolyzer



Hua Li<sup>a</sup>, Akiko Inada<sup>a</sup>, Tsuyohiko Fujigaya<sup>a,b</sup>, Hironori Nakajima<sup>a,b</sup>, Kazunari Sasaki<sup>a,b</sup>, Kohei Ito<sup>a,b,\*</sup>

<sup>a</sup> Graduate School of Engineering, Kyushu University, Motoooka 744, Nishi-ku, Fukuoka 819-0395, Japan

<sup>b</sup> WPI-F2CNER, Kyushu University, Motoooka 744, Nishi-ku, Fukuoka 819-0395, Japan

## HIGHLIGHTS

- Characterization was conducted for a PEWE in a wide temperature and pressure region.
- A large overvoltage at elevated temperature is attributed to concentration overvoltage.
- An increase in operating pressure is a solution for reducing the concentration overvoltage.
- Maintaining water in the liquid phase is critical in operating high-temperature PEWEs.

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## ABSTRACT

Effects of operating conditions of a high-temperature polymer electrolyte water electrolyzer (HT-PEWE) on the electrolysis voltage are evaluated, and the optimal conditions for a high performance are revealed. A HT-PEWE unit cell with a 4-cm<sup>2</sup> electrode consisting of Nafion117-based catalyst-coated membrane with IrO<sub>2</sub> and Pt/C as the oxygen and hydrogen evolution catalysts is fabricated, and its electrolysis voltage and high-frequency resistance are assessed. The cell temperature and pressure are controlled at 80–130 °C and 0.1–0.5 MPa, respectively. It is observed that increasing the temperature at a constant pressure of 0.1 MPa does not increase the ohmic overvoltage of the cell; however, it does increase the concentration overvoltage. It is also found that the increase in the overvoltage resulting from the rise in the temperature can be suppressed by elevating the pressure. When operating the cell at a temperature of 100 °C, pressure greater than 0.1 MPa suppresses the overvoltage, and so does pressures greater than 0.3 MPa at 130 °C. This behavior suggests that keeping the water in a liquid water phase by increasing the pressure is critical for operating PEWEs at high temperatures.

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## 1. Introduction

Global oil production is likely to reach its peak in the foreseeable future, and soon the world may suffer an oil shortage [1–3]. Therefore, renewable energy sources are attracting significant attention, as they may help to solve pressing environmental and energy issues; and lead to a society where the renewable energy sources are the primary sources of electricity. However, the production of the renewable energy depends on time, and the sources are spread heterogeneously. This results in a mismatch between the energy supply and demand.

Hydrogen produced using a water electrolyzer powered by renewable energy [4] is an energy carrier that can help to overcome the supply-demand mismatch issue. Among the various types of electrolyzers being investigated, polymer electrolyte water electrolyzers (PEWEs) are probably good candidates for this role, as they can produce hydrogen at a low power consumption (e.g., 4.5 kWh/(Nm<sup>3</sup>)) at 80 °C and 0.3 MPa [5].

Although PEWEs have already been commercialized, further improvements in their performance are promising. In order to raise the performance of PEWEs, the catalyst layer must be improved. To achieve this goal, new catalyst structures and materials [6–8] have been proposed. In addition, attempts are being made to optimize the hydrophobicity and the pore size distribution of the current collectors [9–13]. Moreover, the impact of the flow behaviors of the

\* Corresponding author. Graduate school of Engineering, Kyushu University, Motoooka 744, Nishi-ku, Fukuoka 819-0395, Japan.

E-mail address: [kohei@mech.kyushu-u.ac.jp](mailto:kohei@mech.kyushu-u.ac.jp) (K. Ito).

vapor and liquid phases on the electrolysis performance is being investigated [14–16]. Simultaneously, some researchers pay attention on the electrochemical stability [17–23]. Furthermore, it has been proposed that increasing the operating temperature should improve the performance of PEWEs. Recent studies have suggested that the operating temperatures of 80–100 °C increase the oxygen reaction rate [24] as well as the exchange current density [25], leading to a decrease in the power consumption from 4.5 to 4.2 kWh/(Nm<sup>3</sup>) at 100 °C and 0.3 MPa [5].

It has also been suggested that the polymer electrolyte membrane dehydrates during the high-temperature operation, reducing the ionic conductivity of the membrane as well as the performance of the PEWE [26]. Higher temperatures may increase the saturation pressure and reduce the relative humidity of the environment around the membrane, causing its water content and ionic conductivity to decrease. The dehydration of the membrane and the resulting decrease in performance have motivated researchers [5,27,28] to develop composite membranes, based on the Nafion series of membranes with new catalyst structures. These composite membranes consist of a Nafion membrane, to which a hydrophilic material such as TiO<sub>2</sub> has been added. Composite membranes can maintain their ionic conductivity even at high temperatures, allowing PEWEs to exhibit higher performances than those operated at normal temperatures.

This study was also motivated by the aforementioned poor performance of PEWEs at high temperatures due to the dehydration of the membrane. Thereby, PEWE cell was electrochemically characterized under a wide range of temperature and pressure conditions. In conclusion, a strategy for improving the performance of PEWEs at high temperatures is suggested.

