Characterization of the overpotential in electrochemical hydrogen compressor with the internal humidifier and anode dead end channel

郝, 彦明

https://doi.org/10.15017/1807028

出版情報：九州大学, 2016, 博士（工学）, 課程博士
バージョン：

（様式２）

氏　　名　: 郝　　　彦明（Hao　Yanming）

論文題名　: Characterization of the overpotential in electrochemical hydrogen compressor with the internal humidifier and anode dead end channel
（内部加湿とアノードデットエンドを組み込んだ電気化学式水素圧縮機の過電圧解析）

区　　分　: 甲

論　文　内　容　の　要　旨

Hydrogen compressor is one of the important devices for the effective development of hydrogen energy society. Electrochemical hydrogen compressor (EHC) similar to Polymer Electrolyte Membrane Fuel Cell (PEMFC) is a device, which can compress hydrogen gas through the electrochemical process. The advantages of EHC, such as less power consumption and lower noise, motivate to replace the traditional mechanical hydrogen compressor (MHC) with EHC. However, dehydration of polymer electrolyte membrane (PEM) embedded in EHC is a serious issue. Since different from PEMFC no water is produced during the reaction in EHC, humidifying the membrane is the priority task for the development of EHC.

In this study, a new humidification method – internal humidification method is proposed. The method improves the hydration state of PEM and thus lowers membrane resistance, leading to higher efficiency of EHC. Also, EHC in this study employs an anode dead end channel to reduce the system cost. The overall efficiency of EHC achieves approximately 50%, which is competitive with that in traditional MHC.

In chapter 1, the research work on EHC is reviewed and summarized. Conventional EHCs utilize external humidifier – bubbler humidifier, embedded at the inlet of EHC, to hydrate the membrane. However, especially under high operation temperature, EHCs suffer from significant water evaporation from the cathode side, resulting in high ionic resistance through PEM. To overcome these water management issues, an internal humidifier is proposed. Also, an anode dead end channel is employed for lowering system cost. Characterization on the proposed EHC, as well as quantitative comparison with the conventional EHCs, is the goal of this study.

In chapter 2, the proposed EHC and the experimental system are explained in detail. The internal humidifier is realized by storing liquid water in a compartment built in the cathode end plate. The outlet of the anode channel is closed, and thus EHC operates with an anode dead end channel.

Evaluation for EHC concentrates both on the voltage and the current efficiency. Measurement apparatus related to the efficiency, such as overpotential and hydrogen back diffusion, is explained. Since the separation of the overpotential originated from each side (the anode and the cathode) should be carefully made, a reference electrode method with intentionally thick PEM and low Pt loading is introduced as a unique method in this study.

In chapter 3, the proposed EHC is characterized. Ohmic overpotential dominates the cell voltage. The ohmic overpotential is originated from ionic resistance through PEM, and is significantly dependent on the water content in PEM. A mathematical model to evaluate the water transport through PEM is established. Analysis based on this model clarify that, under low current density and high temperature condition, the balance of water transport between electro-osmotic drag and diffusion holds, and PEM is kept hydrated. Under the opposite condition with high current density and low temperature, the balance does not hold and PEM is dehydrated.

Comparison clarifies the advantage of the internal humidifier embedded in the proposed EHC. Different from bubbler humidifier employed in conventional EHCs, the internal humidifier can directly supply liquid water, and can sufficiently wet PEM. The liquid water mitigates water transport resistance near the surface of PEM, leading to the high water content and low ionic resistance in PEM. Also, the internal humidifier can compensate water evaporation especially at high temperature.

The anode dead end channel chosen for reducing the cost is examined and clarified to cause no technical problem. Additional concentration overpotential was predicted to be caused by the contamination of liquid water due to the lack of convection effect with dead end channel, but it did not appear even in a long operation of EHC. Moreover, the dead end channel is found to contribute to the well hydrated membrane. Saturated condition of water/vapor mixture seems to be realized at the anode side, resulting in the suppression of the water evaporation there.

A unique characteristic is found in the efficiency of EHC. Voltage efficiency increases with temperature and compression ratio, but decreases with increasing applied current. Against the voltage efficiency, current efficiency shows the opposite dependency on those parameters. Thus, careful examination to determine operation conditions is required for the design and optimization of EHC.

In chapter 4, the non-ohmic overpotential originated from the cathode and the anode is separately analyzed. Hydrogen evolution reaction (HER), progressed in the cathode, shows charge transfer dependence. Hydrogen oxidation reaction (HOR) in the anode does mass transfer dependence. Volmer-Heyrovksy-Tafel mechanism is introduced to explain these features. Analysis based on the mechanisms clarifies that HER obeys the Volmer-Heyrovsky route, and HER does the Volmer-Tafel route. The analysis also elucidates that the reaction rate of HER is smaller than that of HOR, resulting in larger overpotential at the cathode. Furthermore, the analysis reveals that elevating cathode pressure enlarges the coverage of the reaction intermediate (adsorbed hydrogen) and accelerates HER.

Chapter 5 summaries all chapters, and indicates possible future work.