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Polarization characteristics of a low catalyst loading PEM water electrolyzer operating at elevated temperature

Byung-Seok Lee [a, b], Hee-Young Park [a], Insoo Choi [c], Min Kyung Cho [a, d], Hyoung-Juhn Kim [a], Sung Jong Yoo [a], Dirk Henkensmeier [a], Jin Young Kim [a, e], Suk Woo Nam [a, e], Sehkyu Park [f, ***], Kwan-Young Lee [b, e, **], Jong Hyun Jang [a, e, *]

[a] Fuel Cell Research Center, Korea Institute of Science and Technology (KIST), Seoul 02792, Republic of Korea
[b] Department of Chemical and Biological Engineering, Korea University, Seoul 02841, Republic of Korea
[c] Division of Energy Engineering, Kangwon National University, Gangwon-do 24341, Republic of Korea
[d] School of Chemical and Biological Engineering, Seoul National University, Seoul 08826, Republic of Korea
[e] Green School, Korea University, Seoul 02841, Republic of Korea
[f] Department of Chemical Engineering, Kwangwoon University, Seoul 01897, Republic of Korea

HIGHLIGHTS

- IrO_2 anodes with ultralow loading (0.1 mg cm^{-2}) were fabricated by electrodeposition.
- The effect of operating conditions on PEMWE performance was studied at $120\ °C$.
- The current density at 1.6 V was as high as 1.79 A cm^{-2} at $120\ °C$ and 2.5 bar.
- Water transport characteristics in a PEMWE were experimentally examined.

GRAPHICAL ABSTRACT
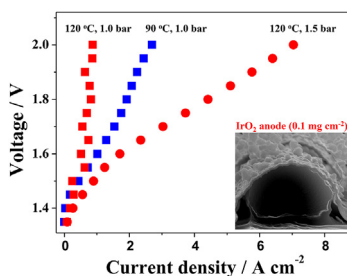
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ABSTRACT

The effect of temperature and pressure, and diffusion layer thickness is assessed on performance of a proton exchange membrane water electrolyzers (PEMWEs) with an ultralow iridium oxide (IrO_2) loading (0.1 mg cm^{-2}) anode prepared by electrodeposition and a Pt/C catalyzed cathode with a Pt loading of 0.4 mg cm^{-2}. Increasing pressure to 2.5 bar at $120\ °C$ enhances the water electrolysis current, so the anode electrodeposited with 0.1 mg cm^{-2} IrO_2 gives a current density of 1.79 A cm^{-2} at 1.6 V, which is comparable to the conventional powder-type IrO_2 electrode with 2.0 mg cm^{-2} at a temperature of $120\ °C$ and pressure of 2.5 bar. The major factors for cell performances are rationalized in terms of overpotentials, water flow rates and thickness of diffusion layers, based on polarization behavior and ac-impedance response.

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* Corresponding author. Fuel Cell Research Center, Korea Institute of Science and Technology (KIST), Seoul 02792, Republic of Korea.
** Corresponding author. Department of Chemical and Biological Engineering, Korea University, Seoul 02841, Republic of Korea.
*** Corresponding author. Department of Chemical Engineering, Kwangwoon University, Seoul 01897, Republic of Korea.
E-mail addresses: jhjang@kist.re.kr (J.H. Jang), kylee@korea.ac.kr (K.-Y. Lee), vitalspark@kw.ac.kr (S. Park).

1. Introduction

Burning fossil fuels that are in finite reserves globally triggers problems because of the emission of carbon dioxide, which is instrumental to the greenhouse effect, and other air pollutants such as sulfur and nitrogen oxides. These issues have stimulated interest in hydrogen as a clean and abundant energy source. To realize a hydrogen ecosystem, the development of an environment-friendly and continuous production of hydrogen by renewable sources [1–8], a method of safe and stable storage [9–14], and the smart utilization through generating electricity by highly efficient fuel cells [15–20] are necessary. Currently, however, the majority of hydrogen is produced by steam reforming (48%), partial oxidation (30%) and gasification of coal (18%) processes [21,22] that consume a variety of fossil fuels such as natural gas, oil, and coal. As an alternative and clean technology for hydrogen production, water electrolysis accounts for only 4% of the total hydrogen production because of the high capital and operating cost associated with the equipment and electricity consumed.

