§10. Extraction Characteristics of Hydrogen into Vacuum from a Proton Conducting Oxide

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In a future nuclear fusion plant, hydrogen isotope recovery for fuel cycle is one of the important issues. The candidate material of the hydrogen isotope recovery is a palladium membrane diffuser for chemical purification of hydrogen. To provide a driving force for hydrogen to permeate through the membrane, the pressure difference is necessary. It is also possible to recovery hydrogen isotope gases from water vapor and hydrocarbons using the cracking catalyst bed.

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, we have proposed to apply a proton conducting oxide instead of the palladium membrane diffuser. In the previous research, $CaZr_{0.9}In_{0.1}O_{3-\alpha}$, which was used for the one-end closed tube of the test pump, was chosen as the proton-conducting ceramic. $CaZr_{0.9}In_{0.1}O_{3-\alpha}$ is excellent in the chemical stability against carbon dioxide at elevated temperature and in mechanical strength. The hydrogen pump performances by the water vapor electrolysis at atmospheric pressure have been reported.[1] On the other hands, in order to recovery pure hydrogen isotope gases and remove impurity gaseous for fuel cycle, hydrogen pump into a vacuum would be required. However, hydrogen extraction characteristics into vacuum are not well-known. In this research, hydrogen extraction characteristics into vacuum atmosphere by water vapor electrolysis were investigated as primary experiments.

We carried out the performance tests of the one-end-closed tube made of $CaZr_{0.9}In_{0.1}O_{3-\alpha}$ with wide electrode area. The shape of the test tube was 12 mm inner diameter, 0.5 mm thickness and 340 mm length. The platinum electrode was attached on both sides of the test tube and the effective area on the cathode electrode was 62 cm^2. In this experiment, wet argon gas, which contained water vapor of 1.2 kPa, was fed to the anode at 300 cm^3/min. The pressure in the cathode could be reduced until around 10 Pa by a scroll pump and then up to 10^5 Pa by supplying argon gas. The test tube was heated up to 973 K by an electric furnace. Then, the constant current was passed through between the electrodes by a galvanostat. Complex impedance measurement was conducted with a LCR meter in a frequency range of 4 Hz to 1 MHz at 0.1 V. The gas components in the anode were measured by an oxygen analyzer.

When the electrochemical hydrogen pump into vacuum was conducted at 0.35 A and 973K, the pressure dependence was exhibited as shown in Fig.1. Therefore, voltage showed a slight decrease with reducing pressure until around 100 Pa. Furthermore, oxygen concentration in the anode component corresponding to the rate of water vapor electrolysis indicated a slight increase. However, when the pressure in the cathode became less than 100 Pa, voltage changed a slight increase and oxygen concentration was decreased slightly. It was suggested that the hydrogen pump performance was improved under the vacuum condition of around 100 Pa. Figure 2 shows the Nyquist plot of the complex impedance measurement under the same condition of Fig.1. Impedance at 4 Hz decreased with lower pressure. In general, the imaginary component to the impedance comes from capacitance and/or inductance. In this experimental condition, only capacitance could come from an electrochemical polarization affected by electrode reaction. If the pressure in the cathode compartment decreases, gas permeation through the electrode could be improved. It supposed that the hydrogen gas flow rate in the cathode electrode might have an effect on electrode polarization.

1) Tanaka, M. et al.: J. Nucl. Sci. Technol. 41 (2004) 61.

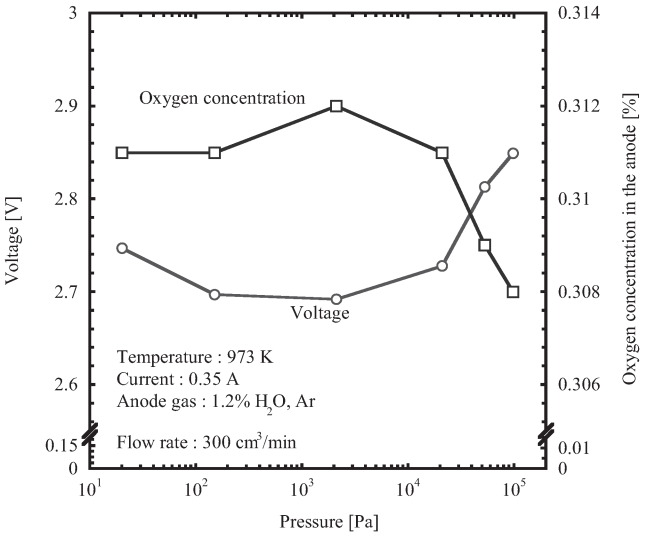

Fig. 1: Pressure dependence of voltage and oxygen concentration in the anode component at 0.35, 973K

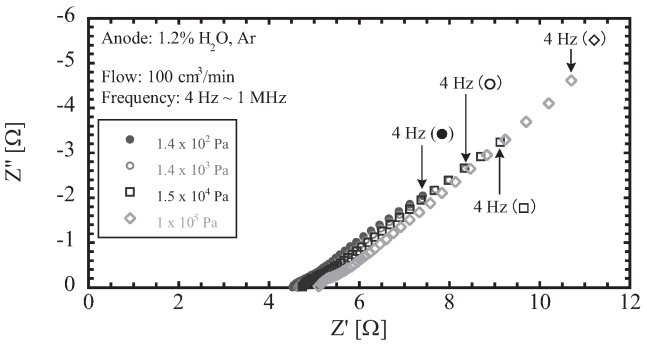

Fig. 2: Pressure dependence of complex impedance measurement in a frequency range of 4 Hz to 1 MHz at 973K