§6. Electrochemical Hydrogen Pump Using a High Temperature Type Proton Conductor under Reduced Pressure

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In a future nuclear fusion plant, hydrogen isotope gas of deuterium (D) and tritium (T) is used as fuel. The developments of hydrogen process technologies for fuel cycle are one of the important issues. An electrochemical hydrogen pump using a proton-conducting oxide is one of the candidate materials for hydrogen recovery. It has attractive advantages such as: hydrogen extraction from both hydrogen molecules and hydrogen compounds; control by electric current; no pressurization, etc. Thus, the application of electrochemical hydrogen pump by a proton conducting oxide instead of the palladium membrane diffuser has been proposed. In our previous research, we have chosen $CaZr_{0.9}In_{0.1}O_{3-\alpha}$, which was used for the one-end closed tube of the test pump, as the proton-conducting oxide. Then, the hydrogen pump performances under various conditions have been reported and shown the possibility of realization. On the other hands, in order to recovery pure hydrogen isotope gases for fuel cycle, hydrogen pump into a vacuum would be required to avoid from being mixed in impurity gaseous as purge gas. However, the hydrogen pump characteristics into vacuum are not well-known. Kato et al. have proposed a small tritium purification and recovery system for muon-catalyzed fusion (μCF) and have evaluated hydrogen pump performance using a high temperature type proton conductor under a high vacuum condition.[1)] As the results, although they showed that hydrogen could be extracted into vacuum, the dependence of total pressure in the cathode compartment has not been studied in detail. In this research, we report the preliminary results of the hydrogen pump characteristics under reduced pressure condition in the pressure range from 10 Pa to 10^5 Pa.

From the viewpoint of the practical use, we carried out the performance tests by use of the one end closed tube made of $CaZr_{0.9}In_{0.1}O_{3-\alpha}$, which was prepared by TYK Co. Ltd. The shape of the test tube was 12 mm inner diameter, 0.75 mm thickness and 340 mm length. Platinum electrodes were attached on both sides of the test tube by paste-baking on the inner surface. The electrode on the outer surface was attached by electroless-plating method. The each electrode of inner and outer surfaces was used as anode and cathode, respectively. The test tube was heated from 873 K to 1073 K by an electric furnace. The gas was humidified by using a bubbler immersed in a constant-temperature water bath controlled at 283K. Wet argon gas was fed to the anode side at 100 cm^3/min and the flow rate was controlled by a mass flow controller. An impedance analysis measurement was conducted by use of two LCR meters with different frequency range (HIOKI, 3522-50 and 3532-80). These frequency ranges are 1 mHz to 100 kHz and 4 Hz to 1 MHz, respectively. The pressure in the cathode was measured by a capacitance vacuum gauge.

Figure 1 shows comparison of the Nyquist plots for different total pressure in the cathode compartment. The left semicircle seems not to be almost affected the total pressure variations in the cathode compartment, while the shape of right semicircle is much changed by the variation of total pressure. The left semicircle would represent the charge transfer control in the electrolyte-electrode interface and the left endpoint of semicircle corresponds to electrolyte resistance. Therefore, the left semicircle would be almost independent of the total pressure. On the other hand, the right semicircle would represent the mass transfer control with diffusion process. The semicircle of diffusion process should become smaller with decreasing total pressure until few kPa, because the pressure dependence of diffusivity of gas is correlated to P^{-1} according to the Chapman-Enskog theory. However, the semicircle became larger at the total pressure below 1 kPa. Although the mechanism of the increase in the impedance spectra less than 1 kPa could not be determined in this report, it might be suggested that the hydrogen pump performance could be improved under the reduced pressure condition of around few kPa and the reduced pressure condition has an effect on the mass transfer process in the electrode.

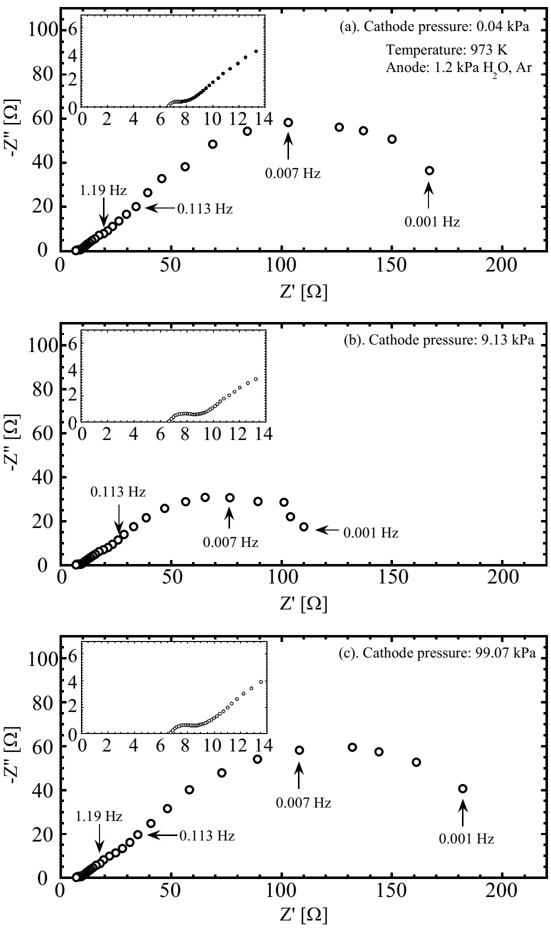

Fig. 1. Comparison of the Nyquist plots for different total pressure in the cathode compartment: (a). 0.04 kPa, (b). 9.13 kPa, (c). 99.07 kPa

1) Kato, M. et al.: Fusion Sci. Technol. **41** (2002) 859