Among various water electrolysis technologies, the polymer electrolyte membrane (PEM) water electrolysis has advantages in high purity of produced H_2, pressurized operation, simple and compact system, and use of neutral water feed [23,24]. The alkaline water electrolysis that utilizes alkaline liquid electrolytes, such as KOH and NaOH, is more commercialized technology with lower system cost, but both current density and energy efficiency is lower. In the case of solid oxide electrolyzer cells (SOEC), high operating temperature (700–1000 °C) enables high efficiency compares to the PEM and alkaline water electrolysis, but there are various technical issues for commercialization. In a PEM water electrolysis system (Fig. 1), water molecules introduced to the anode are separated to generate gaseous oxygen and simultaneously release protons and electrons (oxygen evolution reaction, OER). Protons produced by OER migrate through the membrane and bind with electrons that travel along the external wire to produce hydrogen molecules at the cathode (hydrogen evolution reaction, HER). In PEM water electrolysis, electrochemical processes at the anode that cause oxygen evolution are a substantial portion of the overall overpotential because the OER kinetics are more sluggish than those of the HER [25–27].
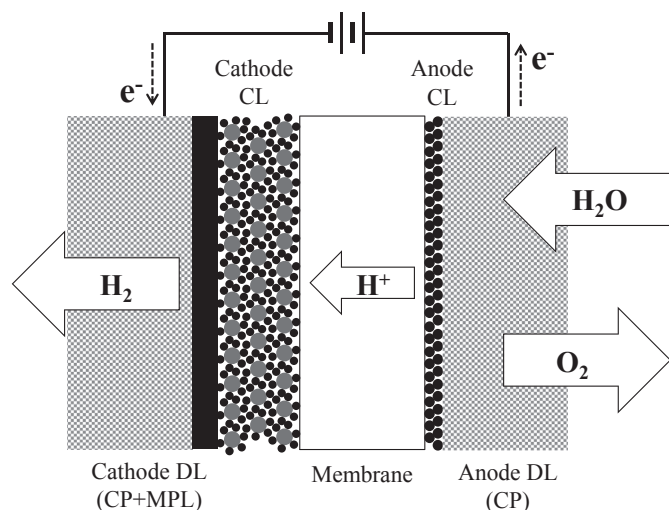
Typically, when an operating temperature of a PEM water electrolysis system increases, the reaction rate is faster and the kinetic overpotential decreases. Additionally, the thermodynamic potential for water electrolysis decreases as the temperature rises [28–30]. Therefore, elevating the operating temperature of a PEM water electrolysis system can improve the efficiency. However, solid electrolytes in PEM water electrolyzers (PEMWEs) are readily dehydrated above 100 °C, the boiling point at ambient pressure. Therefore, the proton conductivity sharply decreases [28,31–36]. To maintain proton conductivity in the membrane at high temperatures, previous investigations have developed (or have reinforced conventional membranes to produce) composite membranes [31,32,34], including a phosphoric acid doped membrane [35,36] and an Aquivion membrane [30]. However, the pressurization of a PEMWE is able to enhance the proton conductivity across the membrane at high temperatures because the water fed into the pressurized cell can be in the liquid phase above 100 °C (depending on the temperature and pressure applied to the system), resulting in a high water uptake in the membrane. This process is the identical principle employed in a pressure cooker to raise the boiling point of the water at high elevations for which the atmospheric pressure is relatively low.

The pressurization of PEMWEs running at a high temperature have been adopted by Antonucci [31] and Baglio [32], who measured the performance of a PEMWE by increasing the pressure at two different temperatures (80 °C and 120 °C). In that study, the PEMWE operating at 120 °C and 1.8 V displayed twice the current at 3 bar compared to that obtained at 1 bar. However, no improvement in the cell performance appeared with increasing pressure from 1 to 3 bar at 80 °C. Xu et al. [34] also studied the pressure effect in a PEMWE that contains a perfluorinated-silica composite membrane as an electrolyte and $Ru_{0.7}Ir_{0.3}O_2$ nanoparticles as an anode catalyst. They reported that the higher PEMWE performance with an increased pressure (i.e., 1.5–3 bar) is attributed to the condensation of water vapor throughout their membrane-electrode-assembly (MEA). The water vapor improved the proton transfer rate over the entire ionic pathway. However, the increase in the performance under pressure is not significantly higher; this is likely because of its relatively low operating temperature (i.e., 100 °C).

When the gaseous oxygen produced at the anode by OER is released towards the diffusion layer (DL), its stream toward the flow channel is unavoidably hindered by the counterflow of the water (the reactant). Furthermore, the liberation of oxygen from the anode can be severely limited under pressure, causing oxygen molecules to accumulate in and near the catalyst layer (CL) and subsequently preventing the water cluster from accessing the active sites. Therefore, the mass-transfer, including the liquid water and gaseous oxygen transport to and from the CL, considerably affects the performance of PEMWE, depending on the materials and porous structure of the DL as a diffuser and a current collector [37].

Assuming that no significant performance degradation appears with a decreased amount of noble metal in the anode CL, the approach to reduce catalyst loading is reasonable in that a relatively thick CL lengthens the proton transport and water diffusion path, thereby decreasing the utilization of the catalyst. Studies covering Ru and Ir-based catalyst loading in PEMWEs at high temperatures effectively decreased the amount of precious catalyst used at the anode in PEMWEs depending on a variety of operating parameters. However, the noble metal content in the anode CL has remained higher than 1.0 mg cm^{-2} [30–32,34–36,38].

Fig. 1. Schematic figure of the PEMWE single cell with an electrodeposited IrO_2 anode.