Optimization of polymer electrolyte membrane water electrolysis nder high temperature condition

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（固体高分子形水電解セルの高温条件下での最適化）

区　　分　：甲

論　文　内　容　の　要　旨

Among a couple of water electrolyzers, Polymer Electrolyte Membrane Water Electrolyzer (PEMWEs) has large advantages on its high conversion efficiency, gas purity, and so on. These superior characteristics meet renewable energy society in the next generation. Hydrogen gas generated with PEMWE and electricity generation with fuel cells fueled by the produced hydrogen gas can compensate the gap between electricity demand and supply. However, PEMWE is rather expensive due to the expensive components, such as catalyst materials, and thus commercialization of PEMWE has not been spread.

High temperature operation is a possible solution to reduce the cost of PEMWE. Increasing temperature generally results in a decrease of activation overpotentials, especially in oxygen evolution reaction process. Less overpotential introduced by raising temperature enables to operate PEMWE under a high current density condition with smaller amount of catalyst, resulting in less cost. However, as a trade off, the high temperature operation tends to cause dehydration of polymer electrolyte membrane (PEM), and raise in ionic resistance (ohmic resistance), resulting in decreasing the performance of PEMWE.

This study elucidates overpotential in PEMWE under high temperature condition with changing operation parameters and embedded components, and challenge to improve electrolysis performance. In addition to the ohmic overpotential directly caused by the PEM dehydration, the other overpotential, such as activation and concentration overpotential, is carefully addressed with electrochemical characterization. Changing operation parameters, such as pressure and flow rate of supplied water, has effect on the water behavior and overpotential, and adequate choice of parameters to suppress the overpotential is explored. Also, different structures of current collector and flow field pattern, which is expected to change the water behavior, are embedded into a PEMWE cell, and try to minimize electrolysis voltage and to maximize electrolysis performance.

In chapter 1 and 2, the researches related to PEMWE are reviewed, and experimental methodology is explained in detail.

In chapter 3, the effect of operation parameters on electrolysis performance are clarified. Electrochemical measurement and characterization reveal that elevating temperature causes a significant increase of the overpotential by liquid water depletion and a rather small increase of ohmic overpotential. In addition to increasing water flow rate as usually treated, raising operating pressure is found to suppress the overpotential by water depletion. This finding suggests that coexistence of vapor and liquid phases in PEMWE cell should be maintained by increasing operating pressure when operation temperature is elevated.

In chapter 4, structural properties of both anode and cathode current collectors (ACC and CCC) are examined in terms of their impact on water transport and overpotentials. Hydrophilicity (contact angle) and thickness of the ACC change the electrolysis voltage mainly through ohmic overvoltage and overpotential by liquid water depletion. Smaller contact angle and thinner ACC in the range of 0-120° and 200-300μm, respectively, enhance water supply to anode catalyst layer and lower the both overpotentials. An optimized pore diameter in ACC is suggested to be 21μm, which can minimize the electrolysis voltage above 100 °C of operation temperature. With the adequate choice of CCC, an optimized pair of ACC and CCC is indicated to be titanium mesh (21μm in pore diameter, 200μm in thickness, zero contact angle) and carbon paper (15μm in pore diameter, 300μm in thickness, 110° contact angle), respectively, and minimize the electrolysis voltage.

Chapter 5 clarifies the impact of flow configurations and flow field pattern on the electrolysis voltage. Anode flow field pattern changes the overpotential by liquid water depletion. Among cascade, parallel, and serpentine pattern, the cascade pattern minimizes the overpotential. Lowest flow velocity appeared in the cascade pattern contributes to decrease the capillary pressure in current collector, leading to the enhancement of water supply through the current collector and less overpotential. Cathode flow field pattern impacts only on the ohmic overpotential slightly. Combination of cascade and serpentine flow field for anode and cathode, respectively, minimizes overpotentials, leading to highest performance. The flow configuration has little impact on the electrolysis performance. Different from PEM fuel cell, water supply in PEMWE distributes water in homogeneous manner, and this nature trivialize the effect of flow configuration.

Chapter 6 summaries all chapters, and indicates possible future works.