Water Electrolysis by the Direct Water Supply to the Solid Polymer Electrolyte through the Interdigitated Structure of the Electrode

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Water supply pass
Separator Catalyst
Hydrophobic gas diffusion layer
Gasacket
Proton conductive membrane
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ABSTRACT

Water electrolysis cell in which the product gases was separated from liquid water on the surface of the electrode was developed. In order to realize the separation between gas and water, interdigitated diffusion layer (GDL) was designed, and the surface of the GDL was covered by catalyst to form electrode. When the pressurized water was supplied, the water directly made a contact to the proton conductive membrane. Due to the hydrophobic surface condition of the GDLs, gas/water separation along the surface of the electrode was completed.

Key Words: Water electrolysis; Polymer electrolyte; Interdigitated gas diffusion layer; gas/water separation
1. Introduction

Today, lots of efforts have been made to the practical usage of the renewable energy. People are going to create energy carrier like methane and ammonium base on the hydrogen generated by the water electrolysis using solar power, windmill, and tidal power.\textsuperscript{1-7} Among the water electrolysis techniques, it is well known that the polymer electrolyte electrolysis cell (PEEC) is an important candidate for the practical applications, since it is operable under the moderate temperature less than 100 °C.

For the manned operation in space, water electrolysis is also very important to generate oxygen and hydrogen. The oxygen is an essential material for the human life support under the closed environment. The carbon dioxide which is produced by the manned operation can be transferred to water and methane by using the hydrogen. Since the recovered water can be used for the manned activities again, the sustainability of mankind in space is improved.

In the case of PEEC system, the gas/water separation is important function. The PEEC system has the water circulation line to supply water to the electrolysis cell. It is very important to realize the continuous operation of electrolysis, the bubble along the surface of the electrode should be removed quickly. The flow of water plays an important role to carry out the bobbles to the outside of the electrolysis cell. After separating gases from water, the water is resupplied to the electrolysis cell. Thus, the water reservoir with the function of gas/water separator should be fixed along the outlet line of the gases. Especially for the terrestrial applications, it is easy to separate gases from the water by the difference of density or weight.
Furthermore, for space applications people cannot expect the effect of gravity. The unique device using centrifuge force combined with water circulator should be applied to separate gas from water in order to make PEEC system functionable under the microgravity condition.

If the water circulator and gas/water separation unit are unnecessary, the simple and compact water electrolyzer can be realized for both terrestrial and space applications. We planned the electrode design which realizes the gas/water separation on the surface of the electrode, and the interdigitated structure of gas diffusion layer (GDL) covered with catalysts were applied as electrode. The pressurized water injected to the electrolysis cell directly touches to the proton conductive membrane. Then the water approaches to the electrode area through the membrane and the interfacial region between membrane and electrode. By using the GDL which has the micro porous surface condition, hydrophobic condition of the GDL makes a separation of gases from water on the surface of the electrode, and only the dry gases like hydrogen and oxygen are introduced to the outlet line.

In this paper, we discuss the PEEC cell design to generate hydrogen and oxygen without containing liquid water by applying the interdigitated configuration of electrode where the pressurized water without circulation was supplied.

2. Experimental

Figure 1 shows the concept design of the water electrolyzer which has the interdigitated configuration of electrodes. The carbon paper, SGL34BC of SGL Carbon co. ltd., was used as the gas diffusion layer
(GDL). On the GDL, the catalyst was applied to form electrode. The GDL has the micro porous structure with hydrophobic surface condition. Each electrode had the width of 5 mm, and total surface area inside the cell was 15 cm$^2$.

Water for the electrolysis was injected through the ‘water supply pass’ which located alternately in between electrodes. The 50 kPa pressurized water was injected to the electrolysis cell without circulation. The water directly touched to the proton conductive membrane, and then accessed to the electrodes. As the proton conductive membrane, NAFION 215® with the thickness of 50 μm was used.

The IrO$_2$ and Pt/C of Tanaka Kikinzoku Kogyo K.K. were used as the catalysts for the anode and cathode, respectively. The catalysts were mixed with DuPont DE520 as ionomer, and the mixture was applied on the surface of the NAFION 215® by the transfer method. The expected amount of catalyst was 1 mg/cm$^2$ for both cathode and anode. The electrodes and proton conductive membrane were sandwiched by the Pt-plated Ti separators without hot-pressing.

For the electrochemical measurements, the temperature of the cell was using the oven. Water was initially stored inside the metallic cylinder as reservoir and the water was pushed by the 50 kPa N$_2$ gas from the gas cylinder. The back pressure of the produced gases was not controlled and exhausted through the outlet port of the electrolysis cell.

The content of the outlet gases was measured by using the Agilent 490 Micro GC with the Pola Pack Q10. The Micro GC was directly connected to the outlet line of the gases from the electrolysis cell and inline measurement was conducted.
3. Results and Discussion

In the case of the conventional water electrolysis, metallic separator has the water flow channel. The back side of electrode consisting of GDL covered with catalyst first touches to the supplied water. The water passes through the GDL, and then access to the catalyst. When the bubble of hydrogen and oxygen is produced, it is carried out by the steam of water. The gaseous materials are then separated by gas/water separator so that people can use the outlet gases like hydrogen and oxygen for their own usage. Thus, the conventional water electrolyzer always consists of water electrolysis cell, water supply pump, and gas/water separator to make the continuous operation.