## 2. Experimental method

### 2.1. Functioning of a PEWE

Fig. 1(a) shows the set up used to evaluate the performance of the PEWE cell. The piping for both the cathode and the anode passes through the cell, a pressure-reducing valve, a dehydrator, and a mass flow meter. A water pump is built into the anode piping. The hydrogen gas produced by electrolysis exits through the back pressure valve and water is removed from the gas at the dehydrator. The hydrogen and oxygen gases boost the pressure in the anode and cathode, respectively, while the back pressure valve controls the pressure. During water electrolysis, the ohmic resistance is measured using a high frequency resistance (HFR) meter (Frequency: 10 kHz; Model: 356E; TSURUGA ELECTRIC CORPORATION, JAPAN). The flow rates of the hydrogen and oxygen gases are also measured per second using a flow meter located in the downstream of the dehydrator in order to confirm the current efficiency. Due to the fluctuations, the hydrogen and oxygen flow rates are determined by averaging the data populations recorded for 10 mins.

Though the existence of preheater will be very helpful for controlling the water temperature at inlet and inside of the cell, deionized water was fed into cell directly without preheated. Specific points in our experiment may clarify this concern. Relatively small flow rates of water are chosen in our experiment. Under 1 A/cm<sup>2</sup> condition, the water utilization defined in dividing electrolyzed water by fed water is 2% and 20% for water feeding of 1.0 and 0.1 mL/min, respectively. This water utilization is much larger than that in conventional operation which is no more than 0.5% [17–20,22,23], and suggests that water flow rate in our experiment is quite small. Moreover, the approach region running from cell inlet to flow channel is long, as shown in Fig. 1(b). Taking into the consideration of small amount of water feeding, the long approach

will give enough time to heat the fed water before the water reach the catalyst region, so that the temperature of fed water can be equilibrium to nominal temperature. The nominal temperature is obtained from thermocouple placed in flow field plate, whose temperature is stable in the experiment. These specific experimental conditions and cell configuration suggest that the nominal temperature is close to the temperature of water in the flow channel without preheating water at inlet.

### 2.2. Cell components

A Nafion117 membrane-based catalyst-coated membrane (CCM) was fabricated by spraying and hot pressing, as described in the following. As for the pretreatment, the Nafion membranes were treated in following processes: (1) The membrane is rinsed in the solutions composed of 5 wt% H<sub>2</sub>O<sub>2</sub> for 1 h at 80 °C, and (2) washed in de-ionized water for 1 h at room temperature, and (3) rinsed in 8 wt% H<sub>2</sub>SO<sub>4</sub> for 1 h at 80 °C, and finally (4) washed in de-ionized water for 1 h at room temperature. Then, a slurry consisting of 67-wt% catalyst (IrO<sub>2</sub> powder, type IV, Tokuriki Co., Japan) and 33-wt% Nafion ionomer (5 wt% Ion Power solution) in deionized water and ethanol was prepared and sprayed onto one face of the membrane to form the anode catalyst layer. Commercial 46% Pt/C (Tanaka Kikinzoku Japan) powder was used as the catalyst for the cathode catalyst layer. In this case, a slurry was also prepared by mixing the Nafion ionomer and catalyst in water in a catalyst/dry ionomer weight ratio of 72:28. This slurry was sprayed onto the other face of the membrane to fabricate the cathode catalyst layer. The sprayed slurry was fixed onto the membrane by hot pressing. The loading rate of IrO<sub>2</sub> was 1.5 mg/cm<sup>2</sup>, whereas that of Pt was 0.5 mg/cm<sup>2</sup>. The area of the catalyst layers for the anode and the cathode was 4 cm<sup>2</sup>.

The other components of the cell are listed in Table 1. An SUS316L mesh and a titanium mesh coated with platinum are used as the current collectors for the cathode and anode, respectively. Carbon and titanium plates were used as the cathode and anode flow field plates, respectively. Both flow field plates had serpentine channels. To form the PEWE cell, all the components were sandwiched between a pair of fastening plates and tightened them by 12 sets of bolt and nut at the torque of 4 Nm. This procedure and the area of flow field plate (64 cm<sup>2</sup>) make the tightening pressure to be about 3 MPa.

### 2.3. Operating conditions

The PEWE cell was operated at temperatures of 80–130 °C and pressures of 0.1–0.5 MPa. Deionized water at 20 °C was fed into the anode at flow rates of 0.1 mL/min and 1 mL/min; these flow rates correspond to the water utilization ratios (electrolyzed water/ fed water) of 20% and 2%, respectively, for at 1 A/cm<sup>2</sup>.

The water flow rates are lower than that used under normal operation for controlling the cell temperature and evaluating the performance of PEWEs at high temperatures. If the water flow rate was high, the cell temperature would either decrease or become unstable because of the sensible and latent heat of the fed water.

The characteristics of the probable flow pattern in the anode channel are listed in Table 2; the values were estimated based on the nominal temperatures and pressures between 80 and 130 °C and 0.1–0.5 MPa, respectively. The assumptions made for estimating the characteristics are as follows: (i) Faraday's electrolysis law is in effect; (ii) the current density is 0.5 A/cm<sup>2</sup> for determining the molar flow rate of oxygen gas, which is assumed to be an ideal gas; and (iii) the water in the channel could be described by the water phase equilibrium diagram at the nominal temperatures and pressures. With these considerations, the superficial velocities of

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