In the case of the cell shown in Fig. 1, electrodes with the interdigitated configuration locates inside the cell. This concept basically comes from the cell design by Matsumoto et al., in which only gases are able to be exhausted by preparing the hydrophobic electrodes.\(^8,9\) Especially in the case of the current design shown in Fig. 1, we targeted the scalability of the concept for the future realization of the large scale water electrolyzer.

The water was pressurized up to 50 kPa, and supplied to the electrolysis cell. The water accesses to the surface of the proton conductive material directly through the slits of the metallic separator. The water migrates along the intermediate area between membrane and electrodes, and then it is decomposed to hydrogen and oxygen. Since the GDL produced by SGL carbon has the hydrophobic surface condition, only gaseous materials can pass through the electrode.
The configuration for the cell operation is shown in Fig. 2. The pressure to inject water to the cell can be created by the gas cylinder. Only the pressure difference between the supplied water and the gas outlet port creates the driving force to release gases through the outlet port of the electrolysis cell.

If people want to use hydrogen to create methane or ammonium as the energy carrier, they must need the dry hydrogen to enhance the catalytic reaction. When we used the water electrolyzer in space, we originally needed the device to create centrifuge force for the separation of gases from water. The cell we invented here will realize the simple and compact system with low consumption of electricity.

Figure 3 shows the relationship between the applied current ($I$) and monitored terminal voltage ($V$) during the electrolysis. The cell was located inside the oven to raise the temperature to the appropriate condition. The temperature of the cell stack was controlled from 25 ℃ to 80 ℃.

As shown in Fig. 3, the electrolysis started at around 1.48 V which is known as the thermally neutral voltage for electrolysis at 25 ℃. With elevating temperature, the voltage at which the electrolysis started, decreased. Especially at 80 ℃, the electrolysis reaction started and maintained at less than 1.48 V up to 0.2 A/cm², which suggested that the endothermic operation of the PEEC should be possible with the current density less than 0.2 A/cm² at 80 ℃. In the case of the conventional water electrolyzer using circulation of water, the heat sometimes carried out of the cell quickly by the stream of water. In the case of the current electrolysis cell, the heat can be exchanged properly along the electrode. The endo-thermic electrolysis area was maintained up to the current density like 0.2 A/cm², which shows the enough supply of water was maintained up to this range of current density. Above 0.2 A/cm², the voltage for the electrolysis increased rapidly with the increasing current. In the case of the current cell design, the water...
migration along the horizontal direction dominate the electrolysis rate. The rapid increase of terminal voltage with increasing current density suggested the tendency of the lack of water for the reaction. Further improvement for the cell structure or the surface area of the interdigitated gas diffusion layer for this electrolysis cell will be needed. However, the terminal voltage was still 1.75 V when we applied the current density up to 1.0 A/cm$^2$. Since the terminal voltage was always between 1.6 and 1.8 V at 80 ℃ for the conventional water electrolysis using polymer electrolyte, the performance we obtained here was appropriate for the PEEC technique.$^{10}$

Table 1 shows the content rate of hydrogen and oxygen obtained through the outlet port of the electrolysis cell. The conventional water electrolyzer carries the gases with the stream of water to the outside of the cell. In contrast, the only driving force for our electrolysis cell to exhaust the gases is the pressure difference between the supplied water and the backside of electrodes. Since the shield of the generated gases is only the proton conductive membrane with the thickness of 50 μm, we should pay attention to the mixture of H$_2$ and O$_2$.

It was reported that the lower explosive limit of hydrogen in oxygen is 4.0 %, and the higher explosive limit is 94 %.\textsuperscript{11} The level of the content rate we observed here was enough low for the explosive limit, while we should make further efforts to improve it.

4. Conclusion

The water electrolysis cell using the interdigitated structure of the electrodes was designed. The pressurized water was supplied without circulation, and it directly approached to the proton conductive
membrane. Since we applied the carbon paper with micro porous condition, the electrode preformed as the gas/water separator. Thus, the hydrogen and oxygen were immediately separated from the liquid water on the surface of the electrodes.

By using the novel water electrolysis cell, we confirmed the proper water electrolysis between 25 °C and 80 °C. The content rate of gases measured through the outlet line of the cell showed the appropriate separation of hydrogen and oxygen with the level lower than the explosive condition.

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Figure captions

Fig. 1  The design concept for the present electrolysis (a), and the photograph of the two-cells-stack (b).

Fig. 2  The experiment condition for the electrolysis cell.

Fig. 4  I-V curves during the water electrolysis using interdigitated GDL structure.
Table 1  The content rat of outlet gases from the water electrolyzer.

| Temperature | Content rate of O$_2$ line | Content rate of H$_2$ line |
|-------------|-----------------------------|-----------------------------|
|             | O$_2$ content | H$_2$ content | O$_2$ content | H$_2$ content |
| 25$^\circ$C | 98.80 % | 0.63 % | 0.03 % | 98.60 % |
| 80$^\circ$C | 98.88 % | 0.76 % | 0.04 % | 99.62 % |
Fig. 1 Y. Sone et al.
Data recorder
DC power supply
Shunt resistance to measure current

Water reservoir
Electrolysis cell
Oven

Fig. 2 Y. Sone et al.
Fig. 3 Y. Sone et al.